SKBF KBS
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80-14

Properties and long-term behaviour of bitumen and radioactive waste-bitumen mixtures

Hubert Eschrich

Eurochemic, Mol, October 1980

SVENSK KÄRNBRÄNSLEFÖRSÖRJNING AB / PROJEKT KÄRNBRÄNSLESÄKERHET

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ABSTRACT

This report consists of two main parts.

Part I represents a survey of the properties and the long-term behaviour of pure bitumens and mixtures of bitumens with radioactive reactor and reprocessing wastes.

This survey includes information on the origin, amounts, and composition of the various wastes considered for bituminization and the different waste bituminization techniques used. The influence of various factors on the quality of waste-bitumen products and on the radiological safety during transport, short- and long-term storage of the final products is described.

Special consideration is given to the most important safety relevant factors associated to the use of bitumen as matrix material for radioactive wastes, such as leach-resistance, radiolysis, chemical and mechanical stability, combustibility, and microbial attack.

Part II consists of a comprehensive bibliography on the bituminization of radioactive wastes, giving about 300 references to literature published from the beginning of the use of bitumen in radioactive waste management in 1960 until the beginning of 1979.

The bibliography serves at the same time as source for the literature referred to in the text of Part I of this report.

Methods for the quality control of bituminous materials and some useful data are given in an annex.

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PART I

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PART 1

PROPERTIES AND LONG-TERM BEHAVIOUR OF BITUMEN AND RADIOACTIVE WASTE-BITUMEN MIXTURES

SUMMARY AND CONCLUSIONS

This report deals with the properties and long-term behaviour of bitumens of importance for their use in the conditioning of radioactive wastes and reviews the present knowledge on the properties of waste-bitumen mixtures obtained by the bituminization of reactor and fuel reprocessing wastes.

Based on that knowledge and furthermore on the short-term behaviour of the various bitumen-waste products (BWPs), the results of accelerated testing and the evaluation of relevant experiments, it has been tried to predict and to estimate the long-term behaviour of the BWPs during their final storage period.

To aid the follow-up of the fate of the respective wastes during their entire life-time and their safe handling in the different waste management phases, some representative information is given on the origin, amounts, and compositions of the wastes, and furthermore on bituminization techniques and installations, and interim and final storage facilities for low- and intermediate-level radioactive wastes.

Technical and safety aspects related to the handling, transport, interim and final storage are outlined.

The presently considered scheme in Sweden for the management of reactor wastes (at least for the intermediate-level category) comprises the following steps:

- bituminization or cementation at the reactor site,
- interim storage at the reactor site in engineered facilities,
- transportation of the waste packages in containers from the interim storage facility to the final repository by a special ship,
- final storage of the conditioned waste in a central bedrock repository for low- and intermediate-level radioactive wastes (called ALMA).

Bituminized reactor wastes derive from the power stations situated at Barsebäck and Forsmark, and bituminized reprocessing wastes would originate from the reprocessing of Swedish nuclear fuels in France.

The successful and safe bituminization of a wide variety of low-level wastes (LLW) and intermediate-level wastes (ILW) generated at reactor stations and reprocessing plants has been demonstrated since more than 10 years on an industrial scale using various pre-treatment processes, incorporation techniques and conditions.

Up to now, several thousands of 200 l drums filled with bituminized wastes of up to about 1 Ci/l have been produced in industrial plants operating in Belgium, France, the Federal Republic of Germany, Finland and Sweden.

The successful application of bitumen as fixation material for certain types of radioactive wastes is due to its favourable chemical and physical properties which are described in this report.

However, some unfavourable properties of bitumen, of which the burnability and the radiation degradation appear to be the most important ones, might motivate further development efforts on the improvement of the final product characteristics as well as the search for or the use of other matrix materials.

An assessment of these properties with respect to the safe production, transport and storage of the various bitumen-waste products (BWPs) has been made.

It has been concluded that the incorporation of adequately pre-treated reactor and reprocessing wastes can be carried out without risking a fire, an explosion or any other dangerous exothermic reaction if the bitumen/waste solids ratio (about 1:1 per weight) and the temperature of the mixture (< 200°C) are controlled and the accumulation of inflammable or explosive gas mixtures is prevented by applying a sufficiently powerful ventilation (especially above the container receiving the hot BWP).

Interim storage facilities and sufficient technical concepts are available showing that a safe interim storage of bituminized low- and intermediate-level radioactive wastes can be accomplished taking into consideration the properties of the stored product, local conditions, activity inventory and storage time. Most important for a safe interim storage is the absence of an ignition source and of a radiolytically generated explosive gas mixture.

The transport of BWPs under normal conditions can be carried out safely by using properly designed individual concrete casks or large freight containers (for the accommodation of a larger number of product drums) ensuring also adequate radiation shielding and protection against impact, temperature and fire; the low leachability of the products provides the required protection against attack of water. From a safety assessment devoted to the sea transportation of BWPs it has been concluded that in view of the low probabilities for accidents causing release of radioactive material and the limited consequences, the foreseen transport system meets very high safety standards.

In Sweden, the final storage of BWPs is foreseen in a bedrock repository (ALMA) at intermediate depth which will comprise storage space within a concrete structure, interlayers of concrete, and a backfilling material consisting of a moraine (sand)/ bentonite mixture which shall function as a mechanical buffer, a seal against ground water flow and a sorption barrier for radionuclides released from the stored waste products.

The detailed structural design features and the depth of the repository will depend on the results of quantitative risk estimates for which the properties and the long-term behaviour of the BWPs are of decisive importance.

In comparison to other conditioning methods bituminization is preferred because it produces in a reliable way durable, homogeneous products of low leachability, and of low volume and weight. The products are well suited for a safe final storage that can be realizable, within limits, independent of local geologic and hydrologic conditions.

The high weight- and volume-percentage of the solid waste components in the final product is highly economical as it reduces the costs for the embedding material (bitumen), transport, handling and storage.

The versatility of accommodating virtually all types of LLW and ILW arising at reactor stations and reprocessing plants and the demonstrated natural stability over millions of years in an underground environment can be regarded as real advantages of bitumens over most man-made materials.

Bitumen is an organic material which consists of mixtures of mainly aliphatic and aromatic hydrocarbons - part of which have combined with nitrogen, sulfur and oxygen - of high molecular weight.

Bitumen is obtained either from distillation of petroleum or from natural deposits. In general terms, bitumens are: thermoplastic substances, readily adhesive, semi-solid to solid at room temperature, easily liquefied upon heating, highly waterproof, durable, resistant towards most acids, alkalis, and salts, not poisonous, highly resistant towards ageing and climatological influences, soluble in many organic solvents, and light (density of about 1.0).

These favourable physico-chemical properties and their longterm durability form the basis for their application as embedding material for nuclear wastes.

Based on the results of experiments and evaluations given or referred to in the various chapters of this report the following conclusions on the properties and long-term behaviour of properly prepared bitumen-waste products (BWPs), - composed of about 50 wt.-% low-level or intermediate-level reactor or reprocessing wastes and about 50 wt.-% of a suitable type of bitumen, - can be drawn:

- The steady state leach rates of "insoluble" or insolubilized radionuclides are normally between 10^{-5} to 10^{-7} g . cm^{-2} . d^{-1} the leach rates of soluble waste constituents (e.g. sodium) are in the order of 10^{-4} g . cm^{-2} . d^{-1}. All these leach rates are extremely favourable for the safe long-term storage of BWPs.

- The radiation from the embedded radionuclides causes the generation of radiolysis products, heat, increased porosity and hardness, however, does not affect changements of the physico-chemical properties of the products which would significantly enhance the release rate of radionuclides from the stored products or endanger the stability of the geologic repository.

  The total radiation dose absorbed at infinite time by BWPs containing initially $\leq$ 1 Ci/β-γ-emitting nuclides per litre product will normally not exceed 2 x 10^8 rad. The amount of radiolysis gases is proportional to the applied dose; their main component is hydrogen which is generated at a rate of approximately 0.01 cm^3/Mrad . g bitumen.

  The specific rate of hydrogen generation is almost independent of the dose rate, type of bitumen, temperature, the composition of the incorporated salt mixtures, and the gas phase above the products.

  The nature and amounts of gaseous radiolysis products released from the BWPs have to be considered during the interim storage phase and the emplacement period of the final repository, however, they will not cause any untolerable physical or chemical attacks on the integrity of the stored BWPs or the geologic bedrock repository (as foreseen in Sweden) after it has been properly backfilled and sealed.

  - The heat output of the stored BWPs of a specific activity of < 1 Ci/l is low, even in the first years of storage; the specific power will be lower than 0.005 W/l, normally about 0.002 W/l or less.

    The heat generated in a BWP block of 200 l is insufficient to cause enhanced creeping, particle sedimentation, migration of radionuclides, dangerous chemical reactions or selfignition.

    The thermal effects from the embedded radionuclides and the prevailing temperature at the repository will neither endanger the long-term stability of the BWPs nor that of the repository.

  - Bitumen-waste products containing up to 60 wt.-% residues from reactor and reprocessing wastes do not explode or detonate during handling, transport and storage; they do also not burn if they are not heated above 300°C and/or an ignition source is absent.

    Although a content of nitrate facilitates and accelerates combustion, it is not sufficient to increase the rate of combustion significantly.

The burnability of radioactive BWPs becomes unimportant with respect to a safe storage or disposal as soon as the product packages are adequately buried or emplaced in a repository that is carefully backfilled.

- Sedimentation of finely divided salt or ion exchange particles within a homogeneous BWP is insignificant (< 5 mm/1000 years) if the storage temperature remains below 60°C or generally more than 20°C below the softening point of the BWP.
- Dimensional changements of the BWPs due to radiation degradation and/or water uptake can be reduced by an appropriate pretreatment of the waste, the application of a suitable type of bitumen and embedding conditions or the coating of the product block by a layer of pure bitumen.
- The mechanical properties of BWPs can be considered to be highly satisfactory under the conditions of the final geological storage.
- Though mixtures of radioactive wastes and bitumen are principally chemical instable systems, the chemical changements are slow and little destructive; they do not endanger the chemical long-term stability of the mixture in the absence of water (leaching). Bitumen shows a good compatibility with the embedded waste residues, also at elevated temperatures.
- Interactions of the stored solid BWPs with the surrounding solid or gaseous substances will not result in physical or chemical alterations which would worsen the ability of the repository to isolate the wastes.
- Though site- and product-specific calculations and studies need to be carried out, one may predict that the rates, amounts and nature of the gases formed from the degradation of the stored waste packages will neither endanger the integrity of a bedrock repository nor enhance the release of radionuclides from the BWPs to any significant extent.
- The possibility that microorganisms destroy the bitumen matrix of BWPs, is extremely small and, if at all, the speed of attack will most probably be very slow.
- There exist no long-term risks associated to nuclear criticality, helium generation by alpha decay, and the sudden release of stored energy in the geologic storage of waste products considered here.

It appears that properly prepared and composed radioactive waste-bitumen mixtures form waste products possessing adequate properties and long-term stability which ensure a satisfactory containment of radionuclides with a sufficient degree of reliability for the requisite storage period.

It is hoped that the data given on the properties of BWPs and the assessments made of the long-term behaviour of bitumen-waste products will aid in the design and construction of a final repository in bedrock that will preclude contact with the biosphere of radionuclides in concentrations which are sufficient to cause deleterious impact on people or their environment.

1. INTRODUCTION

The research and development work on the bituminization of radioactive wastes commenced in 1960 (in Mol, Belgium) and led relatively fast to the realization of a mature process on an industrial scale, namely already in 1967 (in Marcoule, France).

The choice of solidifying or conditioning low- and intermediate-level radioactive wastes by bituminization is guided by the aims to reduce as much as possible the risk of exposing the operation personnel to radiation, to contain the radionuclides within a durable product of low leachability, volume and weight suited for a safe final (geological) storage, and to minimize transport and storage costs.

Up to now, several thousands of 200 l drums filled with bituminized wastes of up to about 1 Ci/l have been produced in the industrial plants operating in Belgium, France, the Federal Republic of Germany, Finland and Sweden. The successful application of bitumen as fixation material for certain types of radwastes is due to its favourable chemical and physical properties which are described in some detail in the next chapter.

Further development work on the improvement of the final product characteristics as well as the search for or the use of other matrix materials are obviously due to the unfavourable properties of bitumen, of which the burnability and the radiation degradation are the most important ones. An assessment of these properties with respect to the safe production and storage of the various bitumen-waste products (BWPs) is also contained in this report.

The technique applied for the final storage (disposal) of solidified radioactive wastes should provide both isolation and containment; this can be realized by providing a series of barriers against the migration of the radionuclides along all the pathways to man. One of these barriers is the form of the waste.

Conversion of all wastes to stable leach-resistant forms will ensure that the release of the radionuclides from the waste will be slow even though other barriers should fail and flowing water should contact it. One of the waste forms, gaining more and more importance in this respect, is the subject of the present review: "bituminized waste".

1.1 Background

The Swedish State Radiation Protection Institute has indicated in its proposal for the guideliness of the radioactive waste management (SSI:1972-019) that the nuclear power plants shall take care of the (on-site) storage of the radioactive waste, generated in connection with the reactor operation, for at least five years. This proposal has been accepted and followed-up by the utilities owning nuclear power plants. Subsequently several proposals were made by the competent institutions on a coordinated management of reactor wastes. One of the proposals concerns the construction of a central facility for the final storage of the conditioned low- and intermediate-level wastes originating from the operation of the various Swedish reactor stations.

Within the framework of the respective R + D-programme, studies have already been carried out on the realization of this "Central Facility for Low- and Intermediate-Level Waste", called "ALMA" (Swedish acronym for "Anläggning för Låg- och Medel-Aktivt Avfall"). A summary of the results of a preliminary study has been published in July 1978 /R-1/, and proposals on the construction of the facility will be published in 1980 /R-28/.

ALMA will be constructed to cope with the final storage of different low- and intermediate-level waste products, originating primarily from reactor operation. Depending on the type of waste and the reactor station the bulk of the waste will be incorporated either in concrete or bitumen. The bituminized reactor wastes derive from the power stations situated at Barsebäck and Forsmark.

In the ALMA project also the storage of final low- and intermediate-level waste products from fuel reprocessing has to be taken into account. In the reprocessing contract between SKBF and COGEMA (France) information about the waste types and amounts are given. However, detailed specifications about the final form of the waste to be received in Sweden after 1990 are presently still unknown. It can be assumed, though, that a large fraction of the waste from reprocessing will be incorporated into bitumen.

It is obvious that for any kind of waste storage one has primarily to consider the properties of the waste material to be stored and the behaviour during the envisaged or required storage period, particularly the possible negative changements with time at the chosen storage place and environment.

It is understandable that the variety of waste products, the wide range of their physical and chemical properties, and their possible interaction with the environmental media after disposal make it difficult to confidently predict their long-term behaviour and their hazard to man. It is the main purpose of this report to review what is already known on the properties and long-term behaviour of bitumen and bituminized low- and intermediate-level wastes from reactors and reprocessing plants to aid in the design and the realization of a safe final storage (disposal) in Sweden (i.e. ALMA) and elsewhere. This review - which is based on publicly available literature and own research and development work - may also serve to identify factors deserving further investigations.

1.2 Scope

This report deals with the properties of bitumens of importance for their use in the field of radioactive waste management and gives a status of the present knowledge on the properties of bitumen- waste mixtures obtained by the bituminization of reactor and fuel reprocessing wastes. Based on the properties of these bitumen- waste products (BWPs) their long-term behaviour during storage in the envisaged environment is assessed.

The time span of concern for the storage of the conditioned reactor wastes is in the order of 500 years; after this time the most active bituminized material (ion-exchange resins from the water purification of the primary circuit and the fuel storage pond) will have reached a specific activity of 2 nCi/g which is considered as the limit for which a permission for possessing radioactive material is required /R-1/. The products from the bituminization of reprocessing wastes may contain larger amounts of long-lived radionuclides (actinides) necessitating to consider this factor in the assessment of the long-term stability and behaviour of these BWPs.

Main emphasis is put on the storage-relevant properties of pure bitumens and the various radioactive bitumen-waste mixtures. To enable a better follow-up of the fate of the respective wastes during their entire "life", some information is given on the origin, amounts, and composition of the wastes and furthermore on bituminization techniques, installations, and interim storage facilities in use. Finally technical and safety aspects related to the handling, transport, interim and final storage are outlined.

The bibliography and the annexes shall permit the interested reader to find quickly detailed information in the original literature or some useful indications and data for his work.

The entire bituminization of reactor and reprocessing wastes forms thus the framework of the present review with its central point the properties and the long-term behaviour of the resulting radioactive waste-bitumen products in view of their final storage within a suited geological formation.

2. THE PROPERTIES OF BITUMENS

2.1 General

2.1.1 Definition of terms

In this report no clear distinction is made between bitumen, asphaltic bitumen, bituminous materials, tar, pitch, etc. The term "bitumen" is used almost exclusively in this report. One should be aware that the term "bitumen" - which is more used in Europe than in the USA - is broader than "asphalt".

The word bitumen (lat.) is derived from "pix tumens" which means "tombs' pitch" as the Egyptians used bitumens from the Black Sea for the embalming of corpses. It is also assumed that the word bitumen comes from the original Sanskrit word Gwitu-men, meaning pitch. The Greek word for native ashphalt was asphaltos.

"Bitumen" is defined by the A.S.T.M as mixtures of hydrocarbons of natural or pyrogenous origin, or combinations of both, frequently accompanied by their non-metallic derivatives, which may be gaseous, liquid, semisolid, or solid, and which are completely soluble in carbon disulfide.

The definitions which follow show the relationship between the various groups of "bituminous substances" /R-2/:

"Bituminous substances" are a class of native and pyrogenous substances containing bitumens or pyrobitumens, or ressembling them in their physical properties. This definition includes bitumens, pyrobitumens, pyrogenous distillates (pyrogenous waxes and tars) and pyrogenous residues such as pitches and pyrogenous asphalts.

"Pyrobitumen" is a generic term applied to native substances of dark colour, comparatively hard and non-volatile, composed of hydrocarbons, which may or may not contain oxygenated bodies; sometimes associated with mineral matter, the non-mineral constituents being infusible, and relatively insoluble in carbon disulfide. (This definition includes the asphaltic and non-asphaltic pyrobitumens and their respective shales).

"Mineral Wax" is a term applied to a species of bitumen, also to certain pyrogenous substances, of variable colour, viscous to solid consistency; having a characteristic luster and unctuous feel; comparatively non-volatile; composed of hydrocarbons, substantially free from oxygenated bodies; containing considerable crystallizable paraffins, sometimes associated with mineral matter, the non-mineral constituents beeing easily fusible and soluble in carbon disulfide. (This definition is applied to crude and refined native mineral waxes, also to pyrogenous waxes).

"Asphalt" is a term applied to a species of bitumen, also to certain pyrogenous substances of dark colour, variable hardness, comparatively non-volatile, composed of hydrocarbons, substantially free from oxygenated bodies; containing relatively little to no crystallizable paraffins; sometimes associated with mineral matter, the non-mineral constituents being fusible, and largely soluble in carbon disulfide; the distillate, fractionated between 300 and 350°C, yields considerable sulfonation residue. (This definition is applied to native asphalts and pyrogenous asphalts).

"Asphaltite" is a species of bitumen, including dark coloured, comparatively hard and non-volatile solids; composed of hydrocarbons, substantially free from oxygenated bodies and crystallizable paraffins; sometimes associated with mineral matter, the non-mineral constituents beeing difficulty fusible and largely soluble in carbon disulfide; the distillation residue, fractionated between 300 and 350°C, yields considerable sulfonation residue. (This definition includes gilsonite, glance pitch, and grahamite).

"Asphaltic Pyrobitumens" are species of pyrobitumen, including dark coloured, comparatively hard and non-volatile solids; composed of hydrocarbons, substantially free from oxygenated bodies; sometime associated with mineral matter, the non-mineral constituents being infusible and largely insoluble in carbon disulfide. /This definition includes elaterite, wurtzilite (depolymerizes on heating, becoming fusible and soluble) albertite, impsonite, and the asphaltic pyrobituminous shales/.

Some distinguishing characteristics of substances mentioned above are given in Table I (based on R-2, p.168).

2.1.2 General composition and properties

Bitumen is an organic material which consists of mixtures of mainly aliphatic and aromatic hydrocarbons (- part of which have combined with nitrogen, sulfur and oxygen-) of high molecular weight. The constituents appear to be enormously variable, complex and subject to rapid change under different methods of treatment, although possessing rather stable physical properties except when heated. No method for accurately analyzing asphalt has yet been developed. Methods of analysis have usually consisted of determining relative molecular weights, general nature of compounds and elemental composition rather than specific chemical structure.

Bitumen is obtained either from the distillation of petroleum or from natural deposits. Bitumens from the two sources resemble each other in all respect except that natural bitumen - believed to be an early stage in the breakdown of organic marine deposits into petroleum - often contains minerals while petroleum bitumen does not. Most crude oils contain some bitumen; the amount may reach up to 50%. Bitumens derived by refining petroleum generally have more paraffinic side-chains than natural bitumens.

TABLE I

BITUMINOUS SUBSTANCES AND SOME OF THEIR CHARACTERISTICS

Substance	Species	Member	Specific density at 25 °C	Penetr. at 25 °C	Fusibility °C	Fixed carbon	Solubility in CS_2	Org. matter insol. CS_2	Mineral matter	Solub. in naphtha 88 °	O_2 in org. matter
Bitumens	Petroleums										
	Native mineral waxes	Ozokerite	0.85-1.00	5-10	60-95	0.5-10	95-100	0-1	0-5	75-95	0-2
		Montan wax	0.90-1.00	5	75-95	2-10	98-100	0-2	0-2	80-100	3-6
	Native asphalts	Cont'g < 10 % mineral matter	0.95-1.12	0-350	15-165	1-25	60-98	0-40	0-10	25-95	0-2
		Cont'g > 10 % mineral matter	0.95-1.15	0-150	15-175	5-25	Tr.-90	0-25	10-95	Tr.-85	0-2
	Asphaltites	Gilsonite	1.05-1.10	3	120-175	10-20	90-100	0-1	Tr.-1	40-60	0-2
		Glance pitch	1.10-1.15	5	120-175	20-30	95-100	0-1	Tr.-5	20-50	0-2
		Grahamite	1.15-1.20	0	175-320	30-55	45-100	0-5	Tr.-50	Tr.-50	0-2
Pyrobitumens	Asphaltic pyrobitumens	Elaterite	0.90-1.05	Rubbery	Inf.	2-5	10-20	70-90	Tr.-10	5-10	1-5
		Wurtzilite	1.05-1.07	5	Inf.	5-25	5-10	80-95	Tr.-10	Tr.-2	0-2
		Albertite	1.07-1.10	0	Inf.	25-50	2-10	85-98	Tr.-10	Tr.-2	0-3
		Impsonite	1.10-1.25	0	Inf.	50-85	1-6	90-99	Tr.-10	Tr.-2	0-3
		Asphaltic pyrobituminous shales	1.50-1.75	0	Inf.	2-25	Tr.-3	15-70	30-85	0-Tr.	0-3
	Non-asphaltic pyrobitumens										
Pyrogenous distillates	Pyrogenous waxes										
	Tars										
Pyrogenous residues	Pyrogenous asphalts	Residual oils	0.85-1.05	100-350	-20-25	2-10	98-100	0-0.5	0-0.5	80-99	0-3
		Blown petroleum asphalts	0.90-1.07	25-200	25-200	5-20	95-100	0-5	0-0.5	50-90	2-5
		Residual asphalts	1.00-1.17	0-150	25-110	5-40	85-100	0-15	0-1	25-85	0-2.5
		Sludge asphalts	1.05-1.20	0-150	25-110	5-30	95-100	0-5	0-1	60-95	3-7
		Wurtzilite asphalt	1.04-1.07	5-10	65-150	5-25	98-100	0-0.5	Tr.-2	50-80	0-2
	Pitches										

Bitumens are complex colloidal systems of hydrocarbons, in which it is difficult to draw a sharp distinction between the continous and the disperse phases. They can be considered as dispersions of micelles in oily constituents (the maltenes) similar to heavy lubricating oils, the micelles being peptized to different degrees. The dispersed particles are called asphaltenes. The asphaltene content of bitumen is determined as the portion insoluble in petroleum naphtha.

Asphaltenes consist of high-molecular hydrocarbons (approximately 10^3 to 10^5), probably predominantly aromatic in character and having carbon/hydrogen (C/H) ratios of about 0.8 to 0.9. When heated they do not soften, but decompose, swell, and sinter. They are practically insoluble in petroleum fractions, but they absorb similar resinous petroleum fractions of lower molecular weight, which are present in asphalt, and which act as the dispersion agent to peptize the asphaltenes. It is difficult to make a clear definition of resins since no standardized method of separation exists, but they are often separated by absorption on clay from the naphta solution after precipitation of the asphaltenes.

There are variable amounts of sulfur, nitrogen, oxygen and entrained salts (from salty oil-well water) in bitumens. These constituents in general do not have an important effect on the uses of bitumen. Bitumen from certain waxy crudes contains wax in varying amounts. Since petroleum bitumen is obtained as the residue on distilling crude petroleum, its composition is naturally dependent on the extent to which the distillation has been carried. The further distillation is carried the lower the penetration. "Distillation residue bitumen" is often also called "steam-refined bitumen".

Oxidized asphalts contain a higher proportion of asphaltene micelles, and have typically plastic properties, as compared with the generally viscous properties of straight-run bitumens. The oxidation process effects a molecular structure, which gives plastic and elastic properties to the material that are valuable where liquid flow is undesirable.

On the other hand, oxidation reduces durability under some conditions and lowers the ductility considerably.

The softer the asphalt the longer the oxidizing or air-blowing time required to reach a given penetration, and the more plastic the oxidized bitumen.

In general terms, bitumens are:

- organic materials

- black or dark brown in colour

- thermo-plastic substances

- readily adhesive

- semi-solid to solid at room temperature

- easily liquefied upon heating

- highly waterproof
- durable
- resistant towards most acids, alkalis and salts
- not poisonous
- "inert" (i.e. do not react visibly with the substances they contact)
- highly resistant towards ageing and climatological influences
- soluble in many organic solvents (e.g. benzene, chloroform, carbon disulfide)
- light (density of 0.99 - 1.2)
- good electrical insulator.

2.1.3 Sources

The earliest known bitumens or asphalts occurred in springs in the east and to some extent in Europe. Later, deposits of solid, semi-solid and liquid bitumens usually mixed with minerals ranging from dust to sand, were found and mined. Lake asphalt from Trinidad was the first large commercial source but natural asphalt has declined in relative importance as petroleum became the major source. Large natural deposits of bituminous materials occur in many parts of the world (e.g. in Cuba, California, Venezuela, Canada, Syria) which are designated as bitumen, asphaltite, pitch, elaterite, albertite, gilsonite, impsonite, grahamite, and wurtzilite.

It is interesting to note that deposits of glance pitch in Utah have been found to contain an average of 1.75% uranium oxide (U_3O_8) and 4% vanadium pentoxide (V_2O_5) /R-5/.

Petroleum bitumens are obtained mainly as the residue on distillation of certain petroleum crude oils, and are produced in all consistencies from light road oils to heavy (high viscosity) industrial types. They represent over 90% of the total production of bitumens. The petroleum - bitumen production in the USA is about 9 000 000 ton per year, which is about 3% of the total production of crude oil. The bitumen content of crude oil varies with the source; some crudes contain practically no bitumen, while in others the bitumen content may be as high as 50%.

2.2 Types of Bitumen

Bitumens can be subdivided into the following types according to the mode of their preparation:

1. Straight-run distillation bitumens
   - obtained as residue after distillation of certain petroleum crude oils.
2. Oxidized bitumens
   - obtained by blowing air through molten bitumen at 150-260°C.
3. Cracked bitumens
   - obtained by pyrogenic breakdown of high molecular-weight compounds (cracked residues).
4. Cutback bitumens (asphalts)
   - obtained by liquefying bitumens by addition of a solvent (e.g. petroleum distillates).
5. Emulsified bitumens
   - obtained by the addition of anionic (alkaline soap), cationic (amine salt) or non-ionic aqueous solutions (emulsifiers).
6. Natural bitumens (asphalts, asphaltic pyrobitumens, rock asphalt, glance pitch)
   - consist of naturally occurring materials obtained by mining.

2.3 Application of Bitumens

2.3.1 History

Bitumen has been known since earliest times and used for thousands of years to cement various building materials (e.g. in Babylon), to caulk boats, for waterproofing, and for other purposes. Probably its earliest use was a water stop between brick walls of a reservoir in the beginning of the third millenium B.C. at Mohenjo-Daro, in India. In Genesis XI, 3, reference is made to the use of asphalt (bitumen) as mortar.

Early Buddhist traditions mention "earth-butter". In the middle east asphalt was extensively used for roads and water works, such as flood control. A king left an inscription saying that he had found his realm in mud and had left it laced with roads glistening with asphalt.

The recovery of objects from Pre-Biblical times in good condition is one of the reasons why bitumen for many years has been thought to be immune to microbial attack.

2.3.2 Conventional applications

Bitumens and bituminous materials are used for a great variety of purposes, e.g. for:

road surfacing, roofings, floorings, insulating varnishes, waterproofing, automobile undercoating, lining of water canals and reservoirs, coating of pipelines, acid-resistant paints, cold-molded products, printing inks, thermoplastics, thermal and electrical insulating material, river and sea revetments, beach erosion barriers, battery manufacture, soundproofing, and injection into sands and fissured rock formations to stop the flow of underground water.

Presently in the USA, about 75% of the bitumen, derived from crude oil, is used for paving, about 15% for roofing and the rest for purposes partially enumerated above.

2.3.3 Applications in nuclear waste management

It seems that the first thorough study on the use of asphalt in the field of nuclear waste management has been carried out in the year 1958 in the United States at the Oak Ridge National Laboratory /R-3/. From this study it was concluded that "asphaltic membranes appear practical for lining earth storage pits for aqueous radiochemical waste provided that the wastes are neutralized and are decayed sufficiently that the self-heating temperature does not exceed 150°F (65°C) and the time for the asphalt to acquire a dose of 10^9 rad is more than 25 years".

As from the year 1960 bitumen has increasingly been used as matrix material for the incorporation of low- and intermediate-level radioactive wastes /104/. The first investigations on the bituminization of radioactive wastes have been carried out at the Research Centre for Nuclear Energy at Mol, in Belgium /2/ and at the Plutonium Production Centre of Marcoule in France /3/.

In Mol a small installation has been built and operated from 1960 to 1964. As from 1964 until today an improved bituminization facility, called "Mummie", has allowed to treat all the radioactive concentrates and sludges producted in the Research Centre at Mol, and this without experiencing incidents or significant problems /189/. The second facility for the bituminization of radioactive wastes started operation in 1965 at Marcoule, France. Since then the bituminization of radioactive wastes has been utilized in many countries, primarily in Europe. Large-scale bituminization plants are operated, e.g. at the Nuclear Research Centre in Karlsruhe, (Federal Republic of Germany)/194/ and at the EUROCHEMIC Company in Mol (Belgium) /167, 193/.

Moreover, radioactive waste bituminization facilities of various treatment capacities are existing at nuclear research centres, power reactor stations, and other industrial nuclear energy institutions in several further countries, e.g. in the Sovjet Union, United States, United Kingdom, Sweden, Denmark, Austria, Switzerland, Finland, Poland, Japan, and Czechoslovakia.

2.4 Chemical Properties

There are many kinds of bitumens, differing widely in chemical composition. depending on sources and treatments. Bitumen may sometimes be mixed with up to 27 wt. -% of minerals /275/. After being heated to 100°C to expel water and gases the mineral content of certain bitumens may be as high as 38 wt. -%. All bitumens are insoluble in pure water and generally solid at temperatures below 70°C. Bitumens free of minerals, gases and water, consist mainly of carbon (80-88%). hydrogen (8-11%), ogygen (1-12%), sulfur (1-7%), and nitrogen (trace to 1,5%). These five elements occur in four major components:

- saturated hydrocarbons

- cyclic hydrocarbons

- resins and

- asphaltenes in various proportions.

The amount of asphaltenes in the bitumens ranges from 10 to 32%. The asphaltenes are insoluble in water and non-polar paraffinic hydrocarbons, soluble in carbon disulfide (CS_2) and chloroform ($CHCl_3$), and only slightly soluble in alcohol, ether, and low molecular-weight paraffinic hydrocarbons. Asphaltenes contain rather high amounts of oxygen (3-11%) and sulfur (7-9%); they are mainly composed of high molecular-weight (greater than 1500) aromatic and heterocyclic hydrocarbons. The asphaltenes melt in the range of 180 to 280°C. By hydrogenation at moderate temperatures, asphaltenes may be partly converted to resinous materials. Hydrogenation at high temperatures and increased pressures results in a low yield of liquid hydrocarbons.

Resins are amorphous solids that are soluble in most hydrocarbon solvents. The compounds forming resins are structurally similar to those forming asphaltenes. Natural bitumens may consist of up to 20% resins, however, generally of less than 10%. The resins (- like the asphaltenes -) contain rather high proportions of oxygen and sulfur; they are mostly naphthenic acids of a molecular weight of about 800.

The paraffinic, napthenic, and aromatic hydrocarbons in bitumens and also heterocyclic compounds (which contain oxygen, sulfur, nitrogen, and small amounts of various metals) have molecular weights ranging from 500 to 5000; compounds of lower molecular weight can also be present in small amounts in bitumens.

Sulfur is present primarily in thiophenes and benz-thiophenes, with smaller quantities in sulfoxides. Nitrogen is present primarily as pyridines and pyrrole-type compounds. Oxygen is present mainly in hydroxyl, carbonyl, and ester groups. About 35% of the hydrocarbon fraction is saturated; about 50% or more is aromatic.

The simplest way by which the chemical character of the bitumen components (mainly asphaltenes and maltenes) can be characterized is by giving the carbon to hydrogen atomic ratio (C : H Ratio) in the material, as obtained from combustion analysis. This ratio stands for its mean aromatic character, a high ratio indicating a strongly aromatic nature; for saturated aliphatic hydrocarbons the C : H = ~ 0.5, for benzene the C : H = 1, and for naphthalene C : H = 1.25.

By blowing air through molten bitumen oxygenation and dehydrogenation reactions take place. About 30% of the bitumen produced by industry is air-blown.

Many varieties of both blown and distilled bitumens have been used for the bituminization of radioactive wastes.

2.5 Chemical Stability

Bitumens are generally very resistant to many reagents at atmospheric temperature. It is therefore applied for protection of more reactive material against chemical influences.

At higher temperatures, bitumen can react with various agents, such as oxygen, sulfur, and chlorine, and this property is made use of in the preparation of special types of bitumen. These reactions effect mainly a dehydrogenation of the bitumen and a formation of asphaltenes.

Chemical changes may also occur when the bitumen - without being in contact with other substances - is exposed to high temperatures (> 300°C). This phenomenon should be considered not only when producing bitumen by means of distillation but also in the bituminization of radioactive wastes.

The chemical and heat stability are influenced by the chemical composition of the bitumen. Differences in composition cause differences in the rate of reaction; however, differences in the type of reaction have seldom been observed.

The reaction of bitumen with oxygen is important in the manufacture of blown bitumen and because bitumen, when applied, is very often in contact with air.

The rate of reaction depends very much on the temperature. Under conditions prevailing in the manufacture of blown bitumen, when air and molten bitumen are in close contact at 250°C or higher temperatures, the reaction is fairly rapid. In its applied form, bitumen is usually in contact with oxygen at atmospheric temperature, when the bitumen has a very high consistency. Under these conditions the reaction proceeds very slowly.

Bitumens can be hardened due to oxidation. The rate of oxidation is much higher in the presence of light /R-4/. Oxidation in the light is promoted and hardening is restricted to a depht of about 5 µ. In the dark, hardening may occur down to dephts of 2.5 - 5 mm.

Different types of bitumen vary greatly in their susceptibility to oxidation in the presence of sunlight, e.g. blown bitumen has a greater resistance towards oxidation.

At a depht of about 50 µ in a bitumen layer the intensity of the light is reduced to about 1%. Oxygen can only reach the interior by means of diffusion at a very low rate, owing to the high consistency of the bitumen. These factors contribute to the resistance of bitumen to atmospheric influences.

Generally bitumens are less resistant towards acidic than alkaline solutions. Bitumen does not seem to be attacked by concentrated alkaline solutions at room temperature, although dilute alkaline solutions react with acidic bitumen constituents to form salts such as sodium naphthenates which serve as excellent emulsifying agents for bitumen.

This reaction is particulary noticeable in soft bitumens of high acid value and at a concentration of 0.1% sodium hydroxide.

Using e.g. 20% sodium hydroxide and 10% sodium carbonate solutions, emulsification is not observed at room temperature, even after five years contact.

However, the surface of bitumens may show signs of emulsification in contact with these concentrated solutions at 60°C.

The resistance of bitumens to acids depends on the concentration of the acids. Generally, concentrated acids attack bitumens. Prolonged contact with dilute acids may cause a hardening of the bitumen, due to the formation of asphaltenes.

Concentrated sulfuric acid (e.g. 96%) attacks the aromatic components of the bitumen (acid tar). The saturated hydrocarbons are not even attacked at 200°C.

Bitumen is resistant to dilute sulfuric acid, and highly resistant to concentrated hydrochloric acid solutions at room temperature.

Bitumen is not resistant to nitric acid.

Concentrated nitric acid effects oxidation and nitration. Dilute nitric acid also attacks bitumen, even at low concentrations and at room temperature. Bitumen is therefore unsuitable for the protection of materials against the action of free nitric acid.

The resistance of bitumen towards various chemical reagents under different conditions of concentration, temperature (and time) is indicated qualitatively in Tables II and III.

TABLE II

RESISTANCE OF BITUMEN TO THE EFFECTS OF CHEMICALS AT ROOM TEMPERATURE

Mexphalt / Chemicals		40/50,20/30,10/20				R85/25,R85/40,R115/15			
		0.5	1	1.5 (years)	2	0.75	1	3.5 (years)	5
Inorganic acids									
Hydrochloric acid up to	10%	+	±		-				+
Hydrochloric acid	10-30%	±	±		-				+
Sulphuric acid up to	50%	+		±	-				+
Nitric acid up to	10%	±	-						+
Nitric acid up to	25%	-							±
Nitric acid up to	50%	-							-
Phosphoric acid	5%					+			
Hydrofluoric acid	40%						+		
Organic acids									
Lactic acid	10%			-					+
Butyric acid	10%			-					+
Formic acid	85%					+			
Acetic acid	20%					+			
Benzoic acid	1%						+		
Picric acid	1%						+		
Salicylic acid	0.1%						+		
Phenol solution	1%						+		
Inorganic alkalis									
Caustic soda up to	30%							+	
Ammonia	25%						+		
Soda solution	10%							+	
Soda solution (sat.)						+			
Salts									
Common salt solution (sat.)								+	
Chlorine-containing brine								+	
Sea water									+
Magnesium chloride	14%					+			
Sodium hydrosulphite sol. (sat.)						+			
Sodium sulphite sol. (sat.)						+			
Sodium hypochlorite	5 g/l					+			
Waterglass						+			
"Teepol"								+	+
Formaldehyde sol.	30%					+			

Not affected:	+
Little to moderate attack:	±
Strongly attacked:	-

(Source: Shell, Brussels, data from Amsterdam Laboratory and literature).

TABLE III

CHEMICAL RESISTANCE OF BITUMEN

- The data are based on continuous exposure to the chemical for one month -

(Source : Shell Centre, London)

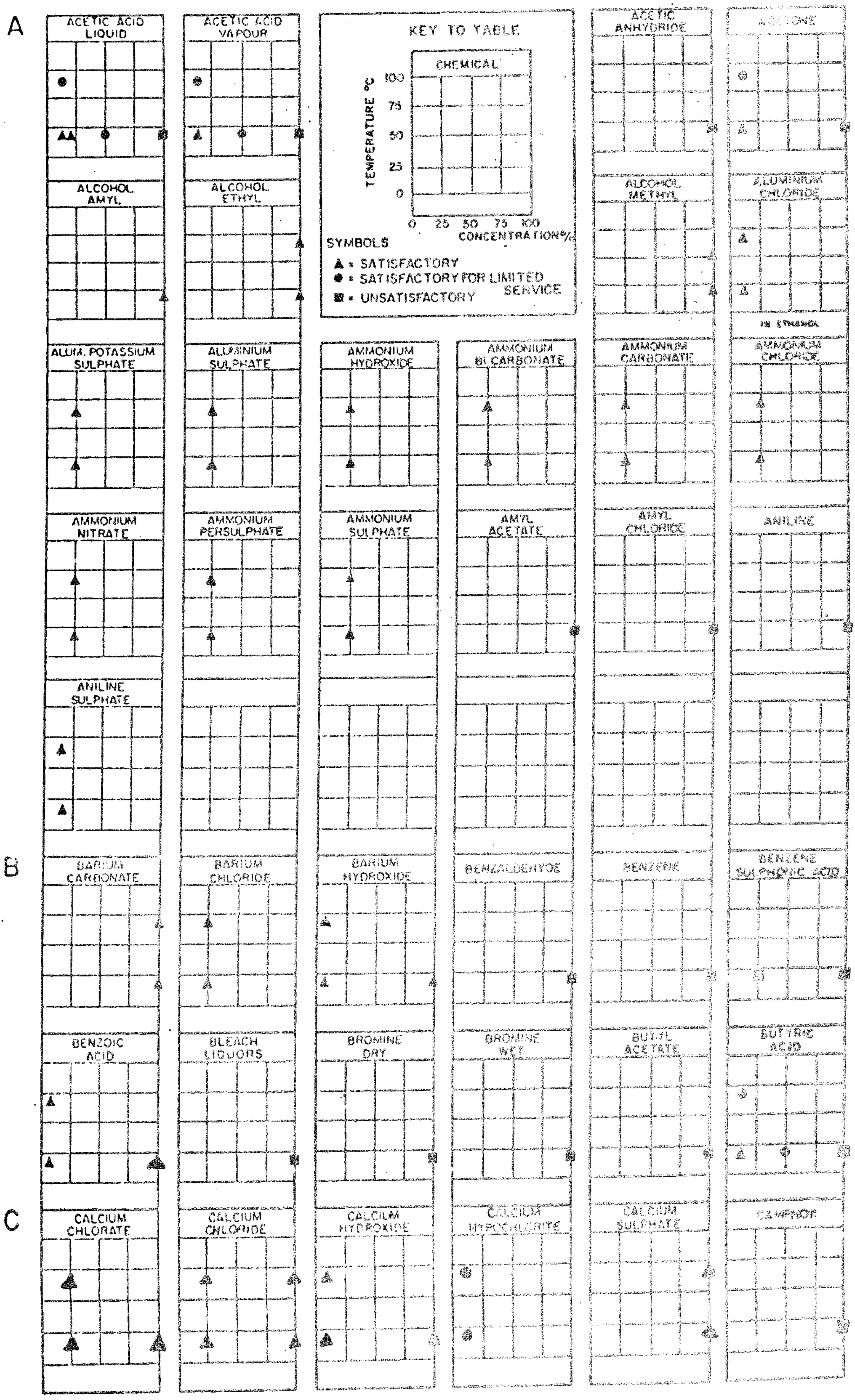


TABLE III

(continued)

CHEMICAL RESISTANCE OF BITUMEN

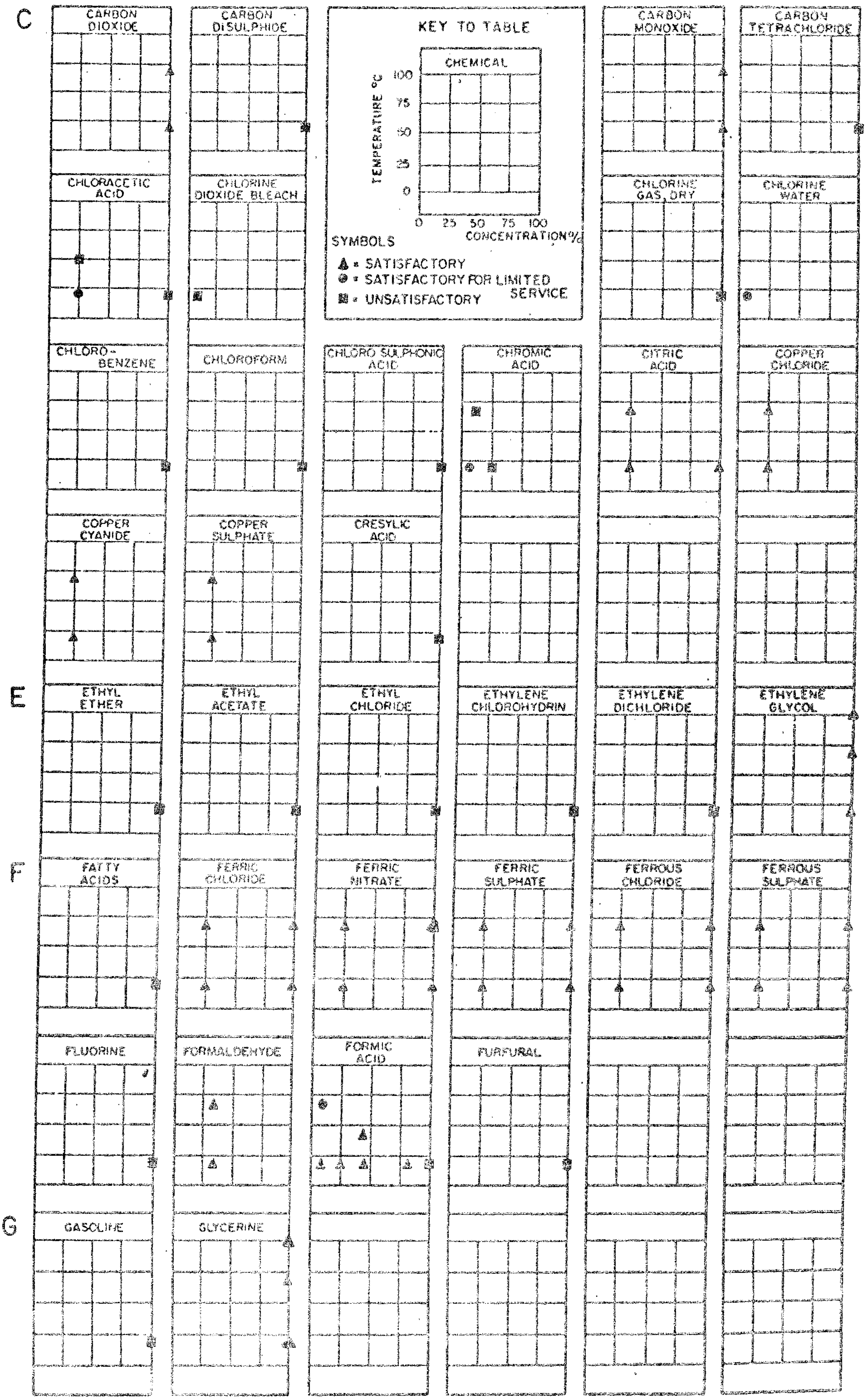


TABLE III

(continued)

CHEMICAL RESISTANCE OF BITUMEN

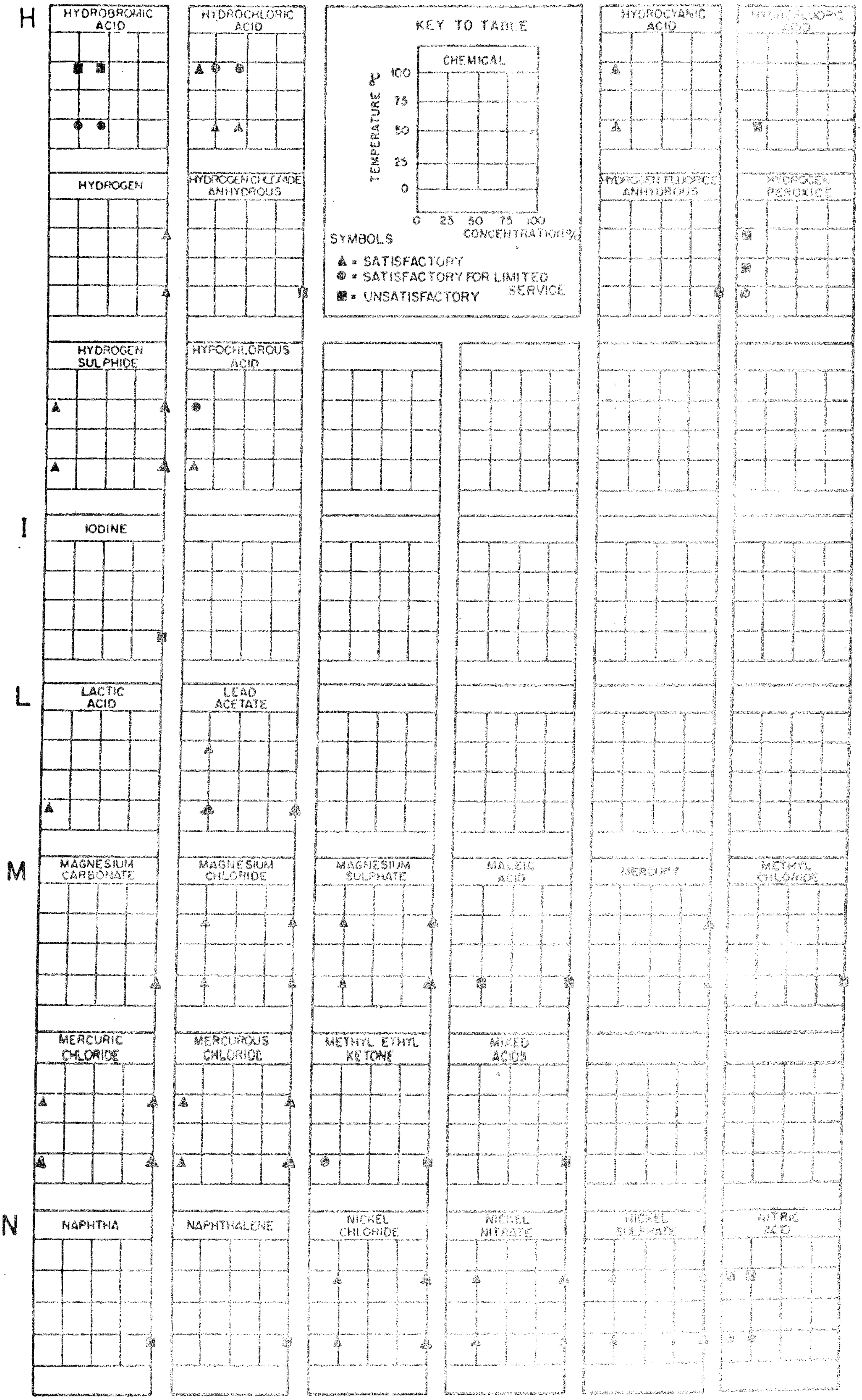


TABLE III

(continued)

CHEMICAL RESISTANCE OF BITUMEN

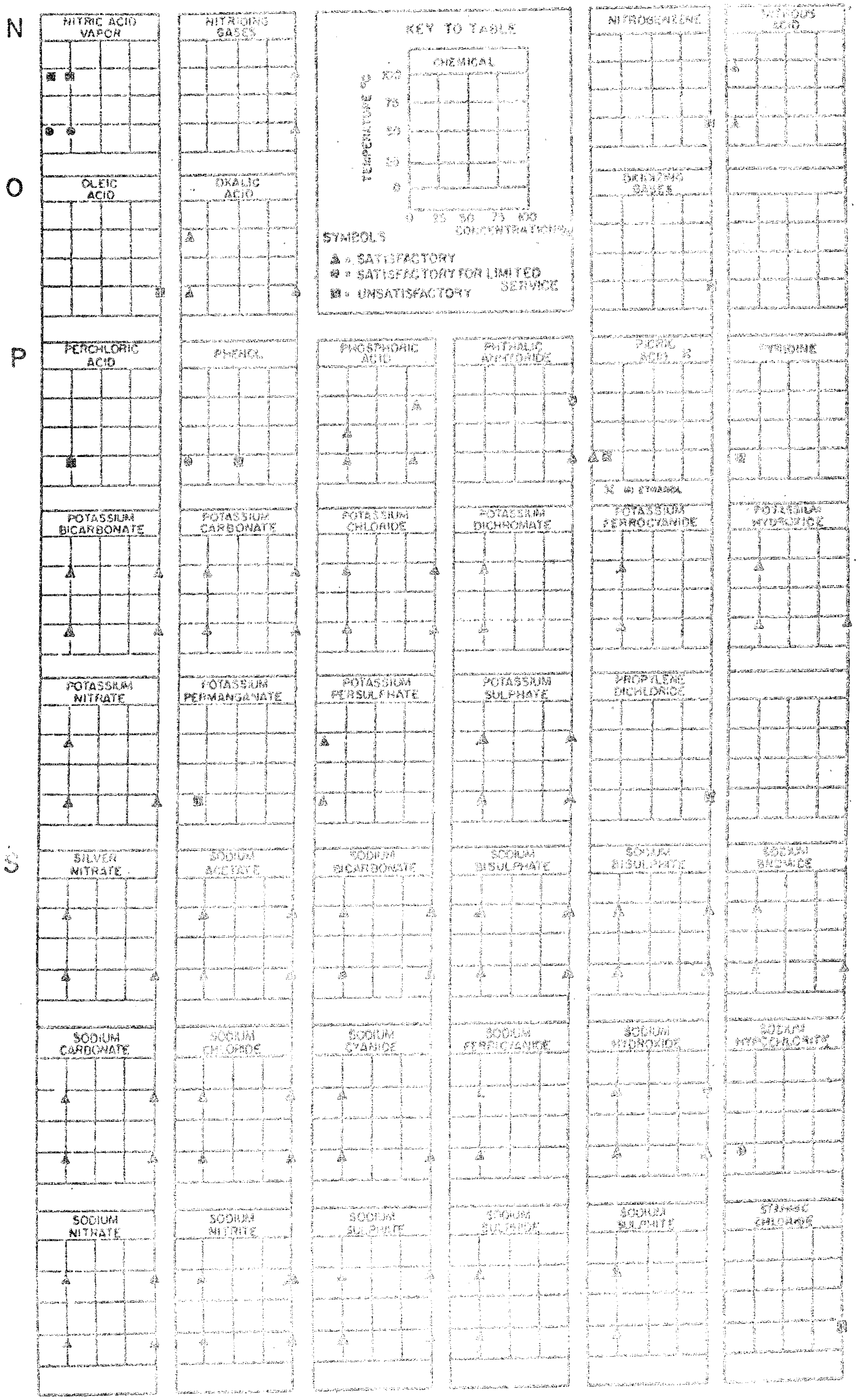
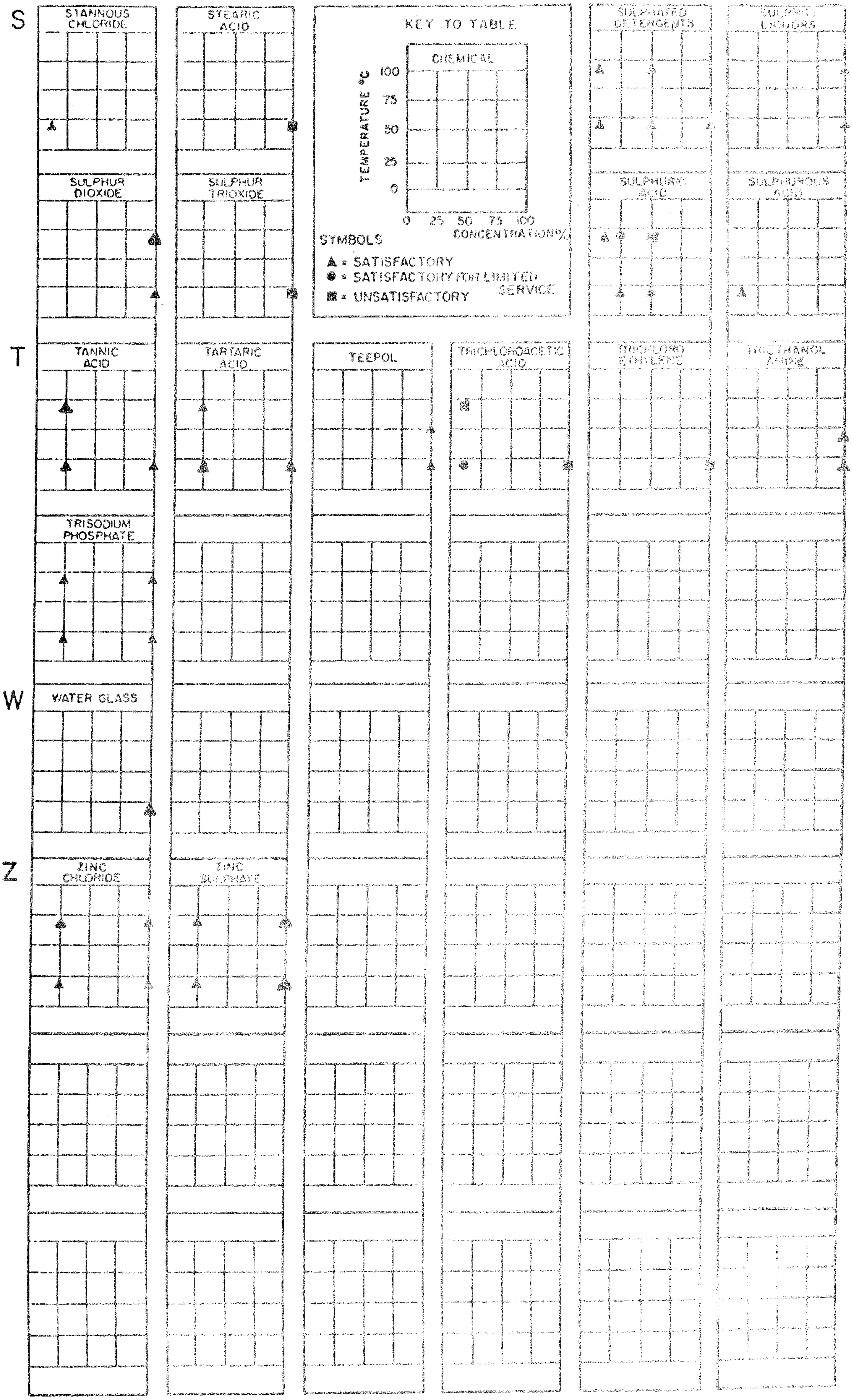


TABLE III
(continued)

CHEMICAL RESISTANCE OF BITUMEN



2.6 Physical Properties*)

2.6.1 General

The physical properties of bitumens are of a decisive importance for their use as a suitable coating material for radioactive wastes. Bitumens are solid or semi-solid substances at room temperature. They can easily be liquefied by heating. The viscosity of bitumens decrease with increasing temperature.

The relationship between temperature and viscosity is not identical for different types and grades of bitumen as can be seen in Table IV and Fig.1.

In most applications, bitumen is heated until it becomes sufficiently fluid for each particular application. If the optimum application viscosity is known, the temperature to which any given grade of bitumen must be heated can easily be read from Fig.1.

The optimum application viscosity in the bituminization of nuclear wastes will be determined by various factors such as the characteristics of the waste to be incorporated, the bitumen to solids ratio, the type of mixer, the evaporation rate of water etc. Due to the variable factors to be taken into consideration, the most suitable viscosity for each specific case must be found by trials.

As the temperature of bitumen is increased, e.g. from 250 up to 500°C, its colloidal and micelle structure is altered, volatile substances are evaporated, and the fluidity of the residue increases until it flows almost like lubricating oil.

When then cooled to room temperature, the bitumen is usually harder and tends to be more brittle than it was originally. Further cooling causes progressively less fluidity, i.e. higher viscosity.

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*) Mainly based on Ref R-6.

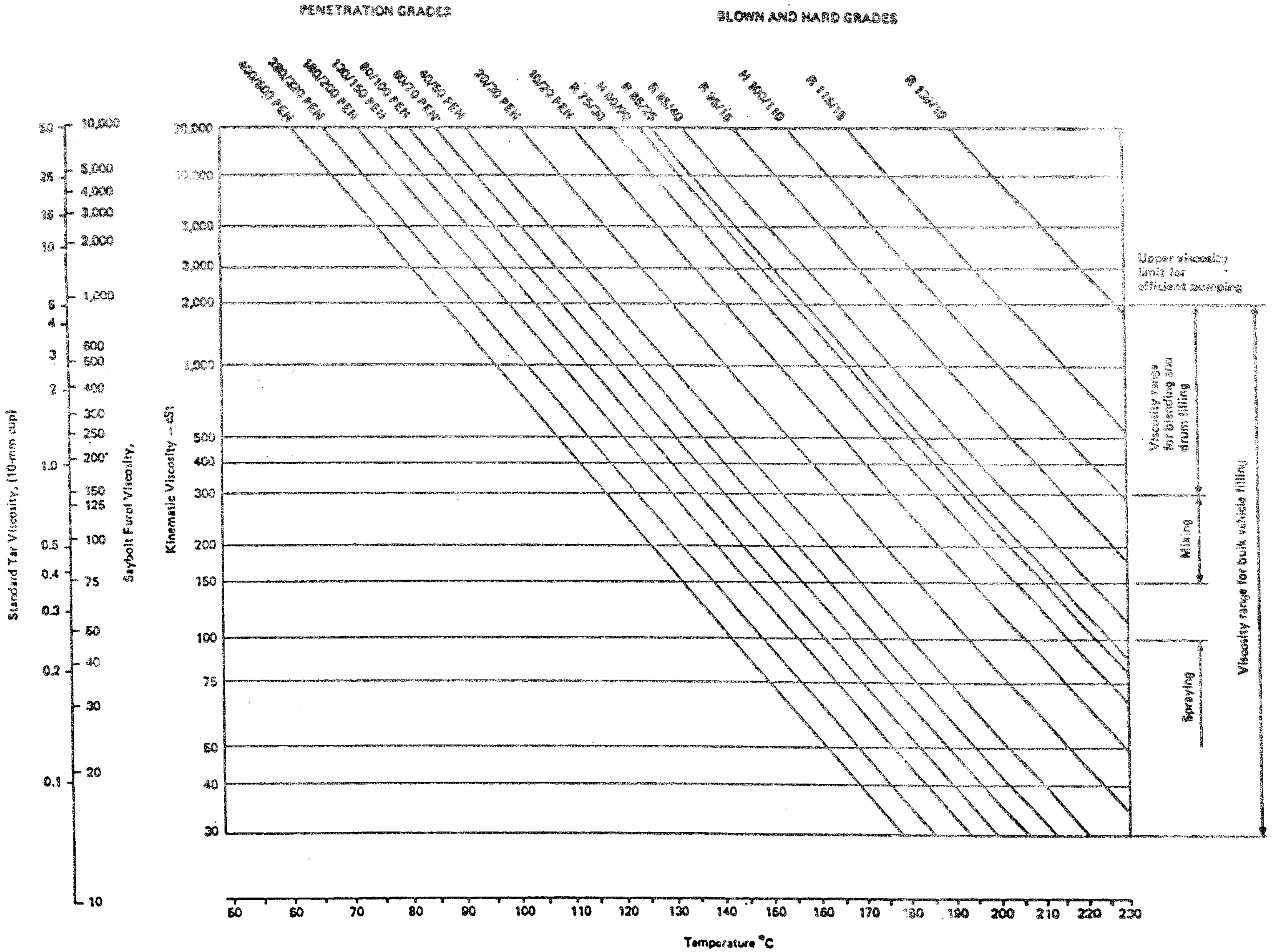


Fig. 1 - TYPICAL TEMPERATURE/VISCOSITY CHART FOR BITUMENS

TABLE IV

EQUIVISCOUS TEMPERATURES (°C)

Mexphalt Grade	Viscosity in cSt						
	20 000	5 000	2 000	1 000	200	100	50
180/200 ("Spramex")	70	85	97	108	138	156	176
80/100	78	94	106	117	149	166	187
60/70	85	101	113	123	155	172	193
50/60	88	104	116	126	158	175	197
40/50	91	107	119	130	162	179	201
30/40	95	111	124	135	168	185	208
20/30	101	117	130	142	175	193	216
10/20	111	128	141	153	186	205	228
H 80/90	133	150	163	172	206	224	246
H 100/110	154	170	184	195	227	245	-
R 85/25	126	144	157	169	205	225	248
85/40	127	144	157	168	201	220	243
95/15	143	161	176	188	225	245	-
115/15	166	186	202	216	(256)	-	-
135/10	190	210	225	240	-	-	-

TABLE V

CONVERSION FACTORS FOR VISCOSITIES

Known Viscosity	To obtain value in unknown units, multiply by:						
	Kinematic (cSt)	Redwood I	Redwood II (sec)	Saybolt Universal	Saybolt Furol	Engler (°E)	STV (sec)
Kinematic (cSt)	-	4,05	0,405	4,58	0,458	0,132	0,0025
Redwood I (sec)	0,247	-	0,1	1,13	0,113	0,0326	-
Redwood II (sec)	2,47	10	-	11,3	1,13	0,326	0,0062
Saybolt Univ (sec)	0,218	0,885	0,0885	-	0,1	0,0287	-
Saybolt Furol (sec)	2,18	8,85	0,885	10	-	0,287	0,0054
Engler (°E)	7,58	30,7	3,07	34,81	3,48	-	-
STV (sec) (10 mm cup)	400	-	162	-	183	528	-
Ford cup (sec)	3,15	-	0,78	-	-	-	-

The viscosity of bitumen is high, ranging from 10^3 to 10^{20} poises at 0 to 50°C.

(The viscosity of heavy machine oil at 20°C is 6.6 poises). The viscosity of a natural pitch was found to be about 1×10^{10} at 15°C and 5×10^{11} poises at 0°C.

It is recommended to transfer bitumen by pumping at a viscosity of about 1000 cSt.

Two shipments of a given type of bitumen are likely to fluctuate in their physical properties and composition, even when derived from the same source.

Each bitumen has a predetermined consistency and some fixed physical properties over which one has no control.

The selection of a suitable bitumen for a given waste bituminization process should primarily be based on the following physical factors which can be determined beforehand by using standard test methods:

a) the penetration which characterizes the relative hardness or consistency of a given bitumen under known conditions of temperature, loading, and time;

   (the penetration is defined as the depth, measured in multiples of 0.1 mm, that a standard needle penetrates into a bituminous surface under a constant load (e.g. 100 g) in a specified time (5 sec) and at a specified temperature (e.g. 25°C))

b) the viscosity which determines the flow characteristics of the bitumen at the temperature applied;

c) the softening "point" which indicates the temperature at which the bitumen gets "slow-flowing";

   (the softening point is determined by means of the ring-and-ball apparatus:

   a ring of given dimension, filled with bitumen, is loaded with a steel ball (3.5 g); the whole is heated in a bath and the temperature at which the bitumen reaches a certain deformation is reported as the ring-and-ball (R + B) softening point)

d) the flash point which indicates the temperature to which the material may be safely heated on an open flame.

Besides these particular properties the knowledge of some physical constants of bitumens might be of value for their use in nuclear waste management.

In the subchapters hereafter a short survey of the following physical properties is given:

- specific density
- coefficient of cubical expansion
- specific heat
- thermal conductivity
- permeability to water vapour
- surface tension
- total surface energy
- resistivity/conductivity
- dielectric strength (breakdown voltage)
- dielectric constant and dielectric loss
- mechanical and rheological properties
- radiation resistance.

2.6.2 Specific density

The specific density of bitumens having a penetration at 25°C greater than 5 was determined by means of a pyknometer, that of harder bitumens by means of a hydrostatic balance.

The results obtained in these determinations are given in Table VI.

TABLE VI

SPECIFIC DENSITY OF BITUMENS

Pen. 100 g/5 sec/25°C	Specific density at 25°C
300	1.01 ± 0.02
200	1.02 ± 0.02
100	1.02 ± 0.02
50	1.03 ± 0.02
25	1.04 ± 0.02
15	1.04 ± 0.02
10	1.05 ± 0.02
5	1.07 ± 0.03
< 5	1.07 ± 0.03

The specific densities of bitumens partly depend on the origin of the crude oil from which they have been prepared, but in general they lie between the limits indicated in Table VI.

For the rest, the specific density is only dependent on the penetration, and is therefore practically the same for bitumens of the pitch type, the normal type, and the blown type of the same penetration.

However, bitumens prepared from cracked residues are generally of higher specific density; bitumens from heavily cracked residues for instance, yielded the values given in Table VII.

TABLE VII

SPECIFIC DENSITY OF BITUMENS FROM HEAVILY CRACKED RESIDUES

Pen. 100 g/5 sec/25°C	Specific density at 25°C
100	1.13 ± 0.02
50	1.13 ± 0.02
25	1.13 ± 0.02
15	1.14 ± 0.02
5	1.15 ± 0.02
< 5	1.16 ± 0.02

Generally the specific density of bitumens ranges from 0.99 to 1.2 g/cm^3.

For convenience and for most technical purposes one may assume that the density of bitumen at 25°C is 1 g/cm^3. (Nearly all crude oils are lighter than water, i.e. they have a specific density of less than 1 g/cm^3).

2.6.3 Coefficient of cubical expansion

The knowledge of the cubical expansion is required e.g. for the calculation of vessel capacities and in cases where shrinkage on cooling is an important factor.

The coefficient of expansion is practically constant in the temperature range of 15 - 200°C and almost identical for all bitumens (of a penetration 100 g/5 sec/25°C from 23 to 196). The coefficient of cubical expansion of bitumens in the range of 15 - 200 °C is 0.00061 ± 0.00001.

2.6.4 Specific heat

The specific heat of bitumens is a linear function of the temperature (in the range 0 - 300°C) which shows that no transition point and consequently no definite melting point does exist.

The specific heat varies somewhat with temperature as indicated in the following Table VIII:

TABLE VIII

SPECIFIC HEAT OF BITUMEN (Blown bitumen, penetr.40 at 25°C, soft.point = 85).

Temp., °C	Specific heat, cal/g °C
0	0.430
100	0.462
200	0.494
300	0.526

Change of specific heat per 1°C = 0.00032.

Changes of specific heat of various bitumens per 1°C were measured which ranged from 0.00032 to 0.00078.

A value of 0.45 cal/g.°C can be considered as mean value for the specific heat of bitumen.

2.6.5 Thermal conductivity

It has been most suitable for technical use to express the thermal conductivity in kcal/m.°C.h, that is the number of kilo-calories flowing per hour through a cross-section of 1 m^2, when the temperature gradient is 1°C per metre.

The values expressed in kcal/m.°C.h are converted into cal/cm.°C.sec. units by multiplication by the factor 0.00278.

The thermal conductivity is practically the same for all bitumens; it changes (decreases) only slightly with the temperature (about 1% per 10°C).

[illegible]

For practical purposes a thermal conductivity of 0.13 kcal/m.°C.h can be used.

[illegible]

0.15 W/m °C

2.6.6 Permeability to water vapour

For determining the coefficient of diffusion of the water vapour within the material, Fick's diffusion law can be used, which is expressed by the following equation:

$$N = \frac{D\,At}{x}(p_1 - p_2)$$

where N = the amount of water that will diffuse in time t in the direction x though a cross-section with a surface area A, $p_1 - p_2$ being the difference in the vapour pressure of the water.

This law applies perfectly to materials which absorb practically no water (polystyrene).

The permeability of bitumen to water vapour can be expressed by the coefficient of diffusion D; this represents the amount of water vapour (in grams) which, at a vapour pressure difference of 1 mm Hg, would diffuse in one hour through a layer of 1 cm thickness having a surface area of 1 cm^2 if Fick's law would hold.

A diffusion coefficient of 1.3×10^{-8} $g.h^{-1}.cm^{-1}.mmHg^{-1}$ has been determined for bitumen at 25°C.

The diffusion coefficient depends on the penetration and the temperature of the bitumen.

2.6.7 Surface tension

The surface tension valid for a great variety of bitumens of various temperatures is given in Table IX:

TABLE IX

SURFACE TENSION OF BITUMENS

Temp., °C	Surface tension, dynes/cm
25	33 ± 1 (extrapolated)
100	29 ± 1
120	28 ± 1
150	25.5 ± 1.5

2.6.8 Total surface energy

The total surface energy E_s can be calculated using the following expression:

$$E_s = \sigma - T \frac{d\sigma}{dT}$$

σ = surface tension

T = absolute temp.

For a great variety of bitumens a total surface energy of 51 ± 1 ergs/cm^2 has been determined.

2.6.9 Resistivity/conductivity

Bitumen has a high resistance (a low conductivity) and is therefore a good insulating material. Harder grades have a slightly higher resistance than soft grades though the differences will not be important in practice. The resistance of all grades decreases with increasing temperature; typical figures are:

Temp (°C)	Resistance (ohm/cm)
30	10^{14}
50	10^{13}
80	10^{12}

The influence of fillers on resistance is negligible, provided that they are present in the bitumen as solids, and that conductive fillers such as coke are not used at high concentrations. Large quantities of conductive filler can, however, reduce the resistance markedly, e.g mastic asphalts containing coke, graphite or metal powder fillers can have resistances of 10^3 - 10^6 ohm/cm and may be used as flooring where some conductivity is required to avoid electrostatic hazards.

2.6.10 Dielectric strength (breakdown voltage)

The dielectric strength is measured in kV/mm, and depends on the conditions of measurement and the shape of the electrodes. Hard bitumens have a higher dielectric strength than soft grades: the strength of all grades decreases with increasing temperature.

a) Bitumen of a penetration of 170/200:

Temp (°C)	Dielectric strength (kV/mm)(flat electrodes)
20	10
50	5

b) Hard bitumens:

Temp (°C)	Dielectric strength (kV/mm)(flat electrodes)
20	30-60
50	10-20

The effect of fillers on these values is not known precisely but they will tend to reduce the above values.

2.6.11 Dielectric constant and dielectric loss

The dielectric constant of bitumen is about 2.7 at 20°C, rising to about 3.0 at 90°C.

The dielectric losses in bitumen increase with increasing temperature but decrease with increasing frequency as shown in the following Tables X and XI.

TABLE X

DIELECTRIC LOSS

°C	tan δ (50 c/s)
20	0.015
50	0.017
80	0.029
100	0.045

TABLE XI

DIELECTRIC LOSS

Frequency (c/s)	tan δ (20°C)
50	0.015
10^5	0.006
10^6	0.003
10^7	0.001

where δ = angle of phase distortion.

2.6.12 Mechanical and rheological properties

Bitumens behave either as viscous liquids or as elastic solids; a behaviour intermediate between the two states can also be observed. Bitumens are thermoplastic and subject to variable mechanical reactions; these properties are affected by the conditions of loading and by the presence of solvents remaining from distillation or which have been added. Mechanical resistance to deformation can be measured in terms of poise, which is an absolute unit of viscosity; resistance can be measured in all ranges of consistency and at all ordinary temperatures. In bitumens deviation from pure liquid, or Newtonian, flow is common, with various forms of plastic behaviour being found.

The deviation also can be measured and expressed in terms of poises. Elasticity appears in bitumen under certain conditions. In the Newtonian or pseudo-Newtonian types it appears as a roentgen effect, or reaction to impact; in oxidized industrial bitumen it is a weak solid-type, partly static reaction. These effects have various applications in practical use. The roentgen effect is of special importance in road use, where dynamic impact is high.

The most important characteristic of bitumen is its flow or rheological properties. Bitumens can be divided into those that have purely viscous flow characteristics and those that have plastic characteristics. Plastic bitumens are higher in asphaltenes, and these are not so well dispersed as in viscous bitumens.

Empirical tests are used in measuring the flow characteristics of asphalt; the most important properties tested are the penetration and the softening point. Penetration and hardness are reciprocally related, i.e. the lower the penetration the greater the hardness. Bitumen is produced in various "penetration grades", which may range from 5 or even lower, up to 300.

The penetration and the softening point are related to the asphaltene content.

The viscosity of viscous bitumens is difficult to measure directly, however, it can be determined roughly from the penetration by the following formula:

$$\text{Viscosity (poises)} = \frac{5.31 \times 10^{9}}{(\text{penetr.})^{1.93}}$$

For many purposes bitumens with purely viscous flow properties are not satisfactory. Even the hardest viscous bitumen will flow in time under small stresses, and there are cases in which there is a stress on the bitumen that needs to be resisted without flow. Roofing bitumen is an example in which the small gravitational stresses set up by the slope of the roof will cause flow and consequently a plastic type of bitumen has to be used. The viscous bitumens are generally made plastic by air-blowing.

This decreases the content of resinous dispersion agent and increases the asphaltene content. The micelles tend to be attracted to each other and set up first a loose structure and, as resins are depleted further, a more firm, gel-like structure. Such systems show complex flow characteristics such as elasticity and thixotropy and will tend to resist stresses.

The knowledge of the compressibility of bitumens might be of importance for the geological storage of bitumen-waste products (BWPs). The change in volume of a certain quantity of pure bitumen under pressure is relatively small compared with that resulting from a change in temperature.

The compressibility of bitumens is of the order of 40×10^{-6} cm^2/kg. As the thermal coefficient of expansion per °C is 600×10^{-6}, the change in volume per 1°C drop in temperature is about the same as that per 15 kg/cm^2 rise in pressure.

The influence of pressure on the viscosity of bitumens depends on the type of bitumen, however, can be enormous.

If one applies 1000 kg/cm^2 pressure onto bitumens that behave almost like Newtonian liquids, the viscosity may increase by a factor of as much as 10^5 and more.

The addition of certain types of fillers permit to build up a structure in a sol-type bitumen which has sufficient yield stress to prevent flow at elevated temperature and which is also sufficiently plastic to permit slow but considerable deformation of the bitumen product at low temperature.

A bitumen emulsion containing a colloidal clay emulsifier offers, upon drying, an extreme example of a bitumen product with that kind of filler structure. An amount of only 1.5 vol -% of colloidal clay (bentonite) in the form of a honeycomb skeleton in the resultant bitumen layer prevents a soft bitumen from flowing off vertical surfaces even at temperatures far above 100°C.

Bitumen products of high mechanical resistivity can also be obtained by mixing bitumen with mineral aggregates resembling road carpet mixtures.

The mechanical strength required for a certain bitumen-solids mixture can be realized by a suitable combination of the viscosity of the bitumen and the structure and amount of the inorganic (mineral) material incorporated in the bitumen.

2.6.13 Ductility and brittleness

Various tests have been suggested to characterize the ductility and the brittleness of bitumen.

The ductility test put forward as early as 1903 by Dow /R-7/ consists in stretching a bitumen briquette narrowed in the middle to a cross section of 1 cm^2, in a water bath of 25°C at the rate of 5 cm/min. Under these conditions most bitumens for road making show very high ductilities, mostly exceeding 100 cm.

Bitumens of the elastic sol-type exhibit a more regular, yet strong increase in ductility with rise of temperature, whereas the ductility of bitumens of the gel-type is little affected by temperature. The ductility is closely related to the rheological properties of the bitumen.

The degree of brittleness is important in many applications of bitumen. Whether a substance is brittle or not, depends on the conditions prevailing or applied. One can distinguish between the static brittleness (a measure of which is the elongation at rupture), the dynamic brittleness, and the "dynamic toughness". The brittleness of bitumen depends on the rate of deformation and on the temperature. Therefore the degree of brittleness is expressed by means of the temperature at which breakage occurs under given conditions. The method evolved by Fraass /R-8/ is widely used in Europe; it is based on the determination of the temperature at which a strip of bitumen breaks when bent slowly (see Annex).

2.6.14 Radiation resistance

In the bituminization of radioactive wastes the stability of the bitumen matrix against α-, β-, or γ-radiation is of decisive importance, especially for the question on the maximum amount of radioactivity (α-, β- or γ-emitting nuclides) within the matrix compatible with a safe storage.

The investigations on the radiation resistance of bituminous materials (using various internal and external radiation sources and dose rates of 1 - 700 rad/sec) allow to draw the following conclusions:

- bitumen is suited (sufficiently stable) for the conditioning of radioactive wastes if the total dose will be less than 10^9 rad;

- bitumen absorbing a dose of 10^{10} rad (and higher) suffer from a break-down of the chemical structure leading to:

  o formation of hydrogen, methan, C_2H_x, C_3H_x, C_4H_x, carbon monoxide

  (ca 0.5 - 1 cm^3 H_2/g bitumen/10^8 rad, measured at 25°C and 760 mm Hg),

  o volume increase (due to the gas evolution by radiolysis),

  - increase of the softening point,
  - increase of the elasticity,
  - slight increase of ductility,
  - decrease of penetration,
  - increase in asphaltenes and resins (and decrease in oils), and a
  - decrease of the flash point;
- irradiation of bitumen of 10^7 rad causes only minor changements of its properties;
- for the same total dose the changes of bitumen decrease with the decrease of source intensity (dose rate);
- hardening of bitumen becomes apparent at doses exceeding 10^8 rad;
- the increase in volume due to radiation degradation is less pronounced for oxidized (blown) bitumen than for straight-run distillation bitumens;
- the higher the dose rate (rad/sec) the lower the speed of gas release (expressed in $cm^3 . d^{-1} . Ci^{-1}$) from the bitumen products /42/.

2.6.15 Summary of physical data

In order to obtain a quick survey on the physical properties and constants of bitumen the mean values or most "practical values" are summarized in the following Table XII.

TABLE XII

PHYSICAL DATA ON BITUMEN

No	Property	Value
1	Specific density	1.04 ± 0.03 kg/l at 25°C
2	Coeff. of cub. expansion	0.00061 per °C (in the range 15-200°C)
3	Specific heat	0.45 cal/g. °C
4	Thermal conductivity	0.13 kcal/m. °C.h
5	Permeability to H_2O-vapour	1.3 x 10^{-8} g.h^{-1}.cm^{-1}.$mmHg^{-1}$ (at 25°C)
6	Surface tension	29 ± 1 dynes/cm (at 100°C)
7	Total surface energy	51 ± 1 ergs/cm^2
8	Resistivity/conductivity	10^{14} ohm/cm (at 30°C)
9	Dielectric strength	20-30 kV/mm (at 20°C; flat electrodes)
10	Dielectric constant	2.7 (at 20°C)
11	Dielectric loss	tan δ 0.015 (50 c/s; 20°C)
12	Radiation resistance	ca 10^9 rad
13	Viscosity	10^3 - 10^{20} poises (in the range 50-0°C)
14	Combustion heat	10 000 cal/g
15	Compressibility	~ 4 x 10^{-5} cm^2/kg
16	Permeability to O_2 (blown bitumen, penetr = 16, R+B = 116°C)	~ 4 x 10^{-10} g.h^{-1}.cm^{-1}.$mmHg^{-1}$ (at 25°C)

2.7 Health Aspects

Pure bitumens, which do not contain additives, can be considered as not health hazardous. Numerous observations could not give any evidence of carcinogenic properties of pure bitumen. When using bitumens of unknown origin one has to take into consideration that they may contain coal-tar which is carcinogenic and very irritating.

When working with bitumen in a closed room a maximum permissible concentration for vapours of pure bitumen of 5 mg/m^3 has been proposed as a mean value for each period of max 15 min; when working with impure bitumen a concentration of 0.2 mg/m^3 should not be exceeded. If those concentrations cannot be kept masks provided with fresh air supply have to be used.

If warm or hot bitumen comes into contact with the skin one should not try to remove the still fluid bitumen; instead the place of contact should be brought into flowing cold water.

For the removal of bitumen from the skin pure toluene can be used. In principle, there is no haste to remove pure bitumen from the skin as it forms a sterile closure of the wound; the bitumen loosens by itself after some days.

The wearing of overalls, protecting gloves and shoes is necessary when working with hot bitumen.

2.8 Safety Aspects (Fire)

Normal bitumen at room temperature is not classified as an inflammable liquid.

Nevertheless, hot bitumen has to be considered as a burnable liquid. Bitumen has a flash point ranging from 200 - ~ 320°C depending on its origin and nature. Therefor the major fire and electrical hazards during the operation stage are basically due to sparks and possible local overheating.

For the fighting of a bitumen fire foam-, powder-, or carbon dioxide (CO_2)-extinguishers must be employed.

The use of water beams should be avoided as it can cause cooking over or even a tearing open of the vessel containing the bitumen.

For certain special cases water spray or water beams may be used for the removal of heat from vessels containing burning bitumen.

3. LONG-TERM BEHAVIOUR OF BITUMEN

3.1 General

The tendency of bitumen to age under atmospheric influences has been known for a long time. However, until recently the ageing of bitumen was not intensively investigated. This is undoubtly due to the fact that even under severe conditions most bitumen constructions have a long service life. During the last 20-30 years the phenomenon of ageing received an ever-increasing interest, especially in connection with protective coatings, bitumen paints, road constructions, and presently also in connection with the long-term storage of bituminized radioactive wastes.

The ageing is influenced by several factors, such as the presence of impurities in the air or in the water in contact with the bitumen, and the kind of solids incorporated in the bitumen.

Besides the ageing of bitumen two further factors need to be considered for the long-term stability and behaviour of bitumen:

- reactions with (solid) materials coming into contact with it and
- its possible microbial degradation.

These factors, which are of importance for the long-term stability of bitumen in view of its use as coating material for radioactive wastes, are treated in the following chapters.

3.2 Durability of Bitumen /R-4/

3.2.1 Physical hardening

As already mentioned, bitumens are colloidal systems in which heavy constituents are peptized by means of the resins (aromatic and polar substances) in a mixture of hydrocarbons.

Cooling of such systems from its application temperature to ambient temperature shifts the absorption equilibria in the direction of larger conglomerates and hence an increase in viscosity.

This phenomenon may be classified as physical hardening. Owing to the high viscosity the shift to new equilibria is a relatively slow process which may last several months.

Another cause of physical hardening may be the presence of paraffin wax, which slowly crystallizes when the bitumen is cooled down. In most cases this process is faster than the one mentioned above. Both processes are reversible in that upon reheating of the bitumen the original viscosity is restored.

A third possible cause of physical hardening is evaporation of volatile material. The rate of evaporation depends on the nature and quantity of the volatile components and on the conditions of exposure, mainly the temperature. The resulting losses vary considerably with the type of bitumen, being dependent on the molecular weight distribution of the maltenes. Commercial bitumens normally do not harden to an appreciable extent due to this cause, e.g. it was found that the viscosity of bitumens usually increases by a factor of less than two after heating at 60°C for 14 days. In some cases the hardening might be of importance. It is then comparatively easy to assess this factor by heating the bitumen under exclusion of oxygen to a temperature about equal to that applied in practice.

3.2.2 Chemical hardening

Like many organic substances, bitumen is slowly oxidized when in contact with atmospheric oxygen. The polar oxygen-containing groups formed tend to associate to micelles of higher micellar weight and thus the viscosity of the bitumen may increase considerably.

It is necessary to distinguish between chemical hardening in the presence and in the absence of light. In the light, the oxidation reaction proceeds at a much higher rate than in the dark and, more-over, is of a different type. The bound oxygen is mainly present in CO groups and the remainder of the oxygen consumed disappears for the greater part as CO_2 and H_2O.

In the absence of light all the reacting oxygen is bound:

- after short times of ageing (e.g. one week) as SO-groups, and
- after prolonged ageing (one year and more) as CO-groups (probably esters).

Presumably the sulphoxides slowly decompose into carbonyl compounds. This reaction is well known from work with model compounds, which showed sulphoxides to decompose into aldehydes and mercaptans.

The ageing (oxidation) of bitumen in the presence of light is mainly activated by the ultra-violet part.

Experiments have shown that oxidation in the presence of light is limited to a depth of about 5 µ. In practice, ageing affects the material to a greater depth, because in the long run the skin that is formed may crack and disintegrate into a powdery substance, which may be removed by rain or wind, a fresh surface thus being exposed to the light. However, for the durability of road constructions this phenomenon is only of minor importance.

The skin formed is very hard and insoluble in benzene.

In the application of bitumen in the field of nuclear waste management, it is the ageing in the dark that has to be considered as a cause of hardening.

The ageing in the dark is governed by two mechanisms:

- oxygen transport by diffusion and
- oxygen consumption by reaction.

A fast diffusion and a slow reaction lead to deep penetration of the oxygen, the opposite to shallow penetration (skin formation). For a distilled bitumen 50/60 the following values could be derived:

diffusion coefficient of oxygen: 1.2×10^{-6} cm^2/sec (at 50°C)

solubility of oxygen in bit.: 0.10 cm^3 (at standard temp. and press)/cm^3 atm. (at 50°C).

The reactivity of bitumen towards oxygen varies with origin, type and temperature. In the range 30 to 50°C the temperature has only little influence on the mechanism of hardening.

It has been shown experimentally and by calculation that the maximum depth of oxygen penetration is in the range of 2.5 to 5 mm. In the long run, however, the depth of the oxygen penetration will increase somewhat; in any case, the rate of hardening drops considerably in the course of time.

There are no appreciable differences in the type of ageing between most bitumens.

An increase in temperature and pressure accelerates the oxidation of bitumen.

3.3 Reactions with Various Materials

In view of the use of bitumen as immobilisation matrix for solid radioactive residues the possible changements of bitumen properties with time, due to materials contacting it, have to be taken into consideration. Besides the contact with the incorporated waste and the container material, the bitumen can or will come, during (geological) long-term storage, in contact with air, hydrogen (present as hydrolysis product), water solutions (ground water, salt solutions, other mineralized water solutions), and the surrounding geological media and man-made structural materials (mainly concrete and metal alloys).

The general conclusion to be drawn from available data on the reaction of bitumen with these substances at the prevailing storage temperature and pressure, and during the envisaged times is that the bitumen properties of importance are mainly affected at and near the surface and are not influenced in a way which would endanger its function as protective coating material.

Besides the already mentioned ageing effects the information given hereafter might be useful in the assessment of the long-term stability of bitumen and bitumen-waste products (BWPs).

The degree of impermeabilization obtained by bitumen coatings depends on:

- the compactness of the bitumen coating layer,
- the rate of diffusion of water (vapour) and air through the bitumen, and
- the nature of the coated materials.

There are two reasons why only small amounts of water will pass through a homogeneous bitumen layer:

- firstly, the solubility of water in the bitumen is low (in the order of 0.001 - 0.01%),
- secondly, the viscosity of the bitumen is high at normal ambient temperatures.

If the bitumen contains drops of a salt solution or water-attracting particles in considerable quantities, the water permeability can be substantially increased.

The diffusion constants of bitumen for water and air (oxygen) is of the order of 10^{-8} and 10^{-10} g . cm^{-1} . h^{-1} . $mmHg^{-1}$, respectively.

One can calculate that even thin tight layers (~ 0.5 mm) provide an adequate barrier against water and air in the protection of materials against corrosion and water attack.

The effectiveness of the protection depends on the absence of hydrophilic materials in the bitumen and of pinholes and cracks through which water could penetrate.

The lower the penetration of the bitumen, the slower the diffusion of water through it; distillation bitumens are less permeable to water than blown bitumens of the same penetration.

A 5 mm thick layer of blown bitumen after 10 years contact with tap-water will absorb 3 - 4% water and a hard residual bitumen will absorb less than 1% water.

The ability of bitumen to resist attacks by acidic, alkaline, and salt solutions let assume that the bitumen will show a high resistance against the (usually) less agressive aqueous solutions and the various solid materials occurring in nature and in the waste repository.

3.4 Microbial Degradation

In 1972 Drent /105/ gave a literature review on the "Effects of Microorganisms on Bituminous Materials".

Recently a "Survey of Microbial Degradation of Asphalts with Notes on Relationship to Nuclear Waste Management" has been published by ZoBell and Molecke /275/. It might therefore suffice to cite some important information only and the conclusions contained in the latter publication.

It seems that virtually all kinds of bitumen are biodegradable by bacteria and fungi at rates varying from extremely slow to fairly rapid. The rates of biodegradation are influenced by the contact of microbial enzyme systems with the bitumen, the chemical composition of the bitumen, its physical state, and environmental conditions.

The waste bituminization temperatures in the range of 80 - 200°C are high enough to kill most bacteria and fungi contained in the waste itself. However, bacterial degradation of the bitumen waste from external sources is still quite possible.

Most soil and aquatic bacteria, including asphalt oxidizers, grow best within the range of pH 6 and 8.

Even under the most favourable conditions for the microbial degradation of bitumens, the rate is generally very slow.

The rate of degradation appears to be directly proportional to the surface area exposed to oxygenated water per unit weight of asphalt. The biodegradation of asphaltenes and other hydrocarbons by a Pseudomonas Species was found /R-9/ to be promoted by bioemulsifiers or surfactants.

Pseudomonas asphaltenicus, a new species recently isolated from asphalt deposits in Ontario (Canada), utilizes a wide range of hydrocarbons, including asphaltenes. The highest temperature at which microbial activity occurs is about 100°C.

Relatively few microbial species survive prolonged exposure at temperatures higher than 60°C.

Hyperbaric oxygen, carbon dioxide, and certain other gases are bacteriostatic or bactericidal at pressures of only 5 to 10 atm. It has not been determined whether asphalt (bitumen) is attacked by microorganisms in the absence of free oxygen.

If certain anaerobes attack bitumen, the rate is much slower than aerobic degradation.

It was shown by Harris /R-10/ that certain physical properties of paving asphalts were altered by microbial activity at ambient temperatures within a few weeks; later he noted /R-11/ a steady increase in the microbial population in soil surrounding asphalt coatings on buried pipes and essentially no microbial growth around coal-tar coatings. Mainly pseudomonads, corynebacteria, and actinomycetes were involved in the destruction of asphalt.

Depending on the kinds of bacteria present and various environmental conditions, the paraffin fractions of asphalts seems to be attacked more rapidly than the heterocyclic compounds of asphaltenes. If so, destruction of paraffins may render the remaining fractions more vulnerable to enzymatic and chemical action.

In view of using bitumen as embedding material for radioactive wastes the following statements or conclusions - derived from the relevant literature can be made:

- microorganisms, widely present in nature, are capable of attacking bitumen under aerobic (and most probably also anaerobic) conditions;

- in general, microbial attack seems to be fastest for steam-refined bitumens, followed by air-blown and finally coal tar pitches;
- little is known about the use and effectiveness of products that inhibit microbial action;
- many of the components of bitumens, such as asphaltenes, resins, and paraffinic, naphthenic, and aromatic hydrocarbons can be more resistant to chemical reagents, such as strong acids and alkalines, than to microbial enzyme systems;
- the few species attacking bitumen are most abundant in soil or bottom sediments that have been in contact with crude oil or bitumen;
- the speed at which microbes attack asphalt is very slow (under optimum growth conditions only about 0.025 mm thickness of bitumen was found to be affected in 3 years) /R-12/,
- the conditions have to be perfect for a very long time to produce any noticeable damage;
- the possibility that microorganisms destroy the bitumen coating (of radwaste) is extremely small.

3.5 Conclusions

The application of bitumen in nuclear and non-nuclear fields is based on its favourable physico-chemical properties and its long-term durability.

Various bitumens may show differences in this respect, dependent on origin and manner of production, particularly, with respect to the oxidation stability.

The demonstrated natural stability of bitumen over millions of years in an underground environment must be regarded as a real advantage over man-made materials.

4. WASTE CONSIDERED FOR BITUMINIZATION

4.1 General

For the bituminization only those wastes are considered here which:

- are generated at light-water reactor stations and nuclear fuel reprocessing plants (wastes from other nuclear activities are therewith included as they do not pose new problems and do not require other waste management techniques),
- are inorganic or organic solutions or suspensions or finely devided solids, requiring solidification and/or fixation prior to their final storage,
- belong to the low- or intermediate-level radioactive waste categories, i.e. having a specific activity in the unconditioned as well as conditioned form of less than 10 Ci/dm^3 (in practice normally less than 1 Ci/dm^3 = 3.7 x 10^{10} Bq/dm^3).

Besides the bituminization of the "wet" wastes, also the coating of certain "dry" wastes by bitumen can be considered.

To place the "bituminizable wastes" in the correct perspective also types and amounts of wastes are mentioned which are conditioned for storage by other means than bituminization.

4.2 Reactor Wastes

4.2.1 Origin

Most of the radionuclides in liquid waste effluents at LWRs originate from the primary coolant or from the fuel storage pond. Four main processes contribute to the radioactivity in the reactor primary coolant /R-13/.

- activation of the coolant and its impurities,
- corrosion of activated reactor components,
- contamination of the fuel cladding with fissile material,
- leakage of radionuclides from defective fuel rods.

The quantity of fission products in reactor coolants depends on the quality of the fuel elements.

The quantity of activation products present depends upon the degree of interaction between the coolant and reactor components.

Radionuclides in the water of the fuel storage pond originate from defective fuel rods and corrosion of the fuel elements.

Approximately equal contributions to the total volume are made by the treated "wet" and "dry" wastes, but most of the activity (up to several thousand Curies) is associated with the "wet" wastes.

A significantly greater volume reduction could be achieved if combustible wastes were treated by incineration.

Radio-cesium is the main activity contributor within ion-exchange resins after a storage time of 20 - 40 years. Typically, about 150 - 500 Ci of Cs-137, < 1 Ci of Sr-90, and 5 Ci of Co-60 are found in the evaporator concentrates accumulated during one year at a PWR station.

The oil waste may contain small amounts of tritium, cesium or corrosion products.

In pressurized water reactors (PWRs) the reactor coolant usually contains 20 - 40 cm^3 of hydrogen per kilogramme of water to inhibit the radiolytic decomposition of the coolant. The control of pH may be done by using lithium-7 hydroxide, or potassium hydroxide (Soviet-type VVER-440 PWRs).

The control of long-term changes in reactivity of the core during plant operation and for cold shutdown reactivity control is effected by means of boric acid as the soluble neutron poison.

Under normal operation conditions the radioactive contaminants of the waste residues are activated corrosion products and tritium (produced by activation of coolant additives, naturally occuring deuterium in the coolant, and ternary fission of U-235).

Under normal operation of a boiling water reactor and with 100% fuel cladding integrity the main (non-gasesous) radionuclides present in the solid waste residues and liquid concentrates are: Cr-51, Mn-54, Fe-55, Co-58, Co-60, Zn-65, Zr-95, Sb-124, i.e. activated metals from the corrosion of the materials used in the primary system, turbine and feed-water plant.

The leakage of radionuclides from the fuel to the coolant influences more the activity level than the amount of the wastes.

The largest fraction of ion-exchange resins from a BWR belongs to the low-level waste category. The largest amount of radioactivity is contained in about 30% of the resins /R-14/.

In Tables XVI to XVIII some further data on reactor waste amounts and constituents are given as examples.

TABLE XIII

APPROXIMATE ANNUAL "WET" WASTE PRODUCTION BY A 900 MWe - BWR

Type of Waste	Volume m^3/a	Activity Ci/m^3
Ion exchange resins:		
- Granular form (reactor water clean-up)	15	< 50
- Granular form (condensate clean-up)	7	< 1
- Powder form (condensate clean-up)	50	< 10
- Powder form (fuel storage pond water clean-up)	2	< 50
Filtration sludges	50	< 10
Evaporation concentrates*)	20	< 1

*) At some BWR plants evaporation is seldom used.

TABLE XIV

APPROXIMATE ANNUAL "WET" WASTE PRODUCTION BY A 900 MWe - PWR

Type of waste	Volume m^3/a	Activity Ci/m^3
Ion exchange resins:		
- Granular form (primary loop)	6	$10^2 - 10^3$
- Granular form (secondary systems)	15	< 1
Effluent concentrates	25	< 2
Boron effluent concentr.	30	< 2
(Filters, solid	6	$10^2 - 10^3$)

TABLE XV

APPROXIMATE ANNUAL VOLUMES OF SOLID RADIOACTIVE WASTES FROM A 1000 MW_e - LWR

Type of waste	BWR		PWR		Treatment Applied
	Before treatm. (m^3)	After treatm. (m^3)	Before treatm. (m^3)	After treatm. (m^3)	
"Wet" wastes	130	65	80	40	Bituminization
Dry compressible	400	100	250	60	Compression
Total (non-compressible wastes excluded)	530	165	330	100	Compression + Bituminization

TABLE XVI

WASTES FROM TWO 590 MW_e - BWRs CONDITIONED BY BITUMINIZATION AT BARSEBÄCK /277/

Waste category	Origin	Activity concentration	Amount per year	Resulting drums per year
Powered resins	Condensate polishing	low	15 tons	250
Bead resins	Reactor water clean-up	medium	10 m^3	120
Powdered resins	Fuel pool clean-up	medium	0.7 ton	10
Mechanical filter aids	Rad waste system	low-medium	1.5 ton	20
Concentrates	decont. chemicals etc.	medium	very small volumes *)	5

*) These small volumes are due to the fact that no demineralizer system is used.

TABLE XVII

CORROSION AND FISSION PRODUCTS IN THE PRIMARY WATER

CORROSION PRODUCTS		FISSION PRODUCTS	
Nuclide	Halflife	Nuclide	Halflife
Cr-51	27.8 d	Sr-89	52 d
Mn-54	303 d	Sr-90	28.1 y
Mn-56	2.58 h	Mo-99	66.7 h
Co-58	71.3 d	I-131	8.07 d
Co-60	5.26 y	I-132	2.3 h
Ni-59	7.5×10^4 y	I-133	20.9 h
Ni-63	92 y	I-134	52 m
Fe-59	45.1 d	I-135	6.7 h
Cu-64	12.9 h	Te-132	78 h
Zn-65	243.6 d	Te-134	42 m
W-187	24 h	Cs-134	2.05 y
		Cs-135	3×10^6 y
		Cs-136	13 d
		Cs-137	30.2 y
		Cs-138	32.2 m

TABLE XVIII

DISTRIBUTION OF RADIONUCLIDES (%) IN AN EVAPORATION CONCENTRATE OF A 670 MWe - BWR /R-15/

Nuclide	March	after				
	1976	7 months	10 months	2 years	3 years	4 years
Cs-137	6.8	8.4	8.0	10.0	11.6	15.4
Co-60	65.4	72.2	76.7	82.5	83.0	80.8
Cs-134	3.1	2.8	2.5	2.8	2.1	1.8
Mn-54	2.0	1.7	1.6	0.9	0.6	-
Zn-65	17.6	13.8	11.0	3.9	2.6	2.0
Co-58	5.0	1.2	0.5	-	-	-
Activity Ci/m^3	0.2	0.17	0.16	0.12	0.11	0.08

4.3 Reprocessing Wastes

4.3.1 Origin

Present-day fuel reprocessing plants apply almost exclusively Purex-type processes - which are characterized by the use of tributyl phosphate (TBP) solutions as extractant - to separate uranium and plutonium from fission products and other impurities. The reprocessing operations generate a large variety of gaseous, liquid, and solid wastes.

The origin of the various categories of waste in a Purex-plant using a chop-leach head-end is shown in Table XIX.

If a plant would apply a chemical decanning of the fuel elements (as e.g. the Eurochemic plant from 1966 - 1974) then the resulting large volumes of decanning solutions would belong to the intermediate-level waste category.

4.3.2 Amounts and composition

In Table XX the approximate specific waste arisings for the various types of reprocessing wastes are given.

As only the intermediate- and low-level waste solutions (besides filter aids and ion exchange resins) are considered for bituminization, the composition of other waste types will not be discussed.

It is difficult to quote a specific production rate of LLW and MLW solutions (in m^3/t fuel material processed) as their amounts generated are not directly linked to the through-put of the plant.

a) Intermediate-level waste

The ILW production rate may vary in certain periods from 1 - 40 m^3/t fuel treated. The main reason for this wide range is that a large fraction of the decontamination waste belongs to the ILW category. The liquid ILW, resulting from normal process operations consist mainly of alkaline sodium carbonate solutions or acidic sodium nitrate solutions, which may furthermore contain aluminium, iron, potassium, fluoride, and sulfate. Their activity level is generally in the order of 10 - 100 mCi/l. Typically, a ILW solution contains 400 g sodium nitrate per litre and 2 moles nitric acid per litre. The main radionuclides present in ILW are Zr/Nb-95, Ru-106, small amounts of other fission products, uranium, and plutonium. A wide variety of alkaline and acidic medium active decontamination waste is generated containing oxidizing or reducing agents, organic and inorganic complexing agents and other reagents such as surface-active compounds (detergents) and corrosion inhibitors.

All these solutions are usually mixed as far as compatible with their subsequent treatment or they are stored separately.

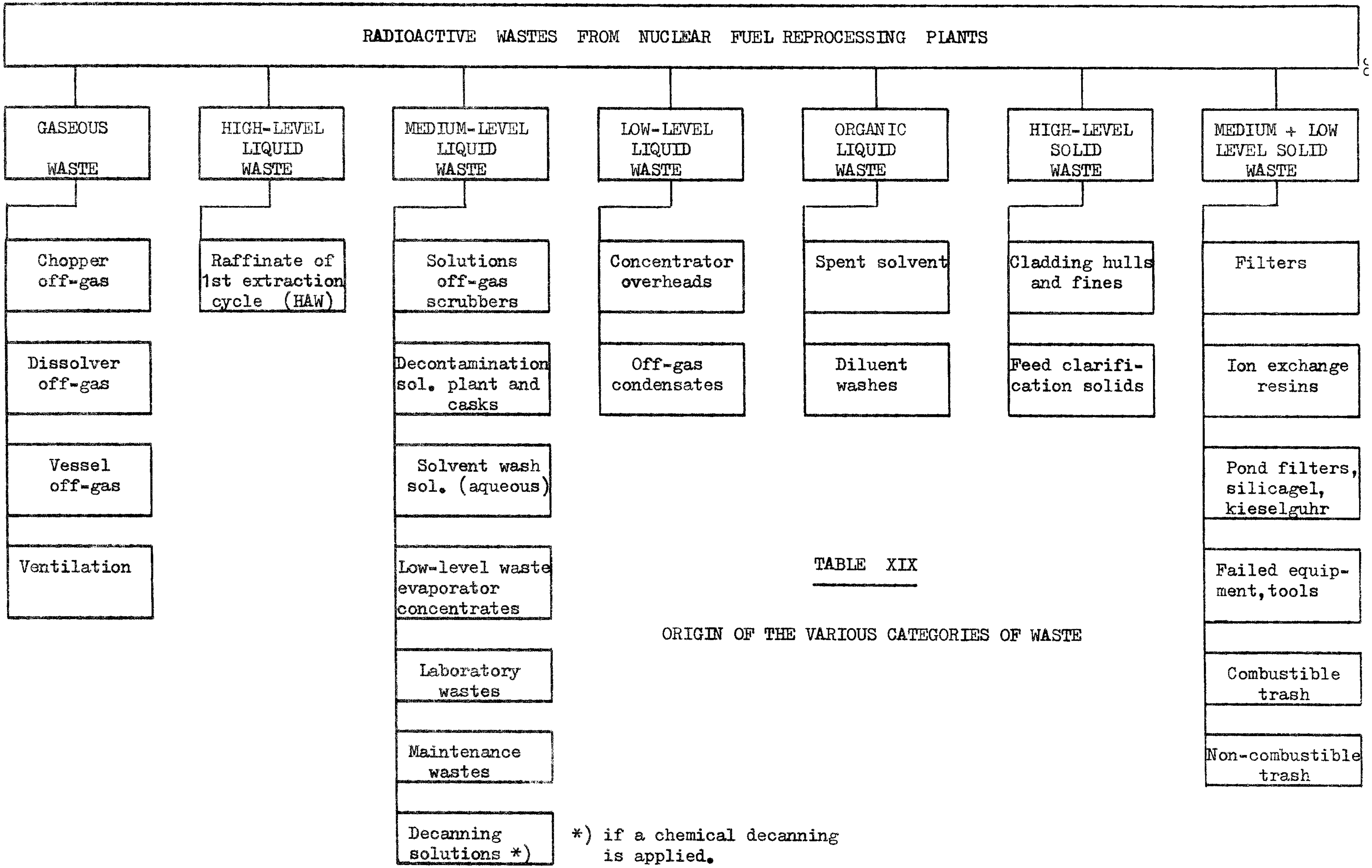


TABLE XIX

ORIGIN OF THE VARIOUS CATEGORIES OF WASTE

*) if a chemical decanning is applied.

TABLE XX

APPROXIMATE SPECIFIC WASTE PRODUCTION OF THE VARIOUS WASTE TYPES
(1000 MWe LWR 33000 MWd/t burn-up)

WASTE TYPE	WASTE VOLUME (m^3/t fuel)	SPECIFIC ACTIVITY (Ci/m^3 waste)
GASES	depends on ventilation	depends on ventilation
LIQUIDS		
(* after concentration)		
High-level* (> 10^4 Ci/m^3)	0.7 - 1.0	3 x 10^6 - 3 x 10^5
Intermediate-level* (10^{-2} - 10^4 Ci/m^3)	1 - 4	10^1 - 10^3
Low-level (< 10^{-2} Ci/m^3)	40 - 100	< 10^{-1}
Organic solvent	0.5	10^1 - 10^2
SOLIDS		
(before treatment)		
Canning hulls	0.5	> 10^3
Combustible	4	< 1
Non-combustible	1	10^{-1} - 10^3

These primary wastes are treated thereby producing normally two waste fractions. One fraction contains a high percentage of the majority of the radionuclides in a small volume whilst the other fraction will be of such a low specific activity that it can be safely disposed of into the environment. The radionuclides are concentrated by different methods according to the activity level and chemical composition of the solutions.

The methods mostly used are:

- chemical precipitation,
- ion-exchange processes, and
- evaporation

According to the technique applied the radioactivity is concentrated in the precipitates (sludges), or on the ion exchange material, or in the regeneration solution, or in the evaporator concentrate. These concentrates can be incorporated into bitumen or another suitable matrix material.

As an example, Table XXI gives the estimated composition of an intermediate-level liquid waste after evaporation generated at a commercial 1500 t U/year plant (Allied-General Nuclear Services at Barnwell, SC, USA) /R-16/.

b) Low-level waste

It is equally difficult to state a relatively precise specific production rate for the LLW arisings, however, the indicated upper limit (Table XX) of 100 m^3/t of fuel processed will normally not be exceeded.

The low-level liquid wastes from fuel reprocessing are chemically not greatly different from natural waters. They contain only very small amounts of inert chemicals and radionuclides.

The radionuclides of greatest importance in these wastes are above all tritium, then ruthenium-106, cesium-137, and strontium-90.

These wastes are very large in volume. Evaporator condensates alone may average 40 m^3/t of fuel processed. Because of their great volume and low concentrations of radionuclides, the LLWs have been suitable for environmental disposal.

If the LLW solutions have not the required purity for safe discharge to the environment they have first to be sufficiently decontaminated. Normally distillation is applied, which produces extremely pure distillates. Also co-precipitation processes are employed in which the solid residues - after their separation by filtration - can be incorporated into bitumen.

TABLE XXI

ESTIMATED COMPOSITION OF LIQUID ILW AFTER EVAPORATION /R-16/

Cations	
Component	Conc. (M)
H^+	2.5
Na^+	1.1
UO_2^{2+}	0.037
Hg^{2+}	0.023
Mn^{2+}	0.009
K^+	0.005
Fe^{3+}	0.003
Ag^+	0.003
Zr^{4+}	0.001
Ru^{4+}	0.0006
Cs^+	0.0001
Sr^{2+}	0.0001

Anions	
Component	Conc. (M)
NO_3^-	2.74
$NO_2^{-*)}$	0.26
PO_4^{3-}	0.24
Cl^-	0.016
SO_4^{2-}	0.009
I^-	0.007

Cleaners	
Component	Wt.-%
Soap	0.5
Detergents	0.5
Na_2 EDTA	0.5
Na_2 Citrate	0.5
Na_2 Tartrate	0.5
Na_2 Oxalate	0.5

*) NO_2^- is not stable in this solution.

4.4 Conclusions

The operation of a 1000 MW_e LWR generates yearly approximately the following low- or intermediate-level wastes which can be bituminized:

- 35 tons of ion exchange resins and filter aids of an average specific activity of < 100 mCi/kg,

- 3 tons of dry matters contained in evaporator concentrates.

These solids are contained in a total "liquid volume" (i.e. solution or suspension volume) of about 100 m^3 which bituminized would result in approximately 75 m^3 bitumen-waste products 60 wt.-% bitumen/40 wt.-% solids) or 375 product drums (containing 200 l BWP/drum).

The reprocessing of fuel unloaded yearly from a 1000 MW_e LWR (ca 29 tons) generates approximately:

- 75 m^3 ILW solutions containing about 400 g salts per litre and an activity of < 1 Ci/l and

  2000 m^3 LLW solutions containing small amounts of salts and an activity of < 10 µCi/l.

The bituminization of the ILW solutions and the sludges or concentrates from the treatment of the LLW solutions results in the formation of 80 tons or 60 m^3 bitumen-waste products (60 wt.-% bitumen/40 wt.-% solids). The 60 m^3 BWPs are usually filled in 180-litre-portions into 220 product drums which results in the generation of about 330 drums. The bituminization of ILW generated in a large reprocessing plant will result in a specific bitumen-waste volume of about 2 m^3/t U processed corresponding to 11 BWP drums per ton uranium processed.

One can conclude that the operation of a 1000 MW_e LWR and the reprocessing of the unloaded fuel per year generates in total about 700 BWP-drums requiring storage or disposal.

5. BITUMINIZATION OF REACTOR AND REPROCESSING WASTES

5.1 General

The solid and initially liquid wastes of the low- and intermediate-level radioactive waste categories from reactor stations and fuel reprocessing plants can principally be bituminized, i.e. incorporated in or coated by bitumen.

Thus, the bituminization is relatively insensitive to the type of waste being processed; it may be operated either as a continuous or a batch process.

The bituminization of liquid waste can be classified as an evaporation mixing process; the bituminization of dry solids is a mixing and/or coating process.

The temperature of the bitumen-waste mixture during the incorporation process does normally not exceed 200°C.

Bitumen-waste products containing up to 80% solids could be prepared. Usually a product containing 40-60 wt.-% solids is chosen as it represents the best compromise of the conflicting requirements of volume reduction, viscosity, ductility, and the flow (casting) temperature.

The aim of the bituminization of liquid and solid wastes is to condition the various wastes in such a way as to obtain a final solid product suited for long-term storage or disposal.

5.2 Process Techniques

For the incorporation of solid and initially liquid wastes three different bituminization techniques have been developed:

1. Mixing of the solid or liquid wastes with molten bitumen at a temperature of 140 - 230°C under simultaneous evaporation of water and casting of the fluid mixture into containers (drums) for storage after cooling down to a solid product.

2. Mixing of the solid or liquid wastes with emulsified bitumen at room temperature and subsequent heating of the mixture obtained to evaporate water; the remaining mixture is filled into containers, cooled down, and stored.

3. Mixing of waste sludges, surface-active agents (for emulsifying), and bitumen to obtain in the first stage a preliminary coating and simultaneously a partial removal of water; in the second stage the final coating of the mineral salts and other solids by bitumen and the removal of water are achieved by heating the mixture to about 130°C. The solids-bitumen mixture is filled into containers, cooled down, and stored.

These three types of incorporation processes can be carried out batchwise or in a continuous manner.

The bituminization of radwastes (especially from reprocessing) by the "sedimentation technique" /227/ can hardly be recommended as the operational principle involved does not assure the required quality of the final product.

5.3 Process Equipment

The bituminization of wastes requires mainly reaction vessels, evaporators, and mechanical mixers; in addition condensers, off-gas clean-up systems, and condensate clean-up systems (e.g. filters for the removal of bituminous oils) are usually needed.

Three types of heating-mixing equipment have successfully been used on an industrial scale:

1. A "pot" provided with a heating mantle or heating elements (inside) and a mechanical stirrer for batch processes.
2. Two - and four - screws - extruder evaporators (diameter of the elements e.g. 83 or 120 mm) effecting continuously homogeneous mixing of the solid waste components with bitumen under simultaneous evaporation of water (and other volatile components) and extruding of the waste-bitumen mixture in product containers.
3. Wiped-film (or thin-film), evaporators permitting also in a continuous way the mixing of the solid residue with bitumen, after evaporation of the water, along the heated evaporator walls and outflow (or pumping out) of the resulting mixture into the product containers.

5.4 Final Product Container

Design and material of the BWP container are first of all of importance for easy and safe filling, handling, transporting, and interim-storing of the entire final product.

In most safety analysis made in the past it has been the tendency to give no credit to the container as a barrier against the dispersion of radionuclides from the waste products to the environment in order to base the analysis on "conservative assumptions". Such a view might be justified in cases where the expected life-time (corrosion resistance) of the container in the forseen storage environment is negligibly short compared to the time the contained nuclides remain radiotoxically harmful, e.g. a relatively thin, steel cylinder filled with vitrified high-level waste from fuel reprocessing placed at a great depth in a bore hole or a salt deposit.

In view of a prolonged interim-storage of bituminized waste, its subsequent handling and transport to the final repository, the design and material of the product container could be of such a quality as to assure dimensional stability, intactness of the container, and a real diffusion barrier over a significant length of time. Whether such an advanced product container is safety-technically desirable and economically defendable or not, depends on the quality and composition of the contained product and on the strategy, technique, and place selected for its interim- and final storage.

At present, the most widely used containers for BWPs are mild steel drums of 170 - 220 l total volume protected against atmospheric corrosion by mineral paints.

In Czechoslovakia even paper drums were used, however, only for the purpose of transport from the place of origin to the place of burial /216, p. 204/.

At Eurochemic, studies have been carried out which aimed either at an improvement of the corrosion resistance of normal mild steel drums by protective coating materials (such as special paints, bitumen, plastics, silicon oil etc..) or the selection of a suitable, relatively inexpensive container material more corrosion-resistant than painted mild steel. As a result of these studies a new type of steel plates, recently developed by Cockerill-Ougré (Belgium), has been selected for manufacturing product drums suited for the engineered surface storage of bitumen-waste mixtures for at least 50 years.

This new plate material is called "chromized steel" (tôle chromisée); it possesses on both sides an 70 - 110 µ thick surface layer with the properties of a 20% chromium containing ferritic stainless steel. The chromized steel plates can be welded by all conventional processes. If the price of a standard painted mild steel drum (60 cm diameter, 90 cm height, 1.2 mm steel plate thickness) is taken as unity, then a drum of identical design and dimensions made of:

- galvanized steel costs 1.6,

- chromized steel costs 2.1, and

- stainless steel (type 18/8) costs 10 times as much (in 1979-prices).

It was concluded that the drums made of chromized steel represent the best compromise between required properties and price.

Fig 2 is a construction drawing of the drum used at Eurochemic for BWPs.

If considered necessary the design of future product containers could take into consideration an improved mechanical stability and a double wall with an appropriate material between inner and outer wall, to increase the number of barriers and to improve at the same time the corrosion-, ignite-, leach-, and mechanical resistance.

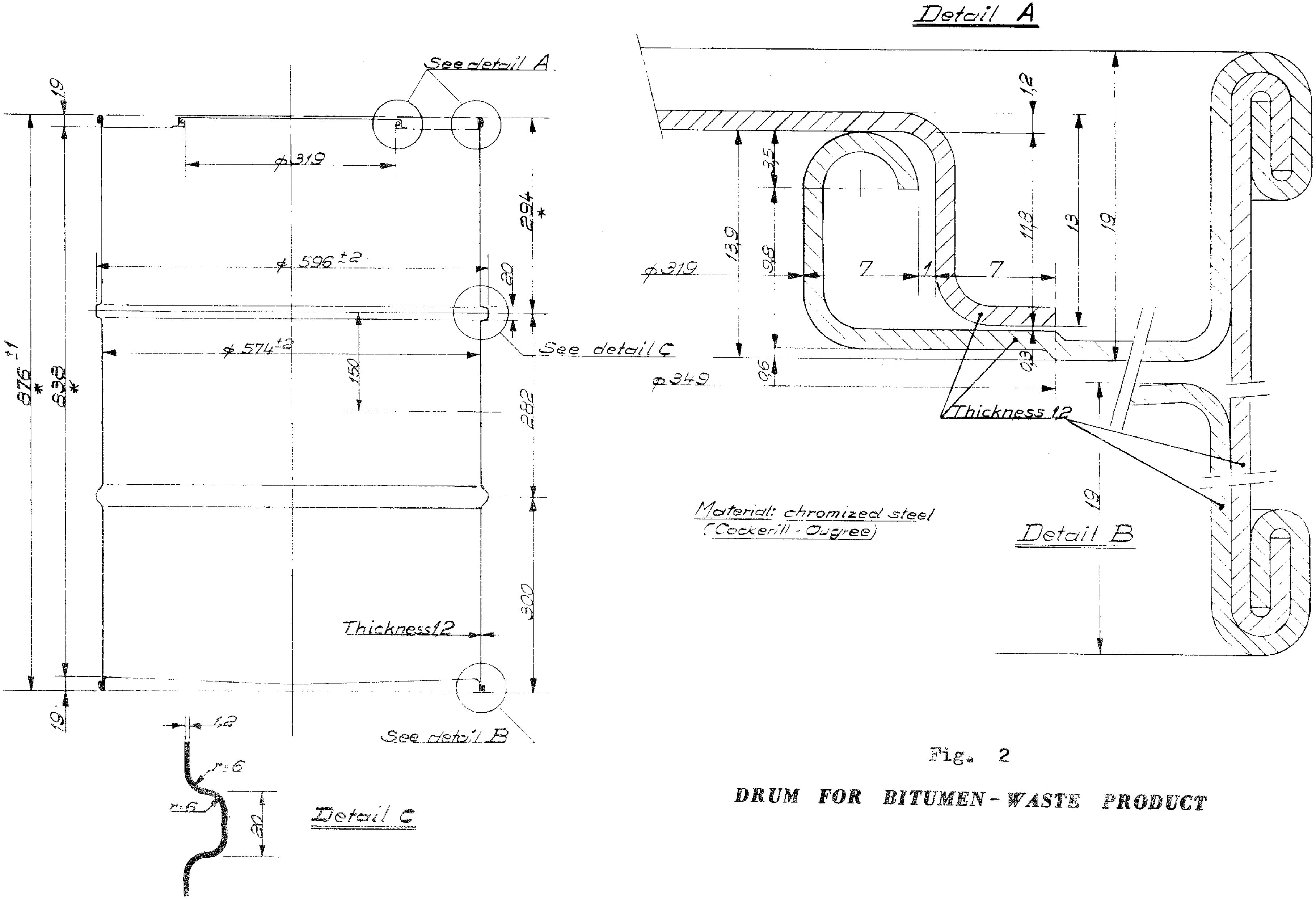


Fig. 2

DRUM FOR BITUMEN - WASTE PRODUCT

5.5 Factors Influencing Final Product Quality

The amount and nature of salts or other waste constituents mixed with molten bitumen may effect chemical and physical changements of components of the mixture and consequently the quality of the resulting solid product.

Certain salts tend to agglomerate or to settle (due to their higher density) causing inhomogeneous products especially if "pot type" coating equipment is used; to these salts belong: sodium iodide, sodium chloride, barium sulfate, calcium nitrate.

Other salts react with the bitumen causing a hardening of the product and sometimes an increased leachability; to these salts belong: sodium metaborate, calcium metaborate, sodium orthophosphate, sodium nitrate, magnesium chloride, magnesium sulfate, and aluminium sulfate.

To the inert, easily mixable salts belong: calcium phosphate, calcium sulfate, calcium carbonate, sodium carbonate, sodium bicarbonate, and sodium sulfate.

During the incorporation of mixed bed ion-exchange resins, a thermal degradation of the basic anion exchanger fraction takes place at temperatures above 100°C. The main gaseous degradation product is trimethylamine, which has an autoignition temperature of about 180°C.

To reduce the deliberation of the amine, the bituminization of anion exchange resins should take place at the lowest possible temperature.

Using a screw extruder the temperature profile should preferably be decreasing from the waste inlet to the outlet, e.g. from 160°C to 120°C.

To obtain waste solutions, concentrates, slurries, and other liquid materials suited for incorporation into bitumen and to assure that the resulting BWP has satisfactory safety-relevant properties, the following pre-treatments and conditions should be taken into consideration:

- the aqueous phase, if acidic, has (except in very few cases) to be neutralized or made alkaline (pH 7 - 10) prior to bituminization;

- as the precipitates, resulting from the alkalinization (mainly hydroxides) are usually not effective enough to insolubilize the most important fission products strontium-90 and cesium-137, and some other long-lived radionuclides, specific insolubilization procedures (using precipitations or sorption on pre-formed predipitates) should be applied as e.g.:

  o coprecipitation of Sr by barium sulfate, or calcium carbonate,

  o coprecipitation of Cs by nickel ferrocyanide,

  o scavenging of Ru by a mixture of Cu and Fe (II) hydroxides at pH ≈ 8 /139 a/, o anions forming water soluble salts and which can bind more than two molecules of water should be converted in less soluble or insoluble compounds to reduce the leach rate and the risk of swelling of the product when in contact with water.

Further measures may effect the product properties in the following way:

- the addition of emulsifying agents (e.g. 4 - 6%) lowers the inflammability temperature, but aids in the preparation of easily castable, homogeneous products by lowering the viscosity of the hot product mixture /280/.
- the incorporation of wet ion-exchange resins at higher temperatures (> 150°C) can result in excessive foaming and may cause products of higher porosity;
- increased incorporation temperatures (> 135°C) may lower the leach rate of waste constituents if they can react for a longer time with components of the bitumen (e.g. Sr);
- an incorporation temperature of 190 - 200°C effects under normal hold-up times in the coating equipment that the bitumen-salt mixtures retain less than 1 wt.-% water (usually less than 0.5 wt.-% water);
- the simultaneous incorporation of ion-exchange resins and evaporator concentrates of high salt contents diminishes the fixation of radionuclides by the resins and favours thereby a higher leach rate (inert materials and aditives improving the immobilization of the radionuclides can be bituminized together with ion-exchange resins);
- the incorporation of ion exchange resins (< 50 wt.-%), salts, minerals, and other fillers increase the form-stability of the BWPs during storage;
- the simultaneous presence of more than about 1% of unhydrolysed Fe (III)-salts and sodium nitrate and/or nitrite lowers the thermostability of bitumen and leads to a considerable acceleration of the incineration /227/;
- the incorporation of larger amounts of hygroscopic substances and more than 50 wt.-% ion exchange resins can lead to a considerable swelling of the BWP in contact with water;
- mixing of higher amounts of sodium nitrate (> 15 wt.-%) with bitumen at temperatures higher than about 220°C leads to strongly hardened (oxidized), incastable mixtures;
- bituminization of strongly alkaline (pH > 13) sodium nitrate solutions can result in hard products of relatively low ignition temperature and higher leach rates.

One could assume that the slow cooling rate of just cast fluid bitumen-waste mixtures (see Fig. 3) could cause inhomogeneities (e.g. by sedimentation) in products of industrial size.

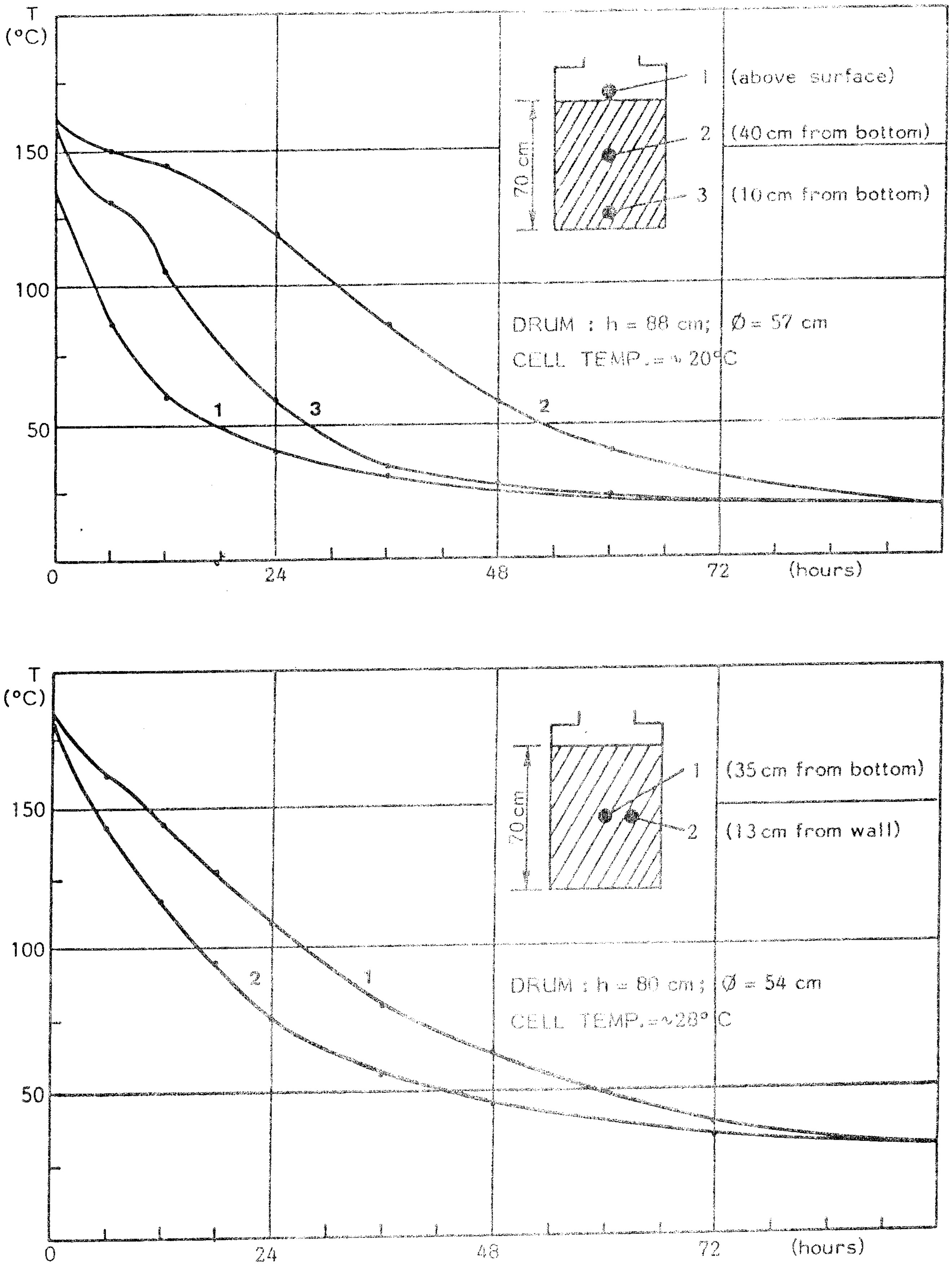


Fig. 3 - COOLING CURVES OF BITUMEN WASTE PRODUCTS

BWPs : 55 % bitumen/45 % $NaNO_3$

Cooling rate in the middle : ≈ 2 °C/h

It has been experimentally verified at Eurochemic that under normal operation conditions an average cooling rate of about 2°C/h from the casting temperature down to near cell temperature did not effect the homogeneity of BWPs containing 50 to 55 wt.-% bitumen (Mexphalt 10/20 and Mexphalt R85/40) and a wide variety of salt mixtures.

As the sedimentation rate depends mainly on the particle size, shape, densities and the viscosity (temperature) of the bitumen, significant differences in the settling rate and homogeneity may occur under certain process conditions.

Worth mentioning is an experiment in which a considerable settling of active ion exchange particles was measured (indicated by an almost double as high activity at the bottom part than at the top part of a "settling tube" placed within the product in the drum) using the normal bituminization conditions /280/; it has been assumed that the relatively high residual fraction of large resin particles remaining in the otherwise finely ground ion exchange material has caused the remarkable sedimentation.

Unfortunately, neither the filling time and temperature nor the cooling rate has been indicated to permit an interpretation of the experimental result. However, also studies carried out in Finland /R-31/ have shown that considerable sedimentations of ion exchange resins in granular form can occur while the sedimentation of resins in powder form under the same bituminization conditions are insignificant.

5.6 Safety Risks During Bituminization

5.6.1 General

The incorporation of waste residues into hot bitumen necessitates to consider two important safety risks: an ignition and an explosion.

The combustibility of bitumen (as well as other organic material) is considered as the main drawback for its application as encapsulation material for radioactive wastes. As the incorporation of certain types of waste into bitumen may not only involve the danger of ignition but also explosion-like reactions much attention has been paid to the safety of the bituminization process.

The waste concentrates from reprocessing plants (and nuclear research centres) can be identified as the critical type of waste in view of fire and explosion risks, especially those containing nitrates and thermally instable or volatile organic compounds. The main component of ILW solutions from fuel reprocessing is sodium nitrate which could oxidize the bitumen under the generation of heat, gas and pressure; such a reaction may damage the bituminization equipment. Therefore, the thermal conditions and the concentration of reactive salts are of prime importance for a safe performance of the bituminization process. The operation conditions have to be chosen in such a way that exothermal reactions of an explosive nature are impossible and that volatile organic substances do not accumulate to form inflammable or explosive mixtures.

Normal bitumen is not classified as an inflammable substance. It has a flash point ranging from ~ 200 - 320°C depending on the origin and nature of the bitumen. Therefore the major fire hazards during the operation stage are basically due to sparks (or other ignition sources) and local overheating.

5.6.2 Investigations and results

Investigations on the hazards caused by incorporating sodium nitrate and sodium nitrite into bitumen /76, 193/ have shown that significant exothermic reactions for all investigated compositions occurred exclusively in the temperature range from 390 - 430°C (i.e. beyond the melting points of $NaNO_3$ and $NaNO_2$). The thermal stability of bitumen/salt mixtures diminishes if the $NaNO_3$ and/or $NaNO_2$ content is increased and the content of bitumen and/or insoluble solids is decreased.

A great number of experiments were performed to establish ever-safe incorporation conditions using Mexphalt R 85/40 and varying proportions of soluble ($NaNO_3$ and $NaNO_2$) and insoluble salts. The elementary analysis of the bitumen type Mexphalt R 85/40 indicated the following composition: 84.0% C, 11.0% H, 2.7% S, and 2.3% O.Thus, the binary mixture of 13.13% Mexphalt R 85/40 and 86.87% $NaNO_3$ contains the theoretical amount of oxygen which is necessary for a complete combustion of the bitumen considered.

This mixture showed no remarkable exothermic reactions until 315°C (fusion of $NaNO_3$) and an "explosion" temperature above 400°C.

It could be demonstrated that other mixtures, though oxygen deficient, are more "dangerous" than the 13/87 %-mixture.

A sample containing 13.13% Mexphalt R 85/40, 41.94% $NaNO_3$, 16.47% $NaNO_2$ and 28.46% insoluble solids showed exothermic reactions as from 250°C and the formation of an explosion-like reaction at about 350°C.

It is evident that such extreme bitumen/salt ratios are unlikely to occur under well-controlled operation conditions.

Differential thermogrammes obtained on a great number of differently composed bitumen/salt mixtures (40-50% blown bitumen/$NaNO_3$ + $NaNO_2$ + water-insoluble salts) revealed that no exothermic reaction takes place below 295°C.

From the investigations performed one could draw the following main conclusions /193/:

- there exists no risk of a spontaneous exothermic reaction during the incorporation of nitrate and nitrite containing waste slurries into blown bitumen (Mexphalt R 90/40 or R 85/40) provided that the bitumen/salt mixture contains at least 40.wt.-% bitumen, that the different salt constituents are homogeneously dispersed and that the temperature at any point of the mixture is kept below 280°C;

- if the homogeneous dispersion of the salt components cannot be assured, a minimum content of 50 wt.-% bitumen is required and the temperature at any point of the mixture should not exceed 230°C in order to avoid hazardous exothermic reactions.

Investigations carried out in the Sovjet Union have shown /273/ that sodium nitrate and bitumen interact at all ratios when a certain temperature is reached; using up to 70% sodium nitrate the lowest interaction temperature was found to be 350°C. In the presence of other components, e.g. boron compounds, iron and manganese hydroxides, alkalis, the nitrate-bitumen interaction temperature drops only in the presence of alkalis (to about 240°C) which is a consequence of the decrease in the melting point of nitrate-alkali mixtures.

Using a temperature below 200°C the bituminization of neutral or weakly alkaline waste does not lead to any complication due to strong exothermal reactions. The bituminization of anion exchange resins should preferably be carried out at temperatures below 140°C (and - exceptionally - from a slightly acidic slurry) to reduce the formation of inflammable gases. Dangerous exothermal reactions can arise only in the bituminization of waste with a high alkali content.

Certain metal nitrates, e.g. iron (III) nitrate, are thermally instable causing not only the formation of nitrogen oxides but generally an enhanced oxidation of bitumen-nitrate mixtures already at lower temperatures. One could fear an explosion-like decomposition of high amounts of nitrates at higher temperatures and in the presence of catalyzing heavy metals.

Using various mixtures of alkali and heavy metal nitrates with bitumen (weight ratio of salts/bitumen = 2 : 1) it could recently again be verified that the incorporation or presence of nitrates in bitumen at temperatures between 180 - 250°C provides no safety problems and does never lead to explosion-like reactions, neither in air nor in a watersaturated atmosphere; the weight loss of the products in the range 180 - 250°C are small, especially if the nitrate salt containing slurry had been adjusted to pH 9-10 prior to its bituminization /274/.

The most important results of the investigations carried out at various institutions can be summarized as follows:

- most bitumens do not spontaneously ignite at temperatures below 400°C;

- if ignited, bitumen-nitrate salt mixtures burn faster than pure bitumen, i.e. nitrates accelerate the combustion;

- bitumen-nitrate waste mixtures containing up to 60 wt.-% solids do not show any explosive potential;

- some substances which can be present in decontamination solutions lower considerably the inflammation temperature of the bitumen-waste mixture, e.g. mixtures containing nitrates and 20 wt.-% sodium citrate have a very low burning temperature (230 - 260°C) /283/;

- apart from few exceptions the burning point of seventy bitumen (B 15)/40 - 60 wt.-% salt mixtures (simulating mixed wastes from reprocessing and research institutions) were found to be above 300°C /283/;

- the burning points of 50 wt.-% bitumen/50 wt.-% salt mixtures of a composition typical for reactor wastes (borates, detergents, sodium sulfate) are higher than 400°C /283/;

- a burning point of about 400°C has been measured for a mixture of 50 wt.-% bitumen (B 15), 40 wt.-% Li/borate-loaded ion exchange beads, and 10 wt.-% residual water; (it has to be observed that anion exchange resins are thermally degraded at elevated temperatures, ca 120°C /283/;

- the self-ignition point of bitumen/60 wt.-% salts from fuel reprocessing were found to be higher than 360°C /283/.

5.6.3 Incidents

To our knowledge, no explosion has ever occurred during the industrial bituminization of radioactive process wastes.

However, three fire incidents are known to have happened during operation which all could be extinguished within a short time; no radioactivity escaped from the plants, no person was injured and only small damages and contaminations occurred in the cell where the hot bitumen-waste mixture caught fire.

In the first incident the fire was caused by ignition of the vapours of organic solvents contained in the waste concentrate /194/.

In the second incident organic compounds contained in the waste (TBP and its degradation products, antifoaming agents, polyethylene-oxide adducts) decomposed - as a consequence of improper pH-adjustment and simultaneous equipment failure - into easily inflammable volatile compounds /186/.

In the third incident a research experiment was executed upon request of an outside client which involved an inactive mixture of distillation bitumen with nitrates, sulfates, fluorides, sodium, calcium, barium, iron, nickel, chromium, aluminium, and magnesium. Upon attempts to re-liquefy the warm mixture after standing overnight in a "pot-type" mixer-evaporator by means of immersed electrical heating elements a temperature well above 400°C was reached locally, leading to coking and finally to fire which could quickly be extinguished; no damage to persons or equipment occurred.

5.6.4 Conclusions

There exist no risk of a fire or an explosion or any other dangerous exothermic reaction during the industrial bituminization of radioactive wastes normally generated at LWR stations and reprocessing plants provided that:

- the bitumen content of the mixture is higher than 50 wt.-%,
- the temperature at any point of the bitumen-waste mixture is lower than 230°C,
- an efficient mixing of bitumen and waste residues and thus a homogeneous product is assured,
- the accumulation of explosive gas mixtures is prevented by applying a sufficiently powerful ventilation (especially above the container receiving the hot BWP),
- substances which are considerably lowering the burning point or are easily volatilized are destructed, converted, or removed in a pretreatment process prior to the incorporation of the remaining waste fraction.

No explosion or fire is known to have occurred in the industrial bituminization of real reactor and reprocessing wastes.

The fire incidents known happened all with the just cast or overheated final product mixture involving wastes from research activities, partly of unknown composition (originating from various activities of a nuclear research centre), and under no or insufficient ventilation.

A fire during the bituminization of wastes cannot be excluded with certainty, however, the probability of its occurrence is extremely low if the above listed requirements are strictly observed.

Anyhow, it is recommended to install an appropriate fire-detection and/or fire fighting system to reduce damages and contamination due to a fire as much as possible.

5.7 Bituminization Facilities in Various Countries

5.7.1 Sweden

As Sweden does not possess a fuel reprocessing plant the radioactive wastes generated in this country originate primarily from reactor stations, and to a minor extent from nuclear research centres, universities, radiochemical laboratories, hospitals, etc..

These wastes belong almost exclusively to the low- or intermediate-level categories and are conditioned for interim storage by means of cementation and bituminization techniques.

Presently, two bituminization facilities exist in Sweden, one at the Barsebäck nuclear power stations (built by Saint-Gobain Techniques Nouvelles, SGN, France), the other at the Forsmark nuclear power stations (built by BelgoNucléaire, Belgium).

a) Barsebäck Facility and Process /281/

The bituminization facility treats the wastes (ca 27 tons per year) generated by the two 590 MW_e - BWR-stations.

These wastes consist to about 99% of ion exchange resins (in form of powder- and beads) and filter-aid material (ca 5% of total weight of solid waste material). The volume of concentrates is at present very small as there are no demineralizer systems installed (which would produce sodium sulfate solutions).

The solid components of the waste have a specific activity prior to their incorporation into bitumen of 0.1 - 100 mCi/kg dry weight.

In Fig. 4 a simplified "standard" flow-scheme is presented according to Harfors /281/.

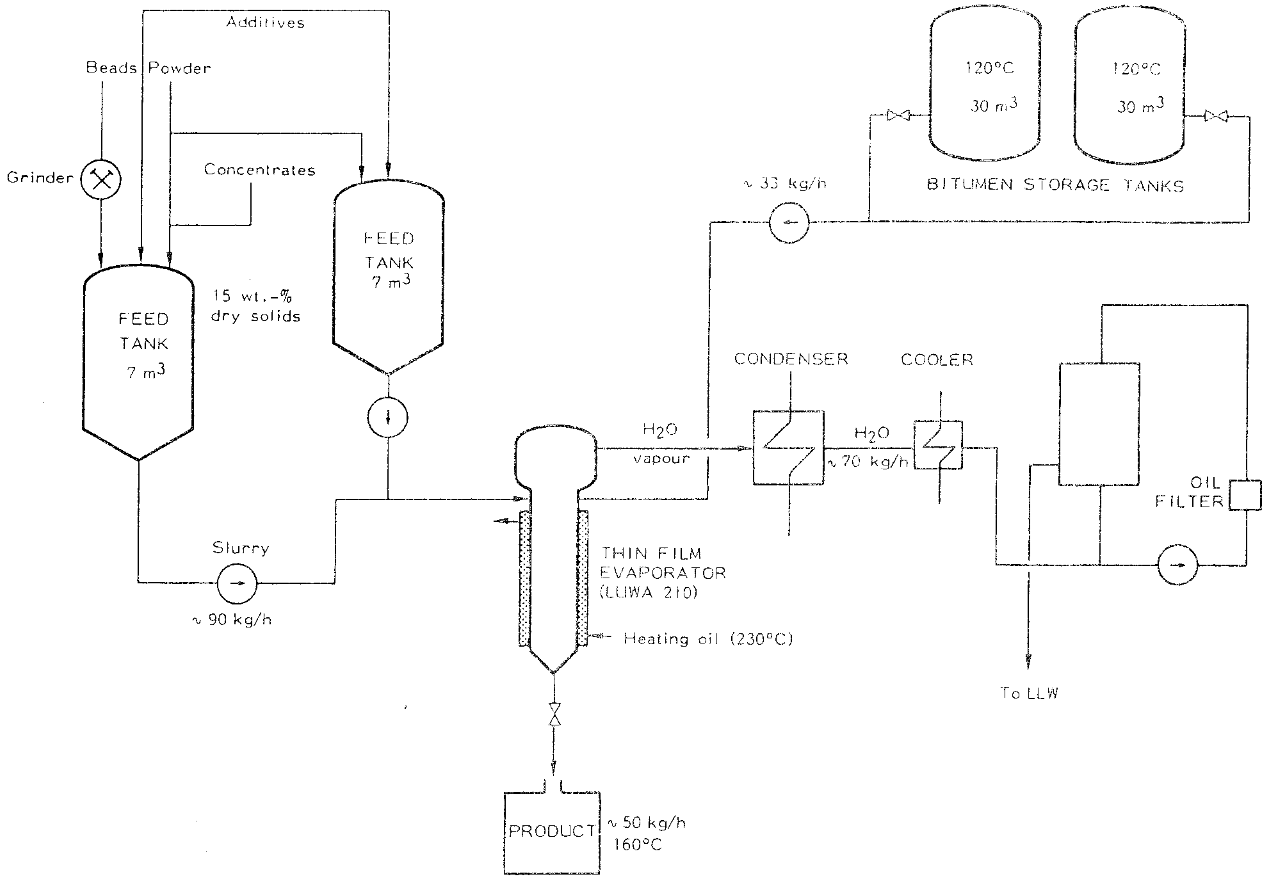


Fig. 4 - FLOWSHEET OF THE BITUMINIZATION AT THE BARSEBÄCK NUCLEAR POWER STATION (according Ref. 281)

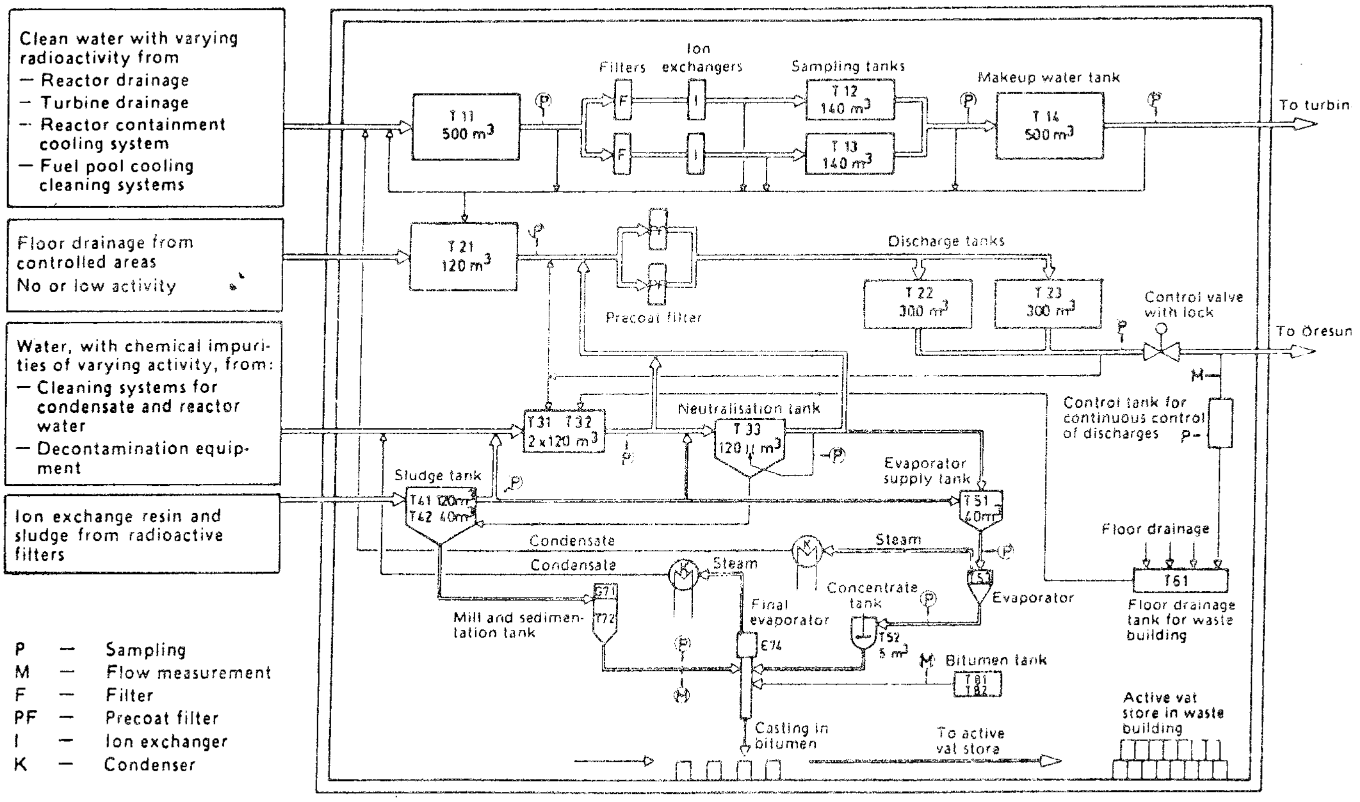


Fig. 5 - SYSTEM FOR PROCESSING RADIOACTIVE WASTE AT THE BARSEBÄCK NUCLEAR POWER STATION

The wastes to be bituminized are collected in a stainless steel tank of 7 m^3 equipped with a stirrer and a coil for steam heating.

By decanting at three different levels a waste slurry (suspension) of 15 - 20 wt.-% dry matter can be prepared.

Ion exchange resins in the form of beads are ground to powder in a wet grinder prior transfer to the feed vessel.

The slurry batch is heated to 60°C, then sodium sulfate is added to obtain a final concentration of 0.1 $\underline{M}$ (effecting a disaggregation of flocculated cation and anion resin particles and a reduction in the release of amines during bituminization), and finally an amount of emulsifying agent (Collemul H4) is added corresponding to about 2 wt.-% of the total dry weight of waste material (improving the homogeneity of the bitumen-waste particle mixture).

Bitumen (120°C), type Mexphalt 40/50, and the pre-treated waste slurry are fed by pumps separately but simultaneously and continuously to the top part of a thin-film evaporator, type LUWA 210, where both streams are mixed and the water evaporated. The evaporator is heated by a circulating thermofluid (Texatherm 320) having a temperature of 230°C.

The bitumen-waste mixture flows at a temperature of about 160°C and a rate of 50 - 55 kg/h from the bottom of the evaporator into drums of 220 l placed at a turn-table with positions for 8 drums.

The vapours from the evaporator are condensed, and the condensate is pumped via a cooler through a cartridge filter to remove entrained bituminous oils (normally < 200 ppm). The filtered condensate is then treated together with low-level liquid wastes from other sources.

The product drums are provided with a lid, monitored for surface dose rate, and then discharged from the filling cell after a minimum cooling time of 16 hours of the filled drums.

The drums with the BWP are transferred one by one by means of a shielded transport container (4 cm of lead) to an engineered interim on-site storage facility.

In Fig. 5 the liquid waste treatment system is shown as it has originally been planned /R-17/; the "as built" system is slightly different.

Fig. 6a shows the drum transfer system and the adjacent interim storage of the BWPs. Fig. 6b shows in more detail the lead bell used for the transfer of the BWP drums.

The Barsebäck facility is in operation since 1975 and had produced at the end of January 1979 approximately 1200 product drums.

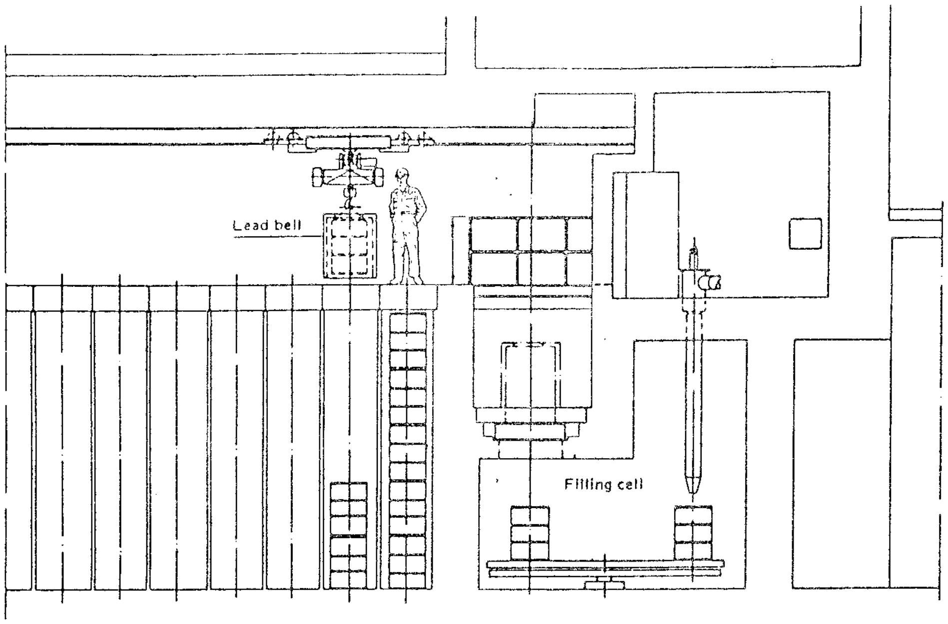


Fig. 6 a - DRUM TRANSFER AND INTERIM STORAGE FACILITY AT BARSEBÄCK

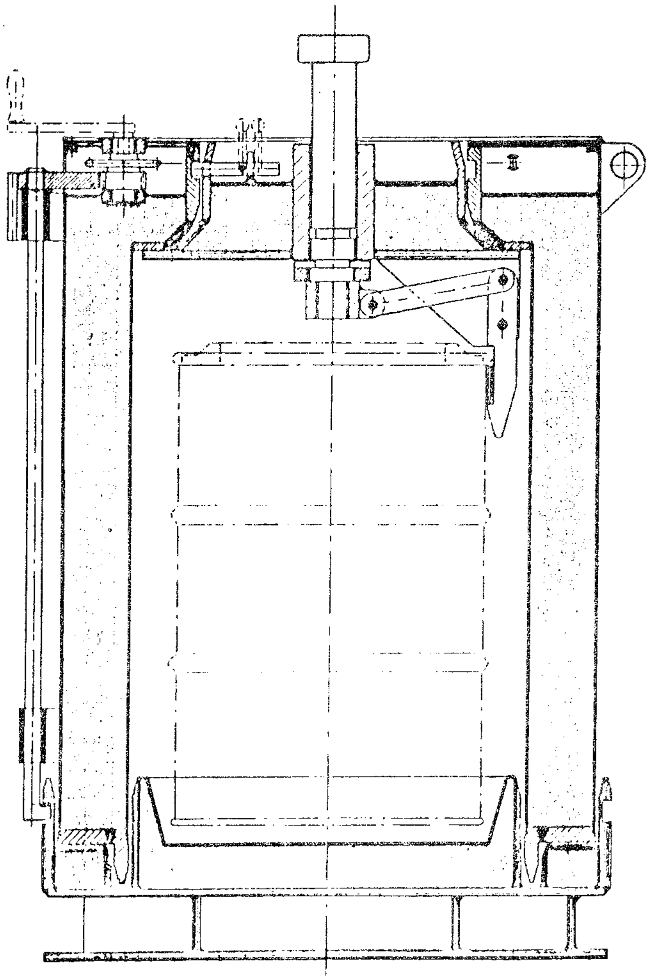


Fig. 6 b - LEAD BELL FOR THE TRANSFER OF PRODUCT DRUMS

b) Forsmark's Facility and Process /163 a/

The waste treatment building at the Forsmark Power Station (presently two 900 MW_e - BWRs, a third 1050 MW_e - BWR is under construction) contains systems for treating liquid, semi-solid and solid wastes and a storage for LLW and ILW. The storage capacity corresponds to 5 years production of ILW.

An extensive waste segregation is practiced in Forsmark.

A scheme of the various lines treating the radioactive liquid and semi-solid wastes is shown in Fig. 7.

The indicated evaporator is of the thermocompressor type permitting to reach a decontamination factor of 10^6 - 10^7.

The ion-exchange resins are stored in the 75 m^3 tanks for 0.5 - 3 years to allow the short-lived radionuclides to decay.

The bituminization facility at Forsmark comprises the following main components:

- waste feed system,
- rotary drum dryer,
- mixing vessel,
- offgas system,
- drum filling station, and
- bitumen storage.

Sludges containing 80 - 90% water are fed to the rotary drum dryer via the waste feed system. Ion-exchange beads are milled prior to the drying step. The dried materials containing less than 10% of water, are conveyed to the mixing vessel and are mixed with a sufficient quantity of bitumen for half an hour to a homogeneous product. The final product is discharged into 220 l steel drums.

After cooling for 20 - 24 hours, the drums are transferred to the storage for medium-active waste.

5.7.2 Other countries

Since the first small waste bituminization unit was built in 1960 at the Nuclear Research Centre at Mol (Belgium) until today, a considerable number of bituminization installations on laboratory, pilot and industrial scale has been constructed in many countries for research and industrial purposes.

(Numbers indicate the tank volume in m^3)

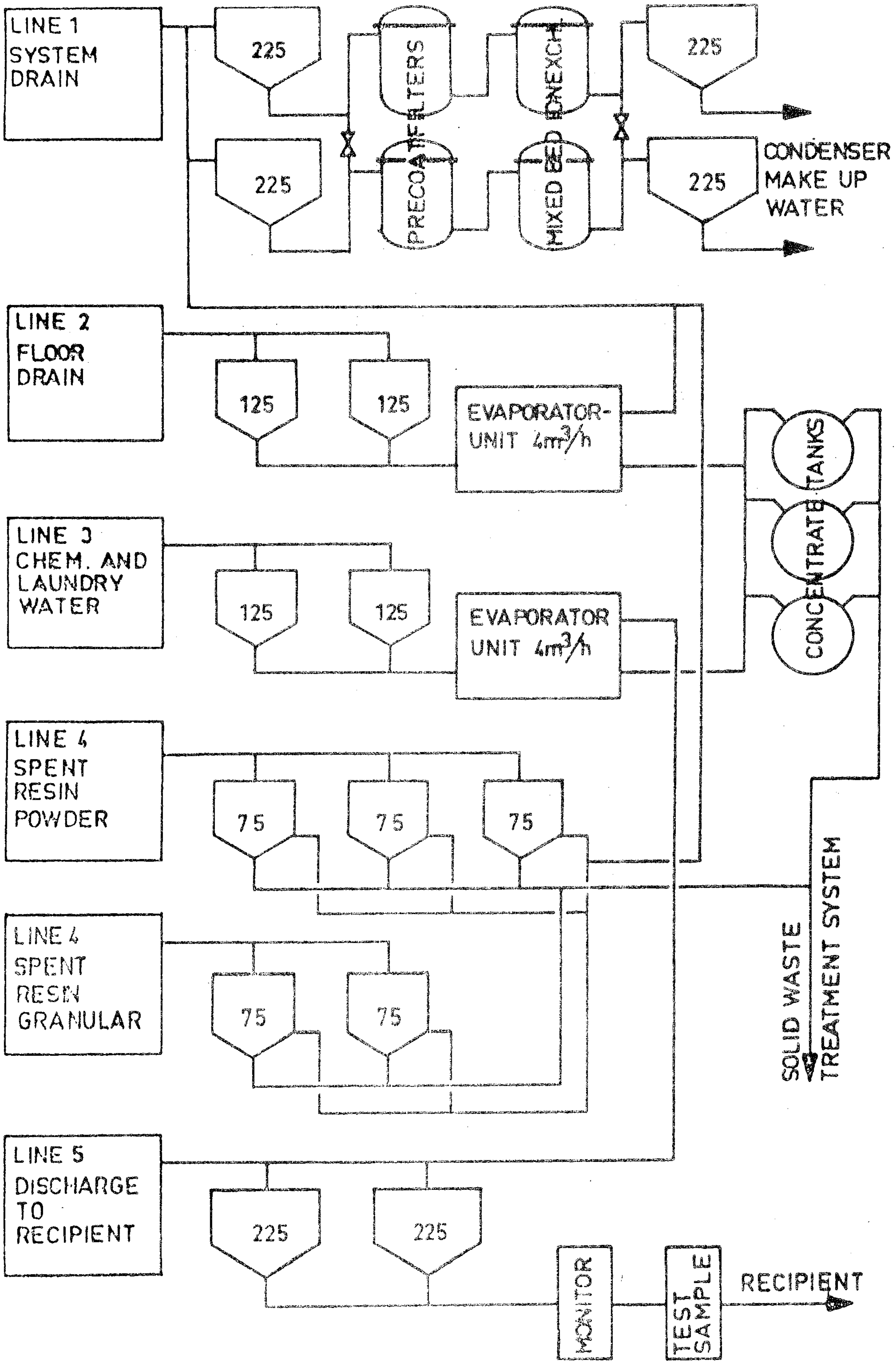


FIG. 7 - LIQUID WASTE TREATMENT SYSTEM AT THE FORSMARK NUCLEAR POWER STATIONS

Plants solidifying ILW solutions on an industrial scale from reprocessing by incorporation of the contained salts into bitumen exist in Marcoule, Karlsruhe (Fig. 8) and Mol (Eurochemic Fig. 9 and Fig. 10). In these plants a more or less elaborated chemical pretreatment of the waste solution is applied before the resulting slurries are mixed with molten bitumen at temperatures between 120 and 200°C.

For the mixing of the waste slurries with the bitumen, the evaporation of the water and the homogeneous incorporation of the residual salts into the bitumen matrix a screw-extruder-evaporator is used at all the three plants.

Moreover, besides these and the above described plants in Sweden, radioactive waste bituminization facilities of varying treatment capacities exist in the following countries:

Austria, Belgium, Bulgaria, Canada, Czechoslovakia, Denmark (see Fig. 11), Finland, France, (Cadarache, Saclay, Valduc), German Democratic Republic, Hungary, Japan (Tsuruya), Poland, Soviet Union, Switzerland (Goesgen), United Kingdom, and the United States.

5.8 Conclusions

All low- and intermediate-level waste solutions, suspensions, ion exchange resins, and sludges from reactor stations and reprocessing plants can safely be incorporated into bitumen by various techniques on an industrial scale, as demonstrated in several countries.

A safe and smooth operation is secured if:

- the weight ratio bitumen/solids is equal to about one or higher,
- a homogeneous distribution of the solids within the bitumen matrix is secured,
- the incorporation temperature for salts and thermally stable solids is below 230°C, and for anion exchange resins preferably below 150°C,
- the accumulation of inflammable gases is prevented (e.g. by a powerful ventilation),
- the generation of sparks is excluded, and if
- any other ignition source is absent.

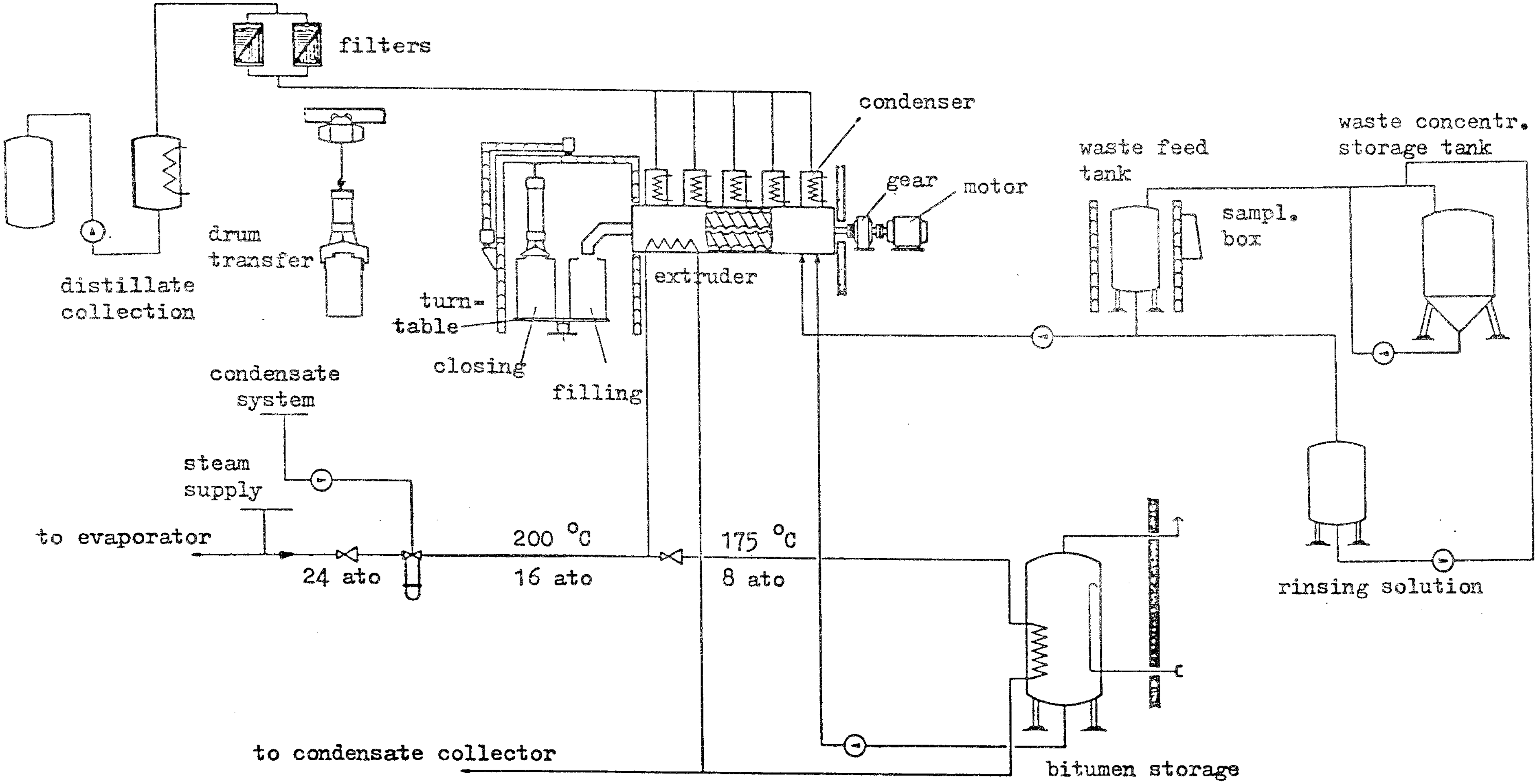
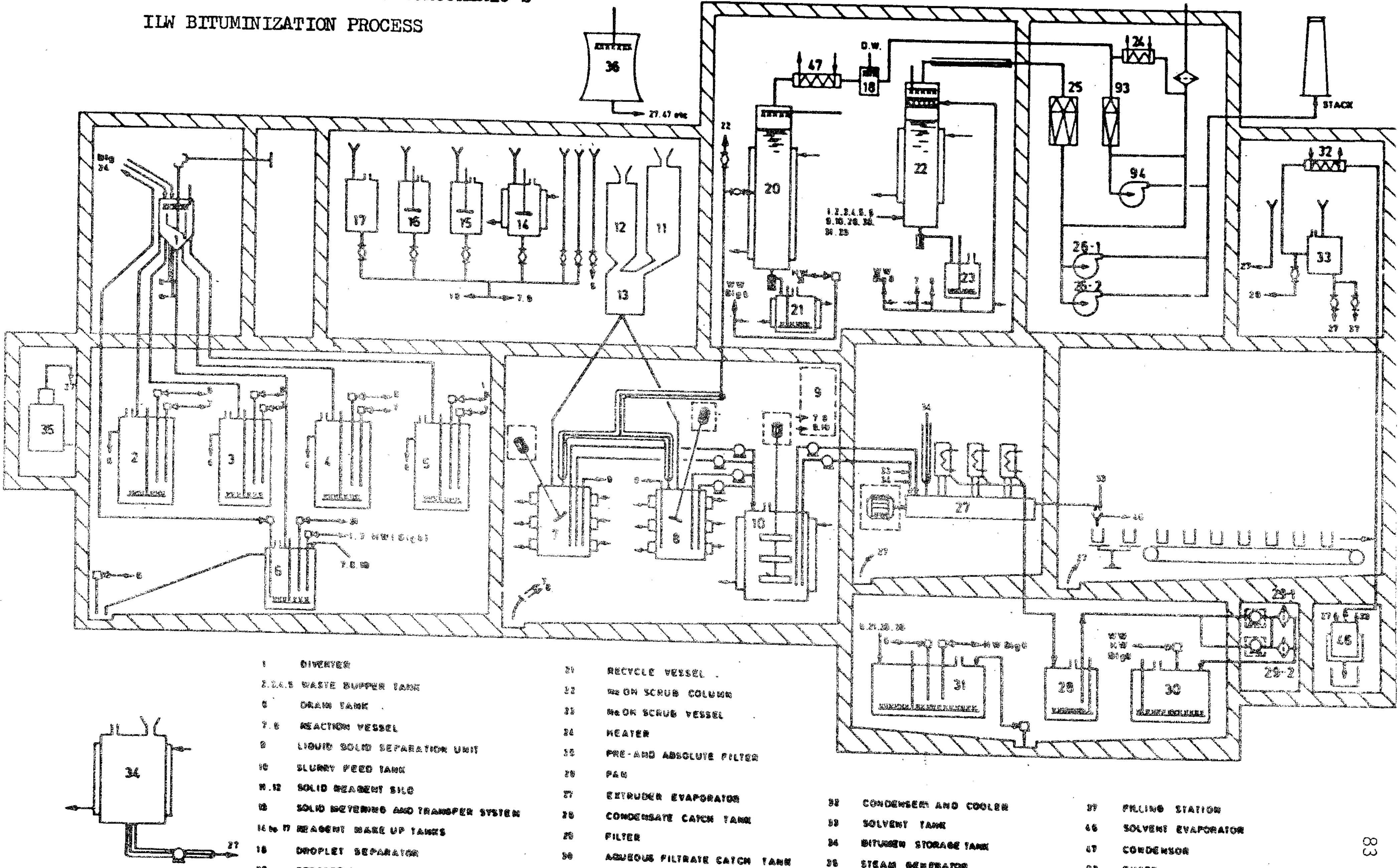


Fig. 8 - BITUMINIZATION INSTALLATION AT THE KARLSRUHE NUCLEAR RESEARCH CENTER [179]

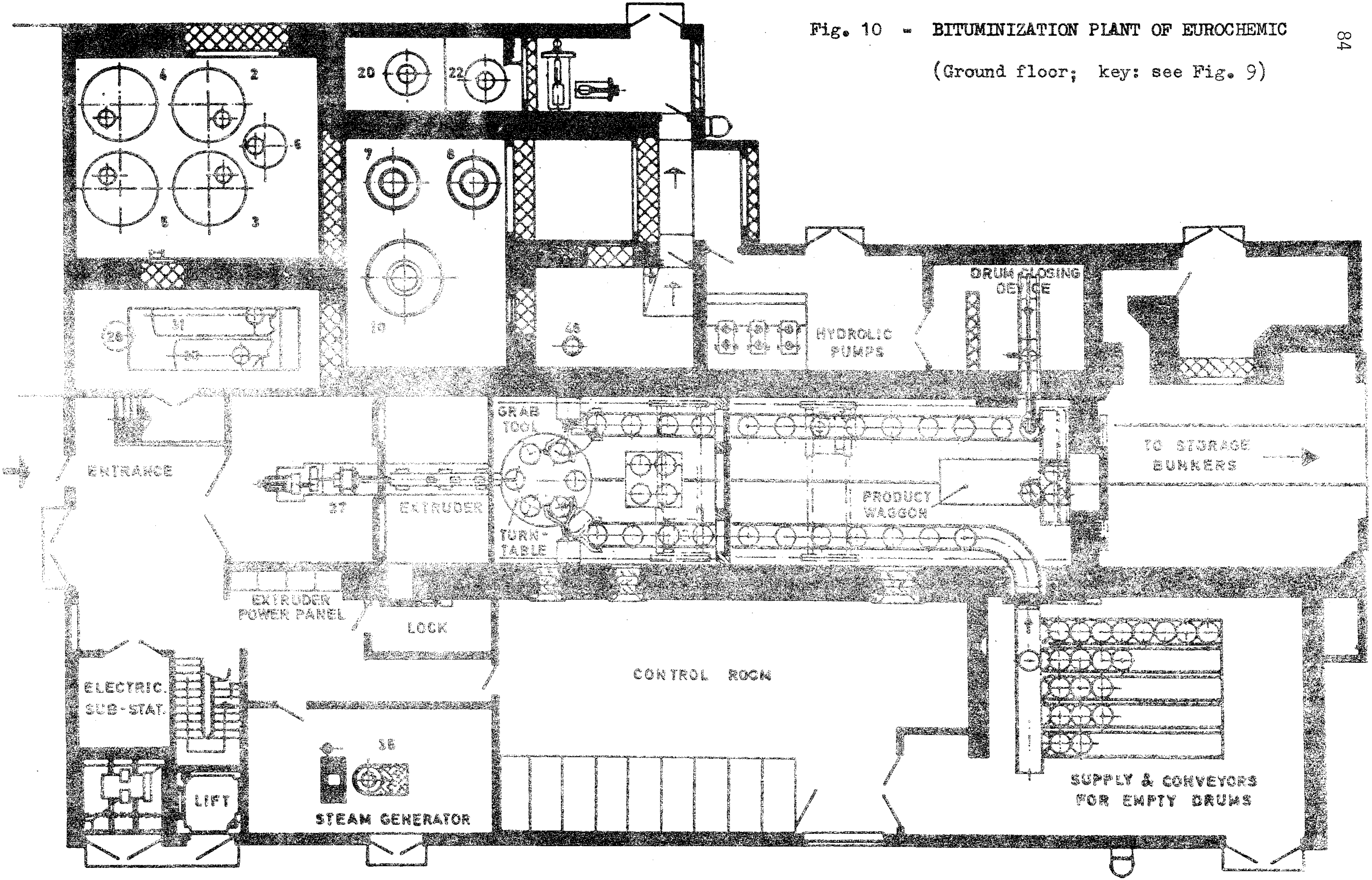
Fig. 9 - FUNCTIONAL FLOWSHEET OF EUROCHEMIC'S ILW BITUMINIZATION PROCESS



1	DIVERTER
2,3,4,5	WASTE BUFFER TANK
6	DRAIN TANK
7,8	REACTION VESSEL
9	LIQUID SOLID SEPARATION UNIT
10	SLURRY FEED TANK
11,12	SOLID REAGENT SILO
13	SOLID METERING AND TRANSFER SYSTEM
14 to 17	REAGENT MAKE UP TANKS
18	DROPLET SEPARATOR
20	DESORPTION COLUMN
21	RECYCLE VESSEL
22	NaOH SCRUB COLUMN
23	NaOH SCRUB VESSEL
24	HEATER
25	PRE-AND ABSOLUTE FILTER
26	FAN
27	EXTRUDER EVAPORATOR
28	CONDENSATE CATCH TANK
29	FILTER
30	AQUEOUS FILTRATE CATCH TANK
31	HOT WASTE TANK
32	CONDENSER AND COOLER
33	SOLVENT TANK
34	BITUMEN STORAGE TANK
35	STEAM GENERATOR
36	COOLING TOWER
37	FILLING STATION
46	SOLVENT EVAPORATOR
47	CONDENSOR
93	FILTER
94	FAN

Fig. 10 - BITUMINIZATION PLANT OF EUROCHEMIC

(Ground floor; key: see Fig. 9)



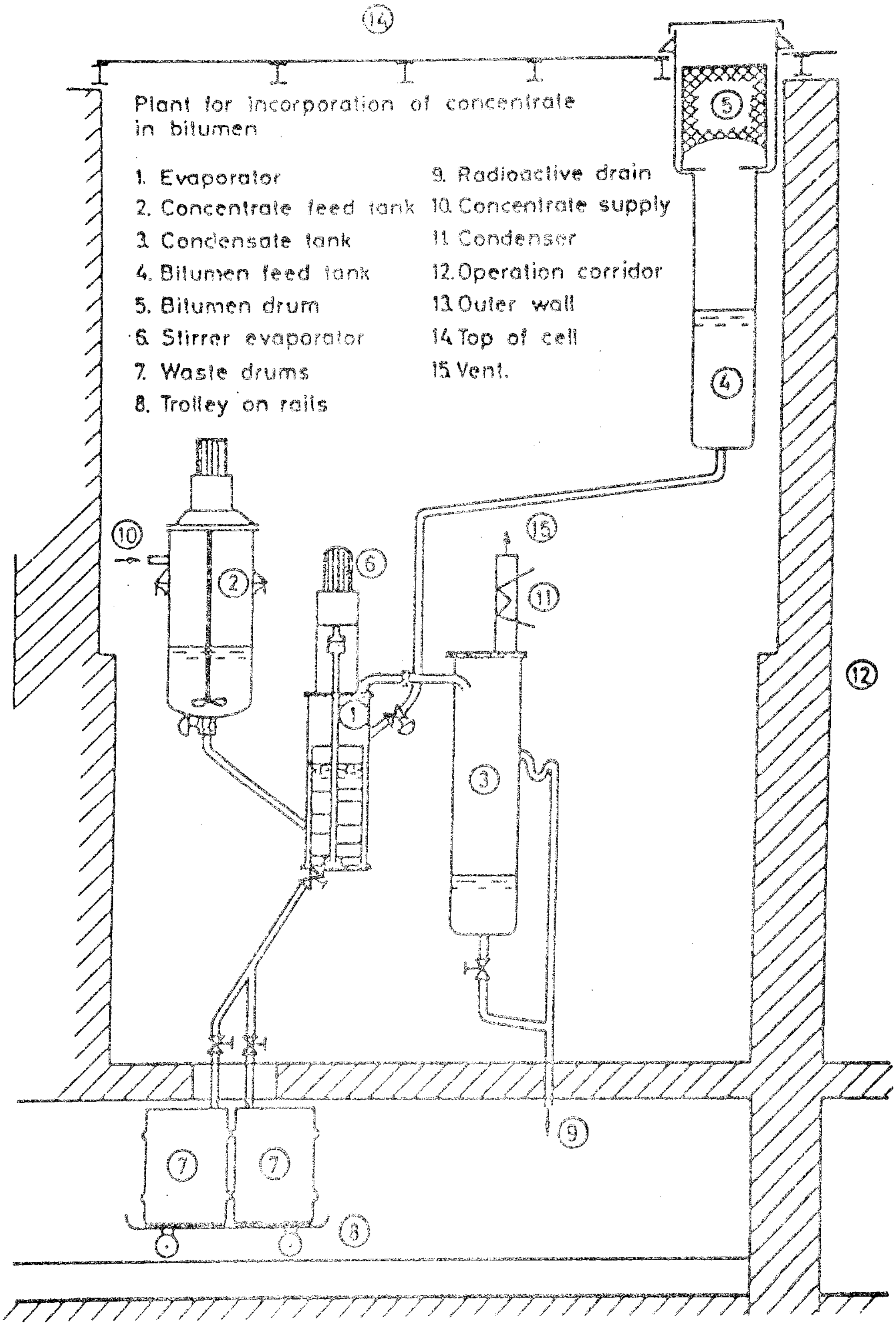


Fig. 11 - BITUMINIZATION FACILITY AT THE RISÖ RESEARCH ESTABLISHMENT [115]

6. THE PROPERTIES OF BITUMEN-WASTE MIXTURES

6.1 General

The properties of mixtures of bitumen with radioactive wastes which can be of importance during the various waste management phases are listed in Table XXII.

In general, the incorporation of solid matters into bitumen causes a considerable decrease of the penetration and the ductility of the bitumen with increasing solids' content and furthermore a significant increase of the softening point and flash point.

In this chapter mainly those properties and factors are considered which influence the safety during the short- and long-term storage of BWPs.

6.2 Composition and Characteristics of Bitumen-Waste Products (BWPs)

6.2.1 Reactor waste-bitumen products

The majority of BWPs generated at reactor stations are composed of about 50 wt.-% bitumen and the solid constituents originally present in the wet LWR wastes (i.e. mainly spent ion exchange resins, filter sludges, and evaporator concentrates).

The ion exchange resins usually applied in Swedish nuclear power plants are:

- Duolite ARM 381 (of a mixed bed equivalent capacity of 1 : 1);
- Duolite ARC 351 (cation exchanger);
- Duolite ARA 366 (anion exchanger).

These are resins in bead form having a cross-linking of 8% DVB. The powder resins are mostly made out of bead resins by grinding. At Forsmark Microionex AOH-CH of 20 - 100 μ particle size is used; the proportion of cation exchange resin to anion exchange resin is approximately 3 : 1.

The water associated with the ion exchange resins is partially or almost completely removed by means of a dryer prior to their mixing with liquefied bitumen whilst the water of the aqueous solutions and slurries is removed during the incorporation process until a residual content of less than 1 wt.-%.

The final products consist thus of a mixture of bitumen with the solid and non-volatilized (at the incorporation temperature and time) constituents of the treated wastes.

TABLE XXII

IMPORTANT PROPERTIES OF BITUMEN-WASTE MIXTURES IN THE VARIOUS WASTE MANAGEMENT PHASES *)

Property	Waste Management Phase **)			
	P	I	T	L
Density	+		+	
Penetration		+	++	+
Viscosity	++	+		+
Softening point	++	++	++	++
Plasticity			++	+
Homogeneity	++	+	+	++
Porosity		+	+	++
Compressive strength				++
Shock resistance		+	++	+
Cold resistance		++	++	++
Content of solids	++	+	+	++
Specific activity (α, β, γ)	++	++	++	++
Thermal conductivity	++	+	++	++
Thermal expansion	+	+	++	++
Ignition point	++	++	++	+
Flash point	++	++	++	+
Burning point	++	++	++	+
Burning rate	++	+	++	+
Phase separ. during burning	++	+	++	+
Radiation stability		++		++
Dose rate	++	++	++	++
Gas generation		++		++
Swelling (due to radiolysis)		+		++
Swelling (due to water)		+	+	++
Water absorption		+	+	++
Water content	+	+	+	++
Leaching		+	+	++
Effect of micro-organisms				++
Ageing				++

*) Based on Ref. 219, however, somewhat modified.

**) P Solidification process

I Interim storage

T Transportation

L Long-term storage

\+ important

++ very important

The weight or volume fractions of bitumen and the waste components are chosen beforehand (according to proven flowsheets and required product properties) and are carefully controlled during the bituminization by accurate flow or weight measurements of the inactive bitumen and the radioactive waste "streams".

The spent ion exchanger beads from PWR stations are loaded to a high degree by lithium and borate ions.

The evaporator concentrates from BWPs have normally a high content of sodium sulfate whilst those from PWRs contain high amounts of sodium borate (or boric acid) and detergents. Because of solubility limitations (e.g. the low solubility of boric acid in water) the total salt content does not exceed 300 g/l, and is usually about 200 g/l.

Approximate data on the properties of three "typical" BWPs prepared from LWR wastes are given in Table XXIII.

TABLE XXIII

SOME PROPERTIES OF "TYPICAL" LWR WASTE-BITUMEN MIXTURES

BWPs Properties	BWR/PWR 50% Bitumen* 48% Ion exch. ~ 2% H_2O	PWR 53% Bitumen* ~ 47% Salts ~ 0.3% H_2O	BWR 50% Bitumen* ~ 35% Salts ~ 15% Fillers ~ 0.5% H_2O
Specific Density (g/cm^3)	1.18	1.40	1.35
Softening point (°C)	90	90	> 85
Flash point (°C)	> 300	> 300	> 300
Ignition point (°C)	~ 440	~ 400	~ 425
Leachability in dist. H_2O $g.cm^{-2}.d^{-1}$	3×10^{-5}	8×10^{-4}	1×10^{-3}
Specific activity (Ci/l)	0.05	< 0.01	< 0.02
Heat generation (W/l)	$< 3 \times 10^{-4}$	$< 5 \times 10^{-5}$	$< 10^{-4}$
Total integrated dose (rad)	$< 10^7$	$< 2 \times 10^6$	$< 4 \times 10^6$
Spec. H_2-gen. rate (cm^3/Mrad . g BWP)	⩽ 0.005	⩽ 0.005	⩽ 0.005

*) Mexphalt 15.

6.2.2 Reprocessing waste-bitumen products

The majority of the BWPs are composed of 40 - 60 wt.-% bitumen and the nitrate salts contained in the neutralized evaporator concentrates of the ILW-category, i.e. mainly sodium nitrate. The decontamination wastes are either contained in these concentrated sodium nitrate solutions or are bituminized separately. Process and decontamination wastes undergo usually a chemical pretreatment to insolubilize long-lived radionuclides and, eventually, to eliminate or to convert substances which might interfere with the bituminization process.

A small fraction of BWPs may contain - as in the reactor waste products - ion exchange resins and granular filter-aid materials.

The beta-gamma activity in the ILW-solutions (from the processing of short-cooled fuels) stems mainly from Zr/Nb-95, Ru-103, and Ru-106; the alpha-activity is due to plutonium isotopes, americium-241 (grown-in from Pu-241) and uranium.

The specific activity of the bituminized ILW is in the order of 0.8 Ci/l BWP.

If a plant would use a chemical decanning process (e.g. the Zirflex-process for Zircaloy-jackets or the Sulfex-process for stainless steel jackets) the salts contained in the decanning solutions and about 1% of the fission product activity (due to leaching) would finally be present in the BWPs.

a) A typical BWP from waste of a plant using a mechanical head-end has the following approximate composition:

55 wt.-% blown bitumen,

40 wt.-% sodium nitrate,

~ 4 wt.-% salts of heavy metals and organic acids (oxalic, tartraric, citric acid)

0.5 wt.-% water,

0.5 wt.-% various substances,

~ 0.8 Ci/l fission product activity, $\leq$ 3 mg Pu/l; $<$ 1.0 g U/l.

b) A typical BWP from waste of a plant using a chemical decanning has the following approximate composition:

55 wt.-% bitumen (distillation or blown),

20 wt.-% salts of the canning components (sulfates, fluorides),

20 wt.-% sodium nitrate,

4 wt.-% solids from decontamination wastes,

0.5 wt.-% water,

0.5 wt.-% various substances,

< 2 Ci/l (depending on cooling time);

< 5 mg Pu/l; < 1.5 g U/l.

The specific density of the product is between 1.3 and 1.4 g/cm^3.

c) In principle, spent organic solvents from extraction processes can also be incorporated into bitumen.

The spent 30 vol.-% tributyl phosphate (TBP) - kerosene solution originating from the Purex process can be separated into its TBP and kerosene components /R-18/ and the TBP fraction can be incorporated into bitumen.

As the bitumen-TBP mixture is an unusual reprocessing waste product and as certain general considerations in the following chapters are not necessarily valid for this mixture, some more information on its properties will be given already here.

TBP incorporated into bitumen exhibits a plastification action, it lowers considerably the softening temperature, increases the penetration, and decreases the viscosity. Consequently TBP should be incorporated in hard bitumens only and in a relatively small amount.

To obtain a better product quality it is advantegeous to incorporate TBP together with inorganic wastes or inorganic filling materials (absorbents).

A product of a quality comparable to inorganic waste-bitumen mixtures should contain up to 40 wt.-% inorganic filling material and not more than about 5 wt.-% TBP.

It was shown by differential thermoanalysis that up to the distillation temperature of TBP (289°C), no significant interactions take place if one heats TBP-bitumen products containing inorganic wastes, e.g. sodium nitrate /209 a/.

The leach rate after one month of a product composed of a hard bitumen, 40 wt.-% filler and 10 wt.-% TBP (ca 3×10^{-4} g/cm^2.day) is higher than a product composed of a softer bitumen and 40 wt.-% sodium nitrate (leach rate: approx. 1×10^{-4} g/cm^2.day).

The BWPs are usually collected in 220 l painted mild steel drums (in some cases more corrosion resistant drums are used), filled to about 80% of the total volume (ca 180 l) to allow for an eventual expansion during storage.

The total weight of one BWP package (product plus drum) is approximately 280 kg. In Table XXIV some data of "typical" intermediate-level BWPs are given which, of course, have to be considered as very rough values only.

TABLE XXIV

SOME PROPERTIES OF "TYPICAL" INTERMEDIATE-LEVEL REPROCESSING WASTE-BITUMEN MIXTURES

BWPs Properties	Mech. head-end 55% blown bitumen* ~ 45% salts 0.3% H_2O	Chem. head-end 55% blown bitumen* ~ 45% salts 0.3% H_2O
Specific density (g/cm^3)	1.36	1.35
Softening point (°C)	~ 115	> 110
Flash point (°C)	> 290	> 290
Ignition point (°C)	~ 380	> 360
Leachability in dist. H_2O, average after 1 year ($g.cm^{-2}.d^{-1}$)	~ 10^{-4}	~ 10^{-5}
Specific activity (Ci/l)	~ 0.8	~ 1
Heat generation (W/l)	< 0.005	< 0.005
Total integrated dose (rad)	$< 2 \times 10^8$	$< 2 \times 10^8$
Spec.H_2-gen.rate (cm^3/Mrad.g BWP)	$\leqslant 0.005$	$\leqslant 0.005$

*) Mexphalt R 90/40.

6.3 Storage- and Safety-Relevant Properties of Bitumen-Waste Products

6.3.1 Introduction

The storage of radioactive waste products is safe when the release of radionuclides from these products to the environment does not lead to a hazardous radiation dose to man.

Apart from the storage place and storage conditions the quality of the waste product itself plays an important role in preventing or retarding the dispersion of waste components towards man's biosphere.

The quality of a waste product in view of its safe storage is given by its composition which determines the stability towards various physical and chemical (sometimes biochemical) impacts.

The properties of a great number of BWPs have been examined at various research institutions thoughout the world.

The properties and factors - which are judged to be of importance for a safe storage of BWPs - are:

- resistance against leaching media;
- radiation stability and effects;
- thermal effects and stability:
- sedimentation of incorporated solids;
- combustibility and explosiveness;
- mechanical stability;
- swelling caused by radiolysis gases and uptake of water.

Some of these factors are of greater importance during the temporary storage (in engineered storage facilities), some of them during the final (geological) storage of the BWPs.

The essential results of investigations on the above mentioned properties and other characteristics of BWPs are dealt with in a concise manner in the sub-chapters that follow.

For detailed information on the experimental procedures applied and other particulars the original literature should be consulted.

6.3.2 Leach resistance

a) General

The most important property of radioactive waste products to be stored (or to be disposed of) is the resistance against the leaching of the contained radionuclides by water (rain water) and aqueous solutions (e.g. ground water, sea water, saturated sodium chloride or other brines).

The aqueous solutions coming in contact with the waste products may slowly dissolve components of the waste and transport them away from the product into the environment.

The leachability of a substance is characterized by its "leach rate" L (expressed in $g \cdot cm^{-2} \cdot d^{-1}$) which can be determined - using a well defined test method - according to the following formula:

$$L = \frac{A_t}{A_o} \cdot \frac{m}{S \cdot t}$$

Where A_t = amount or radioactivity of substance A removed (leached) from the sample in time t

A_o = initial total amount or activity of substance A in the sample

m = mass of sample (g)

S = geometric surface area of sample exposed to leachant (cm^2)

t = leach time (days)

The "substance A" can be a specific element, a radioisotope, a compound, or the entire sample; it must therefore be clearly indicated for which "substance" the leach rate has been determined.

It should be noted that the correct SI-unit for the leach rate is $kg \cdot m^{-2} \cdot sec^{-1}$; this unit has, however, up to now seldom been used in reports dealing with the leach rate of waste products.

The results of leach tests can also be expressed by a plot of incremental leach rates, R_n (in $m \cdot s^{-1}$), as a function of leach time, t (in s), using the following formula:

$$Rn = \frac{a_n}{A_o \cdot S \cdot \rho \cdot t_n}$$

where a_n = radioactivity (s^{-1}) leached during each leach period t_n

A_0 = specific radioactivity ($s^{-1}.kg^{-1}$) initially present in the sample

S = surface area of sample exposed to leachant (m^2)

ρ = density of the sample ($kg.m^{-3}$)

t_n = duration of each leach period (s)

t = cumulative leach time (s).

A_0 and a_n are to be corrected for decay time.

The leach rate, R_n, of suitable products becomes constant after a certain time of leachant renewal periods as indicated by the plot of R_n versus t. It is recommended to report the mean value of R_n (together with the accuracy) for the virtually constant range.

When comparing the results from leach tests one should take into consideration:

- the possible sources of errors in the measurements;
- whether the experimental conditions have been carefully controlled;
- whether the methods, conditions, and the types of products used are really comparable;
- whether the sample can be considered as representative for the product generated in the bituminization process.

The leach rate of waste constituents from BWPs is influenced mainly by the following factors:

- composition of the BWP (waste/bitumen ratio, soluble and insoluble salt content, etc...);
- particle size of the incorporated waste solids;
- homogeneity of the product (dispersion of the waste within the bitumen matrix);
- residual water content of the BWP;
- bitumen type;
- bituminization conditions;
- radiation dose;
- composition of leaching liquid (salt concentration, pH);
- temperature.

A determining factor of the leach rate of radionuclides is the solubility of the chemical compound they form part of, or the solid species in which they are incorporated, and consequently all factors affecting this solubility.

The most important results of experimental investigations on the various factors influencing the leach rate of different species from BWPs are summarized hereafter.

b) Influence of the amount and composition of the incorporated wastes

- The leach rate is affected more by the percentage of solid waste residues in the bitumen and the species of isotopes being leached (Cs and Na > Sr) than by the conditions of the experiments /5/.

- The leach rate increases with the increase of the salt content in the BWP.

- Amounts of alkali exceeding 5 wt.-% decrease the leach resistance of BWP /216/.

- BWP containing from 20 to 60 wt.-% solids from evaporator concentrates reach a constant leach rate in about six months /14/.

- BWPs with 60 wt.-% solids from evaporator concentrates showed the following constant leach rates:

  3×10^{-4} g . cm^{-2} . d^{-1} for static conditions,

  5×10^{-4} g . cm^{-2} . d^{-1} for dynamic conditions;

  (BWPs with 20 wt.-% solids showed a leach rate of 1.5×10^{-4} g . cm^{-2} . d^{-1}) /14/.

  The leach rate for Ru-106 was 7×10^{-6} g . cm^{-2} . d^{-1} /8/.

- BWPs prepared from emulsified bitumen and three typical aqueous intermediate-level reprocessing wastes, namely:

  o alkaline evaporator concentrates,

  o aluminium cladding solutions, and

  o neutralized second Purex-cycle solvent extraction raffinates (2 CW),

  containing about 60 wt.-% solids (including up to 35 wt.-% nitrates) and 13 Ci/l were subjected to static leach tests with demineralized water for about 2.8 years.

At the end of the leach period the accumulated dose amounted to max 1.5 x 10^8 rad (- which corresponds approximately to the dose that would be received by a BWP containing 5 Ci of Cs-137 per litre of BWP after ten years of storage -) and the total amount of sodium leached amounted to 1.4, 3.8, and 2.5% respectively of the total sodium in the sample /51, but p. 46-60/.

- The leach rate of Cs-137 after 1 year from bitumen BN-III
  - o containing 40 wt.-% sodium nitrate was ca 10^{-4} g. cm^{-2} . d^{-1},
  - o containing 40 wt.-% calcium nitrate was ca 10^{-5} g. cm^{-2} . d^{-1},
  - o containing 40 wt.-% calcium carbonate was less than 10^{-6} g . cm^{-2} . d^{-1} /43/.
- BWPs with about 45 wt.-% bitumen show an optimal leach resistance compared to those with 40, 50, and 60 wt.-% bitumen /139 a/.
- Mixtures of bitumen B 15 and various amounts of sodium nitrate (~ 500 µ particle size) showed the following sodium leach rate (mean values) for 1 year's leaching in distilled water /283/.

$NaNO_3$-cont wt.-%	1	5	10	20	38
mean leach-rate (g . cm^{-2}. d^{-1})	not detectable	4x10^{-6}	7x10^{-6}	2x10^{-5}	9x10^{-5}

- Most of the BWPs prepared from evaporator concentrates (from reprocessing plants) containing about 40 wt.-% sodium nitrate have leach rates (in stagnant distilled water) of about 5×10^{-4} g . cm^{-2} . d^{-1} /283/.

- BWPs containing compounds which can bind a high amount of water (e.g. as crystal-water or by hydratization) and which are soluble in water show a low leach resistance towards water, i.e. leach rates of the compounds considered of 10^{-3} to 10^{-1} g . cm^{-2} . d^{-1}; typical examples are:

  o sodium carbonate forming $Na_2CO_3 \cdot 10H_2O$ (decahydrate)

  o sodium sulfate forming $Na_2SO_4 \cdot 10H_2O$

  o sodium phosphate forming $Na_3PO_4 \cdot 12H_2O$

  o disodium hydrogen phosphate forming $Na_2HPO_4 \cdot 12H_2O$

  o sodium tetraborate forming $Na_2B_4O_7 \cdot 10H_2O$

  o sodium chromate forming $Na_2CrO_4 \cdot 10H_2O$.

  If an anhydrous salt takes up water within the BWP the salt volume increases, causing an expansion of the product and finally its disintegration (cracking) when the limit of the elastical expansion of the bitumen matrix is exceeded. As the disintegration of the product is accompanied by a considerable increase of the surface exposed to the leachant, the leachability of the waste consituents is also considerably increased.

  The conversion of anhydrous sodium sulfate (density of 2.68 g/cm^3) into its decahydrate form (density of 1.46 g/cm^3) increases the molar volume by a factor of four.

- The leach resistance of bitumen-sodium sulfate mixtures (typical products from BWR-waste) in a saturated sodium chloride solution and a quinary brine (consisting of 31.2% $MgCl_2$, 1.85% $MgSO_4$, 0.56% KCl, 0.18% NaCl, 66.21% H_2O) was found to be high (for 50% bitumen - 50% $NaSO_4$): 1.7 x 10^{-6} g . cm^{-2} . d^{-1} in the saturated NaCl-solution and 1 x 10^{-6} g . cm^{-2} . d^{-1} in the quinary brine /283/.

  This result is remarkable (- though explainable -) considering the high bulk leach rate of 10^{-1} g . cm^{-2} . d^{-1} of similar product samples (18 - 44 wt.-% sodium sulfate) leached in distilled water and ground water which, in addition, degraded completely within seven days (for 5 cm Ø x 10 cm long specimen) or fourteen days (for 7.5 cm Ø x 15 cm long specimen) /265/.

- The leach rate is little influenced by up to 6 wt.-% of emulsifying agents in the BWP /280/.

- The bituminization of PWR-waste concentrates containing borates resulted in strong alkaline salt-bitumen mixtures (50 : 50 wt.-%) having a mean leach rate of 7 x 10^{-3} g . cm^{-2} . d^{-1}; products containing besides other solids more than 30 wt.-% detergents showed a leach rate of 1.5 x 10^{-2} g . cm^{-2} . d^{-1} /283/.

c) Influence of the particle-size and distribution (homogeneity)

The bituminization of ILW solutions containing a high percentage of dissolved salts ($NaNO_3$) by means of a screw-extruder evaporator resulted in products of good homogeneity in which the dispersed salt crystals were found to have a size of $\leq$ 40 µ /76/. When slurries (containing up to 40 wt.-% solids) from a chemical pretreatment of decanning solutions were bituminized, a little less regular distribution of the solids - which had particle sizes from "very fine" up to200 µ and more - was observed, however, no agglomeration of crystals.

The bituminization of evaporator concentrates using an extruder-evaporator results in extremely homogeneous products with particle sizes of the waste solids of 10 - 30 µ /283/.

Bitumen-sodium chloride mixtures with 38.5% NaCl of 500 - 800 µ and of 50 - 80 µ particle size showed clearly that the product with the smaller NaCl-crystals were less leachable (up to one order or magnitude).

One may conclude: the smaller the particle size and the more homogeneous the distribution of the incorporated waste solids the higher the overall leach resistance of BWPs.

d) Influence of the water content

BWPs prepared from wastes originally containing water (e.g. waste concentrates, ion exchange resins, filter aids) contain a certain amount of residual water, usually less than 0.5 wt.-%.

A higher water content indicates a higher porosity of the product and thus a higher leachability of its content, especially of its water-soluble parts; however, the residual water content of some few weight percent does not significantly increase the leaching rate.

If the BWP has been prepared from an acidic slurry (e.g. ion exchange resins) the residual water may remain acidic causing a considerable increase (ca 5 - 50 times) of the leach rate compared to an equal product prepared from a neutralized slurry liquid.

BWPs immersed in water absorb different amounts of water depending on their composition:

- the up-take of water is much higher in bitumens containing soluble salts than in bitumens containing insoluble ones /43/;
- the highest absorption of water occurred when surface-active agents were present in bitumen /43/;
- BWPs with 43 and 60 wt.-% bitumen took up 56% water during 1500 days' (4 years') immersion into water /139 a/.

e) Influence of the type of bitumen

- Generally, leach rates of nuclides from products with soft bitumens are lower than those composed of harder ones.
- Samples containing evaporator concentrates in straight-run distillation bitumens (especially Mexphalt 40/50) showed a slightly lower leach rate than those prepared with oxidized bitumen /47/.
- The bitumen surface layer of the BWP is much more resistant to the penetration of water than the bitumen inside the product.
- Experiments, in which distilled bitumens Spramax 180/220, Mexphalt 40/50, M 80/90, and the blown bitumens R 90/40 and 115/15 were used, have shown that the type of bitumen has only a small influence on the leach rate of radionuclides. BWPs with Mexphalt 40/50 showed the lowest leach rate /139 a/.

f) Influence of the bituminization conditions

The following conditions may influence the leachability of waste components:

- incorporation temperature,

- mixing-evaporating equipment and conditions applied, and

- bitumen type.

At higher incorporation temperatures (> 150°C) and longer contact time, waste components can react with components of bitumen (e.g. Sr-90 and Ca) which normally reduces the leachability of these waste components considerably. However, the bituminization of anion exchange resins at temperatures beyond 120°C may result in the deliberation of volatile degradation products (amines) which may have an adverse effect on the leach resistance of the resulting product. If another coating equipment than an extruder-evaporator is used, processes in which aqueous concentrates are dispersed into emulsified bitumen, produce usually more homogeneous BWPs than processes in which the direct incorporation into hot "non-emulsified" bitumen is applied.

Longer mixing of the bitumen-waste fractions at higher temperatures (190 - 210°C) reduces the residual water content in the resulting product (usually below 0.5 wt.-%) and thus the leach rate of the incorporated inorganic waste solids.

The coating of radioactive salts by hot, or just fluid bitumen, or by the emulsified form (anionic and neutral) has no influence on the leach rate /139 a/.

g) Influence of the radiation dose

Radiation doses higher than 10^7 rad cause the formation of radiolysis gas bubbles having diameters of up to several millimeters. As this increase of the porosity increases the overall surface of the product accessible by the leachant, one could predict that high radiation doses increase significantly the leachability of BWPs.

However, numerous experiments have shown, that radiation doses of up to 10^8 rad do not influence the leach resistance of BWPs towards various leachants to any appreciable extent:

BWPs containing different types of bitumen and 57 wt.-% solids (including 33 wt.-% nitrate) were irradiated to 1×10^{10} rad (at 720 rad/sec); the leach rates of the irradiated and unirradiated samples were found to be almost the same /65/.

From the consistent results obtained at different institutions using BWPs of widely different compositions one may conclude that the radiation dose (specific activity) has only an insignificant influence on the leach rate of BWPs (of less than 1 Ci/l specific activity), and that the maximum permissible specific activity of BWPs is limited more by the radiolysis gas generation than by the leach rate.

h) Influence of the leachant

The leach rates decrease as the pH of the leachant increases.

For "identical" samples the leach rates are somewhat lower in sea-water than in ordinary water. The leach rates are normally lower in stagnant water than in running water; leaching of BWP of a low content of solids (up to about 20 wt.-%) may show the same leach rate under static and dynamic conditions.

BWP prepared from BWR-wastes are less leachable by ground water which has been in "equilibrium" with concrete than by drinking water or demineralized water /280/.

As examples of possible leachants in a repository a probable composition of a ground-water (in Sweden) and the composition of a natural water after 1 weeks' contact with granular concrete are given in Table XXV and Table XXVI respectively.

TABLE XXV

PROBABLE COMPOSITION OF GROUND-WATER IN CRYSTALLINE ROCK AT GREAT DEPTH /R-21/

Analysis	Probable range mg/l	Max value mg/l
pH	7.2 - 8.5	9.0
$KMnO_4$ consumpt.	20 - 40	50
COD_{Mn}	5 - 10	12.5
Ca^{2+}	25 - 50	60
Mg^{2+}	5 - 20	30
Na^+	10 - 100	100
K^+	1 - 5	10
Fe-tot.	1 - 20	30
Fe^{2+}	0.5 - 15	30
Mn^{2+}	0.1 - 0.5	3
HCO_3^-	60 - 400	500
CO_2	0 - 25	35
Cl^-	5 - 50	100
SO_4^{2-}	1 - 15	50
NO_3^-	0.1 - 0.5	2
PO_4^{3-}	0.01 - 0.1	0.5
F^-	0.5 - 2	8
SiO_2	5 - 30	40
HS^-	< 0.1 - 1	5
NH_4	0.1 - 0.4	2
NO_2	< 0.01 - 0.1	0.5
O_2	< 0.01 - 0.07	0.1
(Conductivity	400 - 600 µS/cm	1100 µS/cm)

TABLE XXVI

COMPOSITION OF A NATURAL WATER BEFORE AND AFTER 1 WEEK CONTACT WITH CONCRETE /280/

Analysis	Before contact mg/l	After contact mg/l
pH	6.8	12.2
Ca^{2+}	33	576
Mg^{2+}	8.6	0.04
Al^{3+}	0.014	0.055
HCO_3^-	275	2260
Cl^-	140	160
CO_4^{2-}	(< 1)	(2)
SiO_2	16.1	1.3
(Conductivity	700 µS/cm	7050 µS/cm)

i) Influence of the temperature

An increase in temperature will normally significantly increase the leach rate of soluble substances as both the solubility and the diffusion rate of these substances are influenced by the temperature.

There exist only a few data on the leach rate of nuclides from "senseful" bitumen-waste mixtures which, unfortunately, appear to be unreliable. In tests, using various sodium chloride-bitumen mixtures (mainly 70 wt.-% NaCl/30 wt.-% bitumen), it was found that the leach rate was nearly twice as high at 40°C as at 15°C /R-19/.

Using 60% Mexphalt 10/20 and 40% calcium phosphate (as coprecipitant for Sr-90) and also copper ferrocyanide (as coprecipitant for Cs-137) as sample mixture for leach tests no difference of the leach rate at 50°C and at room temperature could be observed /66, p. 49/.

j) Improvement of the leach resistance

Apart from the factors already mentioned in the foregoing subchapters (e.g. chemical pretreaments, high bitumen to waste ratio, low water and soluble salt content, etc) two additional measures can reduce the overall leachability of BWPs:

- the design and material of the container (see chapter 5.4), and

- an additional protective coating of the product.

The considerable reduction of leachability of BWPs by their coating with pure bitumen has been verified at various institutions (Eurochemic, KfK, CEA-Marcoule).

It could, for example, be shown that a 6 mm thick layer of pure bitumen H 80/90 and H 120/130 prevented completely the leaching of radionuclides from BWPs during 1220 days (3.3 years) immersion in water /139 a/. In other leach tests using BWPs coated with a pure bitumen layer of 5 mm thickness no leaching of salts took place during 2.5 years /113/; after 5 years a leach rate of 3×10^{-8} g . cm^{-2} . d^{-1} was found as compared to 10^{-4} to 10^{-5} g . cm^{-2} . d^{-1} for samples without this coating by pure bitumen /186/.

k) Conclusions

The steady-state leach rates of the major "insoluble" fission products (Ru-106, Sr-90, rare earths) and of the actinides from well-prepared BWPs containing up to 60 wt.-% solids are normally between 10^{-5} to 10^{-7} g . cm^{-2} . d^{-1}.

The leach rates of "soluble" waste constituents (sodium, Cs-137) are in the order of 10^{-4} g . cm^{-2} . d^{-1}.

If a special chemical pretreatment is performed to insolubilize the long-lived radionuclides one may obtain for the most important nuclides the following leach rates in demineralized water (according IAEA-standard procedure) /82, 139 a/:

Cs-137 : $1.5 \times 10^{-7} - 3 \times 10^{-8}$ cm/day

Sr-90 : $1 \times 10^{-5} - 3 \times 10^{-6}$ cm/day

Ru-106 : $1 \times 10^{-5} - 3 \times 10^{-6}$ cm/day

Co-60 : $3 \times 10^{-4} - 1 \times 10^{-5}$ cm/day

Pu-239, Am-241 : $1 \times 10^{-8} - 1 \times 10^{-9}$ cm/day

All these leach rates can be considered to be sufficiently low for the safe long-term storage of BWPs.

6.3.3 Radiation Stability and Effects

a) General

In judging the validity and reliability of the results given hereafter one has to keep several facts in mind; some of the more important ones will shortly be mentioned to avoid misinterpretation of the results expecially those given in a concise form.

Factors which influence the radiation stability of bitumen (and high polymer substances in general) are:

- chemical structure (bitumen type),
- additives (salts, other fillers, solvents, etc..),
- type of radiation,
- integral dose,
- dose rate,
- temperature,
- surrounding medium (air, nitrogen, vacuum, minerals etc..),
- specific surface,
- mechanical forces during irradiation.

The absorption of high-energetic radiation effects ionization and excitation of molecules of the irradiated products resulting mainly in the formation of highly reactive free radicals.

These free radicals cause changements of the physical and chemical properties due to the following reactions:

- degradation of large molecules,
- cross-linking of molecules,
- chemical reaction with the incorporated waste solids and the environmental medium, (e.g. air), and the
- formation of low molecular weight products, e.g. gaseous products.

Radiolytic gases are thus secondary reaction products of radiation induced radicals.

The gases (H_2, CH_4, C_2H_x etc..) formed effect inner stresses and fissures, and sometimes swelling, especially at elevated temperatures. The reactions with the surrounding (gaseous) medium during the irradiation is of great practical importance.

Irradiations up to about 10^8 rad may result in products of improved properties whilst at very high irradiation doses ($> 10^9$ rad) not only bitumen but almost all polymers are converted to technically useless materials. Results on the radiation stability based on short-term irradiations at high dose rates are of limited value for the evaluation of long-term radiation effects of the same integral dose achieved at much lower dose rates. The results of short-term irradiations lead usually to wrong conclusions about the real radiation stability of the products tested. The presence of oxygen (air) during the irradiation may inhibit the cross-linking and enhance the degradation (e.g. by formation of ketogroups). The presence of distilled water instead of oxygen at low dose rates has no measurable influence on the mechanical properties of the irradiated products.

Generally, the content of radiation-resistant waste components (e.g. inert salts), increases the radiation resistance while the incorporation of non-radiation-resistant material decreases the overall radiation stability of the BWP.

The chemical interaction of radiolysis products with the waste components can lead to further radiation-chemical effects which can improve as well as worsen certain physical and chemical properties of the BWP.

One should aim at to define the radiation stability not for the product as a total but in relation to each of its properties of importance for its short- and long-term storage.

Statements about the radiation stability are only senseful for practical purposes if the conditions applied are clearly specified.

For practical purposes the radiation effects can most easily be characterized by the gas formation and the swelling (or shrinkage) of the BWP.

The irradiation of distilled as well as oxidized bitumens results mainly in the generation of hydrogen: besides hydrogen small fractions of hydrocarbons (with 1 - 4 C-atoms) and carbon oxides are formed. Hydrogen is known to be a product of the cross-linking mechanism, whilst hydrocarbons would predominate if degradation were the main process. Carbon oxides are formed when primary aliphatic radicals react with oxygen. Therefore, in an argon atmosphere no carbon oxides but a higher fraction of hydrocarbons are formed; the degree of hydrogen formation remains unchanged by the irradiation in air or argon atmospheres /289/.

The G-values* for the total gas evolution from bitumens are between 0.4 to 0.5, i.e. relatively low so that part of the radiation energy seems to be absorbed by excitation without chemical transformations.

b) Generation of radiolysis gas and swelling

The main results obtained at various institutions and conclusions derived from published works, are listed hereafter:

- the principal radiolysis products are:

  o hydrogen (H_2)
  } ≈ 75 - 95% of the total gas evolution
  o methane (CH_4)

  o ethane (C_2H_6) ≈ 2 - 8%

  o ethylene (C_2H_4)

  o $C_3H_x+C_4H_x$ ≈ 1%

  o carbon monoxide (CO)

  o carbon dioxide (CO_2)

  o water (H_2O) ≈ 0 - 20% /230 a/;

- the type of radiation (α, β, γ) has no significant influence on the generation rate of radiolysis gases and swelling rate;

- the generation rates of hydrogen for internal radiation (by Sr/Y-90) are two to three times higher than those for external irradiation, however, of the same order of magnitude /92/;

- the results of external irradiation tests are in most cases transferable to radioactive BWPs (i.e. internal irradiation) /92/;

- there seems to be a slight tendency that products with harder bitumens show a smaller rate of generation of radiolysis gases and swelling than softer ones under comparable conditions;

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* G-value = moles produced per 100 eV absorbed.

- the influence of particular substances (e.g. nitrates) is not well defined;
- the amount of radiolysis gases (mainly hydrogen) generated in samples placed in an inert atmosphere (argon) increases proportional with the dose absorbed and has been found equal /289/ or higher than in the presence of air /92/;
- the hydrogen generation rate is a linear function of the integral dose between 1 - 100 Mrad and almost independent of the dose rate;
- irradiation with 10 MeV-electrons of bitumen yielded 5.6 x 10^{-3} cm^3 H_2/g . Mrad and bitumen/50 wt.-% $NaNO_3$ 3 x 10^{-3} cm^3 H_2/g . Mrad, similar values were obtained for γ-irradiations /139/;
- the swelling rates of bitumen B 45 (distillation bitumen) were found to be approximately 20% of the generation rate of hydrogen, whilst the swelling rates of bitumen B 45 - $NaNO_3$ (30 - 40%) mixtures were of the same order of magnitude as the generation rates of hydrogen /283/;
- bitumen B 45 and bitumen B 45/30-40% $NaNO_3$ mixtures showed a hydrogen generation rate of about 10^{-2} cm^3/g . Mrad and a swelling of blocks of about 2 x 10^{-3} cm^3/g . Mrad /92/;
- BWPs containing 40% Li- and borate-loaded ion exchange resins, 50% bitumen (B 15), and 10% water showed, after irradiation to a dose of 80 Mrad, a porosity*) change of 1 - 4% and a hydrogen generation rate of 0.3 cm^3/g . 80 Mrad (at 760 torr, 25°C) /283/;
- partially dried, uncoated ion exchange resin, loaded with lithium and borate and containing 24% residual water showed a hydrogen generation rate of 0.14 cm^3/g . 100 Mrad (at 760 torr, 25°C) /283/;
- when bitumen-nitrate salt mixtures are irradiated also nitrogen oxide (NO) is found among the radiolysis gases, while a wider spectrum, NO, NO_2, methyl amine and trimethyl amine, is formed in mixtures containing anion exchangers, however, at a very low rate so that relative to the pure matrix the total amount of hydrogen is not affected by the additives /289/.
- bitumen (B 15)/42% $NaNO_3$ mixtures irradiated with 10 MeV-electrons to a dose of 80 Mrad resulted in a change of porosity of about 18% /283/;

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*) porosity P (in %) is defined as

$$P = 1 - \frac{\text{product density (irradiated)}}{\text{product density (unirradiated)}} \times 100$$

- the absorbed integral dose of a BWP of 1 Ci/l (containing fission products typical for this waste from reprocessing plants, obtained 2 years after reactor discharge of the fuels processed) amounts to 90 Mrad (9×10^7 rad) after 200 years; after this time the integral dose approaches asymptotically the end value of total decay of the fission products /283/;

- the release rate of radiolytically generated gas for a given integral dose is the lower the higher the dose rate (specific activity), e.g. 0.28 $cm^3/Ci \cdot d$ for 10 rad/s and 0.01 $cm^3/Ci \cdot d$ for 600 rad/s using 60% bitumen/40% $NaNO_3$ mixtures /42/;

- a G-value of 0.41 for H_2 formed from distilled bitumen (60/80) and 0.46 for H_2 formed from blown bitumen (20/30) has been determined /230 a/, (G_{H2} = 2.4 - 2.7 for polyethylene; G_{H2} = 0.044 for benzene; G_{HCl} = 0.3 for polyvinylchloride);

- from the point of view of radiation resistance the tested bitumens can be used to process liquid waste with a specific activity of at least up to 10 Ci/liter without any significant effect on the water resistance, leachability, and changes in the structure of the BWPs during long-term storage /R-20/.

c) Changement of the leach rate and other properties

The irradiation of a wide variety of BWPs have shown that:

- irradiations up to about 10^8 rad do not lower the leach resistance of the products or only to a small extent; based on the highest observed changes waste-bitumen blocks with a specific activity of 1 Ci/l need at least 50 years to observe a leach rate increase by a factor of 5 - 10 /66, p. 23/;

- doses up to 10^8 rad do not change the inflammation temperature of BWPs;

- a significant change (increase) of the softening points and consequently of the visco-elastic properties of bitumens and BWPs (60% bitumen/40% salts) takes only place at doses exceeding 10^8 rad /283/;

- an increase from 1×10^8 to 5×10^8 rad increases the "ring and ball" softening point (hardening) by 10 - 30°C /66, p. 17/;

- for a product composed of 42% $NaNO_3$, 0.05% NaOH, 0.75% H_2O, rest bitumen (Ebano 15) the porosity change amounted to less than 1% at a dose of 10 Mrad and about 26% at 500 Mrad /283/.

d) Conclusions

From the numerous investigations on the radiation-chemical stability of bitumen and bitumen/salt mixtures one may conclude for all practical purposes the following:

- the irradiation of bitumen and waste-bitumen mixtures by alpha-, beta-, or gamma-radiation results (- as far as gas formation is concerned -) almost exclusively in the generation of hydrogen at a rate of 0.3 - 1.0 cm^3/g . 100Mrad, nearly independent of dose rate, atmosphere (air, argon, nitrogen) and bitumen type;

- the swelling rate is in the same order or lower, i.e. less than 1 cm^3/g . 100 Mrad;

- for most estimations and products containing 45% solids a hydrogen generation rate of 0.6 cm^3/g . 100 Mrad can be used and 0.1 cm^3/g . 100 Mrad for other gases formed;

- the total amount of gases generated during infinite time is proportional to the total exposure dose /230 a/;

- the absorbed integral dose of most BWPs containing initially less than 1 Ci/l β-γ-emitting fission and activation products will be lower than 10^8 rad;

- absorbed doses of $\leqslant 10^7$ rad cause negligible swelling, gas evolution, hardening, or increase in leach rate;

- the presence of solid or solidified waste components does neither worsen the radiation stability of bituminous materials nor improve it, i.e. they act as "inert" diluents, and the amount of total gas evolution is governed solely by the quantity of bitumen contained in the BWPs;

- upon irradiation under comparable conditions bitumen generates less hydrogen than polyethylene;

- inspite of differing results of some investigators one may state that oxidized bitumens are more radiation resistant than other types of bitumen;

- internal radiation doses of up to 1 x 10^9 rad cause no significant changements of blown bitumen-waste mixtures, i.e. that from the point of view of radiation resistance all types of liquid ILW can be solidified by incorporation into blown bitumen.

6.3.4 Thermal stability and effects

The question of the combustibility and explosiveness of BWPs has to be considered in relation to external heat and ignition sources; it is therefore of special importance during the preparation of the BWPs, their interim-storage and transport to the final storage or disposal place.

For the long-term storage the internal heating of the BWPs caused by the decay of the incorporated radionuclides, has expecially to be taken into account besides the formation of radiolytic gases, i.e. the maximum allowable specific activity has to be defined in relation to the storage conditions in order to avoid the formation of an ignitable radiolytic gas/air mixture and an intolerable heating (fluidity) of the BWPs.

It is essential that the temperature difference between the centre and the wall of a BWP block is low. This means that the specific activity permissible in the final product depends mainly on heat transfer conditions between the BWP and the container nature and thickness of the container and the environmental conditions.

The heat conductivity changes slightly with the temperature and increases with the increase of solids in bitumen, it is in the order of 0.25 W/m.°C.

Calculation of decay heat shows that for BWPs of a specific activity of 10^3 Ci/m^3 the specific heat release is 3-5 W/m^3 and that the self-heating will not practically limit the method of disposal.

The temperature of BWPs containing fission products will not surpass 70°C in the centre of a 200 l drum at a specific power of 0.05 W/l.

The storage temperature should keep the elasticity of bitumen and limit the segregation of bitumen and incorporated solid materials. For the system bitumen-sodium nitrate the storage temperature should not exceed 60°C /43/.

Considering BWPs of the ILW category (≤ 10 Ci/l) contained in 200 l drums one can state that the power release of the radionuclides will be of the order of 0.05 W/l causing a temperature in the centre of the block of less than 70°C. At temperatures below 70°C no significant segregation and migration of radionuclides exist for almost all BWPs and also no spontaneous inflammation has to be feared.

If the BWP in a 220 l standard drum possesses a specific activity of 10 Ci/l the temperature difference between the centre and the outside surface of the product would not exceed 4°C; when storing such a product in a bunker the temperature in the centre will be lower than 35°C.

The ignition points as well as the appearance of exothermal reactions are far beyond the bituminization temperature of reactor and reprocessing wastes.

6.3.5 Sedimentation and homogeneity

a) General

The solids in BWPs tend to settle to the bottom as the bitumen mass liquefies at higher temperatures.

The slow cooling down of hot mixtures of bitumen and waste solids (2 - 3°C/h) could cause inhomogeneous products due to particle sedimentation. The settling could lead to the formation of concentrated salts-bitumen mixtures.

Thus, the incorporation of oxidants such as nitrates may be suspect since they could be concentrated by settling.

b) Experiments and results

Experiments carried out at Eurochemic showed that there is no significant concentration change of sodium nitrate in the bitumen matrix upon cooling down 190 l of a mixture of 55 wt.-% distilled bitumen/45 wt.-% sodium nitrate in a 220 l product drum from 180°C down to room temperature at an average rate of 2 - 3°C/h.

Experiments carried out in Sweden /280/ showed clearly settling of ion-exchange resins during cooling down of a cast bitumen-resin mixture. The problem of sedimentation of salts during the storage of BWPs has been studied in the United States /51/, in France /139 a/, in the Federal Republic of Germany /283/, and at Eurochemic.

In the experiments carried out in France different BWPs were stored at 30°C, 62°C, and 105°C for one year. The particle size of the incorporated solids was in the range of 1 - 40 µ. It could be shown that the salts do not settle if the storage temperature remains more than 20°C below the softening point of the BWP.

For a large number of BWPs the softening point (R + B) is around 115°C which usually increases with time due to the radiation and other effects causing hardening.

Depending on the type of bitumen used and the radiation dose absorbed the softening point of BWPs may be in the range of 60°C - 160°C. Calculations have shown that the sedimentation rates of 30 µ particles in bitumens (of softening points of around 100°C) at temperatures up to 40°C are $\leq 5 \times 10^{-3}$ mm/year; i.e. the particles settle not more than 1 mm in 200 years /283/.

c) Conclusions

Homogeneous bitumen-waste products should be stored at temperatures below 60°C or at least 20°C below their softening point in order to avoid settling of the incorporated solids and therewith significant inhomogeneities within the product.

6.3.6 Combustibility and explosiveness

In view of the long-term storage of large volumes of BWPs the knowledge about their inflammability and explosiveness is required.

Especially the safety aspects of producing and storing BWPs containing high concentrations of nitrate must be carefully assessed.

Therefore, relevant investigations were carried out in various countries part of which are summarized hereafter.

a) Sovjet Union: test results

In the Sovjet Union teste were made on the explosiveness of bitumen-sodium nitrate mixtures using 7, 15, 20, and 30 wt.-% bitumen (type BNK-2 and BN-3) /273/.

The aim of the tests was to establish the limit of the bitumen content at which the mixture becomes incapable of detonating.

The results showed that mixtures containing 15 wt.-% or less bitumen are capable of detonating whereas mixtures containing more than 15 wt.-% bitumen do not detonate.

This required ratio does not change when the mixture is heated to 60 - 80°C (to take into account the possible heating by the decay of the radionuclides).

The consistency of the mixtures has a substantial influence on their detonability. The potentially most powerful mixtures of stoichiometric or near-stoichiometric composition (15% bitumen) which constitute dense viscous pastes were shown to be either at the detonation limit or incapable of detonating. At the same time, a mixture of 7 wt.-% bitumen and 93 wt.-% sodium nitrate was considerably weaker in respect of energy index but, having a powdery consistency, detonated more easily.

This dependence of detonability on consistency is evidently characteristic of mixtures with various aggregate states of fuel and oxidant.

In addition the explosiveness of mixtures of nitrates of sodium, potassium, and calcium with other organic substances such as TBP, kerosene, toluene, naphthalene were studied. The results showed that all systems are characterized by narrow limits of detonability: from 6-7 to 10-15 wt.-% content of organic substances. The lower limit of detonation is evidently restricted by energy factors (specific content of latent energy in the system) and the upper limit, by the consistency of the system which hinders the propagation of the detonation wave. It may, therefore, be expected that variations in the character of the organic binder and the salt composition of nitrate wastes do not significantly affect this limit of detonability.

Furthermore the combustibility of bitumen (BNK-2) - waste mixtures containing 40 and 60 wt.-% sodium nitrate or 40 and 60 wt.-% of a mixture simulating a reactor waste has been determined. It was found for all these mixtures that the flash point and the burning temperature are close to the corresponding values for the pure bitumen (293°C and 345°C respectively), and that the self-ignition temperature was higher than 500°C.

It has been concluded from these investigations that the incorporation of fillers (waste residues) into the bitumen does not increase its fire hazard and that the probability of fire and explosion is small if the established conditions for the safe preparation and storage are observed.

c) United States: test results

Tests performed in the USA by the Bureau of Explosives (New York City) on bitumen products containing 60 wt.-% solids (including 35 wt.-% nitrate) indicated that the material did not fall within any of the classes of dangerous goods as defined by Interstate Commerce Commission regulations /24, 35/.

At ORNL /24, 35/ the following observations were made:

products containing 60 wt.-% salts heated in air to 550°C did not ignite. Even with an ignitor wire the product could not be ignited in the presence of nitrogen up to 535°C.

The burning tests on the product have shown that it does burn vigorously but does not explode or show a burning rate significantly higher than that for pure bitumen.

d) Federal Republic of Germany: test results /69, 194, 283/

The investigations were performed mainly with BWP containing 60 wt.-% nitrates of various metals and other additives that were expected to be potential constituents of evaporator concentrates. Generally, the ignition temperatures of the products were found to be above 400°C; only some samples with an increased fraction of calcium nitrate and transition metal nitrates showed ignition temperatures between 360 - 380°C.

In a red hot steel pan (700°C) BWPs ignited in a shorter time than pure bitumen (ca 10 times faster), however, the burning periods were of the same order of magnitude.

Pure bitumen burns smoothly with the formation of black smokes and a relatively large residue of coke whilst the BWPs burn partly vigorously generating only a small residue.

Rapid heating in a closed system showed that there is no hazard of explosion to be feared. The BWPs were not sensitive to mechanical or detonation impacts. The products could not be ignited by exploding hydrogen/air mixtures.

The total combustion time of 211 kg BWP composed of 57% bitumen and 43% sodium nitrate placed in a 175 l drum was 85 minutes; about 27% of the sodium nitrate was carried away with the fumes.

Carbon dioxide was demonstrated to be the most suitable extinguishing material.

The burning points of 50 wt.-% bitumen and LWR-wastes (spent mixed-bed ion exchange resins, evaporator concentrates with a high content of detergents or boric acid) were well above 400°C.

BWPs from reactor stations burn smoothly (as usually nitrates are absent) and comparable to pure bitumen.

The investigations demonstrated clearly that the BWPs tested do not belong to the category of explosive or easily inflammable substances. Fire tests, in which 175 l drums with BWPs were placed in concrete containers of 20 cm wall thickness and then put for 50 minutes in a vigorously burning oil fire, showed that a concrete shielding container represents an efficient protection against exterior fires.

e) France: test results /176/

All BWPs show a normal combustibility and are not explosive and do not detonate.

The spontaneous inflammation temperature of the majority of BWPs is higher than 350°C.

The risks of exothermic reactions between oxidizing components of the waste concentrates and bitumen were studied by means of differential thermal analysis (DTA); it has been found that:

- in the absence of ammonium ions, the temperature at which the decomposition of the majority of the BWPs starts is 290°C; this temperature remains unchanged if the following weight percentages of the various salts-related to the total amount of the dry, incorporated residues - are not exceeded:

  25% ammonium nitrate,

  37.5% potassium chromate,

  37.5% potassium chlorate,

  45% potassium dichromate.

The presence of the following constituents does not influence the thermal stability of the bituminized waste concentrates: nitrite ions, sodium nitrate, sodium borate and calcium borate.

Note: Tests carried out upon request of the Eurochemic Company have shown that explosion-like reactions have been observed at 70 - 130°C when more than 12 wt.-% of each of ammonium nitrate, sodium nitrate, and sodium nitrite were simultaneously present in the solid BWP. However, such mixtures cannot be obtained under real process conditions; it is mentioned only to avoid misinterpretation of the results obtained in France.

f) Conclusions

Bitumen-waste products containing up to 60 wt.-% residues from reactor and reprocessing wastes do not explode or detonate during handling, transport and storage; they do also not burn if they are not heated above 300°C and/or an ignition source is absent.

Although a content of nitrate facilitates and accelerates combustion, it is not sufficient to increase the rate of combustion significantly. Maximum values will possibly be obtained only with an even oxygen balance, i.e. a sodium

6.3.7 Mechanical Stability

a) General

The mechanical properties of bitumen-waste mixtures are of importance during handling, transport and storage.

After incorporation of the waste into bitumen at elevated temperatures, the resulting hot mixture is poured into the final product container, usually a 220 l drum. The cooling rate of the BWP in the middle of a 220 l drum from the temperature of the filled drum (ca 160-190°C) down to near cell temperature (ca 20-30°C) will be between 1.3 - 3°C/h, or about 2°C/h on the average.

As the softening point of 50/50 wt.-% BWPs is in the order of 100°C, it takes about 1.5 - 2 days until a completely solidified product is formed. After less than 24 hours cooling at ambient temperature the thickness of the solidified outside layer will normally be large enough (> 10 cm) to permit drum transfer operations without risking a flowing out of BWP in case the drum should fall.

The mixing of the waste solids with bitumen effects an increase of the softening point and a decrease of the penetration and ductility of the bitumen. The reactions that occur during mixing, binding and adhesion are exceedingly complex, involving viscosity, plasticity and the structure of the solids.

The mechanical stability of BWPs can be influenced within a relatively wide range by the choice of the bitumen type, nature of waste solids and additives, and the solids to bitumen (volume) ratio.

b) Results

The mechanical behaviour has been investigated by storing BWPs under static pressures at 25, 30, and 40°C /54/; it was found that all samples were more or less deformed in dependence of the softening point of the various mixtures. Also mixtures of softening points above 110°C showed no form-stability during longer periods of storage under pressure.

The incorporation of ion exchange resins strongly increase the viscosity and the form stability of bitumens, especially of blown bitumen. It has been observed that the standard "penetration" and "Ring and Ball" methods, do not give consistent results for bitumen-resin mixtures; therefore tests (the "cylinder bending test" and the "hole migration test") were developed to measure the form stability of bitumen-waste products /187/.

The effects of continued exposure to - 40°C and of temperature cycles between - 40 and + 20°C on the mechanical properties and leachability are also investigated /187/; so far no adverse effects of this temperature treatment could be observed.

In connection with the sea disposal of conditioned LLW and the standardization of BWP packages (200 l) research work is carried out in Japan /R-32/ to produce waste packages of sufficient strength towards various impacts including hydraulic pressures of up to 700 kg/cm^2 (equivalent of the hydraulic pressure at a sea depth of about 7 000 m).

Using BWPs containing 50 - 70 wt.-% bitumen and the respective amount of waste solids contained in LWR wastes (20 wt.-% spent ion exchange resins, 10 wt.-% filter aids, rest consisting of 20 - 50 wt.-% solid residues from evaporator concentrates) the following preliminary results were obtained:

- ion exchange resins and filter aids have to be mixed with evaporator concentrates to reach the required specific density of at least 1.2;
- the BWP packages are not destroyed by up to 700 kg/cm^2 hydrostatic pressure (provided that proper bitumen/solid ratios and manufacturing are applied);
- free drops in air of BWPs from a height of 1.2 and 9.0 m have shown that BWPs are more resistant against drop impact (than cement-waste products) due to their plastic nature and will resist to the impact (at the water surface and the sea bottom) when the package is sea-disposed;
- packages dropped from a height of 9 m were deformed, but no dispersion of waste could be found;
- certain bitumen-waste mixtures would collapse and swell when sea-dumped; to avoid this the fraction of sodium sulfate, sodium borate, or sodium nitrate should not exceed 40 - 50 wt.-%.

c) Conclusions

The mechanical properties of BWPs depend on the volumetric composition of the mixture (i.e. solids-, bitumen-, and void content), the type of bitumen (asphaltene content and penetration), the temperature, and some characteristics of the incorporated solids (e.g. particle size, porosity).

Thus, the mechanical stability can be influenced within a wide range by the proper choice of the components and the conditions of mixing and storage.

6.3.8 Swelling

a) General

There are three main causes for the swelling (volume increase) of stored BWPs:

- formation of gases inside the product due to radiolysis,
- chemical reactions, and
- uptake of water from the surrounding by the product.

The degree of swelling is determined in the first case by the specific activity, and in the second and third case by the chemical composition of the product and its extent of contact with water of the immediate surrounding.

The rate of swelling caused by radiolysis gases is lower than the generation rate of radiolysis gases, usually by a factor of 1.5 to 7. The rate of swelling of BWPs is in the order of 0.2 $cm^3 \cdot g^{-1} \cdot 100\ Mrd^{-1}$. It has to be emphasized that the volume increase cannot be used as a measure of the radiation stability of a BWP.

Incorporated substances causing swelling of the BWPs are:

o anhydrous salts capable of binding larger amounts of water (e.g. crystallization water) under volume increase;

o detergents in larger amounts (> 6 wt.-%);

o dry ion exchange resins exceeding 50 wt.-% in the product.

It is possible that (some) swelling may occur due to chemical reactions leading to the evolution of gaseous reaction produts (e.g. acidic residual water reacting with incorporated carbonates).

b) Results

It has been observed that:

- samples containing 38 wt.-% sodium carbonate swelled to twice its original volume after 15 days immersion in water /66, p. 111/;
- samples containing 17 - 44 wt.-% sodium sulfate swelled in distilled water and ground water under formation of the decahydrate so much that complete destruction occurred within one to two weeks /265/;
- products containing 60 wt.-% salts (principally Na_3PO_4) led to gradual swelling after 30 days and subsequent unacceptably high leach rates /R-38/;
- even a product containing only 11 wt.-% salts (mainly sodium phosphate swelled after about 6 months immersion in water at 25°C /R-38/;

- samples containing 70 and 80 wt.-% dry ion exchange resins swelled even upon storage in the air /187/.
- samples containing 50 - 60 wt.-% dry ion exchange resins swelled to twice its original volume after 2 - 3 months immersion in distilled water /R-31/.
- the maximum swelling in water of ion exchange resins of a cross-linking of ⩾ 8% DVB is about 1.4 times higher for irradiated resins (cation and anion exchanger) than for non-irradiated resins /R-30/.

c) Conclusions

Dimensional changements of stored BWPs should be as low as possible. Among others, the following measures can reduce the risk of an excessive volume increase:

- anions forming alkali salts of a high crystal water content should be converted into water-insoluble salts of no or low water content,
- the content of water-attracting (hygroscopic) substances should be kept low,
- a layer of inactive bitumen retards and reduces the penetration of water,
- blown bitumen could be used if the integral radiation dose will be higher than 5 x 10^7 rad.
- an extended contact time of ion exchange resins with bitumen at the incorporation temperature leads to partial thermal destruction and "dissolution" of the resins by bitumen resulting in a decreased swelling in water.

7. INTERIM STORAGE

7.1 General

A safe interim storage on-site or off-site of the conditioned waste has to be assured until final storage (disposal) facilities accepted by regulatory authorities are available and put into operation. The interim storage facilities are usually man-made structures which have to assure that

- the stored waste products must remain easily retrievable for the further transport to the final storage place;
- dispersion of radionuclides to the environment by air, water, or animals is prevented;
- the accumulation of a hazardous amount of an explosive gas (radiolysis gas) mixture is excluded;
- sufficient shielding towards the accessible environment is provided;
- the required safety is guaranteed during the actual interim storage period.

Bituminized LLW and ILW placed in 220 l drums require no forced cooling when stored in a properly designed interim storage facility.

7.2 Storage Facilities

The construction of the interim storage facility has to be made according to the waste products to be stored, their amounts and the envisaged storage time.

The retrievable storage structures for bituminized waste include engineered storage facilities, - usually located on or near ground surface, - and individually shielded (concrete) casks which can be placed in a light building structure.

Near surface storage facilities require special safety considerations against damage from hostile acts of man and nature.

The storage facility must have the capability to receive, to transport, and to position at the forseen place the product package; it should be possible to monitor, to inspect, and to repack the waste products, if required.

Interim storage facilities of varying capacity exist on all sites where intermediate-level radioactive wastes are bituminized, e.g. in Marcoule (France), at the Nuclear Research Centre in Karlsruhe (FRG), at Barsebäck (see Fig 6 a) and Forsmark (Sweden), and at Eurochemic in Mol (Belgium).

Eurochemic's interim storage facility for solid and solidified intermediate-level wastes from fuel reprocessing, named Eurostorage, appears to be at present the largest and most elaborated engineered surface storage of its kind.

The Eurostorage facility, shown in Fig. 12 with two bunkers only, is built adjacent to Eurochemic's bituminization plant: it consists essentially of:

- a series of concrete storage bunkers, each 64 m long, 12 m wide and 8 m high, capable of storing up to about 5000 drums of 220 l;
- a corridor connecting the bituminization plant and the storage bunkers;
- a cell for each two bunkers housing the bunker ventilation equipment;
- a transfer station for solid and solidified ILW arriving by truck transport and coming from other conditioning installations than the bituminization plant;
- a control station housing the control room and rooms for auxiliaries (fans, filters, heating, electrical switch boards, etc..).

The control room is situated on the first floor above the junction corridor permitting a direct visual control through lead-galass windows of the remote operations in the corridor and the transfer station.

The storage bunkers are constructed in reinforced and partly in prostressed concrete. The wall, floor and roof thickness is dimensioned according to the required structural strength or radiation shielding; special care is being taken to ensure tightness of the bunkers against rain and ground water.

The majority of the products to be stored consist of bituminized ILW solutions from the chemical decladding process and concentreted waste solutions from the Purex process and decontamination operations. The product drums are filled to 80% of their capacity and are made of "chromized steel" (see chapter 5.4 and Fig 2).

The waste-bitumen mixtures contain $\leq$ 45 wt.-% salts or other waste solids, $\geq$ 55 wt.-% blown bitumen (Mexphalt R 85/40), $\leq$ 0.5 wt.-% water; they have a density of 1.30 - 1.40 g/cm^3 and a specific activity of maximum 1 Ci/l. The radiation exposure rate at the drum surface is about 220 R/h, while the total decay heat produced in one storage bunker, containing 5000 BWP drums, amounts tomaximum 2.7 kW.

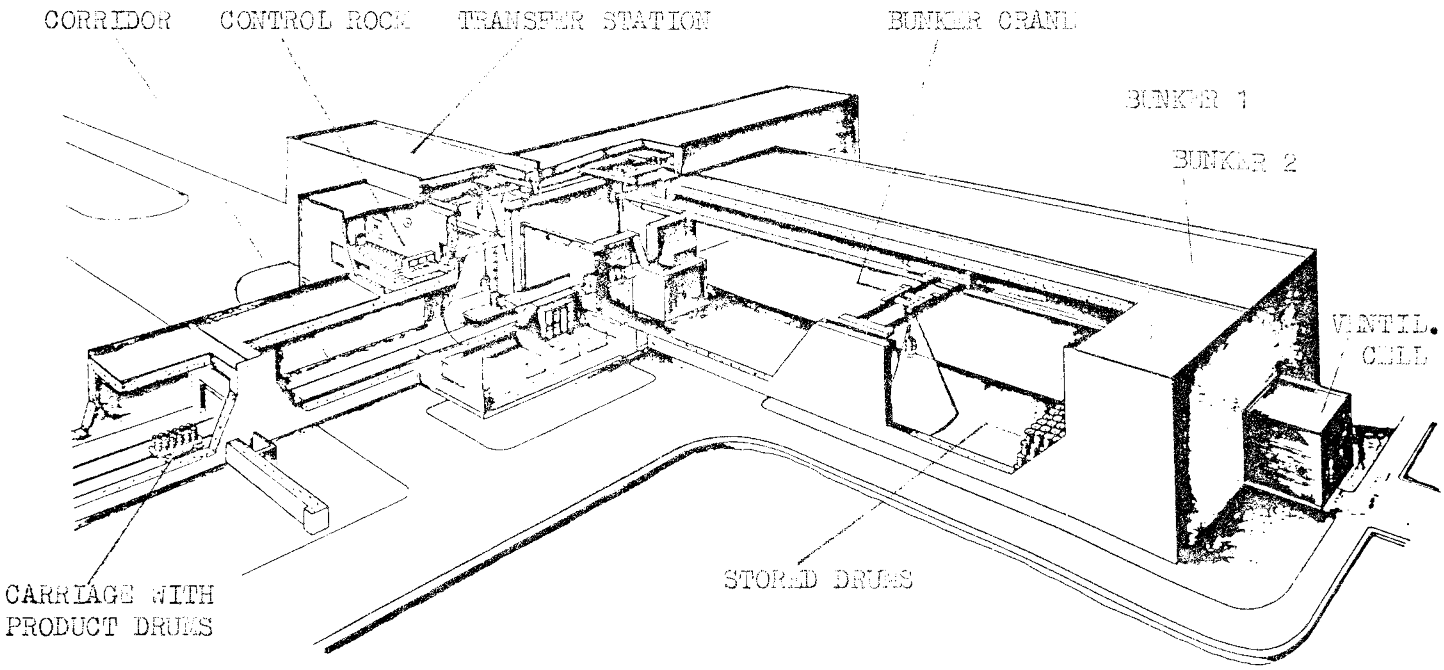


Fig. 12 - EUROCHEMIC'S INTERIM STORAGE FACILITY FOR SOLID AND SOLIDIFIED INTERMEDIATE-LEVEL RADIOACTIVE WASTES

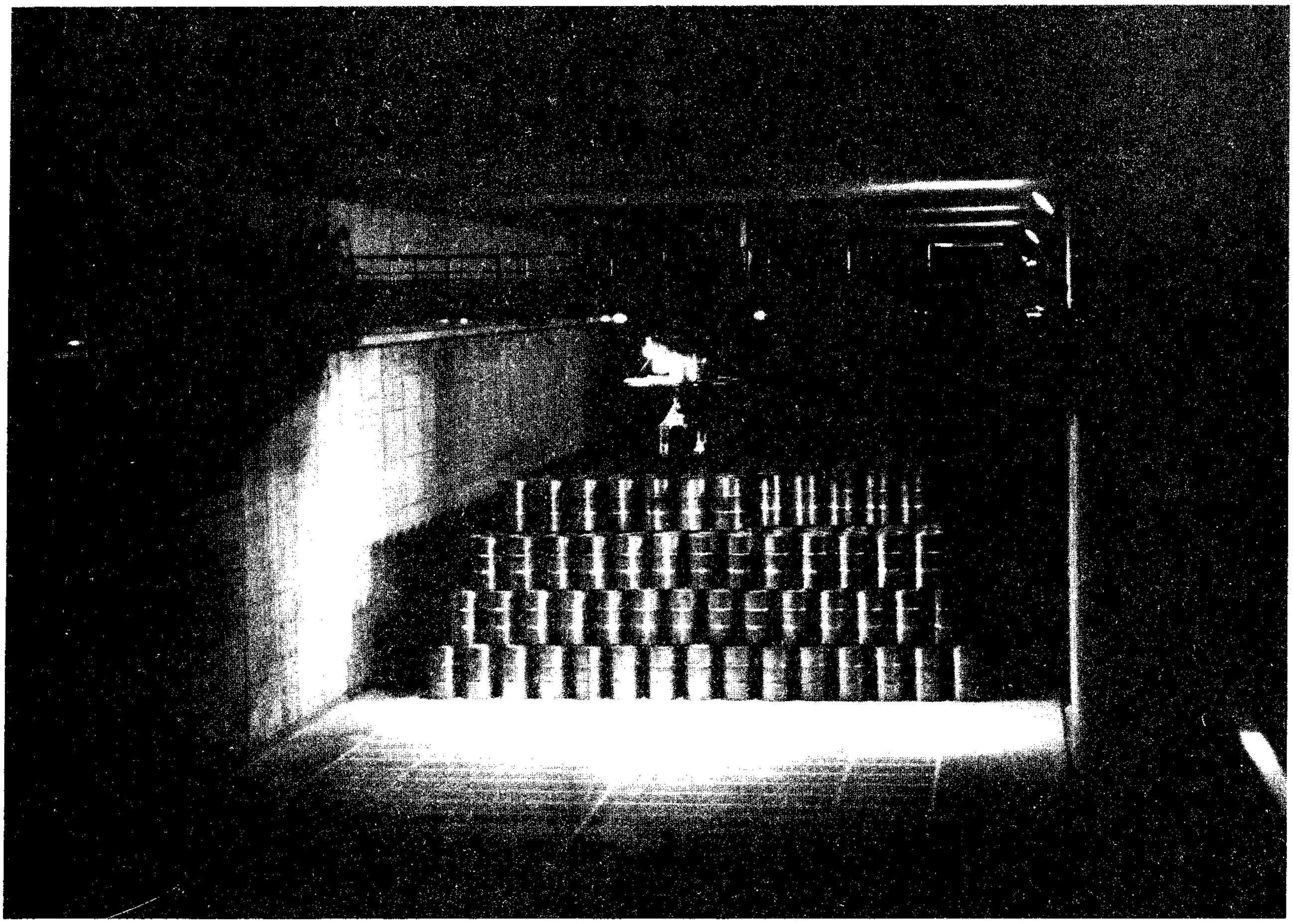


Fig. 13 - INTERIM STORAGE OF RADIOACTIVE WASTE-BITUMEN PRODUCTS IN A CONCRETE BUNKER

The BWP drums are transported by an electrically powered carriage (loading capacity of 12 drums) from the bituminization plant along the corridor to a predetermined position in front of the storage bunker to be filled. The bunker crane, equipped with a telescopic hoisting system, picks up the drums one by one from the carriage in the corridor and transfers them into the storage bunker to a preselected place with a precision of ± 3 mm. The drums are stored in a vertical position and in four layers as can be seen in Fig 13.

All movements of the transport carriage and the bunker crane are automatically controlled and can be followed visually either directly or on television screens. Manual control of the operations is also possible.

Safety devices and procedures are forseen to cope with all foreseeable emergencies and maintenance requirements avoiding radiation exposure to the personnel.

The conditioning of the wastes stored in the facility is such that any loose contamination, particularly airborne contamination, is extremely unlikely. Ventilation is only required, therefore, for the evacuation of hydrogen produced by the radiolysis of bitumen. Calculations based on conservative assumptions have shown that the atmospheric renewal rate required to keep the hydrogen concentration below the lower explosive limit can be ensured by natural air circulation if some openings are present. Because of the limited experience available with such storage conditions, however, a low-flow forced ventilation system with filters has been installed.

The material of the drums, properties of the products, and the conditions of storage at the facility have been selected with the aim of maintaining conditions compatible with future retrieval operations. If the drums should exhibit severe corrosion prior to retrieval, however, handling of the BWPs for repackaging is feasible.

As far as the risk of inflammation of the stored BWPs is concerned, emphasis has been put on fire prevention rather than on fire fighting. Thus, once the bunkers are filled and closed, everything is removed which could cause an ignition of the products stored.

Measures have been taken to enable quick and efficient fighting of a fire approaching the storage facility from the outside.

If once considered necessary or in times of warfare additional protection measures can be taken such as covering the drums by a suitable, non-burnable, granular material (e.g. sand), which, when it is required, can easily be removed.

The Eurostorage facility has been designed and constructed to last for a period of at least fifty years. At present four bunkers have been built of which the first one has already been loaded, without any difficulties, with about 4000 BWP drums.

7.3 Conclusions

Interim storage facilities and sufficient technical concepts are available showing that a safe interim (temporary, provisonal) storage of bituminized low- and intermediate-level radioactive wastes can be accomplished taking into consideration the properties of the stored products, local conditions, activity inventory, and storage time.

Though interim storage of conditioned LLW and ILW will in most cases be a necessary step in an optimized waste management strategy (e.g. for operational or economic reasons or lack of availability of a final repository) its duration should not unnecessarily be prolonged in order to reach as early as possible the ultimate objective of waste management: to dispose of the waste in a safe manner, thus minimizing the need for continuing surveillance by future generations.

8. TRANSPORTATION

8.1 General

The transport of radioactive waste cannot be avoided since it is not always possible to arrange for storage and disposal facilities to be located on the same site as the plants (e.g. reprocessing plants and reactor stations) producing the waste.

The general framework of regulations for the transport of radioactive materials is adequate for all categories of wastes. The model regulations published by IAEA /R-22, R-23/, serving also as basis for national regulations, were developed to protect the public, transport workers and property from hazards due to radioactivity during the shipment of radioactive materials. Protection is achieved by a combination of:

- limitations on the content of a package according to the quantity and type of radioactive material;
- standards of package design;
- simple handling, storage and stowage precautions to be taken during transport.

In general, more reliance is placed on "design safety" than on "operational safety".

Packages are divided into two classes, type "A" and type "B". Type "A" packages are designed to provide economical transport for the large number of small activity consignments, while simultaneously providing a high degree of safety. The quantity of radioactive material which may be transported in a type "A" package is limited so that there will be no serious radiation hazard, even if the package is completely destroyed.

Type "B" packages (type B(U) and type B(M)) are used for transporting greater amounts of activity. They are designed to reduce to a very low level the possibility of release of activity as the result of an accident during transport. Containment standards for type "B" packages are specified in terms of activity loss after tests to simulate damage in normal and accident conditions.

The regulations require certificates of approval for type "B" package designs to be sent to national competent authorities before the first shipment. Although no upper limit is prescribed for the activity in an type "B" package, there are limits beyond which the consignor must notify the competent authority of each country through which a package is to be transported.

In the IAEA transport regulations, there is nothing which prohibits the transport of any form of radioactive materials as such. More hazardous consignments are simply subject to more restrictions and the packaging requirements are more rigorous. Such "special arrangements" have to be agreed by the competent national authorities of all the countries concerned.

In general, radioactive waste conditioning and packaging represent a compromise to meet various requirements concerning handling, transportation, storage and disposal.

For the transport (as well as for the storage) the quality of the barriers formed by the conditioning and packaging have to be evaluated with regard to the attacks to which it can be subjected and the consequenses that it can have on humans and the environment.

The "attacking agents" to be considered in the safety of transporting BWPs are irradiation, temperature, fire, and water.

As it is the main purpose of this paper to consider the importance of the properties of bitumen and bitumen-waste mixtures in the various waste management steps from a safety point of view, no extensive treatment of all factors involved in the transport of these materials can be given, but only those factors can be pointed out which are most significant for the present purpose.

8.2 Definitions

With regard to the IAEA transport regulations definitions have been etablished of which some are given hereafter /R-22/.

Competent authority

Competent authority shall mean any national or international authority designated or otherwise recognized as such for any purpose in connection with these Regulations.

Containment system

Containment system shall mean the components of the packaging specified by the designer as intended to retain the radioactive material during transport.

Design

Design shall mean the description of special form material, package, or packaging which enables such items to be fully identified. The description may include specifications, engineering drawings reports demonstrating compliance with regulatory requirements, and other relevant documentation.

Fissile material

Fissile material shall mean plutonium-238, plutonium-239, plutonium-241, uranium-233, uranium-235, or any material containing any of the foregoing. Unirradiated natural and depleted uranium do not come under this definition.

Freight container

Freight container shall mean an article of transport equipment designed to facilitate the carriage of goods by one or more modes of transport without intermediate reloading. Small freight containers are those which have either any overall outer dimension less than 1.5 m, or an internal volume of not more than 3.0 m^3.

Multilateral approval

Multilateral approval shall mean approval by both the relevant competent authority of the country of origin and of each country through or into which the consignment is to be transported.

Non-fixed radioactive contamination

Non-fixed radioactive contamination shall mean contamination that can be removed from a surface by wiping with a dry smear.

Package

Package shall mean the packaging together with its radioactive contents as presented for transport.

Packaging

Packaging shall mean the assembly of components necessary to ensure compliance with the packaging requirements of the IAEA Regulations. It may, in particular, consist of one or more receptacles, absorbent materials, spacing structures, radiation shielding and devices for cooling, for absorbing mechanical shocks and for thermal insulation. These devices may include the vehicle with tie-down system when these are intended to form an integral part of the packaging.

Type A packaging shall mean a packaging that is designed to withstand the normal conditions of transport as demonstrated by the retention of the integrity of containment and shielding, to the extent required by the IAEA Regulations, after the tests specified in Section VII of Reference /R-22/, as appropriate.

Type B packaging shall mean a packaging that is designed to withstand the damaging effects of a transport accident as demonstrated by the retention of the integrity of containment and shielding, to the extent required by the IAEA Regulations, after the tests specified in Section VII of Reference /R-22/, as appropriate.

Radiation level

The radiation level shall mean the corresponding radiation dose-equivalent rate expressed in millirem per hour.

Radioactive contents

Radioactive contents shall mean the radioactive material together with any contaminated solids, liquids and gases within the package.

Radioactive material

Radioactive material shall mean any material having a specific activity greater than 0.002 μCi/g.

Specific activity

The specific activity of a radionuclide shall mean the activity of the radionuclide per unit mass of that nuclide. The specific acitivity of a material in which the radionuclides are essentially uniformly distributed is the activity per unit mass of the material.

Transport

Transport shall be deemed to include any operation incidental to the whole course of carriage, such as loading, unloading and storage in transit. The term includes both normal transport and that under accident conditions.

Vehicle

Vehicle shall mean a road vehicle (including an articulated vehicle, i.e. a tractor and semi-trailer combination) or railroad car or railway wagon. A trailer shall be considered as a separate vehicle.

8.3 Safety Aspects

In the transport of radioactive materials, any hazardous characteristics of these materials such as explosiveness, inflammability, chemical toxicity, and corrosiveness must be taken into account in such a manner as to be in compliance with the relevant transport regulations for dangerous goods of each of the countries through or into which the materials will be transported as well as in compliance with the IAEA regulations for the safe transport of radioactive materials /R-22/.

The transport regulations apply to the transport by land, water or air. The majority of bituminized liquid and wet reactor wastes (evaporation concentrates, filtration sludges and spent ion exchange resins) can be classified according to the IAEA regulations as "low-level solid radioactive material".

Though not always required, it is advisable to use freight containers for the transport of this type of waste to ensure sufficient resistance to impact and fire. If necessary freight containers can be designed to meet the requirements for type "B" packages.

Bituminized waste of the intermediate-level category have to be placed in transport shells or in large shielding freight containers for the accommodation of a large number of product drums.

Transport of BWP drums in properly designed individual concrete casks or large freight containers ensures adequate radiation shielding and protection against impact, temperature and fire; the low leachability of the product itself provides the required protection against the attack of water.

The bitumen-waste mixtures are normally contained in metal drums of 100 - 600 l capacity (mostly 200 ± 30 l). When solidified, these mixtures can be handled and transported by any means (e.g. crane, grab-tool, conveyor, lift truck, carriage, waggon, ship) under normal circumstances in a completely safe manner, provided that two conditions are fulfilled:

- presence of a sufficient radiation shielding, and

- absence of a hazardous amount of an explosive mixture of radiolysis gases (hydrogen) and air.

Consideration of the risks involved in transport accidents will lead in most cases to the requirement to use for the transport of BWPs an additional transport container of suitable design and materials to cope with the accident criteria in the IAEA transport regulations.

Taking into account the hazardous characteristics and physicochemical properties of the BWPs and furthermore the protection aims to be reached by the bituminization itself and the application of an additional transport container one comes to the conclusion that essentially only one risk needs to be considered: the fire risk.

Due to the relatively high flash point of BWPs it is impossible that a spontaneous fire occurs in a transport accident if an external fire source is absent.

The only realistic scenerio for getting BWPs ignited and burned is an external fire of a longer duration as a consequence of one or several of the following accidental conditions:

- collision with vehicles (or ships, where applicable) loaded with inflammable or explosive materials;

- burning of the kerosene or gasoline of the transport vehicle itself (less likely);

- collision with airplanes;
- collision with containers and other equipment containing inflammables or explosives.

In Sweden it is forseen to transport solidified reactor wastes in special containers by means of a ship and special land-vehicles to the vicinity of the final repository for low- and intermediate-level wastes ("ALMA", see Fig. 14); it can be assumed that solidified, intermediate-level wastes deriving from the reprocessing of Swedish fuel in a foreign country will be shipped in the same way to the Swedish repository.

A safety analysis of the sea transportation of solidified reactor wastes (including bituminized wastes) has recently been edited /279/.

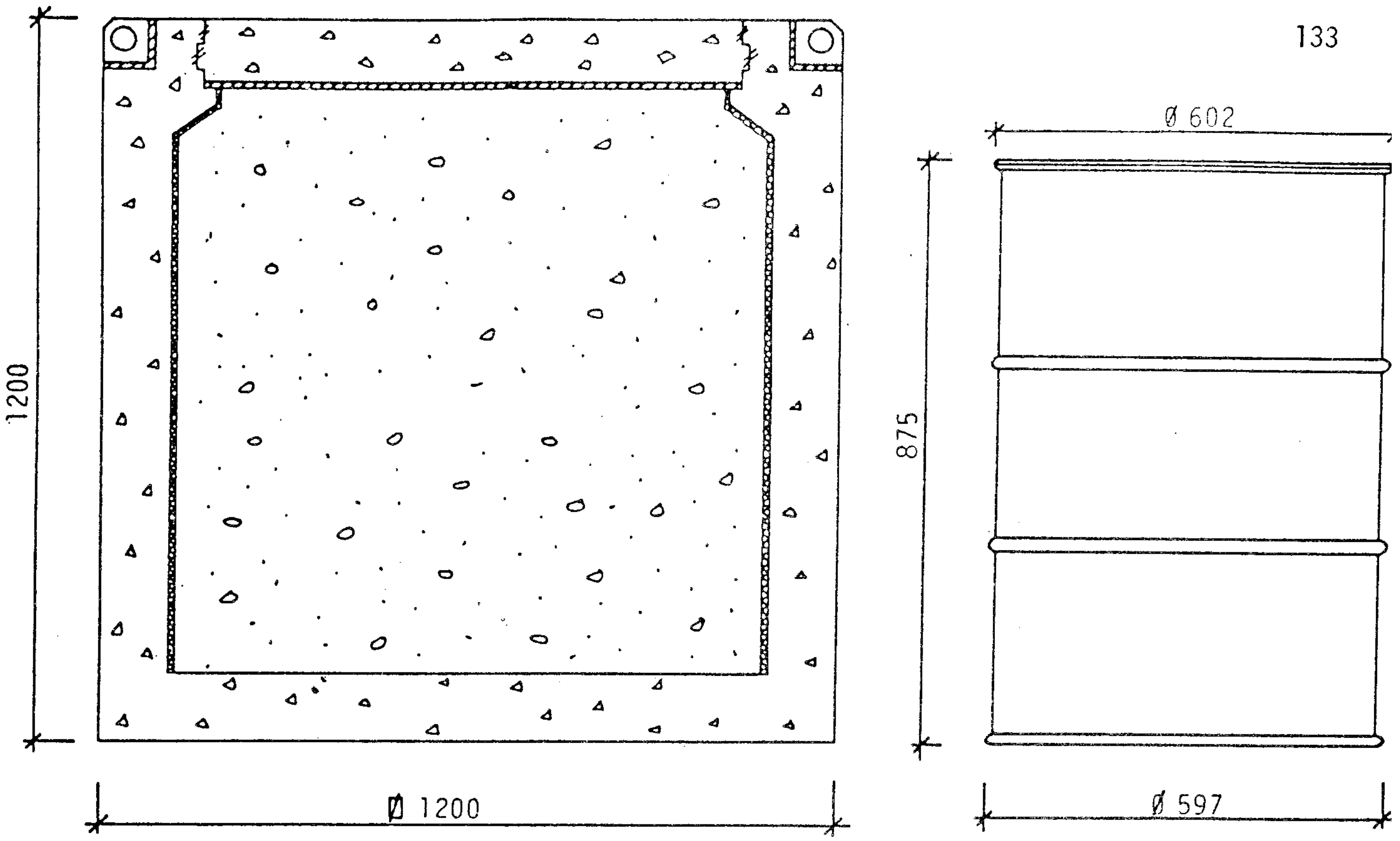
8.4 Experiences and Experimental Results

A great deal of experience has been accumulated to date in the transport of radioactive materials. Standards are well established and the safety record is excellent.

In the Federal Republic of Germany approximately 2000 drums of 175 l each containing BWPs of an average salt content of 50 wt.-% and a specific activity of 100 Ci/m^3 were transported from Karlsruhe to the ASSE II salt mine packed individually in socalled lost concrete containers (ca 20 cm wall thickness), or in a shielded transport container, or in a special cask with a capacity of seven drums by means of railway and vehicles without experiencing any incident or accident during handling and transport.

In the following some experimental results (in addition to those already given in chapter 6.3.6) are compiled which are relevant to the risk assessment of transport accidents.

- Drop tests carried out (in the ASSE II salt mine, FRG) with fourteen 175 l drums containing 50 wt.-% bitumen and 50 wt.-% sodium nitrate showed that a free fall of 86 m onto a heap of coarse salt and onto already fallen drums caused cracks in the metal drum, sometimes dispersion of product pieces until a maximum distance of 2 m and a slight temperature increase, however, no formation of sparks, explosion, fire or dust; the speed on the bound was calculated to be 38 m/sec and the kinetic energy 1.47 x 10^4 kpm.
- In full scale tests 200 l drums filled with pure bitumen only were subjected to impact and puncture forces /288/. The dropping tests of the bitumen drums were performed as corner drops from 20 m height. A similar deformation was caused to drums containing either BIT-45 or BIP-85/40; the deformation was about 80 mm, but the steel drum remained intact. In puncture tests the drums were dropped axially and radially on the top of a standard steel cylinder (Ø 150 mm). In 1.2 m drops the damage was very small. The 9 m drops resulted in small failures in the steel cover. The case of reinforced



concrete container

metal drum

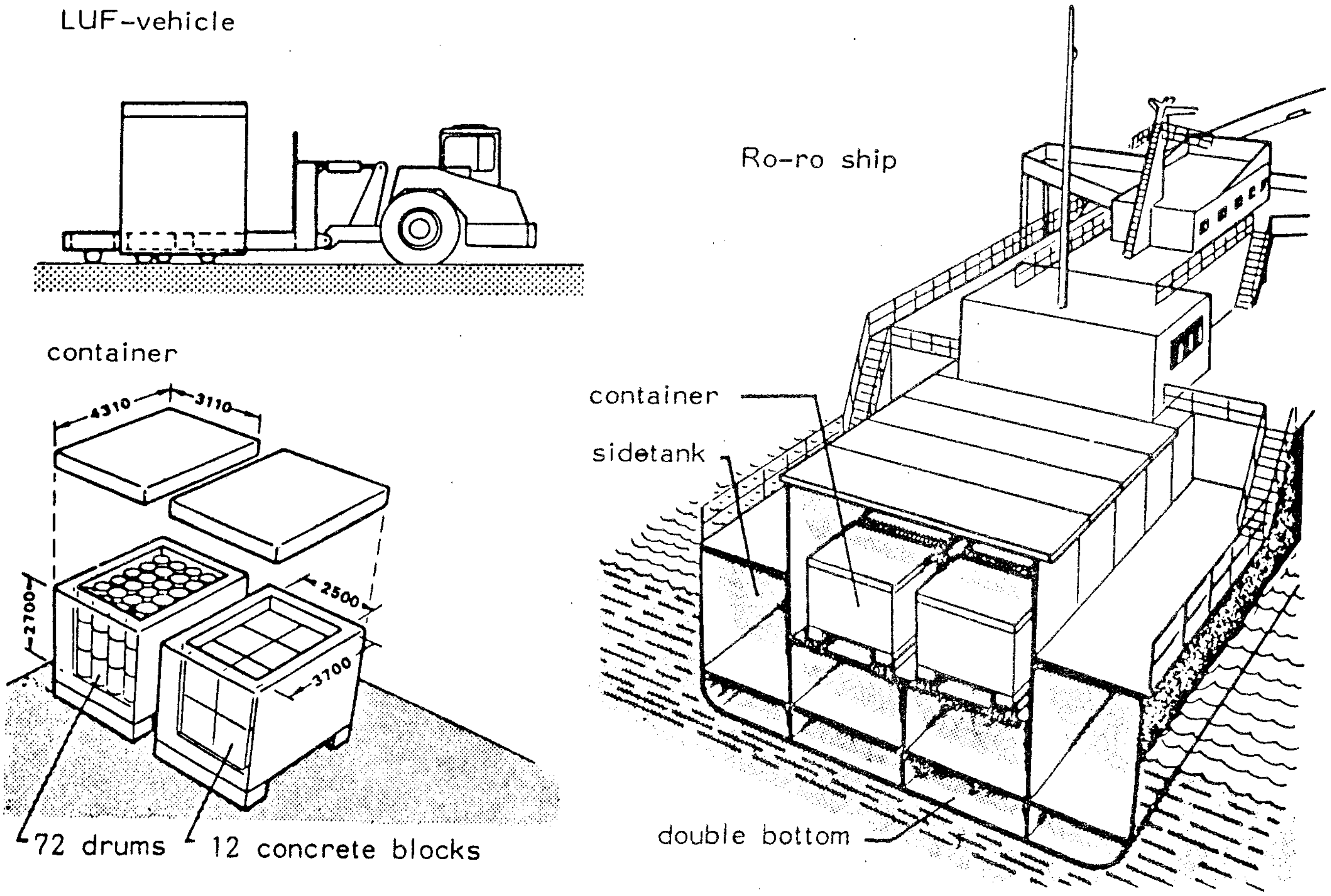


Fig.14 - THE "ALMA" WASTE PACKAGES AND TRANSPORT SYSTEM (R-28)

concrete containers was simulated by removing the steel cover from two concrete drums which were reinforced by iron net In 9 and 20 m drops the inside part of concrete remained intact.

From the tests performed it was concluded that reactor waste solidified by incorporation into bitumen and packaged into steel drums has a very high mechanical impact resistance. It is almost impossible to imagine an accident situation where a considerable part of the contents could be released in dispersable form as a result of mechanical damage.

Auxiliary transport containers increase the mechanical resistance substantially. In the case of fire and water immersion the release of activity depends to a high degree on the mechanical damage on the containers and drums. The worst accident was evaluated to be a mechanical failure of transport container and simultaneous long-lasting fire, which could cause a release of nearly all the activity contents of a bitumen drum.

By using sufficiently strong transport containers, and by solidifying the waste into steel drums or reinforced concrete containers, the probability of significant activity releases can be made very low.

- Using mixtures of 40 wt.-% bitumen/60 wt.-% alkali-, alkaline earths-, heavy metal nitrates, organic acids, and rare earths (in practise the nitrate content will be much lower) it was shown /69/ that:

  - the BWPs can be handled and transported in a safe manner,

  - the BWPs are not able to detonate,

  - the presence of nitrate enhances the burning rate, however, does not lead to explosion-like reactions,

  - a free fall of 40 m of a drum containing 300 kg BWP does not lead to a detonation.

Heating of drums containing a bitumen/salt mixture of 40/60 wt.-% in a fire causes bursting of the drums (due to the generation of gases) and subsequently the ignition of the contents which burns with an enhanced rate compared to pure bitumen.

A slow heating-up can segregate the mixture by sedimentation, or the sodium nitrate can melt and settle-down.

The ignition and burn-up of the mixture show the characteristics of a difficultly inflammable substance and at higher temperatures the characteristics of a substance burning well in the precense of air.

There is no danger that the reactions of the violently burning BWPs enhance to a deflagrating burn-up or even a detonation. There exists no explosion risk during the transport of BWPs originating from reactor stations and reprocessing plants.

- In fire tests involving a bitumen/ion exchange resin product a considerable volume increase occurred causing lifting-off of the lid and an overflow of the product /255/.
- Recently some experiments on the effect of fire on pure bitumen (ca 160 l BIT-15 in a 200 l drum) were carried out /287/ showing that:
  - bitumen started burning after 15 - 20 min,
  - the temperature rise inside the drum was very rapid from room temperature to about 400°C which corresponds approximately to the distillation temperature of bitumen BIT-15,
  - the max.temperature observed outside the drum was about 1200°C which can be considered as the max.temperature of a bitumen fire,
  - irradiated bitumen speeds up the response to fire,
  - most of the bitumen burned in a period of 10 min, the maximum burning rate being 0.25 kg/s.
- BWPs in 175 l drums and surrounded by a 20 cm thick layer of normal concrete were placed for 50 min in a vigorously burning oil fire. After 4 - 5 hours temperatures of max 73°C were measured in the inner part of the concrete shielding; after 24 hours these temperatures had decreased to the ambient temperature.

  The experiments demonstrated the efficient protection of BWPs against external fires by placing the product drums into a concrete container of an adequate thickness /283/.

8.5 Conclusions

Attention to safety in the transportation of nuclear waste is of special importance when bituminized waste is involved.

This concern for safety is centered around the burnability of bitumen-waste products.

When transport takes place at very low temperatures, also the brittleness of the products deserves consideration in the possible dispersion of radionuclides as a consequence of an accident.

An inadvertent release of radionuclides will not occur during normal operation nor in most serious accidents if the BWPs are transported in appropriately designed shipping containers. While complete assurance is impossible, it has been demonstrated that it is highly unlikely that a significant release can occur under any credible accident environment.

The safety assessment devoted to the potential environmental impacts from the sea transportation of BWPs from Swedish nuclear power plants to the final storage facility (ALMA) /279/ showed that:

- no release of radioactive materials will occur to the atmosphere or to the sea under normal conditions;
- the frequency for the "ALMA-ship" to be involved in a serious accident while carrying radioactive cargo at sea is estimated to about 10^{-3} per year;
- a collosion causing a fire in BWP-drums transported in concrete containers is deemed to be inconceivable;
- even an assumed occurrence of a fire would not lead to severe radiation doses to members of the public.

It has been concluded that in view of the low probabilities for accidents causing release of radioactive material and the limited consequenses, the sea transportation system analysed can be deemed to meet very high safety standards.

9. FINAL STORAGE

9.1 General

All radioactive wastes have to be stored or to be disposed of in such a way that the appearance of radionuclides from these wastes in hazardous concentrations in the biosphere is excluded.

The term "storage" is commonly used to denote the emplacement of waste with the intention of retrieving it at a later time /R-13/. Storage is a temporary measure which requires continuing surveillance.

The term "disposal" means the release or emplacement of waste without the intention of retrieval /R-13/. Disposal can be totally irreversible, as for example in the case of environmental release of effluents. Retrieval may be possible, as in some geologic disposal schemes for solid waste, but it is the absence of the intention to retrieve which implies disposal. Disposal concepts do not require continued surveillance. However, in particular cases, surveillance over limited time periods may be desirable.

To avoid the impression that (geologic) disposal of solid and solidified waste would principally mean the technical impossibility to retrieve the waste the terms "final", "ultimate", "permanent", or "terminal" storage are used herein synonymously with the term "disposal".

As far as the final storage of conditioned high-level waste and transuranic wastes are concerned there is generally agreement that their placement in a suitable deep geologic formation can guarantee the required long-term protection of man and his biological environment against hazardous radiation doses originating from these types of waste.

Also the geological storage (at a reasonable depth) of properly conditioned low- and intermediate-level wastes appears to be the safest way to provide the required long-term isolation of the contained radionuclides from the biosphere and to protect them against attacks of man and nature.

Existing regulations in some countries allow the disposal of LLW and certain ILW by either land burial (near the surface) or sea dumping.

Studies on problems connected to the geological disposal of the different categories of waste and assessments concerning the suitability of various geological formations for repositories are carried out in all countries having a significant nuclear power programme.

The three most relevant geological formations are:

- bedrock (granite)
- salt deposits, and
- argillaceous sediments (especially some clays and shales).

9.2 Options in Various Countries

The investigations on the geological disposal of radioactive wastes in the various countries are primarily directed to the disposal of high-level waste (solid and solidified HLW from fuel reprocessing or spent fuels); the large volumes of LLW and ILW can often be disposed of in a safe manner at much lower costs by using other disposal techniques and environments than are required for HLW.

a) In the USA the current means of disposing of LLW and ILW from nuclear power plants and interim fuel storage facilities is through shallow-land burial. Intermediate-level wastes from reprocessing were also disposed of by shallow-land burial initially. Due to the relatively high content of alpha-emitting transuranics (> 10 nCi/g) of these wastes, most are now being placed in interim storage pending selection of an acceptable alternative disposal method. Improved shallow-land burial and intermediate-depth burial are being examined primarily for non-transuranic low-level wastes while disposal in deep geologic structures is being considered for transuranic wastes.

   A pilot plant repository for defense wastes may be established in salt in New Mexico by the mid 1980s, and repositories for commercial wastes may be operative in the 1990 time frame.

   Retrievability of all wastes will be maintained until necessary confirmatory data concerning safety are obtained.

   Regulations presently under consideration by the Nuclear Regulatory Commission (NRC) will define waste according to the type of disposal required and will identify the quantity of each radionuclide that will be disposed of in a

   - sanitary landfill,
   - shallow-land burial site,
   - intermediate-depth burial site, and
   - deep geologic structure.

b) In Canada exists interest to find suitable geologic formation for both secure retrievable storage and terminal storage. An expert group commissioned in 1977 by Canada's Department of Energy, Mines and Resources, considered underground disposal in igneous rock as the most promising Canadian option for the disposal of spent fuels and radioactive wastes.

c) In Japan, the land burial of radioactive wastes is presently not permitted. A survey of Japan for potential disposal sites (for HLW) mainly) showed that geological bodies of granite and zeolite rock are attractive possibilities; under further consideration are formations of limestone, diatomite and shale.

The Japanese are interested in sea-bed as well as island disposal concepts. As Japan does not possess terminal storage capability, low-level waste is currently conditioned by cementation and bituminization and stored in drums in warehouses and underground concrete trenches.

Several European countries have ongoing programmes (sometimes in the framework of international cooperation) to evaluate concepts of terminal isolation of radioactive wastes in geological formations.

d) In the United Kingdom it seems likely that there are two options: granite and clay formations. Furthermore, studies are carried out on the deep sea disposal (on-the-bed deposition) of ILW.

e) In France the planned disposal method for wastes containing alpha-emitters is emplacement in deep geological formations. For fission products, either geologic disposal or long-term storage in engineered facilities are considered as possible solutions.

The Infratome Company carried out final storage operations by placing LLW contained in concrete blocks and drums encased in concrete directly into the ground and LLW in ordinary metal drums were placed in the ground in trenches. The permanent storage centre for waste is situated in the Department of Manche, near the La Hague centre, on a plot of land belonging to the CEA.

Infratome classified "low-level waste" as waste whose specific activity in Ci/m^3 is less than 1000 times the MPC/water (maximum permissible concentration in drinking water) of the radionuclides contained.

For geologic disposal granite and rock salt formations are presently studied, however, a significantly greater effort will be devoted to granite formations.

f) In Belgium (and in Italy) the investigations on the disposal of ILW and HLW in deep, stable geologic formations are focused on the use of argillaceous sediments (clays). Like France, Italy, Switzerland, The Netherlands and the UK, Belgium disposes of part of the low-level waste (after conditioning by cementation or bituminization) by sea-dumping under the London Dumping Convention rules and within the consulation/surveillance mechanism of the Nuclear Energy Agency (NEA) of the OECD.

g) In The Netherlands exist salt domes in the east of the country, and it can be assumed that one of these will be used for the construction of a waste repository.

h) In the Federal Republic of Germany the former salt mine ASSE II at Wolfenbüttel near Braunschweig has been operated since 1965 by the Gesellschaft für Strahlen- und Umweltforschung mbH (GSF) as a national research and development centre for the final storage of radioactive wastes.

In this mine, consisting of about 130 large chambers on 10 different levels, low- and intermediate-level wastes incorporated in bitumen, concrete and plastics have been stored. Approximately 125 000 packages (200 l and 400 l drums and 15 000 concrete containers) containing low-level wastes have been placed in seven chambers at a depth of 725 m and 750 m from April 1967 until 1978.

In August 1972 test disposal of intermediate-level waste commenced, using a technique specially developed for this purpose. In the following six years of operation, about 1300 ILW packages (200 l drums) have been deposited at the 490/511 m level.

The experiences made up to now have been very satisfactory in spite of the small capacity of the transport facility and the impractical transport roads in the mine itself as it was originally designed for the mining of rock salt and carnalite, and not as a test repository.

i) In Sweden, the studies related to the geological disposal of high-level wastes, which are carried out within the KBS-project (Kärn-Bränsle-Säkerhet meaning Nuclear-Fuel-Safety), are described in numerous KBS-Technical Reports.

The main findings together with relevant considerations were published in two main parts under the title "The Fuel Cycle's Final Step":

Part I (5 volumes) entitled "Vitrified Waste from Reprocessing" was published in November 1977 /R-26/,

Part II (2 volumes) entitled "Handling and Final Storage of Unreprocessed Spent Nuclear Fuel" was published in June 1978 /R-27/.

Major efforts have been concentrated on bedrock investigations in the closed-down Stripa iron ore mine.

The first disposal of HLW will not take place prior to the year 2020.

The plans on the final storage of low- and intermediate-level wastes in Sweden are outlined in a separate chapter hereafter.

j) In some other European countries also studies on the geologic disposal of radioactive wastes are carried out among these countries are Switzerland, Denmark, Austria, German Democratic Republic Spain, and Finland.

9.3 Repository for Low- and Intermediate-Level Wastes in Sweden (ALMA)

9.3.1 Conceptual design and costs

Recently a study on the construction of a central repository in bedrock for LLW and ILW ("ALMA") has been carried out /R-28/.

The study is based on earlier investigations within the ALMA-project /R-1/ and the following assumptions·

- the final storage facility shall have a capacity sufficient to take up the LLW and MLW generated during 30 years' operation of Swedish nuclear power stations with a total power of 10 000 MWe;
- the waste to be stored are packaged mainly in drums or concrete containers and have a total volume of approximately 124 000 m^3, of which about 80 000 m^3 are intermediate-level wastes;
- the transport of the waste will be in 25 m^3 containers first by ship and then from the harbour to the repository by hydraulic lift-cars.

In the study three different structures of the storage rooms in bedrock have been considered:

- horizontal vaults (mountain halls) of a cross-sectional area of 600 - 700 m^2, (schematically shown in Fig. 15):
- vertical (standing) circular rooms (silos) of ca 30 m diameter and ca 60 m height;
- a series of parallel tunnels of a cross-sectional area of ca 100 m^2.

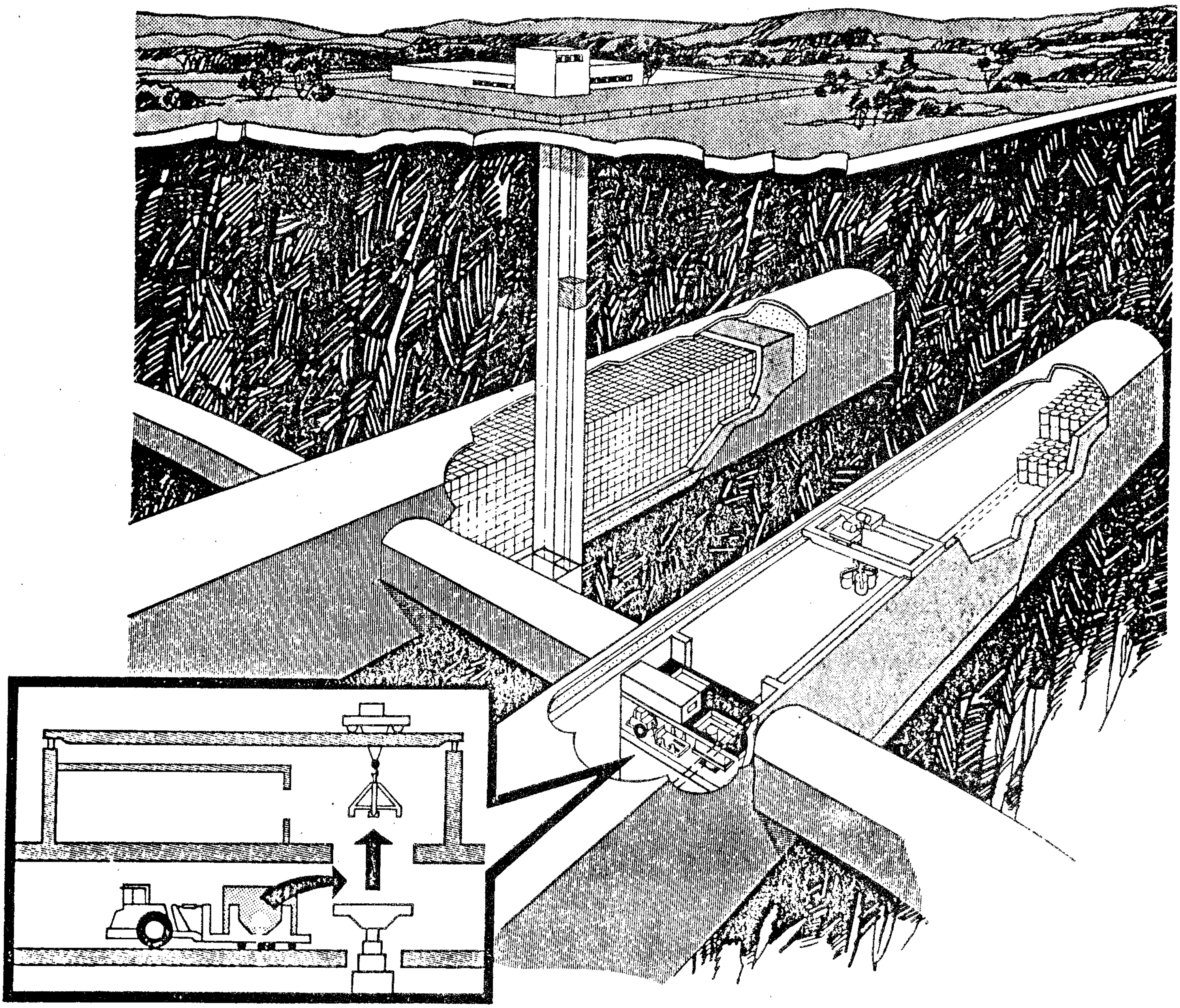


Fig. 15 - A DESIGN PROPOSAL FOR A GEOLOGIC REPOSITORY FOR LLW AND ILW (ALMA) [R-28]

To aid in the isolation of radionuclides from the biosphere the following additional barriers between the waste and the rock have been evaluated:

- a barrier of concrete;
- a barrier of a moraine/bentonite mixture, and
- a barrier of both concrete and moraine/bentonite mixture.

Cost and construction time estimates were made for various storage room/barrier combinations and the influence of the depth (in the range 50-300 m) and the mode of access to the storage place (shaft or tunnel) have been evaluated.

For the three different forms of the storage room, all provided with a concrete and a moraine/bentonite barrier, the following costs in Swedish Crowns (1979-value and price level) have been calculated:

Alternative	Level: - 50 m	Level: - 300 m
Hall	275 x 10^6 Skr	378 x 10^6 Skr
Silo	300 x 10^6 Skr	406 x 10^6 Skr
Tunnel	364 x 10^6 Skr	477 x 10^6 Skr

These costs correspond to 2400 - 4000 Skr/m^3 waste and to 0.015 - 0.025 öre/kWh. (1 US \$ ≈ 4.20 Skr).

If it is not required to apply the additional man-made barriers the costs (given in the table) diminish for all three cases by 130 - 145 million Skr for the level minus 50 m and by 200 - 215 million Skr for the level minus 300 m.

The construction time has been estimated to be 5 - 6 years for the level - 50 m and 2 - 3 years more for the level - 300 m.

The final choice among the different construction possibilities of the repository (possibly even one not yet considered in the study) will depend on the results of a safety analysis still to be performed.

Considering the different characteristics of the waste products to be stored it could well be that a combination of the various structural features will be the most adequate solution.

9.3.2 Waste products and amounts

For the preliminary repository studies only the following waste packages normally generated at Swedish reactor stations. are considered:

- concrete blocks of 1.2 x 1.2 x 1.2 m = 1.7 m^3 and 4 t weight
- concrete tanks of 3.3 x 1.3 x 2.2 m = 9.4 m^3 and 15 t weight
- BWP drums of Ø 60 cm and 87 cm height
- metal boxes of 600 l and 600 kg
- drums of Ø 60 cm and 87 cm height containing ash or non-burnable solids.

The contents of radionuclides, which is important for release, migration and dose calculations, is different in the various packages as already described previously.

The estimated amounts of waste packages from Swedish reactors of a total power of 10 000 MWe during a 30 years' period are given in Table XXVII /R-29/.

TABLE XXVII

ESTIMATED AMOUNTS OF REACTOR WASTE FROM SWEDISH REACTORS (10 000 MWe) ACCUMULATED DURING A PERIOD OF 30 YEARS

Type of Package	No of Packages	Total Storage Vol. (m^3)
Concrete blocks	36 000	63 000
Concrete tanks	1 300	12 000
Drums cont. BWPs	66 000	20 000
Drums cont. non-burn. solids	19 000	6 000
Others (mainly drums)	70 000	23 000
Total	192 300	124 000

9.3.3 Transport system

The preliminary concept foresees transport of the final waste products in transport containers. The transport system consist of:

- a ship provided with a socalled roll on/roll off-system;
- 50 transport containers, and
- 6 socalled LUF-vehicles (of which two are stationed at the repository).

One transport container of 25 m^3 inner volume can take up 12 cubical concrete blocks or 72 standard drums (ca 220 l).

Shielding containers are made of concrete and non-shielding containers are made of steel. Depending of the contents and the shielding thickness the loaded containers will weigh between 25 and 95 t.

Fig. 14 shows the main components of the proposed transport system /R-28/.

9.4 Conclusions

All waste disposal concepts are aimed at isolating the waste from the biosphere for a period sufficient to ensure that any subsequent return of radionuclides to the environment would not lead to unacceptable levels of exposure of man or other biological species. This objective can be attained either by providing containment with an adequate degree of reliability for the requisite period, or by ensuring an adequate delay mechanism and the availability of a dispersal medium to dilute to acceptable levels any radionuclides returning to the biosphere.

The experience up to now gained on the experimental geologic disposal of low- and intermediate-level wastes (in rock salt) is excellent. In Sweden, it is presently foreseen to place the conditioned (by cementation or bituminization) low- and intermediate-level reactor and reprocessing wastes for their safe final storage in a geologic repository situated in a stable hard rock formation.

There exists great experience in Sweden in the excavation of both small and large cavities in bedrock so that the storage rooms can be built with presently available technology.

The structural design features and the depth (distance from the surface) of the repository for the various waste products as well as the eventual need to provide additional barriers will have to be based on the results of quantitative risk estimates.

10. THE LONG-TERM BEHAVIOUR OF BITUMEN-WASTE PRODUCTS

10.1 General

The long-term behaviour of bitumen-waste products (and of other long-lived wastes too) is of importance for the safety assessment of the final storage (or disposal) system.

This system is considered safe if it protects man and his biosphere from hazards originating from the stored waste.

Considering disposal of solid and solidified wastes in a stable geological formation the required protection (mainly radioprotection) is achieved by using manmade barriers in addition to natural barriers against the spreading of radionuclides (and possibly other hazardous substances) originally immobilized in the waste product.

As barriers are usually considered:

- the waste product itself (the fixation compound or the embedding material or both);
- the container of the waste product;
- the eventually applied overpack(s) of the waste product container;
- the backfilling materials;
- the engineered structure of the repository;
- the geological formations (geosphere) between repository and biosphere.

The required nature, quality, dimension, sequence, and number of barriers to achieve the protection aim depends on their effectiveness to retain water or the hazardous waste components.

The main function of the barriers, seen from the biosphere towards the waste product, is the hindrance of water to reach the waste product; if contact of the waste product with water cannot be prevented the main function of the barriers is the retention of the leached radionuclides to such an extent, that, if at all, only insignificant concentrations of radionuclides can appear in the biosphere not affecting public health and safety. Thus, the barriers of the product package and the engineered and natural barriers of the geologic repository should effect the required long-term isolation of the radionuclides from the biosphere; certain deficiencies of the barrier system can be outweighed by choosing a larger distance (time) to be travelled by the radionuclides until the biosphere.

The radiotoxicity of the contained radionuclides is the basic factor determining the hazard of the waste and consequently its treatment, storage and disposal.

When radioactive wastes have been bituminized and the resulting bitumen-waste products are placed in a final repository a number of changements occur or can occur in the course of time by product-internal and - external phenomena capable of influencing the retention or liberation of the encapsulated radionuclides.

These phenomena can be of a chemical, physical and biological nature and depend as much on the properties of the BWPs as on the characteristics of the repository and its environment.

It is therefore necessary from the radiation protection point of view to forecast with a reasonable safety margin the behaviourof BWPs of known starting characteristics in a given environment during its hazardous lifetime to enable in the framework of a safety analysis the determination of the radiation which can reach man.

The safety analysis of the entire waste disposal system appears to be the only reasonable approach of demonstrating its adequacy as finally the effectiveness of all components of the system cannot be verified experimentally in the time span of concern.

10.2 Scope

This chapter on the long-term behaviour of bitumen-waste products is confined to the stability or possible changements with time of those properties which are esteemed to be of decisive importance for their safe disposal in a bedrock repository (ALMA).

The principal factors deserving consideration in this respect are all those affecting directly or indirectly the leachability (release rate) of radionuclides from the waste package and/or the stability of the repository and its environment due to the instability of the stored products.

The long-term behaviour of bitumen and the physico-chemical stability of BWPs, in particular the various factors influencing their leach resistance, have already been described in former chapters of this report (Chapt. 3 and 6).

Much of the findings reported there relate to the long-term stability of BWPs in general.

In the following sub-chapters it has been tried to present as far as possible only additional results, new aspects and preciser statements on the safety-relevant long-term behaviour of BWPs under the envisaged normal repository conditions.

Only those factors are considered which are directly related to both the BWPs themselves and the ultimate aim of radioactive waste management: the radiological protection of man and his biosphere.

Though the product container, - which provides a contamination free surface, additional mechanical strength and containment during handling, transport and storage, - is an integral part of the waste package its barrier effect and long-term behaviour under repository conditions will not be treated here in depth as this will be subject of another report Beyond the scope of the present chapter is furthermore the consideration of the properties and long-term behaviour of engineered and natural barriers and their effectiveness against the spreading of radioactive material to the environment.

It is obvious that the long-term behaviour of waste products cannot be strictly verified experimentally, and that therefore some uncertainty will remain on their behaviour over very long periods of time.

The predictions and judgements made on the future behaviour of the BWPs are based on the knowledge of the properties after their production, their short-term behaviour, accelerated testing, and the results of appropriate experiments and their evaluation.

For the reliability of some predictions the confirmation of certain results or the availability of more precise data would have been desirable; such identified shortcomings have been subject of proposals for an R + D programme.

10.3 Waste Products and Final Storage Time

The final waste product constitutes one part of the total waste disposal system. Its primary role within this system is to provide the initial barrier against the release of the contained radionuclides. The composition and the main characteristics of the BWPs, to be considered here, have already been described (see Chapt. 6.2).

The long-term behaviour of these BWPs are of interest as long as the radionuclides immobilized in them have not decayed to innocuous levels. The time span of concern will thus primarily depend on the amount, half-life and the radiotoxicity of the contained radionuclides.

This time span is for most BWPs from power stations (which are almost free from actinides) in the order of 500 years after which the specific activity has reached the value of $\leq$ 2 nCi/g.

Considering the total amount of waste packages (bituminized and cemented wastes) in ALMA generated by 13 power stations (corresponding to 10 000 MWe) during a period of 30 years, it has been estimated that the risk index of all radionuclides (except Ni-63) is, after a period of 500 years, far beyond the risk index of a corresponding amount of arable soil having the natural radium content /R-28/.

The time span of concern for BWPs from reprocessing plants is more difficult to define as here several approaches can be considered; due to the much higher content of actinides (particularly uranium, plutonium and their daughter products) the behaviour over a longer period than 500 years has to be taken into consideration.

After about 500 years almost all fission products, including Cs-137 and Sr-90, have decayed to non-hazardous levels.

Based on the actual content of actinides and the risk index envisaged, further time has to pass to allow for further decay of the very long-lived radionuclides.

One solution to formally assess the remaining long-term risk and possible environmental impacts of the stored wastes is a comparison of the amounts and radiotoxicity of the radionuclides in the waste with those of naturally occurring radioactive substances existing everywhere in the earth crust in dispersed form, and at many specific locations in more concentrated form.

In any case, for the long-term behaviour of the BWPs, - forseeable or uncertain -, it remains important to determine either the retention quality of the BWP itself or to evaluate the required quality of additional barriers to achieve the protection aim of the disposal system.

The effects of radiation on living organisms are now understood sufficiently well to permit quantification of their impact on human health within the limits of accuracy needed for the protection of man.

After decay of nearly all radioactive fission and activation nuclides, all properties affecting the leaching of actinides from BWPs and especially their migration through the environmental media become the dominant factors of concern.

In estimating the size of the problem related to the long-term behaviour of all reactor waste products to be stored in ALMA in view of their potential long-term radiation risk one should keep in mind that the total inventory of radionuclides in ALMA is about 130 000 Ci (contained in about 120 000 m^3 waste) of which 0.5 Ci are due to Pu-239.

Thus, the total activity in the repository for low- and intermediate-level waste from 30 years' operation of the presently existing and planned Swedish reactors (or about 10 000 MWe installed nuclear power) will be lower than the activity of one single high-level waste product package to be returned from France to Sweden.

After a cooling time of 30 years one glass block (150 l) contains 2×10^5 Ci (corresponding to 9 wt.-% fission products or the HLW generated by the reprocessing of 1 ton uranium) producing a heat of 525 W.

It is obvious that the long-term behaviour of BWPs from reprocessing plants are of concern for a longer period of time than that of products from reactor stations. The time span to be taken into account has to be evaluated on a case by case basis, however, in all cases the result will be that the radionuclides present or originally present in a repository for bitumen-waste products do not form a problem lasting a million of years.

10.4 Physico-Chemical Long-Term Stability of Bitumen-Waste Products (BWPs)

It is difficult to predict all effects and mechanisms by which radioactive material can be destructed and transported to the biosphere.

However, it is generally recognized that changements of physico-chemical properties of BWPs by internal and external effects may ease the availability of the waste radionuclides to leaching by aqueous solutions (ground water or "modified" ground water). Once mobilized (leached) by the action of the ground water stream the most likely mechanisms for further spreading into the surrounding is transport by the flowing ground water or diffusion in the ground water medium.

Consequently all changements of the BWPs not affecting directly or indirectly the spreading of radionuclides by environmental liquids are finally of no or secondary importance for the quality of the "product barrier" and its radiological safety-relevant long-term behaviour.

This chapter assesses the influence of internal and external effects on the long-term stability of container-less BWPs and the consequences on the leachability of hazardous nuclides and the stability of the repository (engineered and geologic environment).

10.4.1 Radiation effects and stability

Radiation from the embedded radionuclides effects:

- generation of radiolysis gases (and other radiolysis products),

- change (normally increase) of product volume,

- hardening of the bitumen, and

- heating of the bitumen-waste mixture.

The generation of radiolysis gases can cause inside the product an increase of the porosity and thus swelling of the product (pressurization of the container), and outside the product an accumulation of explosive gas mixtures and an increased gas pressure, if the gas cannot escape from the repository or react with media of the environment.

The decay heat may effect the consistency of products of higher specific activity.

The extent of the various radiation effects depends on the bitumen-waste ratio the incorporated radionuclides and their concentration within the product.

Theoretically, all the changements of the product properties caused by radiation can have an influence on the leachability, mechanical strength, physical form and dispersibility.

Based on the latest results of investigations on the radiolysis of BWPs /220, 289, 230 a 230 b/ relatively reliable predictions on the longterm changements caused by radiation can be made.

For assessment of the long-term behaviour of BWPs in the repository the following results and data appear to be important:

- The amount of gases generated is proportional to the applied dose, their main component is hydrogen.
- The specific rate of hydrogen generation is approximately 0.01 cm^3/Mrad.g bitumen. (In Ref. 220 a value of 0.003 - 0.0065 cm^3/Mrad.g BWP is given and in Ref. 230 a a value of 0.0055 cm^3/Mrad.g BWP); it is almost independent of the dose rate, type of bitumen, the composition of the salt mixtures ($NaNO_3$, NaCl, Na_2SO_4, sodium borate), the gas phase above the products, and the temperature (20 - 95°C).
- Besides hydrogen other radiolysis gases are formed in minor quantities, namely C_1 - C_3 hydrocarbons, NO and CO (if anion exchange resins are present also methyl amine and trimethyl amine are generated); the amounts formed are also proportional to the exposure dose Methane constitutes the main fraction of the light hydrocarbons generated.
- The following dependence of the mode of exposure has been evaluated /220/:

  a) external γ-irradiation (Co-60) -

  for H_2: 0.003 cm^3/Mrad.g

  for CH_4: 0.0004 cm^3/Mrad.g

  b) internal α-and β-irradiation (Po-210 and Sr-90/Y-90, Cs-137) -

  for H_2: 0.0065 cm^3/Mrad.g

  for CH_4: 0.0001 cm^3/Mrad.g

- For most safety considerations with respect to long-term storage or disposal other gases than hydrogen are of secondary importance.

- The H_2-sorption capacity of 50% bitumen/50% salt mixtures (sodium nitrate) is low; it amounts to 3.2 x 10^{-3} cm^3/g. The rate of H_2-permeation in these BWPs is high (compared with its rate of generation): it amounts to 2.3 x 10^{-9} $cm^3.s^{-1}.cm^{-2}$.

  The influence of the dose rate is insignificant.

- Assuming the mechanism of radiation induced hydrogen generation to be independent of the applied gamma-dose, the sorption capacity of bitumen is comparable with the amount of hydrogen generated in the case of gamma-doses lower than 10^6 rad. It can be assumed that only a minor fraction of the hydrogen gas induced by irradiation remains in the bitumen matrix of BWPs of higher specific activities /230 b/.

The rate of hydrogen generation from 1 cm^3 BWP for incorporated radionuclides of an average half life of 30 years and an integral dose of 10^8 rad is smaller than the hydrogen permeation through an area of only 1 cm^2, even in the first few years of storage /230 b/.

In order to assess the effect of radiation on the long-term behaviour (as well as on other safety aspects during the storage) of BWPs, the cumulative radiation dose until "complete" decay of the contained radionuclides has to be calculated first; based on the doses absorbed (of β- and γ-radiation in the case of bituminized reactor wastes and of α-β-γ-radiation in the case of reprocessing wastes) and the weight fraction of bitumen within the homogeneous BWP, the total amount of radiolysis gases and the portion of hydrogen (75 - 95% of the total gas evolution) to be generated can be calculated and the influence on other safety relevant factors predicted.

In /R-24/ cumulative radiation exposure doses are given as a function of time for waste products of 0.00035 to 3.5 Ci/l based on the relative percentage of each radionuclide present in BWR and PWR solid wastes.

- The cumulative dose absorbed at infinite time by a BWP containing 5 Ci Sr-90 and 5 Ci Cs-137 per litre product will be approximately 6 x 10^9 rad; products of such a high specific activity (10 Ci/l product) were up to now not prepared on an industrial scale.

- As a guiding value one may assume that a BWP containing a two years' aged fission product mixture from fuel reprocessing of a specific activity of 1 Ci/l BWP will absorb during its entire life-time a dose of about 9 x 10^7 rad (which is accumulated almost completely after the first 200 years). Due to the shorter half-lives of many radionuclides in bituminized reactor wastes the total dose absorbed at infinite time of BWPs from reactor stations with an initial specific activity of 1 Ci/l product will usually not exceed 2 x 10^8 rad. The cumulative dose of "typical BWPs" from reactors with a specific activity of 0.1 Ci/l amounts to about 10^7 rad over 100 years.

Based on the experimental results on the radiation-chemical stability of BWPs (see also chapt. 6.3.3) one may predict that:

- absorbed doses of $\leq 10^7$ rad will cause only negligible gas evolution, swelling, hardening, heating, decrease in leach resistance, or other adverse effects on the physico-chemical properties;

- absorbed doses of 10^7 to 2 x 10^8 rad generate an amount of radiolysis gases ($\sim$ 1 $cm^3/10^8$ rad.g bitumen) to be considered in packaging the product (to avoid pressurization) and during the filling period of the repository; if the product container and the immediate environment permits the radiolytically generated gases, mainly hydrogen, diffuse to a large extent out of the product and away in the surrounding; the generation of radiolysis gases cause an increase of the porosity (probably < 10%) and a certain swelling which can easily be accommodated by the voids within the container or between the products;

  There will furthermore be only insignificant changes in the leachability, thermal and mechanical properties of the BWPs and in the chemical stability of incorporated waste solids except the degradation of organic ion exchange material (which is outlined below);

- absorbed doses between 2 x 10^8 and 10^9 rad cause the generation of an increased amount of radiolysis gases (which can be estimated), an increase of BWP volume reaching from zero to over 100% (depending on the bitumen-type/solid mixture and dose rate), a hardening of the product, and most probably a slight increase in the leachability which, up to now, could not be confirmed experimentally.

As mentioned above not only the bitumen matrix but also incorporated organic waste materials suffer from radiation induced chemical changements.

Thus, the radiation induced degradation of cation exchange resins leads to the formation of "sulfuric acid" (or sulfates from the - SO_3H groups), hydrogen and carbon dioxide, while anion exchange resins are degraded in decreasing amounts to trimethylamine > dimethylamine > methylamine > ammonia, and moreover to carbon monoxide, carbon dioxide and nitrogen /R-30/. Apart from an increased swelling of BWPs containing ion exchangers the solid, liquid and gaseous degradation products will not diminish the long-term physico-chemical stability of the BWPs and the repository structure to any significant extent. If the free sulfuric acid has not disappeared by neutralization reactions or reaction with bitumen components it could cause an enhanced corrosion of the product container and certain concrete structures.

The influence of complexing agents possibly formed by radiation (chemical or biological) reactions on leach- and migration rates remains to be investigated in more detail; however, one should keep in mind that the leach rates of BWPs containing considerable amounts of a wide variety of strong organic and inorganic complexing agents have already been determined, and that a purely diffusion-controlled leach mechanism will be little influenced by the (additionally formed) complexing agents and that the amount of all β-γ-emitting nuclides in BWPs of ≤ 1 Ci/l is less than 10^{-3} wt.-%.

As already mentioned, the influence of complexing agents on the migration or retardation of leached radionuclides in various environmental media is beyond the scope of this paper.

In summary, based on the results of accelerated radiation tests and the evaluation of relevant complementary experiments the following conclusions and predictions appear to be justified:

- The integral radiation doses absorbed by BWPs containing about 50 wt.-% waste solids and initially ≤ 1 Ci fission and activation products per litre product (ca ≤ 0.7 Ci/kg BWP) will normally not exceed 2×10^8 rad which, however, cause the generation of radiolysis gases (e.g. ≤ 280 l H_2/200 l BWP), an increase in the porosity and hardness and eventually a swelling of the BWP.

- Neglecting synergistic effects the radiation effects alone will not worsen the physical and chemical properties of the original product to such an extent that the individual leach rates of the radionuclides and the integral leach rate of the BWP is increased by more than a factor of 10 until "complete" decay of the beta- and gamma-emitting isotopes.

  For most products (i.e. products of lower specific activity) the change of the leachability will be negligibly small as the radiation induced enlargement of the leachable surface and porosity as well as the changes in consistency, homogeneity and chemical composition are insignificant.

- The incorporation of higher amounts of alpha-emitters which would contribute an integral dose far beyond 10^9 rad should be avoided as the present state of knowledge is insufficient to predict their influence on the integrity of the product and the leach rate in the very long term.

- The radiation degradation of BWPs caused by doses of up to about 10^9 rad can be considered as acceptable in view of their physico-chemical long-term stability and the leachability of the contained radionuclides.

  The nature and amounts of gaseous radiolysis products released from the BWPs into the surrounding will not cause any untolerable physical or chemical attacks (pressurization, fire, explosion, chemical reactions) on the integrity of the stored BWPs and/or the geologic bedrock repository after it has been properly backfilled and sealed.

10.4.2 Thermal effects and stability

The impact of heat generated by the embedded radionuclides has to be limited to assure that no premature degradation of the BWP occurs and that no thermochemical and thermomechanical interactions endanger the structural stability of the repository.

Temperatures of BWPs causing their melting, decomposition and combustion must be avoided.

The specific power of BWPs depends on the amount of the incorporated radionuclides, their half-life and the kind and energy of their radiation.

The heat output of the stored BWPs considered is low - even in the first years of storage - and will have an higher impact on the waste material itself than on the near-field waste package environment.

Assuming that 200 l of a BWP contain 200 Ci Cs-137 (or 2.3 g Cs-137), the heat generation would be a little less than 1 Watt or the product would have a specific power of 0.005 W/l.

In case a BWP would contain 1 Ci Sr-90/l the specific power would be 0.0067 W/l, and if the BWP would contain 1 Ci Pu-238/l, the specific power would be 0.032 W/l.

Usually, the mixture of radionuclides in actual BWPs of ≤ 1 Ci/l generates a specific power of about 0.002 W/l or less.

The heat generated in 220 l BWP blocks of an initial specific activity of ≤ 1 Ci/l is insufficient at any time to raise the centre temperature of the block above 60°C. Consequently, there is no risk of rapid deformations, sedimentation, migration of radionuclides, dangerous (exothermal) chemical reactions, or self-ignition.

In a properly backfilled and sealed repository burning of bitumen-waste mixtures or explosions are highly unlikely due to the lack of an ignition source and/or oxygen or the presence of water. Even if an explosion of a gas mixture would take place no inflammation of BWPs or serious damages of the repository would occur.

The entire activity of reactor wastes to be stored in ALMA has been estimated to be about 130 000 Ci generating a decay heat of about 600 W.

BWPs stored in ALMA will attain the ambient repository temperature (5 - 15°C); the temperature difference between centre and outside of a free-standing BWP block will be less than 0.5°C.

The thermal effects from the embedded radionuclides and the prevailing temperature at the repository will neither endanger the long-term stability of the BWPs nor that of the repository.

Dimensional and mechanical stability

10.4.3 The dimensional stability can be changed by the influence of temperature, radiation, water, pressure, chemical reactions and biological degradation. The dimensional changement of BWPs stored in a repository influences the surface/volume ratio of the individual product block as well as of a larger mass of bitumen-waste mixtures which could be formed under certain storage conditions (great depth) due to container damage and the thermoplasticity of bitumen.

The BWPs show a considerably lower fluidity (kinematic viscosity) than the pure bitumen matrix at a given temperature.

The dimensional instability of BWP-blocks is usually of greater importance during extended interim-storage periods (e.g. in case of perforated product containers as an easy retrievability is desirable) than during final storage. A container-less monolithic BWP-block will creep, due to the plasticity of the bitumen matrix, in the course of time and adapt a shape imposed by the surrounding.

The creeping rate diminishes with time due to the various hardening effects (radiation, oxidizing agents, etc) and can increase by the temperature prevailing in the repository and the decay heat of the incorporated radionuclides.

If disposal of BWPs of specific activities of greater than about 1 Ci longlived radionuclides per litre are considered in large repositories (caverns) with a diameter to height ratio of about unity the fluidity of BWPs can pose a heat removal problem due to their low heat conductivity.

The fluidity of BWPs can have a negative or a positive effect on the leachability of radionuclides at the repository depending on whether the creeping of the bitumen matrix enlarges the surface exposed to the leachant or whether the leachable surface and the access of water (by tightening cracks and pores) are decreased.

In the proposed ALMA-concepts /R-28/ it is forseen to orderly emplace the BWP-drums in the repository in several layers and to cast concrete (or another material) of a thickness of about 10 cm between the individual drums and each layer of drums; in this way a high dimensional stability is guaranteed. Proper choice of the concrete composition and execution of the casting ensures not only the form stability of the BWPs for more than 500 years but also a protection against container corrosion, water penetration and radionuclides' spreading.

Thus, the measures foreseen in ALMA (or equivalent ones) and the low storage temperature eliminate or reduce to a negligible level the adverse consequences of the fluidity of BWPs.

Radiolytic gas formation and uptake of water by incorporated ion exchange resins and certain dehydrated salts (Na_2SO_4, Na_2HPO_4, Na_2CO_3 etc) enlarge the product volume by swelling phenomena and eventually the leachable surface by formation of fissures. The positive effect of the radiation and chemical induced swelling is the contribution in minimizing the interstitial volumes between solid materials in the repository in order to reduce ground water movements by convection, and in supporting the mechanical stability of the repository. The negative effect of the swelling by water uptake, which is the more significant one, consists in a decrease of the leach resistance of the BWP. This negative effect can be avoided or reduced by a suitable pretreatment of the waste material to be embedded into bitumen.

The maximum swelling in water of ion exchange resins of a cross-linking of ≥ 8% DVB has been reported in /R-30/ to be about 1.4 times higher for irradiated resins (cation and anion exchanger) than for non-irradiated resins. The actual increase in "water-swelling" of resins embedded in bitumen, due to radiation degradation, will be lower.

The force and swelling pressure exerted by ion exchange resins has recently been reported /R-30/.

Long-term storage in air and/or water of the BWPs containing inorganic salts (especially oxidizing agents and hardening accelerators such as unhydrolyzed Fe-III) will principally result in hardening which is the faster the higher the storage temperature (up to a certain limit).

Thus, the mechanical stability of BWPs will normally increase with time without loosing the ability to deform plastically, even at lower temperatures.

In summary, several internal and external effects lead to dimensional changes of the original BWPs causing usually an increased leachability of embedded waste solids and, if swelling is involved, a decrease of larger void spaces between solid matters in the repository. Dimensional changes do not endanger the overall stability of a bed-rock repository. Long-term storage in air or water causes a hardening of the matrix material.

The mechanical properties of BWPs can be considered to be highly satisfactory under the conditions of the final geological storage.

10.4.4 Long-term sedimentation and homogeneity

Theoretical and experimental studies permit to estimate the long-term settling of solids within the BWP under final storage conditions. The results obtained show that the settling of incorporated particles is slow if the temperature of the BWP in the repository is at least 20°C below its softening point.

The softening point of BWPs containing 40 - 50 wt.-% ion exchange resins or salts, or a mixture of resins and salts which are either unirradiated or irradiated up to a dose of 5×10^8 rad have normally a softening point rangning from 80 - 130°C, a break point (measured according to Fraass) ranging from -10 to +15°C and a penetration of about 0 - 20 /R-31/.

If the BWP is stored at temperatures below 40°C, which normally means much more than 20°C below its softening point, and if the BWP remains a monolithic block, the settling rate of salt particles (up to 30 µ) and of ion exchange resin particles will be less than 5 mm per 1000 years.

As in a storage period of 1000 years all long-lived fission and activation products have decayed to non-hazardous levels, and as the leach rate of eventually present actinides is very low, the influence of the settling of waste particles in BWPs stored in ALMA (at temperatures below 15°C) on the homogeneity of the products and the release of radionuclides is insignificant.

10.4.5 Chemical stability

Radioactive waste/bitumen mixtures constitute principally chemical instable systems. However, the chemical changements are usually small and slow and little destructive, provided that the attack of water is excluded and the temperature is relatively low. Nature itself has proved the very long-term stability (millions of years) of bitumens and the materials embedded in them.

The incorporation of adequately pretreated wastes from reactor and reprocessing plants into a suitable type of bitumen results in mixtures which appear to be chemical stable for the required long-term storage considering that:

- the changement of chemical species into others by the decay of contained radionuclides has no influence on the chemical long-term stability of BWPs as the weight fractions of radionuclides in LLW and ILW is very low.

- the bitumen matrix is resistant against the embedded waste constituents at the storage temperature prevailing at intermediate depth (see chapter 2.5), ·

- the ageing of the bitumen matrix, the oxidation reactions at the surface, and the "solid state" reactions within the product are all slow processes and will not necessarily be detrimental for the long-term stability of the BWP as a whole.

A socalled "compatibility test" (gas generation in vacuum at 100°C) on BWPs containing 40% bitumen and 60% nitrate salts showed a good compatibility of bitumen and embedded substances also at elevated temperatures /69/.

It should be noticed that many authors define the chemical stability of a waste product as its leach resistance against various liquids.

If the leach rate of the radionuclides and inactive waste components are known under the actual conditions of tempera- ture, pressure, leachant composition and velocity existing in the repository the "chemical stability" can be determined, i.e. leach-test data can be extrapolated to long periods of time to predict the loss of the various waste constituents from the BWP in the repository environment.

Unfortunately, those leach-test data are presently not available. However, there is experimental evidence that the leach rates determined at room temperature andatmospheric pressure using distilled water are quite pessimistic compared to the actual leach-rates to be expected in a repository of the foreseen ALMA type.

In the Soviet Union burial tests in 2 m deep trenches of clay were carried out to select optimal conditions for the final storage of BWPs using blocks of 1 - 2 tons containing various bitumens, 27 - 60 wt.-% salts, and an activity concentration of 0.09 mCi/kg BWP.

From the results obtained it has been concluded that large BWPs with a specific activity of up to 1 Ci/kg(having a leach rate of 10^{-4} - 10^{-5} g/cm^2 . day) and up to 10 Ci/kg (having a leach rate of 10^{-6} g/cm^2 . day) can be stored directly in the ground.

In France a BWP block was burried in the soil and leached by a permanently recycled amount of water for 1015 days; the activity found in the leachant was negligible: 2 x 10^{8} of the initial activity /78/.

Assuming that the integrity of the BWPs is ensured and that the coefficient of diffusion remains unchanged the quantity of the radionuclides leached after about 100 years will be insignificant.

10.4.6 Interaction with surrounding storage media

The stored product form determines not only how soluble by aqueous media it will be but also how reactive it will be with other environmental media.

The waste product package or the container-less BWPs can, in the long-term come in contact within the repository with concrete, backfilling materials (sand-bentonite mixture), the host rock, mineralized ground water, air, and possibly hydrogen, methane, carbon dioxide, and smaller amounts of other gases.

Interactions of the stored solid BWPs with the surrounding solid or gaseous substances will not result in any physical, chemical, or thermal alterations which would worsen the ability of the repository to isolate the wastes. The interaction between BWPs and the ground water medium is discussed in other sub-chapters.

The adverse effects imaginable on the integrity of the repository would be related to gases generated and to the interaction of the BWPs with the ground water.

The ground water composition (Table XXV) will change after having been in contact with the engineered barrier materials (see e.g. Table XXVI) and the BWPs.

Though not experimentally verified it can be assumed that the modified ground water solution will not endanger to any significant extent the function of the surrounding materials as physical barriers against the spreading of the leached radionuclides.

In how far the efficiency of the repository materials and the surrounding rock as a physico-chemical barrier is influenced is an important aspect, however, its discussion is beyond the scope of the considerations of this report.

It is obvious that the design and construction of the engineered structures and barriers of the bed-rock facility have to match the function of the artificial barriers with the properties and the long-term changements of the BWPs; in this respect the synergistic effects of ground water modification, swelling phenomena and gas generation on the retention function of the various barriers and on the leachability of the BWPs are of primary importance.

10.4.7 Gas generation and effects

In the assessment of the post-closure safety of the BWP-storage in a geologic repository all the potential events that might effect product stability or disrupt the integrity of the repository and provide or favour pathways for radionuclides to reach the biosphere (primarily through groundwater transport) have to be evaluated.

In the framework of the present report only those phenomena will be outlined which are directly related to the properties and the long-term behaviour of the BWPs themselves.

In this respect also the generation of gases from the degradation of BWPs deserves consideration.

The potential consequenses of gas generation from waste products degradation on the overall repository integrity and safety will not be treated in depth as it goes beyond the scope of this report.

The short-term concern over gas generation is the potential risk associated with the accumulation of flammable, explosive, and toxic gases which effect the design and operation of the repository.

The long-term consequenses of gas generation - i.e. after the repository is filled, decommissioned, and finally sealed - can be breaching of the containment integrity and pressurization of the repository /R-33/.

Gases can be generated from BWPs by:

- radiolysis,
- alpha decay (helium),
- heat,
- chemical corrosion and reactions,
- microbial degradation.

The primary gases which can be formed are:

hydrogen, carbon dioxide, carbon monoxide, methane, water, oxygen, nitrous oxides, helium, nitrogen, and - from the degradation of anion exchange resins - trimethylamine, dimethylamine, methylamine, and ammonia.

The time after which the lower explosion limit of a radiolytically generated hydrogen-air mixture (4 vol.-% H_2) will be reached in a gastight enclosure can be calculated according to the formula given in /92/;

from results obtained it has been concluded that BWPs of a specific activity exceeding 1 Ci/kg should not be stored in gas-tight enclosures because a considerable overpressure on hydrogen would develop and the lower explosion limit would be exceeded in a relatively short time. The combustibility of BWPs presents no hazard any more in a properly refilled and sealed repository, however, the generated gases may concentrate and pressurize the still remaining interstitial space if the gases cannot diffuse away sufficiently fast.

The amount and nature of gases generated by radiolysis can be calculated relatively exact.

Gas generation from thermal BWP decomposition at temperatures below 20°C and helium formation from emitted alpha particles are negligibly small.

Gases can be produced by bacterial decomposition, by the metabolic process of anaerobic or aerobic respiration fermentation, denitrification, sulfate reduction, methanogenesis, etc...

Studies carried out on the gas generation from transuranic waste (mixtures of organic material contaminated by plutonium-239) indicated that bacterial degradation of this type of waste yielding gas generation is quite significant and must be taken into account /R-33/; a preliminary gas generation rate from the bacterial degradation at 25°C of 150 kg bitumen in a 210 l drum of 1.3 moles CO_2/year/drum (aerobic) and 0.3 moles CO_2/year/drum (anaerobic) were estimated from small laboratory tests under conditions not directly comparable to a bedrock repository for BWPs.

The only long-term chemical degradation mechanism of importance generating gaseous reaction products is the corrosion of the BWP-drums usually made of mild (carbon) steels.

Mild steels are susceptible to atmospheric, aqueous, and soil environment corrosion.

The mechanism and rate of corrosion depend on the oxygen availability, redox conditions, humidity, pH soluble salt content, presence of certain types of bacteria, temperature, and several other factors.

During storage in a dry air atmosphere the steel corrosion would consume oxygen and not generate hydrogen, i.e. no gaseous corrosion products would be generated.

The emplacement of BWPs in a bedrock repository as foreseen in ALMA, will finally cause an electrochemical corrosion of the mild steel containers.

The anodic reaction is the oxidation of the metal material and dissolution of the formed ions in the electrolyte:

a) $M \rightarrow M^+ + e^-$

$Fe \rightarrow Fe^{+2} + 2\ e^-$

Simultaneously the cathodic reaction in neutral and basic solutions is the reduction of dissolved oxygen which has diffused to the steel surface:

b) $O_2 + 2H_2O + 4\ e^- \rightarrow 4\ OH^-$

If oxygen (air) is absent or kept from the metal surface, then the cathodic reaction is the reduction of water or of hydrogen ions depending on the pH of the electrolyte; these cathodic reductions lead to the formation of hydrogen:

c) $2H_2O + 2\,e^- \rightarrow H_2 + 2OH^-$ (in neutral sol.)

d) $2\,H^+ + 2\,e^- \rightarrow H_2$ (in acidic sol.)

In the presence of oxygen the reaction (b) always takes place preferentially over reaction (c) and is up to 100 times faster /R-33/; in the absence of oxygen the aqueous corrosion rate of mild steel at 25°C would be about 0.005 mm/year, with this rate doubling with each increase of 10°C.

From the studies referred to in /R-33/ the following maximum gas generation/consumption rates were estimated for a 210 l drum of a total surface area (inside and outside) of 4 m^2, immersed in a saturated rock salt brine at 25°C:

aerated - 22 moles oxygen (O_2) per year and drum consumed,

deareated - 2 moles hydrogen (H_2) per year and drum produced.

If in a sealed repository the available oxygen is consumed cathodic reduction of water will commence generating hydrogen at a maximum credible rate of 2 moles H_2/year/drum.

If oxygen is depleted and water is present, a total of about 670 moles of hydrogen (H_2) can theoretically be produced from the complete corrosion of a 210 l (55 gallon) drum.

Any oxygen present will substantially reduce the generation of hydrogen. Hydrogen will not be produced if moisture does not contact the steel drum (e.g. at the inside surface protected by the bitumen matrix). In a dry air environment the only reaction would be the extremely slow oxidation of the mild steel.

It appears that the radiolytically generated gases will form the main contribution to the total gas amount produced from the degradation of BWPs stored under the conditions foreseen in ALMA.

Adverse effects from the generated gases on the BWPs and the repository can in any case be prevented by an appropriate repository design and backfilling technique; the host rock itself presents most probably not a gas-tight enclosure.

Though site- and product-specific calculations and studies remain still to be carried out one may confidently predict that the rates, amounts, and the kind of gases formed from the degradation of the stored BWP-packages will neither endanger the integrity of a bedrock repository nor enhance the release of radionuclides from the BWPs to any significant extent.

10.4.8 Effects on the leach resistance

The various factors influencing the leachability of radionuclides from BWPs have already been discussed in several preceding subchapters. In addition to those factors it remains to point out that the composition of the ground water /R-34/ is of special importance for the leach rate of the waste constituents and furthermore also for the corrosion rate of structural materials of the repository and the container material, the changements of the backfill material, and the maximum solubility of chemical compounds and elements.

Prior to its contact with the "artificial" barrier materials the ground water has a low-mineral and salt content and is either slightly acidic or slightly alkaline (e.g. the ground water found in the Stripa mine - which obviously is disturbed by the drainage system applied - has a pH of about 8.5 /R-26, Part I, Vol. II, p. 68/.

Contact of ground water with repository materials increases most probably the salt load and the pH. Both effects would have a favourable overall influence on the integral leach rate (and the migration rate of actinides), i.e the overall retention of radionuclides will not be worsened by the "modified" ground water composition.

In principal, almost neutral ground water of a reducing nature, lacking strong complexing agents has the least destructive effect on most waste products.

10.5 Microbial Stability

The possible microbial degradation of waste-bitumen products is an important factor to be considered in the selection of the proper mode of their terminal isolation, which could include placement in more or less deep geologic formations and near-surface trenches.

ZoBell and Molecke /275/ have pointed out that the bacterial degradation of bitumen used for the embedding of radioactive wastes has up to now received negligible study.

This might be due to the opinion of some investigators /25 86/ who consider bitumen as being sufficiently resistant towards bacterial degradation

However, within the framework of radwaste bituminization, only some French reports have been published /4. 177, 212/ in which studies on the microbial attack of bitumen/salt mixtures are described.

It seems that presently no results are available from experiments on the microbial attack of radioactive waste-bitumen products and waste-bitumen products containing ion exchange resins.

ZoBell and Molecke /275/ compiled among others the following observations and suggestions on the title subject (complementing those cited in chapter 3.4)·

"Many microorganisms have the ability to produce alcohols, esters, ketones and other fermentation products in which bitumens are soluble. Such solubilization may be of negligible significance during a period of several years. but could result in the perforation or partial matrix solubilization of bitumen encapsulants or coating within decades or centuries In an environment where water and organic matter are present, along with elements essential for microbial growth, some solubilization of bitumen by fermentation products can be expected.

The microbial activities of greatest concern for nuclear waste disposal are those involving metal metabolism, particularly metabolism of the heavy metals (actinides).

Bitumen-oxidizing bacteria have caused deterioration of bitumen-paved streets and bitumen coatings on buried pipelines and other steel structures. This let suggest that under certain conditions of storage where microorganisms, minerals essential for their growth, and a little moisture are present, bituminous coatings might be breached by bacteria within a few decades.

One should be aware that waste-bitumen products (- which are free from bacteria and fungi just after their generation -) may become contaminated by bacteria:

- prior to burial in the final waste repository,

- present in other (organic) wastes buried in the same repository,
- introduced into the repository during excavation and emplacement operations".

Colombo and Neilson, Jr. report /278/ that microbial degradation by microorganisms can result from surface attached growth on the solid BWPs or growth supported by (solubilized) organic nutrients leached from the BWPs.

They carried out an experiment to ascertain whether organic compounds are removed from Portland type II cement, Borden Casco Resin 2 urea-formaldehyde, vinyl ester-styrene, and Pioneer 221 bitumen specimens during leaching in distilled water no organic carbon was detected in the leachant from the cement specimen, and only a small amount in that of the bitumen (0.0010 g/300 ml water after 10 days from a sample of 103 g, vol./surface ratio = 0.75 cm) and vinyl ester-styrene specimens (0 010 g/300 ml water after 10 days from a sample of 198 g, vol./surface ratio = 1.1 cm), but the urea-formaldehyde specimen leachant had an organic carbon content of 2.85 g C/300 ml H_2O.

The form of the organic carbon present in the leachant and its ability to support microbial growth were not determined If microbial growth were possible, its extent would depend on the amount of organic carbon available. The above mentioned experiment concerned the potential biodegradability of the solidification matrix materials but did not consider the effect of the incorporated waste.

The authors point out:

- that the waste itself, such as cellulosic filter media or organic ion exchange resins, may be biodegradable;
- that synergistic interactions between the solidification matrix and the waste may either promote or impede biological degradation of the waste form;
- that biodegradation may promote dispersion and biological concentration of contained radionuclides increased leachability, generation of gas (potentially explosive), and deterioration of mechanical properties

In connection with the long-term stability of various BWPs towards microbial attack Brodersen /277/ recently carried out a critical review of the relevant literature of which the main findings and views are cited hereafter·

The concern of the microbial attack of BWPs is based on the assumption that it may lead to an increased leachability of the embedded radionuclides.

Some general conditions must be fulfilled to enable the growth of microorganisms on the BWPs or nearby them. These are:

- the presence of germs of bacteria or fungi which can use the hydrocarbons in the bitumen as source of carbon;

- excessibility to an exposed surface of the BWP;

- the presence of water and sufficient amounts of nutrients (N, P, etc) needed.

Low temperatures (e.g. zero to minus 10°C), extreme pH values (e.g. water having been in contact with concrete), and the presence of toxic metal ions (e.g. Hg and Cu) or other substances (e.g. aliphatic amines, dihydroxy-hexachloro-diphenylmethane) will hamper or prevent the growth of microorganisms.

Studies on the biodegradation of plastics have shown that many organisms seem to be unable to attack hydrocarbons having a side chain. Usually polyethylene is not attacked. Polystyrene and copolymers of styrene seem not to be attacked by microorganisms, however, a very small biodegradation of UV-irradiated material could be detected using the C-14 method (it can be assumed that gamma-radiation has the same effect as UV-radiation).

The growth of bacteria in ion exchange resins is a well-known phenomenon but the growth medium is most probably not the resin material itself but organic material introduced from the outside by the aqueous phases in contact with the resins: amine groups can possibly also play a role. The literature review has shown that microorganisms can degrade bitumen under conditions favourable for their growth, however, the degradation of thick bitumen layers will proceed slowly.

Based on the present knowledge on the microbial degradation of bitumen-waste mixtures it cannot be excluded that the microbial attack of bitumen effects an increased leach rate of radionuclides present in BWPs. If one needs more assured and detailed information on the microbial degradation of BWPs further experiments have to be carried out.

Current R and D programmes on nuclear waste management in Europe and the United States /275/ include studies on the biodegradability of different types of waste forms under various storage conditions.

The microbial resistance of some typical BWPs is already being investigated in France since 1973 /212/:

- An experiment in which BWP-samples of evaporator concentrates embedded in three different types of bitumen (M 40/50, H 80/90, and R 90/40) were buried in the ground was begun in Cadarache in 1973 and lasted 31 months. The samples were buried at a depth of 50 cm in two different soils; one soil was a meadow with natural drainage, the other was a marshy area permanently damp and flooded for five months of the year. The microbial examinations of the samples buried for 31 months revealed no attack by the microorganisms existing in the two types of soil (i.e. total germes, pseudomonas, nitrate- and sulfate reducing bacteria).

- A 100 l BWP-block composed of evaporator concentrate residues (from Cadarache) and bitumen M 40/50 was buried in 1974 in the ground in a marshy zone: the first examination after 18 months showed the same microbial stability of this block as that of the laboratory samples.

- Small samples of the same nature as those mentioned above were placed in columns filled with sand under aerobic and anaerobic conditions for a period of 8 months; no particular proliferation of germs was detected on the surface of the BWP-samples though an intense bacterial culture had been developed in the sand columns.

In conclusion one may state that:

- in the past the possible microbial attack of BWPs received insufficient attention;

- at present a tendency can be noticed to overemphasize the significance of the microbial attack of wastes and its possible negative consequences, particulary in the United States /275, 278, R-33/; (the negative consequences of microbial degradation or metabolic action could be an enhanced leaching of radionuclides, gas evolution, enhanced migration of radionuclides due to bacterially produced chelating agents, solubilization of the bitumen matrix due to bacterially generated organic solvents, bioconcentration of fissionable radionuclides to a sufficient extent to result in a criticality incident /275, p. 10/; the consideration of a criticality occurrence by bioconcentration in a final repository containing adequately conditioned waste is one of the typical examples of little realistic scenarios which contribute in delaying the realization of safe waste management schemes);

- microbial actions can enhance or significantly retard (by absorption or precipitation) the migration of radionuclides in an aqueous phase /275/;

- the temperatures of up to 200°C used in the bituminization of radwastes are high enough to kill most bacteria and fungi contained in the original waste material;

- hyperbaric oxygen, carbon dioxide, and certain other gases are bacteriostatic or bactericidal at pressures of only 5 - 10 bar;

- the few species attacking bitumen are most abundant in soil or bottom sediments that have been in contact with crude oil or bitumen;

- the conditions for microbial attack of bitumen must be perfect for a very long time to produce any noticeable damage, thus the speed at which microbes attack bitumen is very slow.

With respect to the storage of BWPs in ALMA one has to take into account that - with the exception of the presence of water - the conditions most probably prevailing in the repository (high pH of the "modified" ground water, low temperature, increased pressure, presence of hyperbaric oxygen and carbon dioxide) are not favourable for the growth of microorganisms.

If further studies and experimental results should convincingly prove that a microbial degradation of BWPs under the given storage conditions will take place and lead to a significant increase of the leachability or spreading of long-lived radionuclides, it will be relatively easy to design the final repository and to create conditions in such a way that the microbial degradation is prevented or at least reduced to a negligible level.

10.6 Other Long-Term Aspects

10.6.1 Criticality risk

It is required for all steps in nuclear waste management to insure that a nuclear criticality accident is possible only if at least two unlikely, independent and concurrent or sequential changes have occurred in the conditions essential to nuclear criticality safety.

A system is said to be nuclearly critical when the fission rate in the system is constant in the absence of extraneous sources of neutrons. For a system to become critical, it must contain fissile material, e.g. U-233, U-235 or Pu-239, in such a composition and configuration that the production rate of neutrons as a result of fission is equal to the loss rate of these neutrons. The loss mechanisms for neutrons are capture (non-fission absorption) and leakage (escape) from the fissile system.

The ratio between the number of neutrons available for fission in two successive generations is called the multiplication factor k; the condition necessary for the maintenance of the chain is obviously $k = 1$. If $k > 1$, the number of neutrons increases in each generation and the chain is said to be divergent, which implies the system runs the risk of becoming explosive. If $k < 1$, the chain is convergent, and reaction ceases after a limited number of generations.

With increasing mass of fissile material, the loss of neutrons by escape, which is proportional to the surface area, is more than compensated for by the increase in the number of neutrons produced in the bulk, and for a given system, one reaches a critical mass for which the effective multiplication factor k_{eff} becomes equal to 1;

$$k_{eff} = k\,(1 - P),$$

where P is the probability of escape from the surface.

The probability of escape, and hence the critical dimensions are reduced if the system is surrounded with a neutron reflector. Any material which acts as a moderator can equally well serve as a reflector (e.g., graphite, BeO, D_2O).

If $k_{eff} > 1$, the system is supercritical; alternatively for $k_{eff} < 1$, it is subcritical and the neutron density tends to decrease.

Criticality safety has to be demonstrated under normal and accident conditions by calculating the effective multiplication factor (k_{eff}) which must be sufficiently below unity.

Considering the criticality risk during the long-term storage of BWPs containing wastes from the operation of LWRs and the reprocessing of LWR-fuels the total amounts and concentrations of the fissile isotopes U-235 and Pu-239 have to be taken into account in the first instance.

Knowing the amount of U-235 and Pu-239 (or the isotopic mixture of U or Pu) the maximal possible amount of the fissile isotopes in the repository is given by its storage capacity.

The concentration, total amount, and the isotopic composition (enrichment) or uranium in the bituminized LLW and ILW from light water reactors and from the reprocessing of LWR fuels are such that a criticality accident cannot occur in a geologic repository, also not in the very long-term.

As a very conservative estimate for the critical concentration of plutonium a limiting value of 1 g Pu-239/l BWP can be assumed, i.e. approximately 200 g Pu-239 per 200 l package (drum).

Considering the homogeneous Pu-239/water system the critical plutonium concentration is estimated to be about 7.2 g/l, the critical infinite layer thickness 5.5 cm (Pu-concentration 400 g/l), and the critical mass at full water reflection is 510 g.

The smallest theoretical possible critical mass in spherical form in a repository system of the ALMA type (flooded by ground water) can be calculated to be in the order of several kilogrammes Pu-239.

It has been estimated /R-28/ that the total amount of Pu-239 in ALMA at the time of repository closure will be 0.3 Ci which corresponds to 4.86 g Pu-239. This amount is far below the critical mass of the fissile plutonium isotopes Pu-239 and Pu-241.

The highest plutonium concentration will be found in BWPs from reprocessing plants; it is in the order of a few milligrammes per litre BWP, typically 5 mg Pu/l. This concentration is a factor of about 200 below the smallest critical concentration of 1 g Pu-239/l BWP.

There is no credible mechanism thinkable which could effect a local concentration inside or outside the BWPs to an amount of fissile plutonium causing an uncontrolled chain reaction.

The following conditions are required to effect a significant reconcentration of plutonium which has originally been present in a homogeneous BWP:

- Pu has to be mobilized (preferably in a soluble form),
- the transport medium (ground water) must have a defined flow direction,
- the Pu must be deposited (e.g. by sorption or precipitation at a well-defined place.

Independent of whether a criticality incident will be possible or not the consequenses of an assumed excursion shall be outlined.

It may be stated that any significant mechanical effect on BWPs, the repository, or the host rock can be excluded. as a stable local inclusion of fissile material would be required besides the formation of a rapid excess reactivity. A stable (fixed), local inclusion of fissile material cannot exist, if one has to assume an increase of the concentration of fissile nuclides.

The reconcentration of fissile material by geochemical mechanisms would take place very slowly during geological periods of time so that the reactivity increase (i.e. the relative increase of k_{eff}) and the formation of a critical system will be approached very slowly.

Even if a great excess reactivity of the fissile material assembly would be introduced within seconds which in reality is impossible, the mechanical effects or damages would be negligibly small. This conclusion can be drawn from experimental results and from criticality incidents which occured in the past /R-35, R-36/.

When the fissile material system has become critical it heats up and the radiolysis and formation of vapour bubbles stop the power increase. The heating effects that the system becomes subcritical again.

There are reconcentration mechanisms thinkable which would stabilize the system for a certain period of time until the loss of reactivity predominates due to fissile material consumption and accumulation of fission products. The occurrence of excursions in a sealed geologic repository for BWPs would cause a total energy release and radiation doses which do not endanger the stability of the repository or the health and safety of people.

One may conclude that the nuclear criticality risk associated with the geologic storage of BWPs containing reactor wastes can be neglected. Reprocessing wastes of unnormally high plutonium concentrations or contents will not be conditioned by incorporation into bitumen.

The concentration and amounts of fissile and fissionable actinide isotopes in BWPs, containing LLW and ILW from the reprocessing of irradiated LWR-fuels, are such that only uncredible conditions and reconcentration mechanisms could lead to a nuclearly critical system. Even if an excursion would occur there would be no harmful consequences for people or the repository.

10.6.2 Helium generation

Helium generated as a result of the alpha-decay of actinide isotopes remains either in the BWP or diffuses out and away in the surrounding. It should be recalled that the BWP-containers are normally not gas-tightly closed to avoid their pressurization by gases generated during storage.

As the amount of alpha-emitting isotopes in BWPs is either negligibly low (BWPs from reactor stations) or relatively low (BWPs from reprocessing plants) the contribution of the helium generated (about 2×10^{-6} moles He/Ci/year) to the total gas inventory in the products or in the repository is exceedingly small and will not have any deleterious influence on the long-term stability of the various BWPs or the repository.

10.6.3 Energy storage

The stored energy (Wigner-energy) within BWPs will not lead to a dangerous temperature rise as this fraction of latent energy is small due to the relatively low radiation dose rate and dose and the nature of the waste products considered here.

Any stored energy which might be accumulated in the repository media, surrounding the BWPs (e.g. moraine, bentonite, concrete, granite) - due to gamma radiation - will also not cause serious thermal excursions or other undesirable effects endangering safe operation or storage in the final repository.

One can state that the amount of energy possibly stored in the BWPs and the surrounding materials and the conditions under which it could be released (annealed) are such that no safety hazards are involved.

10.7 Conclusions

It appears that properly prepared mixtures of low- and intermediate-level radioactive reactor or reprocessing wastes with bitumen form products which possess adequate properties and long-term stability for final geologic storage ensuring a satisfactory containment of radionuclides with a sufficient degree of reliability for the requisite storage period; their emplacement in a bedrock repository at a suitable site and sufficient depth will exlude that any return of radionuclides to the biosphere would lead to unacceptable levels or exposure to people or their environment.

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PART II

BIBLIOGRAPHY OF THE BITUMINIZATION OF RADIOACTIVE WASTES

PART II

BIBLIOGRAPHY ON THE BITUMINIZATION OF RADIOACTIVE WASTES

This part of the survey report presents a comprehensive bibliography covering the relevant literature from 1960 (the year of the first application of bitumen for the conditioning of radioactive waste) until 1978. As far as known and available the papers published in 1979 were included.

All titles of publications are given in English.

If the publication has not been written in English the original language is indicated in brackets at the end of the reference. Due to space limitations abstracts of the papers have been omitted.

The references are given in a year by year sequence; within the year of publication the papers are listed according to the alphabetical order of the authors' name or the first name (senior author), if more than one author is given.

Identical texts published in different papers have sometimes been listed two times (under a different reference number) to ease the availability of the publication for the reader.

Literature on the bituminization of radioactive wastes referred to in Part I by a number only is contained in this bibliographical part, whilst references to other literature, indicated by the letter "R" (Reference) and a number, are listed at the end of the text of Part I under the heading "References".

References compiled after the typing of the bibliography was terminated are given in a "Supplement"; they are indicated by a number and a letter (mostly "a").

The table hereafter gives the number of publications per respective year.

Year	60	61	62	63	64	65	66	67	68	69
Number of publications	2	1	0	1	6	8	3	5	24	15

Year	70	71	72	73	74	75	76	77	78	79
Number of publications	20	15	33	21	12	25	47	30	19	17

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ANNEXES

ANNEX I:	Quality control methods for bitumens
ANNEX II·	Characteristics of "SHELL" bitumens
ANNEX III:	Composition of natural waters
ANNEX IV·	Typical composition of granitic rock

ANNEX I

QUALITY CONTROL METHODS FOR BITUMENS

- General Description*) -

A. PENETRATION

B. SOFTENING POINT

C. PENETRATION INDEX (P.I)

D. DUCTILITY

E. BRITTLENESS

F. FLASH POINT

G. SOLUBILITY IN PETROLEUM NAPHTHA

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*) Note: For the execution of the "Standard Test Methods" the procedures described in detail in the respective national or international normes have to be followed.

A. PENETRATION

The penetration is an empirical measure of consistency; it is measured by means of a penetrometer. The method is very simple.

A standardized needle is allowed to penetrate under standardized conditions of time, temperature and load in a bitumen sample placed in a container; the depth of the needle penetration is measured in 1/10 mm.

Usually, the following conditions are applied:

- time: 5.0 ± 0.2 sec
- temperature: 25 ± 0.1°C
- load: 100 ± 1 g.

There is a close relationship between the penetration and the viscosity (and elasticity). For all bitumens the relation between the logarithm of the penetration and the logarithm of the time (t) remains linear; thus:

$$\log \text{penetr.} = B \log t + C$$

For purely viscous bitumens $B = 0.5$, for elastic bitumens $B = < 0.5$.

B being the slope of the log penetration - log time line, can be considered as a measure of the elasticity and consequently of the type of bitumen.

Penetration Test Methods have been standardized e.g. by

- ASTM (D 5 - 52)
- IP (49 - 56)
- NF (T 66 - 004)

B. SOFTENING POINT

Bitumens have no real melting point. When heated they gradually become softer changing from brittle or exceedingly thick and slow-flowing materials to softer and less viscous liquids. Any determination of a softening point is therefore quite arbitrary. Normally, the softening point is determined either:

a) according to the Ring and Ball (R + B) method, or

b) according to Krämer and Sarnow *).

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*) Krämer, G., and Sarnow, C., Chem. Ind., (Berlin) 26 (1903) 55.

a) Ring and Ball Method

A brass ring of given dimensions is filled with molten bitumen and cooled to room temperature. The bitumen is loaded with a steel ball (weight 3.50 ±0.05 g, diameter 9.5 mm) and the whole is heated in a water bath at the rate of 5°C/min.

The temperature at which the disk of the bitumen sample within the horizontal ring is forced downward a distance of 25.4 mm (1 inch) under the weight of the steel ball is reported as the Ring and Ball softening point.

For bitumens having R + B softening points above 80°C glycerin is used as heating liquid instead of water.

The Softening Point Determination has been standardized e.g. by ASTM (D-36-26); DIN 1995/U 4.

b) Softening Point According to Krämer and Sarnow

The softening point according to Krämer and Sarnow is the temperature at which under standardized conditions a bitumen layer is penetrated by mercury.

The test is carried out as follows:

A small ring is filled with bitumen and fastened to a tube. An amount of 5 g of mercury is deposited on it.

The tube is immersed in a water bath (or at temperatures above 80°C in glycerin) and heated at a uniform rate till the mercury drops through the bitumen and reaches the bottom of the bath. The temperature at which this takes place is recorded as the softening point.

There are several variants of this method in existence, producing discrepant results.

Determination procedures have been standardized, e.g. by DIN (1995) and KVBB-N 1013 (Netherlands).

No direct conversion is possible from the R + B softening point to the softening point according to Krämer and Sarnow (- the latter one is found to be about 10 - 20°C lower -).

The viscosity of bitumens at their R + B softening point is on an average 12 000 poises and the penetration at the R + B temperature is 800 ±200.

C. PENETRATION INDEX (P.I.)*

This is not a test method, but an index figure, introduced by Pfeiffer and Van Doormaal** to indicate the temperature susceptibility of the penetration of a bitumen.

If the logarithm of the penetration is plotted against the temperature (T), an almost straight line is obtained, and this suggests that the slope of this line might be used as a susceptibility index. This would require two penetration tests to be carried out at two temperatures for each bitumen. If, however, the straight line is extrapolated to the temperature of the ring-and-ball softening point, the corresponding penetration proves to be approximately 800. Therefore the P.I. can be calculated from one penetration and the ring-and-ball softening point, the slope of the penetration line being given by

$$\frac{\log 800 - \log \text{penetration}}{T_{R+B} - T}$$

where T refers to the temperature in °C at which the penetration test is carried out.

The P.I. of bitumens varies from - 2.6 to + 8. The lower the P.I., the higher the temperature susceptibility.

A close relationship exists between the P.I. and the rheological type of bitumen, and this is the real importance of this index figure.

D. DUCTILITY

The ductility is determined according to Dow /R-7/, accepted as ASTM Standard Test (D 113-35) in 1935.

The method consists in stretching a bitumen briquette narrowed in the middle to a cross section of 1 cm^2, in a water bath of 25°C at the rate of 5 cm/min.

Various modified procedures have been suggested, or inserted in the specifications.

The modifications concern mainly the temperature and the rate of elongation.

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E. BRITTLENESS

The brittleness can be determined according to the method of Fraass /R-8/

A thin layer (0.5 mm) of asphaltic bitumen is spread on a steel plaque. This plaque is bent under standardized conditions at decreasing temperature. The temperature at which the bitumen film breaks is recorded as the Fraass breaking point. It indicates the temperature at which a given slight elongation causes breakage.

The rate of deformation is slow.

F. FLASH POINT

The flash point of a substance is the lowest temperature at which application of a test flame causes the vapours above a sample of the substance to ignite.

A number of flash point testers are in use, including the Pensky-Martens (ASTM D93-61), Cleveland Open Cup (ASTM D92-57), and the Tag types (ASTM D1310-59T).

For the determination of the flash and fire points of all petroleum products (except fuel oils and those having an open-cup flash below 80°C) usually the Cleveland Open Cup Method is used.

In this method the test cup is filled to a specified level with sample. The temperature of the sample is increased fairly rapidly at first and then at a slow constant rate as the flash point is approached. At specified intervals a small test flame is passed across the cup. The lowest temperature at which application of the test flame causes the vapours above the surface of the liquid to ignite is taken as the flash point.

To determine the fire point, the test is continued until the application of the test flame causes the product to ignite and burn for at least 5 seconds.

The Cleveland Open Cup Tester consists of the test cup, heating plate, test flame applicator, heater (flame-type or electric), and various supports.

Note: The "ignition point" (self-ignition temperature) is the temperature at which a sample of the product ignites by itself in contact with air.

G. SOLUBILITY IN PETROLEUM NAPHTHA

This solubility test is employed mainly for purposes of identification. It is also used to a certain extent for determining the adaptability of bituminous substances for a given use, for gauging the uniformity of supply, and for purposes of factory control. As a general principle, the harder the bituminous product, the smaller will be the percentage that dissolves in petroleum naphtha. Asphaltites are relatively insoluble in this solvent. Mineral waxes, peat-, lignite- and shale-tars or pitches are largely soluble. The solubility of native and petroleum bitumens varies, depending largely upon their hardness, and also in the case of petroleum bitumens upon the extent to which the distillation has been carried. Coal-tar pitches are relatively insoluble in 88° Baumé petroleum naphtha.

The portion soluble in petroleum naphtha has been termed "petrolenes" by some, and "malthenes" by others, whereas the non-mineral constituents insoluble in naphtha are generally referred to as "asphaltenes". It is important that the petroleum naphtha should be derived from petroleum composed entirely of open-chain hydrocarbons, and test exactly 88° Baumé, equivalent to a specific gravity of 0.638 at 60°F./60°F.

The method has been standardized by the American Association of State Highway Officials under the designation of AASHO T46-35. It is not an ASTM Standard Method.

ANNEX II

CHARACTERISTICS OF "SHELL" BITUMENS

TYPE	Specific density at 25°C	Ring and Ball softening point	Penetration at 25°C - 5 sec 100 g. Needle No 2	Ductility at 25°C	Solubility in CS_2	Flash poin "Cleveland open cup
REFINED BITUMENS	kg/l	°C	0.1 mm	cm	wt.-%	°C
Spramex 280/320	0.99-1.04	33- 38	280-320	> 100	> 99	> 200
Spramex 180/200	1.00-1.05	37- 43	180-200	> 100	> 99	> 200
Mexphalt 80/100	1.01-1.05	45- 52	80-100	> 100	> 99	> 225
Mexphalt 60/70	1.01-1.06	48- 56	60- 70	> 100	> 99	> 250
Mexphalt 50/60	1.01-1.06	50- 58	50- 60	> 100	> 99	> 250
Mexphalt 40/50	1.01-1.06	52- 60	40- 50	> 100	> 99	> 250
Mexphalt 20/30	1.02-1.07	59- 69	20- 30	> 30	> 99	> 250
Mexphalt 10/20	1.02-1.07	65- 75	10- 20	> 5	> 99	> 250
BLOWN BITUMENS						
Mexphalt R. 75/30	1.01-1.06	70- 80	25- 35	> 4	> 99	> 200
Mexphalt R. 85/25	1.01-1.06	80- 90	20- 30	> 3	> 99	> 200
Mexphalt R. 85/40	1.01-1.05	80- 90	35- 45	> 3	> 99	> 200
Mexphalt R. 95/15	1.01-1.06	90-100	10- 20	> 3	> 99	> 200
Mexphalt R. 110/30	1.01-1.06	105-115	25- 35	> 2	> 99	> 200
Mexphalt R. 115/15	1.01-1.06	110-120	10- 20	2	> 99	> 200
HARD BITUMENS						
Mexphalt H. 80/90	1.03-1.08	80- 90	6- 12	-	> 99	> 320
Mexphalt H 110/120	1.04-1.09	110-120	3- 7	-	> 99	~ 320
Mexphalt 135/10	1.02-1.07	130-140	7- 12	> 1	> 99	> 200
Mexphalt 155/7	1.02-1.07	150-160	5- 10	-	> 99	> 200

Source: Belgian SHELL N.V., Cantersteen 47, 1000 Brussels.

ANNEX III

COMPOSITION OF NATURAL WATERS /R-34/

Analysis	Units	Rain-water	Surface-water	Bedrock-water	Ocean-water
Depth	m	0	≤ 50	≤ 500	≈ surface
Age	years	0	10	100	0
pH		4-0	6-7	7-10	7.9
Eh	volts	0.9	0-0.3	-0.05	0.8
O_2	mg/l or ppm	≈ 10		< 0.1	≤ 9
COD_{Mn}	O_2, or ppm	≈ 0	10-20	5-10	1.2-3
Conductivity	μS/cm		120-150	400-600	
Ca^{2+}	mg/l, or ppm	≤ 1	≤ 20	20-60	400
Mg^{2+}	"	≤ 0.5	≤ 8	15-30	1272
Na^+	"	0.3-3	≤ 5	10-100	10561
K^+	"	0.1-0.5	≤ 2	1-5	380
Fe_{tot}	"	industrial contamin.	0.1-0.3	5-30	< 0.02
Fe^{2+}	"			1-30**	
CO_2 (P_{CO2})	"	0.6 (3 10^{-4})	5-10	0-25	28(4.10^{-4})
HCO^-_3	"	≤ 1	50-60	50-400	
Cl^-	"	≤ 5	5-20	> 20*	18980
SO_4^{2-}	"	≤ 20(2)	10-20	1-15	2650
NO_3^-	"	≤ 2	0.5-2	< 1	< 0.7
PO_4^{3-}	"		≤ 0.1	≤ 0.1	≤ 0.1
F^-	"		≤ 0.1	0.5-2	1.4
SiO_2	"	< 1	3-15	5-30	0.01-7
HS^-	"		≈ 0	< 1	
NH_3	"	≤ 0.5	< 0.1	< 0.5	< 0.05

* For relict water, the value may be 500-3000 ppm.

** 5 ppm will be selected as a typical and recommendable value (se main text in /R-34/.

ANNEX IV

TYPICAL COMPOSITION OF GRANITIC ROCK /R-34/

Chemical composition	(%)(*	Mineral constituents	(%)(**
SiO_2	67-75	Quartz, SiO_2	22-29
TiO_2	0.2-1.0	Orthoclase, $KAlSi_3O_8$	18-32
Al_2O_3	13-16	Albite, $NaAlSi_3O_8$	26-33
Fe_2O_3	0.5-2.0	Anorthite, $CaAl_2Si_2O_8$	6-16
FeO	0.5-4.0	$MgSiO_3$	1.3-3.9
MnO	0.05-0.15	$FeSiO_3$	1.7-2.9
MgO	0.2-1.3	Magnetite Fe_2O_4	1.4-1.9
CaO	0.1-4	Ilmenite, $FeTiO_3$	0.8-1.1
Na_2O	2.5-4	Apatite, $Ca_5(PO_4)_3$ (F, OH, Cl)	0.1-0.5
K_2O	3-5	(F, OH, Cl)	
H_2O	0.5		
P_2O_5	0-0.2		
CO_2	0.01-0.1		
S	0-0.04		
U ppm	15-5		

*) Range of values for Western U.S (D. Isherwood) and Sweden (KBS).

**) According to D. Isherwood.

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