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***Technologies for
in situ immobilization and
isolation of radioactive wastes
at disposal and contaminated sites***



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TECHNOLOGIES FOR IN SITU IMMOBILIZATION AND ISOLATION OF
RADIOACTIVE WASTES AT DISPOSAL AND CONTAMINATED SITES

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FOREWORD

Radioactive waste arising as a result of nuclear activities should be safely managed from its generation to final disposal in an appropriate conditioned form to reduce the risk of radiation exposure of technical personnel and of the public and to limit contamination of the environment. Such an approach and supporting technologies did not exist in the very beginning of the nuclear industrial era. In some countries radioactive wastes from past nuclear activities or accidents are still stored on nuclear sites in an unconditioned state, because of difficulties or impossibility involved in their transportation to waste conditioning facilities. There are a few sites around the world like Hanford, Chelyabinsk-40, and the Chernobyl zone, which accumulate a significant amount of unconditioned radioactive wastes.

In situ immobilization of such wastes and containment of contaminated objects as alternatives to more traditional 'ex situ technologies', applied away from the site of their occurrence, are most promising for these cases. A number of Member States are trying to take advantage of in situ immobilization technologies and have adopted or are planning to adopt them in some cases where immobilization facilities and facilities for waste disposal are located on the same site.

Because of potential advantages of in situ technologies the IAEA has decided to issue a report providing Member States access to the worldwide experience accumulated in this area.

Preparation of this report was accomplished through two consultants meetings and a Technical Committee meeting. The final report was prepared by R. Clegg of the United Kingdom, A. Mishra of India, Yu. Kuznetsov of the Russian Federation, and J. Tixier of the United States of America after review of information, data and comments received from the Technical Committee members.

The IAEA would like to express its thanks to all those who took part in the development of the report. The IAEA officers responsible for the report are A.F. Tsarenko and V.M. Efremkov of the Waste Technology Section of the Division of Nuclear Power and the Fuel Cycle.

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1. INTRODUCTION

Around the world there are land based contaminated sites that require remediation. There are also sites which have been used for long term storage where the waste is in need of final disposal, and there are disposal sites where the original waste form or repository are not performing adequately and where remedial actions are required. Some of these problems may be suitable for in situ immobilization or containment technologies. In addition, there may also be new disposal sites where in situ immobilization and containment could be considered as viable waste disposal strategies. This report describes such technologies that have been developed worldwide and the experiences applied to both waste disposal and contaminated sites.

The term 'immobilization' covers both solidification and embedding of wastes. The term 'containment', on the other hand, is defined to cover physical isolation of the waste from the accessible human environment by emplacement of engineered barriers to limit leaching and migration of the radioactivity. The term 'treatment' is also used in the document when the subject of discussion deals with the procedures, in general defined by the IAEA as a volume reduction, removal of radionuclides, or change of a waste composition. The distinguishing feature of the technologies described in this report is that they are carried out in situ. The term 'in situ' is Latin meaning 'in a natural or original position'. For the case of in situ immobilization technologies this means that the process is carried out at the final disposal location and that the immobilized waste becomes the final disposal waste form. In the case of in situ containment the waste, in its final disposal location, is isolated from the accessible human environment, as mentioned above, without any major disturbance. It is emphasized that although this report is primarily concerned with radioactive materials, many of the technologies could be applied to mixed radioactive and chemically contaminated waste sites.

This report is divided into this introduction and three main Sections. Section 2 describes in situ immobilization technologies applied to waste disposal. Both slurry and liquid waste are covered, where the waste is mixed with a solidifying agent (such as a grout formulation) and allowed to harden in situ at the final disposal location to form a monolithic waste form. This location may be an engineered vault, tank or geological repository.

Section 3 describes in situ immobilization technologies applied to sites requiring remediation. Here the radioactivity may be adventitious soil contamination or contained in waste already stored in tanks or trenches awaiting final disposal. For the latter case (existing wastes in tanks or trenches) the waste may be either solid or a slurry. For slurries in tanks, technologies are described whereby the waste is mixed in situ with various grout formulations. For solid waste in trenches the injection of polyacrylamide grout into the interstitial voidage in the waste to stabilize and form a monolithic waste form is described. With respect to in situ immobilization applied to contaminated sites where the radioactivity exists as adventitious soil contamination, technologies are described involving immobilization or embedding of the radioactive contaminants to isolate them from the geosphere or groundwater.

In the case of geosphere contamination, this may involve disturbance of the waste (such as deep mixing with a grout) or collection of widespread deposited contamination at shallow depth into one location (such as by bulldozing) for subsequent immobilization. Technologies are described to immobilize the contamination in the geomatrix material, such as using in situ vitrification.

For contaminated groundwaters, as opposed to contaminated soils, no technologies appear to have been developed to selectively remove the contamination from the groundwater and immobilize it in situ. Technologies have been developed to remove contamination in situ, such as using zeolites, but the material is then removed from the groundwater and treated ex situ. Because of this such technologies do not fit into the scope of this document. There are possible to place in situ barrier materials that also act to decontaminate groundwater, and where appropriate these have been described in Section 4 of this report.

Section 4 covers in situ containment applied to waste disposal and contaminated sites. The containment technologies that are described are designed to isolate the radioactive material from the accessible human environment by reducing dissolution and migration due to infiltrating rainwater or groundwater. The technologies are split into three categories: surface barriers, such as caps; vertical cut-off barriers, such as bentonite walls; and underground horizontal barriers, such as applied to the bottom sealing of existing waste disposal trenches.

As stated above, this report describes a range of in situ immobilization and containment technologies that have been developed worldwide. In some cases, experience with these technologies goes back a decade or more. Examples of technologies have been taken from a number of countries, including China, Germany, India, the United States of America, the Russian Federation and the United Kingdom. Figure 1 gives a technology map showing the grouping of the technologies under the headings of in situ solidification, immobilization and containment. The sections in this document have been structured according to the layout of the technology map. The third row in the technology map corresponds to the three main Sections in this document, with the fourth row corresponding to the various subsections in these Sections.

The technologies in this document are described factually and it is not intended that their inclusion should endorse them or in any way serve to recommend them as favored processes. They are merely a catalogue of former and existent approaches to solve national radioactive waste management and environmental restoration problems in different countries. Some of these technologies, like for example underground hydrofractioning, have rather limited application (USA, China). It is important, if any of the technologies described in this document are to be used for application at a new site to consider the radio-ecological impact of their usage. A systems approach should be used to calculate the overall impact, taking into account both operational and post-closure phases. The radiological assessment should consider not only the environmental performance of the solidified/immobilized waste form but also the engineering design and performance of the site infrastructure. The fundamental point is that the suitability of using in situ immobilization and containment technologies must be adjudged on a site specific basis and should also take into account national socio-economic values.

Due to the broad scope of this report it has not been possible to cover all in situ immobilization containment technologies in detail. Readers of this report considering using any of the technologies are therefore advised to follow up the technical references given in the text to obtain further information. While it is also recognized that not all international examples of in situ immobilization and containment have been covered, it is hoped that enough generic examples have been described.

Finally, in each section of this report future trends in technology development as well as emerging new technological ideas are described. These descriptions cannot be exhaustive because of the lack of published information in the field.

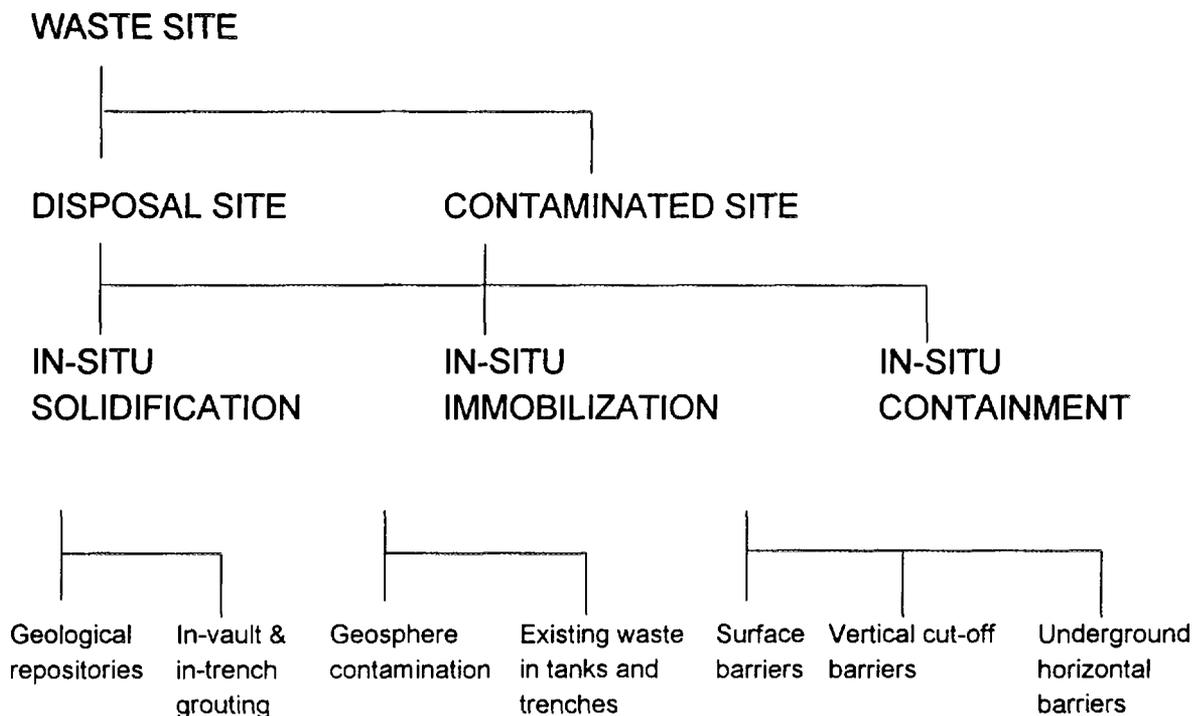


FIG. 1. Process flowsheet for in situ immobilization and isolation of radioactive wastes at disposal and contaminated sites.

2. IN SITU IMMOBILIZATION APPLIED TO WASTE DISPOSAL

It is common practice to solidify liquid radioactive waste into a suitable matrix, containerize it and send the complete package to a waste disposal site. The waste package has to conform to the specifications acceptable to the waste disposal site from a geometry point of view as an out-of-specification consignment may affect the disposal practices at the site and the economics of space utilization.

In situ immobilization at disposal sites located at the point of waste generation eliminates the need to solidify waste into individual packages suitable for transport and gives the advantages of optimization of space, reduction in radiation exposure to operation and maintenance personnel and an improvement in final product quality. In some instances it has been feasible to utilize naturally-occurring mines and hydrofractured shale formations as waste disposal sites. A number of Member States have tried to take advantage of in situ immobilization techniques and have adopted or are planning to adopt different technologies depending on the type of waste, its

2.1. IN SITU IMMOBILIZATION BY IN-VAULT AND IN-TRENCH GROUTING

2.1.1. General description of technologies

In-vault and in-trench grouting is especially applicable in cases where man-made or naturally-formed repositories, which are suitable for permanent disposal, are available near the point of waste generation. This technology has been adopted in India for fixation of radioactive chemical sludge into underground reinforced concrete trenches located at waste disposal sites [2]. In the Chinese Gobi desert, a similar proposal is under consideration for bulk cement grouting of raffinate concentrate and chemical decladding waste into underground vaults [3,4]. In the Russian Federation, in situ immobilization in underground trenches is extensively used for fixing concentrated decontamination solutions into a bitumen matrix [5,6]. The technologies adopted by the countries in the examples above are described in the following section. The types of repository and immobilant used by different countries are tabulated in Table I.

TABLE I. TYPES OF REPOSITORIES AND IMMOBILANTS USED BY DIFFERENT COUNTRIES FOR IN SITU IMMOBILIZATION BY IN-VAULT AND IN-TRENCH GROUTING

Type of facility	Immobilant	Country	Location
Near surface concrete trench	Portland cement, vermiculite	India	Tarapur
Concrete vault	Cement, dehydrating agent	China	Gobi Desert
Near surface concrete tanks	Bitumen	Russian Federation	Leningrad Nuclear Power Plant

2.1.2. Operating experience

2.1.2.1. In situ solidification using a disposable agitator

At Tarapur, India, where the disposal site is near the source of waste generation, in situ cement fixation into underground trenches has been adopted on an industrial scale for fixation of radioactive sludge [2]. Table II gives the characteristics of this chemical sludge.

The subsurface reinforced concrete trenches consist of a series of bitumen-painted compartments fitted with a disposable agitator assembly and provided with nozzles for waste cement and additives inlet and ventilation. The top of the trenches is closed with a concrete cover 440 mm thick to provide adequate biological shielding. A layout of equipment for in situ solidification of sludge in concrete trenches is shown in Fig. 2 and a photograph taken at the time of construction is shown in Fig. 3. The main process steps for this disposal method are:

- 1) Partitioning of the vault into individual compartments using mild steel plates;
- 2) Installation of agitator assembly with feed nozzles into each compartment;

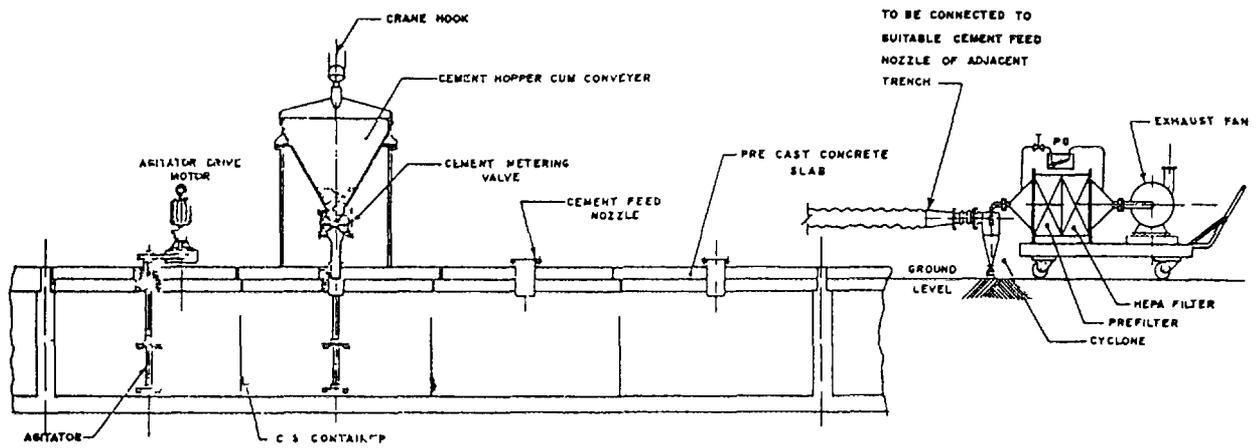


FIG 2 Layout of equipment for in situ solidification of sludge in concrete lined trenches at Tarapur, India

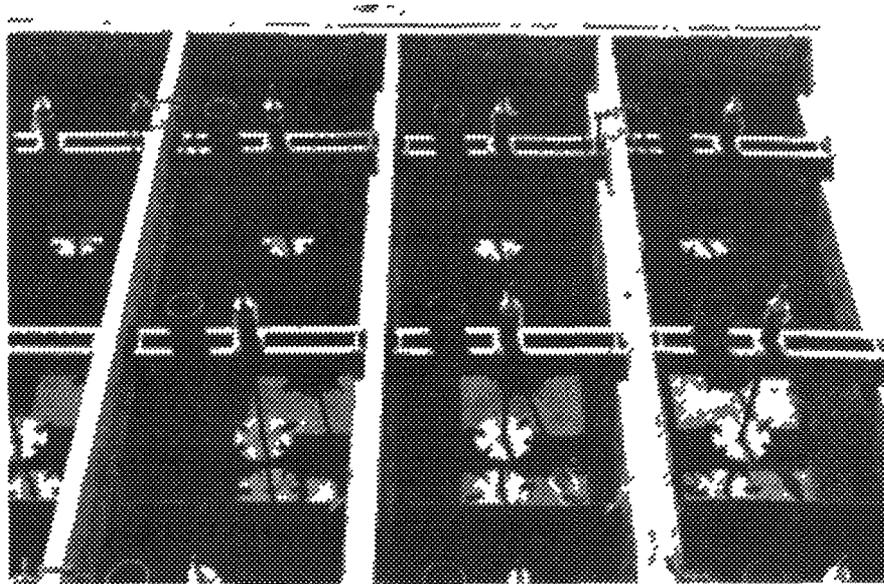


FIG 3 Concrete lined trenches for in situ solidification of sludge at the time of construction at Tarapur, India

TABLE II. CHARACTERISTICS OF THE SLUDGE SOLIDIFIED AT TARAPUR IN INDIA

Parameters	Characteristics
<i>Chemical nature:</i> Fine precipitate (g/L) of: Copper ferrocyanide Ferric nitrate Barium sulphate Sodium sulphate (dissolved)	5 5 30 3
pH	8–9
<i>Solids (g/L):</i> Total solids Suspended solids	40–50 30–40
Activity (Gross , MBq/mL)	37–370
Radioisotopes in sludge from reprocessing and vitrification plants:	^{134–137} Cs, ⁹⁰ Sr, ¹⁰⁶ Ru, traces of isotopes of U and Pu
Radioisotopes in sludge from nuclear power plant	¹³⁷ Cs, ⁶⁰ Co

- 3) Movement of mobile cement equipment into place above the compartment;
- 4) Shielding the top of the vault to minimize exposure;
- 5) Transferring radioactive liquid into the vault;
- 6) Feeding cement and additives into the vault at a controlled rate;
- 7) Preparation of a homogeneous mix by thorough agitation;
- 8) Closing the nozzle openings and providing shielding on top of the openings;
- 9) Waterproofing the top surface to avoid ingress of water;
- 10) Product quality control;
- 11) Surveillance and monitoring via a borehole array installed around the trenches.

The waste is pumped into each compartment from a nearby waste treatment facility by underground pipelines having secondary containment. After thorough homogenization of the sludge with necessary admixtures such as vermiculite, cement is added while continuous mixing of the sludge takes place using the installed agitator. The rate of cement flow is controlled by a rotating valve. The top of the cemented waste inside the compartment is capped with a cement grout after the waste matrix has hardened for several days. After the process is complete, the

detachable apparatus such as the cement hopper, mixer motor and ventilation equipment is moved to a new compartment for the next operation. The nozzles are then welded with required additional shielding and the top of the trench is sealed with a concrete mix, followed by waterproofing treatment.

This process eliminates the need for packaging, transportation and further handling of the radioactive waste. No contact maintenance for active equipment is required, since the agitators are disposable. The exposure to the operator as well as to maintenance staff is negligible as all radioactive items are separated from the working area by the top shielding. This disposal method does not generate secondary wastes as there is no wash down or decontamination of radioactive equipment. Table III gives the properties of the cement matrix formed in situ in the trenches.

TABLE III. DATA FOR THE CEMENT FORMULATION USED FOR IN SITU SOLIDIFICATION OF SLUDGE AT TARAPUR IN INDIA

Characteristics	Value
Waste cement ratio:	1 part of waste by weight/ 1.5–2 part of cement by weight
Vermiculite	10% by weight of waste
Sodium Silicates	0.5 mL per 100 kg of cement
Type of cement	Portland cement
Leaching rate without vermiculite	10^{-2} – 10^{-3} g/cm ² d
Leaching rate with 10% (wt) vermiculite	10^{-3} – 10^{-4} g/cm ² d
Porosity	25–40% by volume
Density	1.3–1.5 g/cm ³
Compressive strength	30–100 kg/cm ²
Volume increase on solidification	50%

2.1.2.2. In situ solidification by in-line mixing

An alternative in situ solidification technique used at Tarapur involves in-line mixing of waste and cement before pumping the mixture into underground concrete vaults [2]. A shielded mobile plant with a facility for withdrawing radioactive concentrates from storage tanks at the disposal site, carrying out, in-line mixing with cement and additives and pumping the radioactive grout to the concrete vaults exists. A schematic diagram is shown in Fig. 4 and a pictorial view is presented in Fig. 5. This method was found to be advantageous for the areas where storage and disposal facilities are located adjacent to each other but the operator dose is reported to be higher compared to the technique detailed in Section 2.1.2.1 due to the requirements of decontamination and contact maintenance of radioactive equipment. The main process steps for this disposal method are as follows:

- 1) Transferring the liquid waste into the shielded mobile mixing plant,
- 2) Homogeneously mixing this waste with cement and additives,
- 3) Transferring the grout into the vault with proper level control to avoid overflow,
- 4) Closing the nozzle openings in the roof of the vault,
- 5) Removing the mobile mixing plant,
- 6) Waterproofing the top surface,
- 7) Product quality control,
- 8) Surveillance and monitoring

2.1.2.3 Bulk grouting process

In the Gobi Desert, China, raffinate concentrates and chemical decladding waste have been accumulated and stored in carbon steel tanks and are proposed for a bulk in situ grouting process into underground concrete vaults [3, 4]

A proposed engineering facility for this bulk grouting process will consist of the following components

- 1) Waste collection and transfer system,
- 2) Cement feed system,
- 3) Waste feed and mixing system,
- 4) Grout pumping system

The wastes will be pumped from the collection and transfer system to the feed system and combined with cement and a dehydrating additive in a mobile mixer located above the concrete vault. The mixed grout will then be cast by gravity into the underground vault. A schematic diagram of the process is shown in Fig. 6. Casting a vault is likely to take about 26 hours. Several days later, a capping layer of clean cement will be put on the solidified waste surface. The vault is built using reinforced concrete 200 mm thick with a structural cover. The structural cover is 5 m thick and includes seven layers, i.e. stone block jointed with cement, backfilling soil, clay, sand, gravel, pebble and clay. A clay layer surrounds the vault which will retard radionuclide migration. A diagram showing the vault and structural cover is shown in Fig. 7. The chemical and radiological composition of these wastes and properties of the solidified grout product based on laboratory studies are given in Tables IV and V.

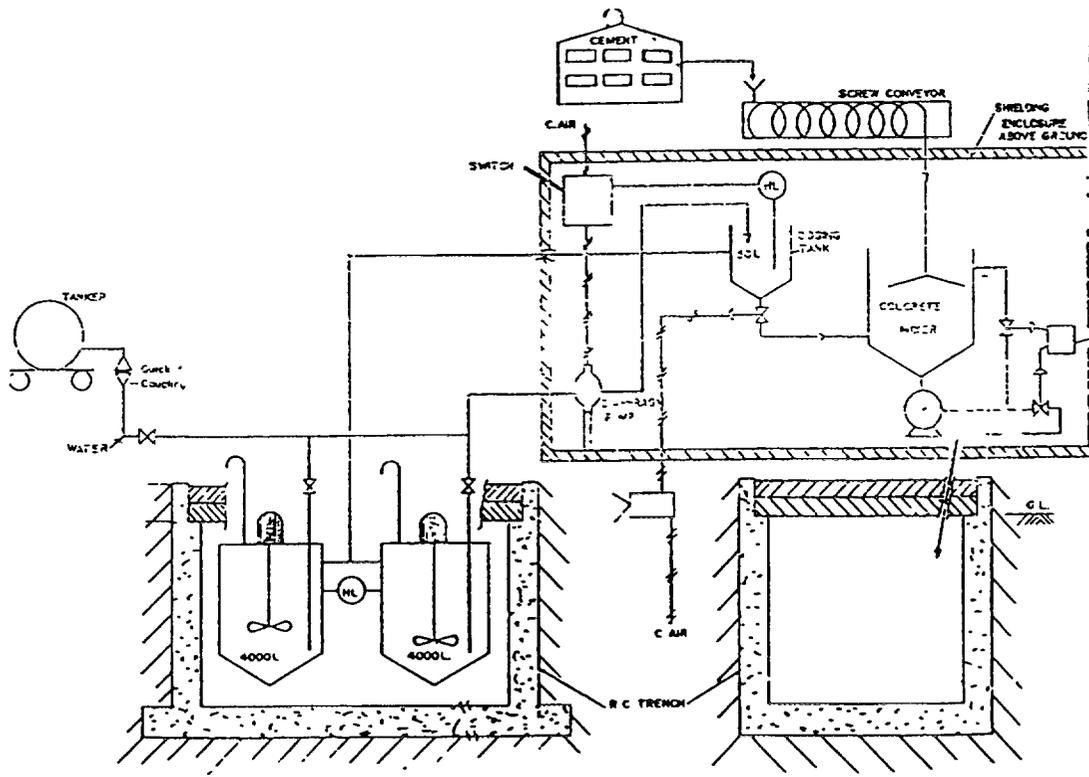


FIG 4 Layout of equipment for in-line mixing and solidification of sludge in concrete vaults at Tarapur, India.

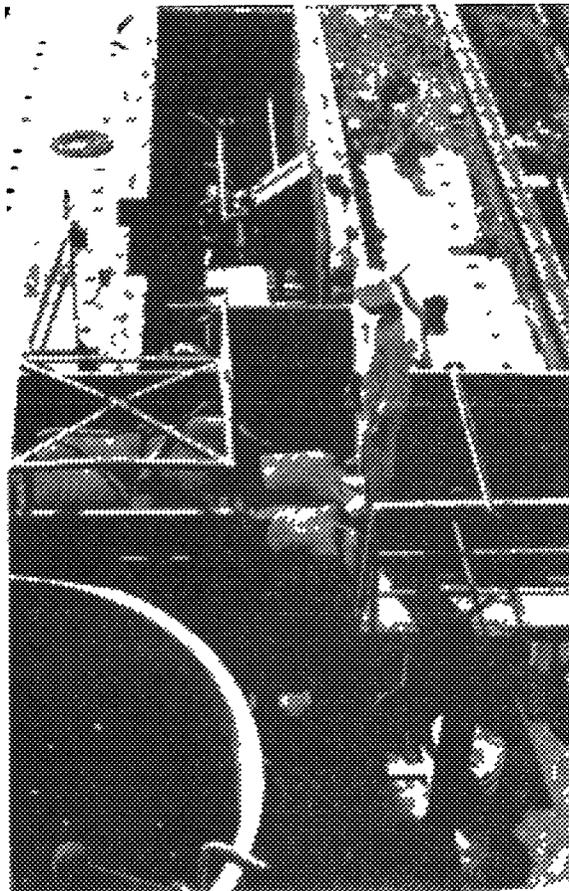


FIG 5 In situ solidification of sludge in concrete vaults at Tarapur, India.

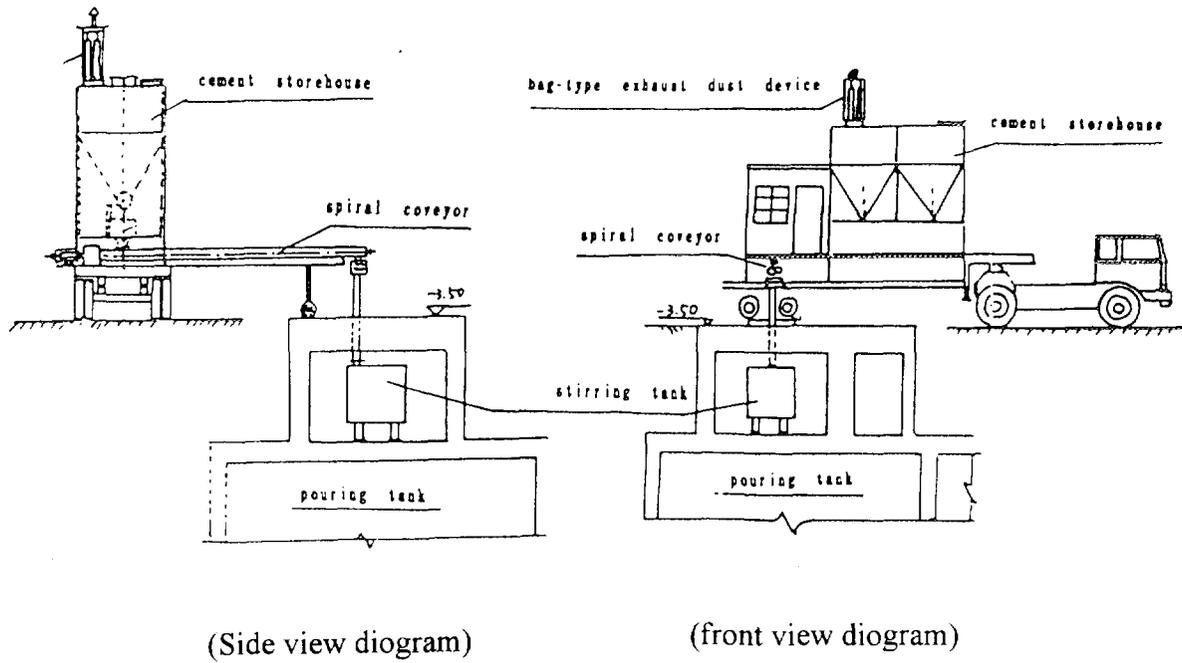


FIG. 6. Schematic diagram of in situ solidification process of liquid waste in the Gobi Desert in China.

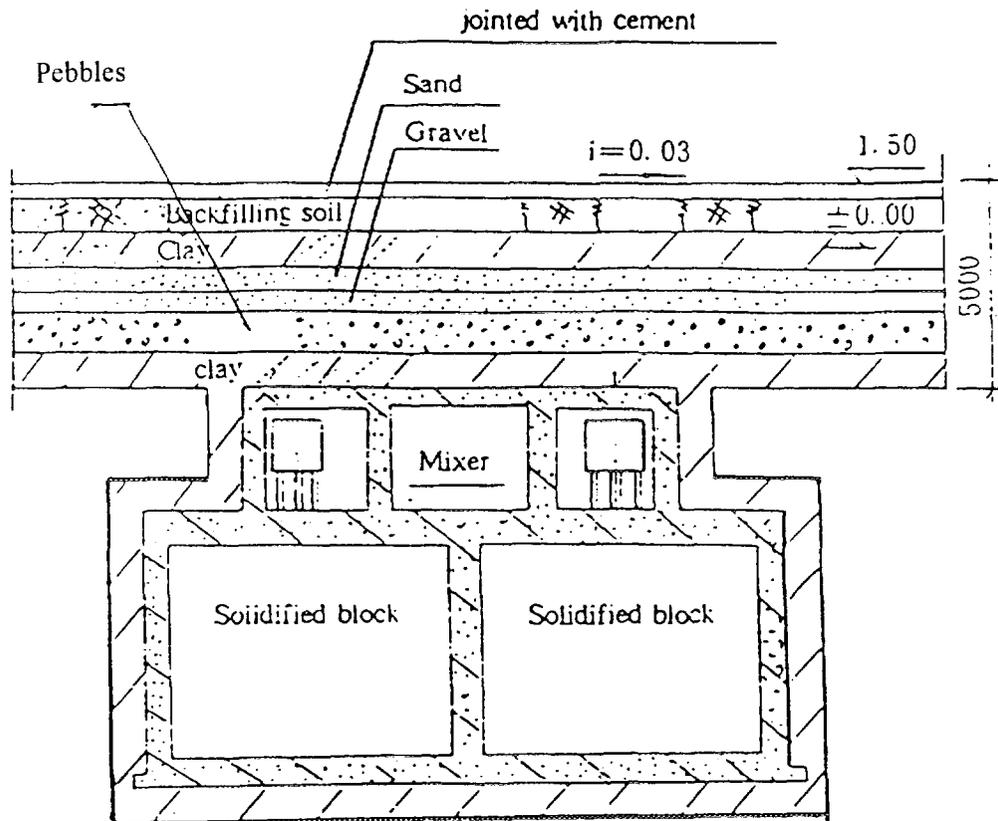


FIG. 7. Site closure arrangement for in situ solidified liquid waste in the Gobi Desert in China.

TABLE IV. COMPOSITION OF WASTES FOR IN SITU IMMOBILIZATION IN THE GOBI DESERT, CHINA

Components	Chemical decladding wastes	Concentrates
NaNO ₃ (g/L)	280	330
Na ₂ CO ₃ (g/L)	40	50
NaOH (g/L)	80	20
NaAlO ₂ (g/L)	100	–
⁹⁰ Sr (GBq/L)	0.056	0.16
¹³⁷ Cs (GBq/L)	1.32	4.25
¹⁰⁶ Ru- ¹⁰⁶ Rh (GBq/L)	0.28	0.58
(Bq/L)	26	67.5
Slurry (MBq/L)	-	51.8

TABLE V. PROPERTIES OF GROUT PROPOSED FOR IN SITU IMMOBILIZATION BY BULK GROUTING AT GOBI DESERT, CHINA

Components	Characteristics
Waste/cement ratio	0.5
Salt/cement ratio	0.23–0.30
Fluidity	≥0.17 m
Initial setting time	>2.5 h
End setting time	<48 h
Compressive strength	>10 MPa
Volume self-expansion coefficient (90th day)	7.310 ⁻⁴
Leach rate (42nd day), g/cm ² d:	
¹³⁴ Cs	1.710 ⁻³ ÷ 3.710 ⁻³
⁸⁵ Sr	510 ⁻⁴ ÷ 110 ⁻³

2.1.2.4. In situ immobilization in a bitumen matrix

At the Leningrad Nuclear Power Plant at Sosnovy Bor near St. Petersburg a continuous process for the bituminization of evaporated concentrates and spent ion exchange resins has been developed. The molten bitumen/waste mixture is transported via a heated pipeline to near surface concrete tanks at the repository operated by the specialized enterprise 'Lensezcombinat RADON', which is located at the Leningrad NPP site [5, 6]. The characteristics of the evaporation waste concentrates is given in Table VI.

TABLE VI. COMPOSITION OF WASTE SOLIDIFIED IN BITUMEN AT LENINGRAD NPP IN THE RUSSIAN FEDERATION

Components	Characteristics
Salts concentration (g/L)	200–300
pH	11–12
Major ions and their concentration (w%):	
Na ⁺	80
Cl ⁻	33
SO ₄ ²⁻	8
NO ₃ ⁻	19
Main radionuclides	¹³⁷ Cs, ¹³⁴ Cs, ⁶⁰ Co, ⁵⁸ Co
Organic substances (g/L)	0.015–0.03
Detergents (g/L)	0.5–0.6
-activity (Bq/L)	10 ⁵ –10 ⁶
-activity (Bq/L)	10 ⁶ –10 ⁷

The main process steps followed at the Leningrad NPP and ‘Lensezcombinat RADON’ are as follows:

- 1) Collection of radioactive liquid waste from stainless steel storage tanks;
- 2) Evaporation of waste for salt concentration and addition of molten bitumen in a thin film evaporator bituminizer;
- 3) Transportation of the molten bitumen/waste mixture via a heated pipeline to near surface concrete tanks;
- 4) Solidification of the molten bitumen mixture in near surface concrete tanks.

The bituminization process includes further concentration of evaporator concentrate waste by evaporating water from the waste and mixing the waterless salts with bitumen at a temperature of 135–145°C in a thin film bituminizer. The schematic diagram of a facility for bituminization of evaporation concentrates at the Leningrad NPP is shown in Fig. 8. The bituminization process data are given in Table VII. The facility also includes a system for heating the equipment and pipelines, for collecting the heating steam condensate and for the control and automation of the process. The repository consists of twelve concrete compartments and is designed for disposal of 27 000 m³ of bituminized waste. The repository is connected via heated pipelines having lengths of 50–150 m depending on the location of the compartment being filled. The temperature of the bitumen mixture being transported is maintained at 120°C. When the mixture cools in the tank a bitumen compound is formed with a uniform distribution of waste particles and good leaching characteristics. The water resistance of bitumen compounds is characterized by a leaching rate of 10⁻⁴–10⁻⁵ g/cm² day for ¹³⁷Cs and ⁹⁰Sr radionuclides.

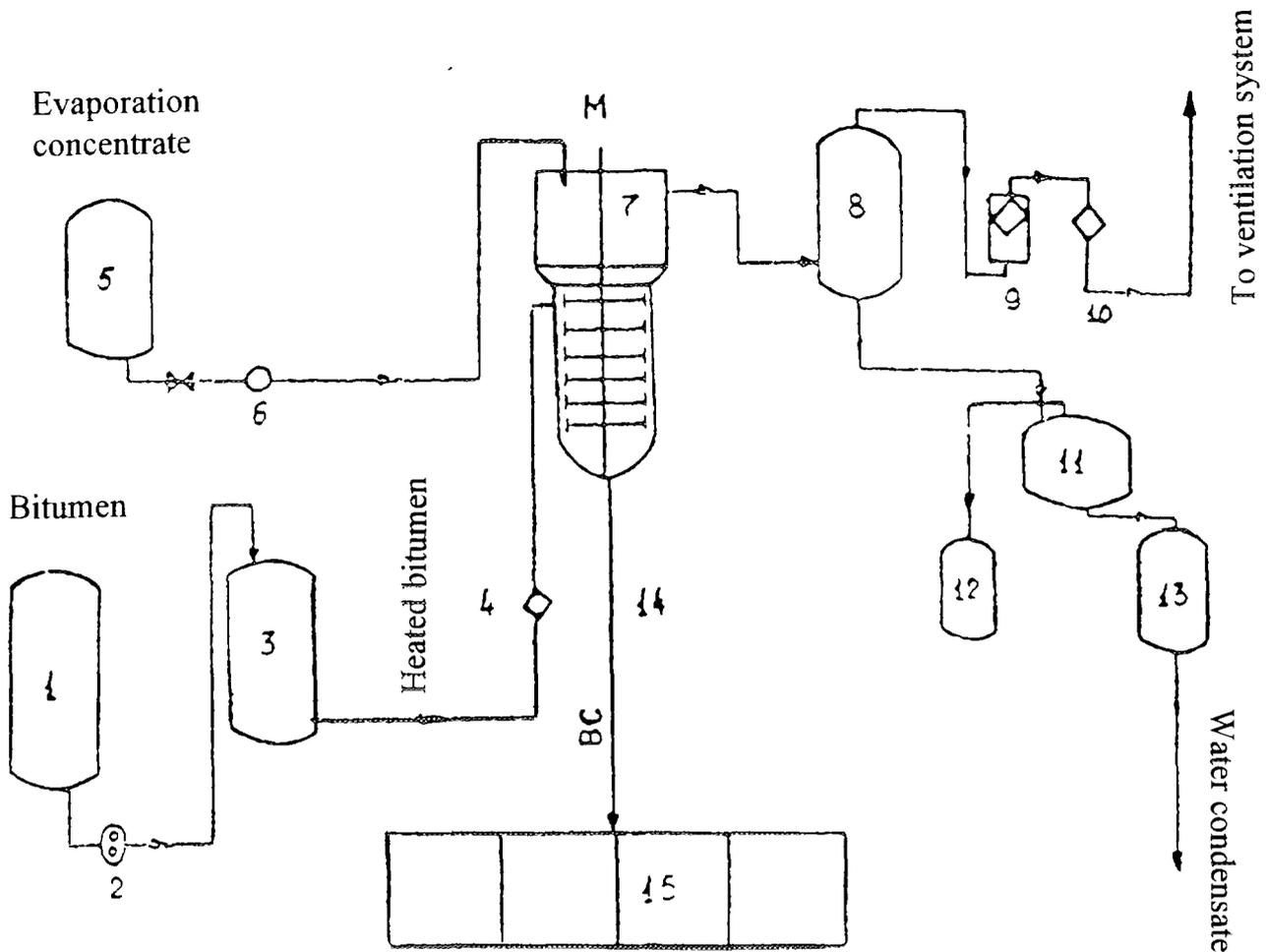
A potential problem with this disposal method is the possibilities of ignition and microbiological destruction of bitumen compounds. However, investigations conducted have shown that even with 45% of evaporator concentrates incorporated into bitumen the possibility of ignition is excluded. Microbiological destruction of the final bitumen compound has not been observed at the storage facility up to now.

TABLE VII. DATA FOR THE BITUMINIZATION PROCESS USED AT LENINGRAD NPP IN THE RUSSIAN FEDERATION

Parameters	Characteristics
Type of bituminizer	RB-1000-14 thin film rotor evaporator
Operation mode	continuous process
Rotation velocity, min ⁻¹	49
Heating media	Steam: 0.52–0.60 MPa
Heating surface	10 m ²
Capacity, L/h of waste	400–500
Type of bitumen used	BND 90/130; BND 60/90; BND 40/80
Operating temperature, °C	150–160
Temperature in the pipeline, °C	110–120
Waste loading factor for the final compound, %	40 ± 5
Water content in the final product, %	< 5

TABLE VIII. TYPES OF REPOSITORIES AND MATRICES USED BY DIFFERENT COUNTRIES FOR IN SITU IMMOBILIZATION IN GEOLOGICAL REPOSITORIES

Type of facility	Matrix	Country	Location
Shale formation at a depth of 200–300 m from ground	Cement	USA	ORNL
Shale formation at a depth of >200 m	Cement	China	Southwest China
Salt caverns at a depth of 900–1000 m	Cemented granules in mixture with cement slurry	Germany	Asse salt mine



- | | |
|---------------------------------------|---|
| 1 – bitumen storage tank, | 8 – condenser, |
| 2 – bitumen feeding pump, | 9,10 – air filters, |
| 3 – bitumen feeding tank, | 11 – air and grease separator, |
| 4 – filter for bitumen, | 12 – grees and oil collection tank, |
| 5 – feeding tank for rad waste, | 13 – tank for condensate, |
| 6 – feeding pump, | 14 – heated pipeline, |
| 7 – thin film evaporator–bituminator, | 15 – concrete compartments for bituminized waste collection |

FIG 8 Schematic diagram of an in situ immobilization process using bitumen at Leningrad Nuclear Power Plant in the Russian Federation

2.2. IN SITU IMMOBILIZATION IN GEOLOGICAL REPOSITORIES

2.2.1. General description of technologies

This technology is particularly applicable where it is possible to have geological repositories near the source of waste generation or near the storage sites for radioactive waste. A geological repository in the form of a nearly impermeable shale formation at a depth of 200 to 300 m at Oak Ridge National Laboratory (ORNL) in the USA was used for immobilization of intermediate level waste by introducing a technology suitable for hydraulically fracturing (hydrofracture) the shale. Similar efforts are being made in Southwest China for adopting a hydraulic fracturing for disposal of intermediate level liquid waste. In Germany, in deep underground salt caverns at a depth of 900–1000 m, in situ immobilization of preconditioned waste granules has been practiced. The technologies adopted by the above countries are described in the following section. The types of repositories and immobilant used by different countries are summarized in Table VIII.

2.2.2. Operating experience

2.2.2.1. *In situ immobilization in hydraulically fractured shale adopted at the ORNL*

The hydrofracture process was developed at the ORNL to dispose of intermediate level waste (ILW) solutions with low concentrations of long lived radionuclides, by fixation of the radionuclides in a stable geological formation well below the level of circulating groundwater [7–9]. This technology has been used at the ORNL between 1959 and 1979 and has been described in detail in a previous IAEA publication [9].

The process adopted at the ORNL involves underground injection of ILW in the form of a slurry containing binding agents (grouts) into a nearly impermeable shale formation at a depth of about 200 to 300 m. Prior to injection of the waste slurry, an initial fracture is formed in the nearly horizontal shale bedding planes by the injection of water under high pressure. The injected grout slurry forms a thin, approximately horizontal, grout sheet parallel to the bedding of the shale and several hundred metres wide. The grout sets a few hours after completion of the injection, thus permanently fixing the radioactive waste in the shale formation.

A sketch of the ORNL disposal facility and flow diagram is shown in Figs 9 and 10, respectively. The process is operated as a large scale batch process; each injection is, however, a continuous operation. Each injection disposes of an annual accumulation of waste solution of about 380 000 L. During an injection, waste solution is pumped and mixed with a stream of dry solids. The resulting grout is pumped into the shale formation at an injection pressure of about 20 MPa through the injection well.

The normal grout injection rate is about 1000 L/min; an injection requires about 8 to 10 h to complete. At the end of an injection, the well is flushed with water so that the slot in the injection well will be free of grout and can be reused for the next injection. A valve shuts the well as soon as the grout sets. Several injections are made through the same slot and form grout sheets that are generally parallel to the first. After approximately four injections have been made through the slot, the bottom of the well is plugged, a new slot is cut into the casing of the well 3 m above the old slot, and a new series of injections is made at the higher elevation. In this manner, maximum utilization of the disposal space is achieved.

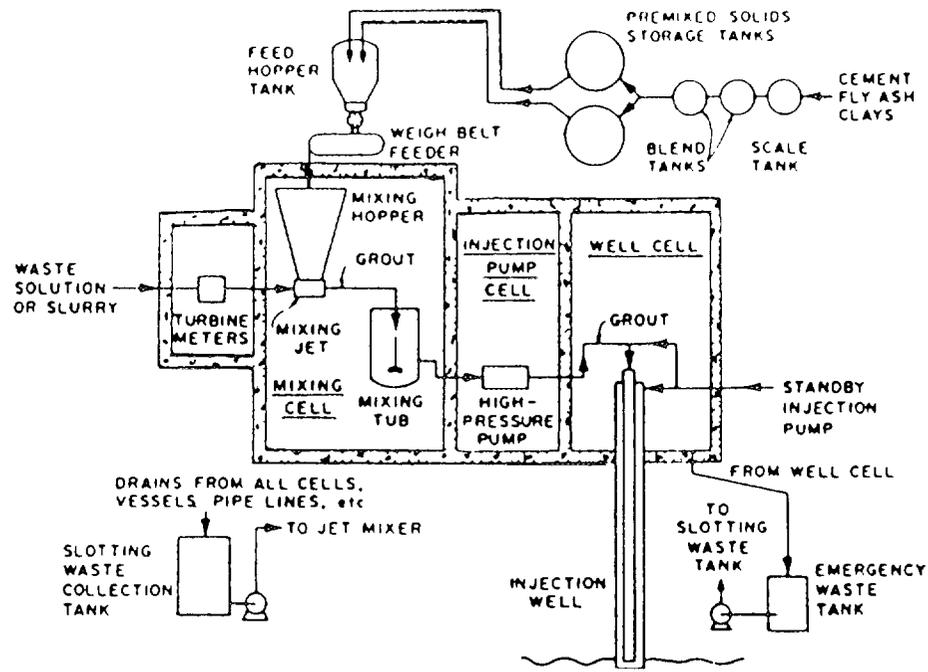


FIG 9. Flow diagram showing the hydrofracture facility at ORNL, USA.

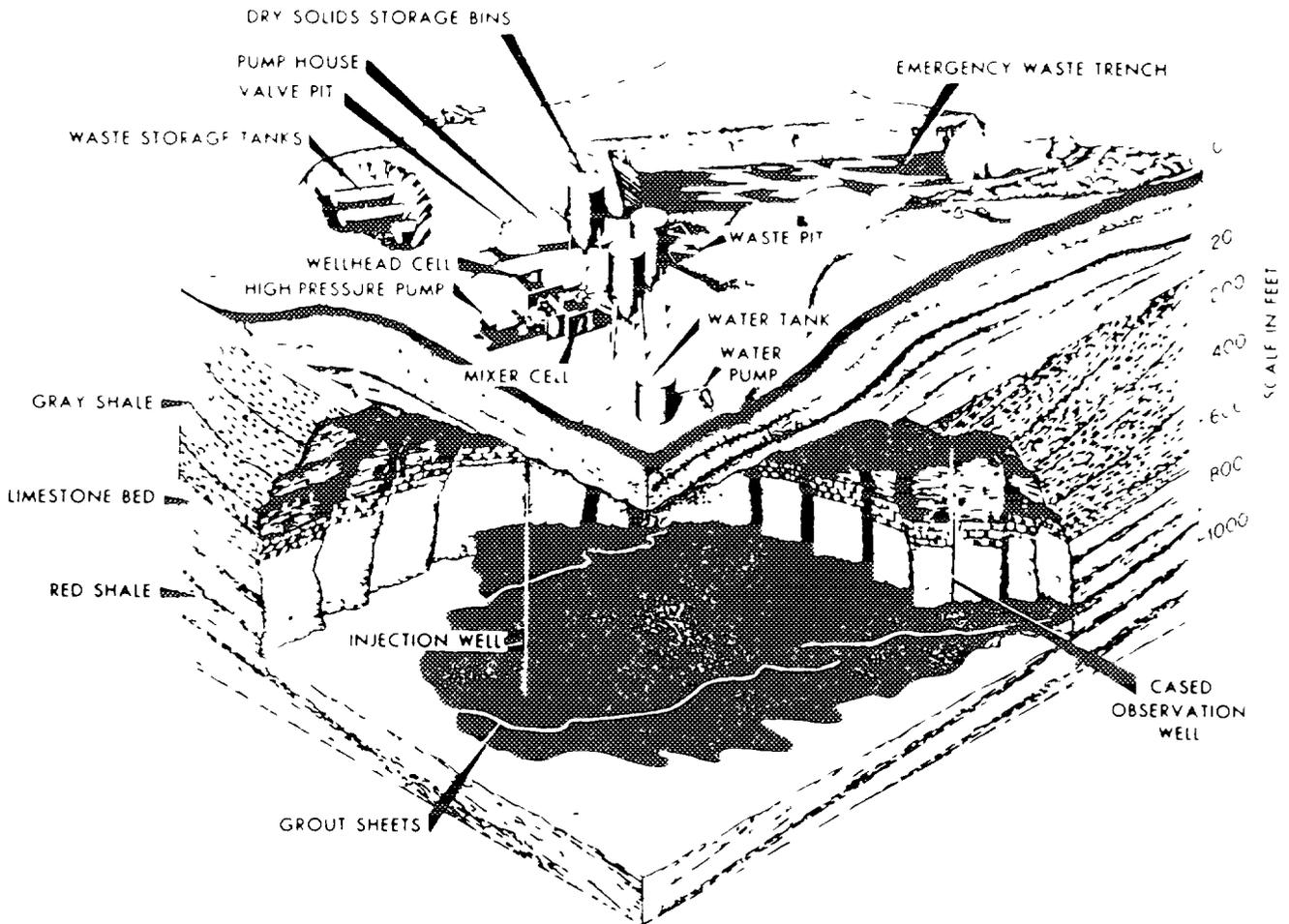


FIG 10 Diagram showing the geological in situ disposal facility at ORNL, USA [9].

The hydrofracture process has been developed to dispose of intermediate level waste solutions generated at the ORNL, but an extrapolation of the ORNL experience suggests that other waste forms could be disposed of by this technique. The probable limitations are:

- 1) Particle sizes in a slurry should be less than 1 mm;
- 2) pH should be neutral or alkaline;
- 3) Chemical compatibility with both the cement in the solids mix and the disposal formation should be considered;
- 4) A waste specific activity should be low enough to be handled in the surface facility and the heat generated underground will dissipate at a temperature that will not cause formation damage. (Material disposed of at ORNL has a specific activity of approximately nine TBq/m³ mostly composed of ¹³⁷Cs).

The essential feature of the shale fracturing process is the fixation of the radionuclides in a geological formation that is known to be isolated from contact with the surface environment. At the ORNL site, the shale formation used for hydrofracture disposal is at 200 to 300 m below ground level which is well below the level of circulating groundwater movement. The permeability of the shale is low, with a calculated rate of water movement less than one cm per 100 years (permeability = $3.2 \cdot 10^{-12}$ m/s).

Additional features of the hydrofracture process provide continued isolation of the radionuclides even if the low permeability of the disposal formation were not considered. For example, the leach rates of significant radionuclides from the set grout are quite low. In addition, any radionuclides that might be leached from a grout sheet would be retained in the disposal zone by the high ion exchange capacity of the shale.

At the ORNL, the principal constituents of the waste were relatively short lived radionuclides (¹³⁷Cs and ⁹⁰Sr) with a low concentration of long lived radionuclides. Hence the required isolation time in the geosphere is only a few hundred years. It is expected, however, that because of the geological stability of shale formations that isolation could extend to time spans measured in millenniums. Environmental impact analysis of the process by the ORNL contained the following conclusions:

- normal operations would have a very low safety impact,
- accident situations were improbable,
- plausible accident scenarios would result in little or no ultimate release of radionuclides.

Calculations of thermal effects resulting from the decay of injected radionuclides predicted a maximum temperature at the centre of the injection zone of about 58°C after 50 years. Mineralogical analysis of the shale formation suggested that a temperature of about 100°C would be tolerable without deterioration of the shale. Since 1966, this facility has been used for 18 operational injections. More than 8 million litres of waste grout containing over $2.2 \cdot 10^{10}$ MBq of radionuclides have been injected. Although operational problems have been experienced, most have been comparatively minor and none have been severe. The general experience has been quite good.

2.2.2.2. *In situ immobilization by a hydrofracture process being adopted in China*

China has been developing hydrofracture technology since the 1980s and has carried out siting, laboratory studies and demonstration tests [3,4,10]. The facility is currently under construction. The full scale engineering operation will be carried out in the near future for liquid intermediate level waste with the following characteristics:

Nature	Alkaline, major component: NaAlO ₂
U	62 mg/L
Pu	26 MBq/L
Salts	183 g/L
Slurry	~3%

For the survey of candidate sites, twelve wells were drilled in Southwest China. Following an extensive geological survey, it was found there was a favourable shale stratum which could be used for hydraulic fracturing disposal of low and intermediate level radioactive waste. The shale stratum is a closed structure with a low water content, extremely low permeability and a high clay content. The shale properties are as follows:

Density	2.8 t/m ³
Effective porosity	0.6–0.9%
Permeability	0.01–0.76 milli-darcy
Clay mineral	70–95% (mainly glimmerton)
Ion exchange capacity	8–20 mg-equivalent /100 g

In order to meet the injection requirements, it is proposed that the waste is incorporated into a grout having the following properties:

Viscosity:	$<4 \times 10^{-2}$ Pa.s, to enable pumping;
After solidification, free water:	< 5 wt%;
Initial setting time:	24–48 h, corresponding
to pumping time; end setting time:	7 days;
Good immobilization of nuclides, leach rate of Sr and Cs (at 102 nd day):	10^{-5} g/cm ² d;
Compressive strength of solidified product:	700 Pa (7 kg/cm ²).

For demonstrating the feasibility and safety of this disposal concept, 300 m³ water was injected into the shale stratum at a depth of 450 m in 1985. After the water injection, 291m³ of simulated intermediate level waste grout was also injected. The water injection was traced by ¹⁹⁸Au (T_{1/2} = 2.696 days, 0,78 TBq); the grout injection was traced using ¹³⁴Cs (T_{1/2} = 2.062 years, 0,36 TBq). The following results were obtained:

Breakdown pressure:	26 MPa, corresponding injection rate: 0.13 m ³ /min;
Prolongation pressure:	20 MPa, corresponding injection rate: 1–1.13 m ³ /min;
Maximum angle of grout sheet:	about 25°;
Maximum distance of grout sheet:	116 m from the injection well.

It was found out that the grout actually expanded horizontally for a distance of 116 m, occupying an area of 14 000 m³. The grout sheet was 2.3 cm in thickness while only 1–2 mm was observed in surface uplift. A schematic diagram of the hydraulic fracturing process is shown in Fig. 11. Such matrix components as cement, fly ash, activated clay and zeolite are weighted and transferred into the high level blended material tank. The waste with the retarder is pumped into the mixer bottom. The matrix components are mixed with liquid waste to form a grout which is then pressurized by the injection pump and flows through the coiled tubing into the injection well. The radioactive grout reaches the shale stratum and is solidified under pressure. The injection was carried out at a pump pressure of 250–300 kg/cm² and an injection rate of 1 m³/min.

The stratum cleaving is carried out using a 360⁰ rotary cleaver by spraying sand. The specifications of the cleaving operation are as follows:

Rotary speed	1–3 r/min
Spray sand size	40–60 mesh
Sand ratio	10–13%
Sand amount	2.3–2.5 t
Spray sand time	30–40 min

To prevent the grout returning to the ground surface from an injection accident, an emergency reception pool of 200 m³ is constructed. Gamma monitoring wells and observation wells are established around the injection well for monitoring the orientation and distribution of the waste grout and for inspection of the earth's surface uplift. A schematic of these facilities is shown in Fig. 12.

2.2.2.3. *In situ immobilization in salt caverns*

The main feature of this concept is in situ immobilization of preconditioned waste granules in deep underground salt caverns at a depth of 900–1000 m [11–14]. Investigation of this technology has been carried out in Germany between 1976 and 1989. The objective was to develop an alternative approach to the conventional method of disposing of drummed low and intermediate level waste in an engineered deep repository. The concept has been proven in Germany only on inactive simulated waste forms. A schematic representation of this concept is shown in Fig. 13.

The main process steps followed in this method are as follows:

- 1) Prefabrication of cementitious waste granules according to the composition given in Table IX. These are kept in an interim storage facility to enable dissipation of hydration heat. This is important to ensure that the operating temperature in the salt cavern is kept low.
- 2) Transportation of granules to the disposal site and mixing of the granules with further cement and water to form a slurry.
- 3) Vertical gravity transportation through a pipeline into the salt cavern.
- 4) In situ immobilization of the cementitious waste form, probably in layers representing separate filling campaigns.
- 5) Plugging of the filler borehole.

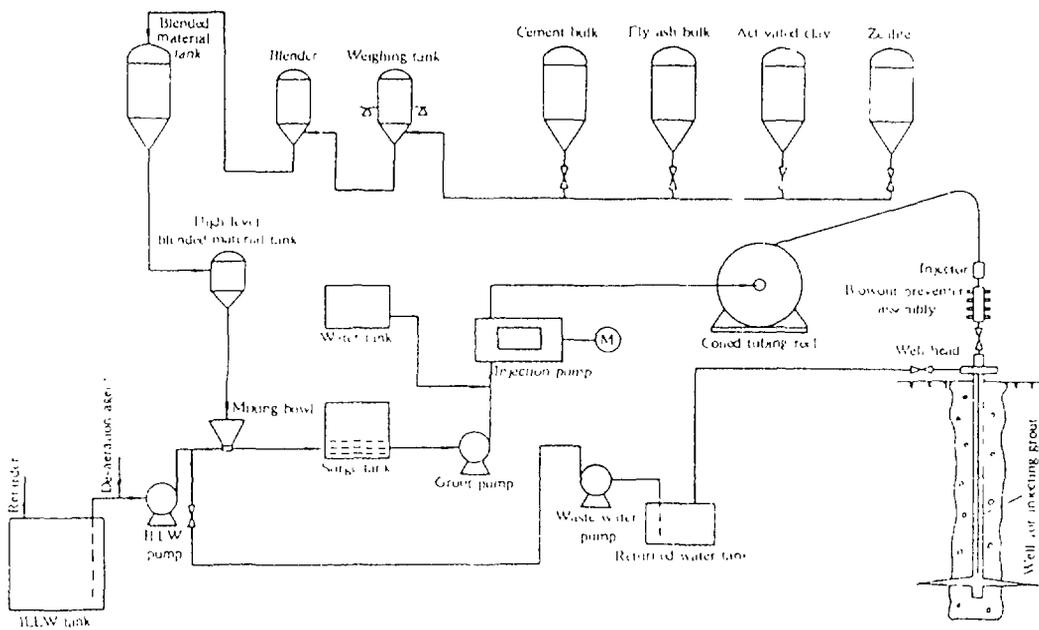


FIG. 11. Diagram of the process for in situ immobilization of intermediate level liquid waste by hydrofracture in shale in Southwest China.

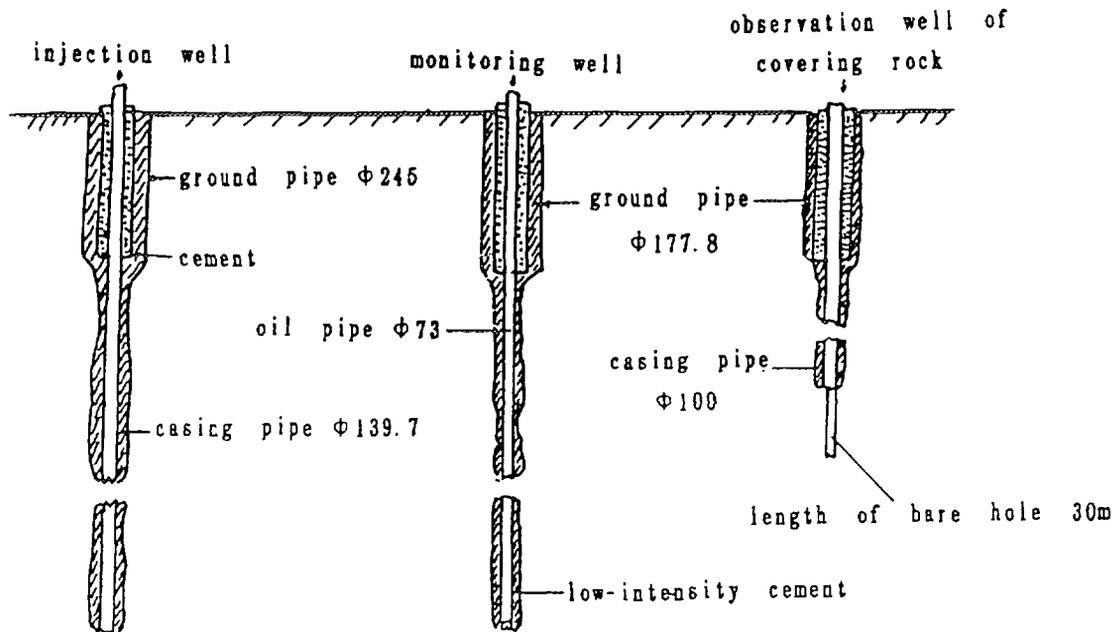


FIG. 12. Schematic diagram showing the injection and monitoring wells for in situ immobilization by hydrofracture in shale in Southwest China.

TABLE IX. COMPOSITION OF WASTE GRANULES AND FILLER SOLUTION USED FOR IN SITU IMMOBILIZATION IN THE ASSE SALT MINE IN GERMANY

Properties	Granules	Filler Solution
Volume ratio in final waste form (%)	60	40
Cement – Portland 35F (% wt)	70	24
Bentonite (% wt)	5	NIL
Waste concentrate	19	NIL*
Water/cement ratio	0.15	0.5
Grain size distribution (mm)	0.3–0.5	
Density, g/mL	2.6	1.6
Porosity, %	18	
Strength, MPa	10	
Salt/cement ratio (%)	6–9	

*Unless H_2O (tritiated water) is used to prepare the filler solution.

A ventilation system is required to treat the displaced atmosphere from the cavern during filling. The cavern is not man operated, hence a higher airborne contamination level can be tolerated during operations. Candidate waste materials for this disposal concept are decontamination solutions with high salt content, ion-exchange resins and ashes from waste combustion

When the preconditioned waste granules have been prepared, setting takes place over a period of 7–10 days before transportation to the disposal site in a shielded container. On arrival at the disposal site, the granules are discharged into a vessel above the salt cavern and mixed with further water and Portland cement, forming a grout slurry with the following reference specification:

Granules:	45–47 wt%
Cement:	53–55 wt%
Water/cement ratio:	0.57
Relative density:	2.0

The slurry is discharged by gravity into the salt cavern, avoiding the use of pumping equipment. After the grout has solidified in situ, subsequent slurries can be placed on top of the existing grout, producing a layered cementitious product which will eventually fill the entire cavern. When the cavern is filled the access borehole will be plugged with suitable material such as bentonite. Three large scale prototype salt caverns (about $10\,000\text{ m}^3$) at the Asse salt mine in Germany were filled or partially filled with the granular cementitious waste. Various measurements and intact samples were taken for analysis, as well as modelling studies carried out of heat transport in the caverns. Initial experiments indicated that the product has good leaching properties; the mean leach rates for ^{137}Cs and ^{85}Sr in saturated NaCl solution were found to be $4.810^{-5}\text{ g/cm}^2\cdot\text{d}$ and $1.110^{-4}\text{ g/cm}^2\cdot\text{d}$, respectively.

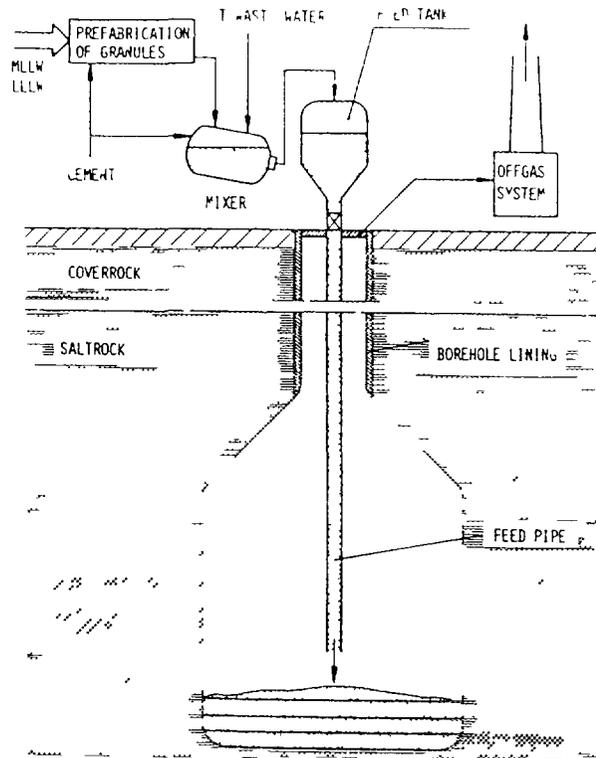


FIG 13 Process diagrams for an in situ immobilization process in a salt mine

3. IN SITU IMMOBILIZATION APPLIED TO SITES REQUIRING REMEDIATION

3.1 IN SITU IMMOBILIZATION OF EXISTING WASTES IN TANKS AND TRENCHES

At some nuclear facilities wastes may be stored awaiting final disposal. In other cases disposal may already have taken place but the disposal facility may not be performing adequately and so remedial action is required. In both these cases if it is not feasible or desirable to retrieve the waste for treatment/conditioning, for either economic or radiological reasons, then in situ immobilization technologies may be an option.

This section describes in situ immobilization technologies for existing wastes in both tanks and trenches. To reiterate, the important feature of the technologies is that the waste is not retrieved for immobilization but is left and immobilized in situ. In this Section a number of examples from different countries are described, including India, USA and Russian Federation. Different immobilants are used by the different countries. These are summarized in the Table X together with the type of facility in which immobilization has taken place (tank or engineered repository).

The following section describes in more detail each of the technologies in the above table and also experience of their usage.

3.1.1. Operating experiences

3.1.1.1. In situ immobilization of sludge in a tank at the Krasnoyarsk site, Russian Federation

This technology is based on the immobilization of a radioactive ferrocyanide sludge directly in a concrete tank by a process involving self mixing of the phosphoric acid and magnesium oxide to form a magnesium phosphate cement [15,16]. Fig. 14 illustrates the process. Table XI shows the chemical composition of the ferrocyanide sludge. The radioactive waste at Krasnoyarsk has arisen from the reprocessing of irradiated natural uranium at the Krasnoyarsk Mining and Chemical Plant. The tank has now completed its service life and is full, but because of evidence of defects in situ solidification of its contents has taken place.

TABLE X. LOCATION OF FACILITIES AND TYPES OF MATRICES USED BY DIFFERENT COUNTRIES FOR IN SITU IMMOBILIZATION OF EXISTING WASTES IN TANKS AND TRENCHES

Type of facility	Matrix	Country	Location
Near surface concrete repository	Portland cement, 10% vermiculite, sodium silicates (0.5 mL per 100 kg of cement)	India	Trombay
Near surface concrete tank	Portland cement, 10% vermiculite	Russian Federation	Sergiev Posad
Near surface burial trenches	Polyacrylamide	USA	ORNL
Near surface concrete tank	Ferrocyanide waste slurry mixed with orthophosphoric acid plus magnesite	Russian Federation	Krasnoyarsk

TABLE XI. COMPOSITION OF FERROCYANIDE SLUDGE SOLIDIFIED IN SITU IN A CONCRETE TANK AT KRASNOYARSK, RUSSIAN FEDERATION

Chemical composition (g/L)							pH	Solid/Liquid	Calculated volume of pulp, (m ³)	Radioactivity	
NaNO ₃	Na-acetate	Na, K-ferrocyanide	Fe	Al	Cr	Mn				GBq/L	¹³⁷ Cs (%)
325	6.0	125.0	6.0	0.5	0.5	0.5	6-7	1:2	80	37	100

One of the important features of the technology developed at Krasnoyarsk is that it is self mixing process. This was imposed by the size of the tank (height 30 m, diameter 12 m) which made mechanical mixing of the contents difficult. Magnesium oxide (caustic magnesite) was finally chosen as the main binding ingredient because of its availability as a byproduct from a local refractory material production plant.

In the tank the weight ratio of ferrocyanide slurry/phosphoric acid/magnesium oxide was 1 : 0.3 : 0.5. About 26 t of orthophosphoric acid and 42 t of caustic magnesite were used. The sequence of adding the binding agents to the tank and the resultant of self mixing process were as follows:

- 1) Introduction of the phosphoric acid to the tank which, because it is denser than the slurry, sinks and disturbs the slurry. After one hour the amount of liquid phase at the bottom of the tank equates to a liquid/solid ratio of about 13.4.
- 2) The tank contents are then left to age. After 24 h the pH of the tank contents equilibrated and because of the reaction heat from the slurry and acid the contents circulate causing mixing. After 24 h the tank contents are mixed to the same degree as would have been achieved if mechanical mixing had been used.
- 3) The magnesite powder is then added. Because of density effects the magnesite again sinks downwards through the acidified slurry and is mixed due to heat circulation in the tank. After 60–80 days a magnesium phosphate cement is formed.

Table XII contains mechanical and physico-chemical analyses results from a sample of solidified material taken from the tank.

TABLE XII. MECHANICAL AND PHYSICO-CHEMICAL DATA FOR SOLIDIFIED FERROCYANIDE SLUDGE AT KRASNOYARSK, RUSSIAN FEDERATION

Density (g/cm ³)	Porosity (%)	Average value of compression strength (kg/cm ²)	¹³⁷ Cs leaching rate (g/cm ² •d)
1.6–1.8	23.1–32.3	61.0	1.810 ⁺

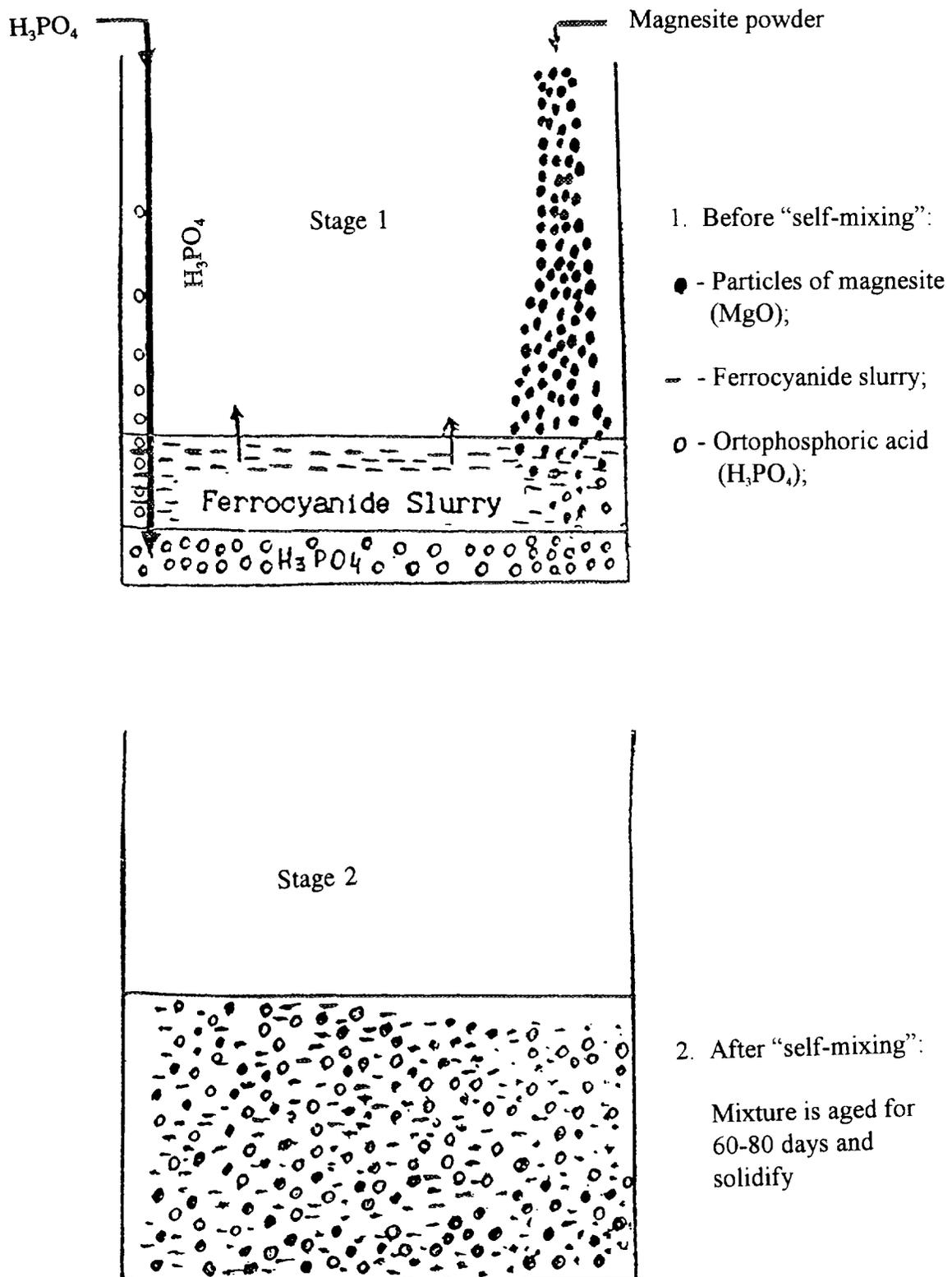


FIG. 14. Process for in situ solidification of sludge in a tank, involving self mixing of the tank contents plus additives to form a magnesium phosphate cement, (Krasnoyarsk, Russian Federation).

3.1.1.2. In situ immobilization of solid and liquid wastes in tanks and trenches at Sergiev Posad, Russian Federation

In situ grouting has been carried out at the facility operated by the enterprise RADON situated at Sergiev Posad near Moscow in Russian Federation [16]. At RADON, solid radioactive waste is received from various sites in the Moscow region and emplaced in shallow land trenches. The void space in the waste is filled with grout which itself is prepared using liquid low level waste. The resultant grouted liquid radioactive waste then immobilizes solid waste in situ. This method enables codisposal of solid and liquid radioactive wastes. The same method is utilized in Belarus at the specialized enterprise ECORES located near Minsk for immobilization of institutional waste [17].

In addition to disposal in trenches, in situ immobilization has also taken place in tanks at Sergiev Posad. The tanks are of a special design and are constructed partially below ground level but above the groundwater table. The tanks are constructed out of reinforced concrete and covered by reinforced concrete slabs. Internally the tanks are partitioned into compartments. The tanks are also lined with a special grout formulation incorporating sodium silicate. Solid radioactive waste was emplaced in the tanks but because of the irregular shape of the waste the voids was about 50%. To stabilize and immobilize the waste in situ grouting was adopted. Figure 15 shows the general layout of the equipment for injection of the grout incorporating the liquid low level waste into both the trenches and tanks with solid waste.

The resulting cement stone has physical properties which depend on the composition of the waste streams, types of inorganic binders used, and the grout/cement ratio.

In 1988 the grouting apparatus shown in Fig. 15 was replaced by a mobile unit built on the chassis of a truck, Fig. 16. This enabled the volumetric rate for production of the grout to be increased from about 5 m³/h to about 30 m³/h.

3.1.1.3. In situ grouting in trenches at the ORNL, USA

Two in situ grouting field demonstrations have been completed at the Oak Ridge National Laboratory (ORNL) [18–19]. Both demonstrations involved injecting polyacrylamide grout into uncompacted burial trenches and were successful in changing the trenches from being permeable (e.g. 10⁻² cm/s) to a condition of unmeasurable permeability (<710⁻⁶ cm/s). The long term stability of the polyacrylamide grout has been partly established by measurement of low rates of microbiological decay.

These demonstrations established (at the time of the work) that the cost of materials alone for in situ grouting with polyacrylamide was quite high, i.e. about \$50 000 per typical (4 m × 5 m x 5 m deep) burial trench, due to the large amount of voids per trench and the cost of the grout materials (about \$530/m³).

Field grout operations consisted of mixing the grout and catalyst solutions. The final formulation based on a 1 : 1 mixture of the two solutions would contain 10% of acrylamide grout, 0.3% triethanolamine, 0.01% potassium ferricyanide, and 0.5% ammonium persulfate. Figure 17 shows the equipment for mixing and injecting the polyacrylamide grout.

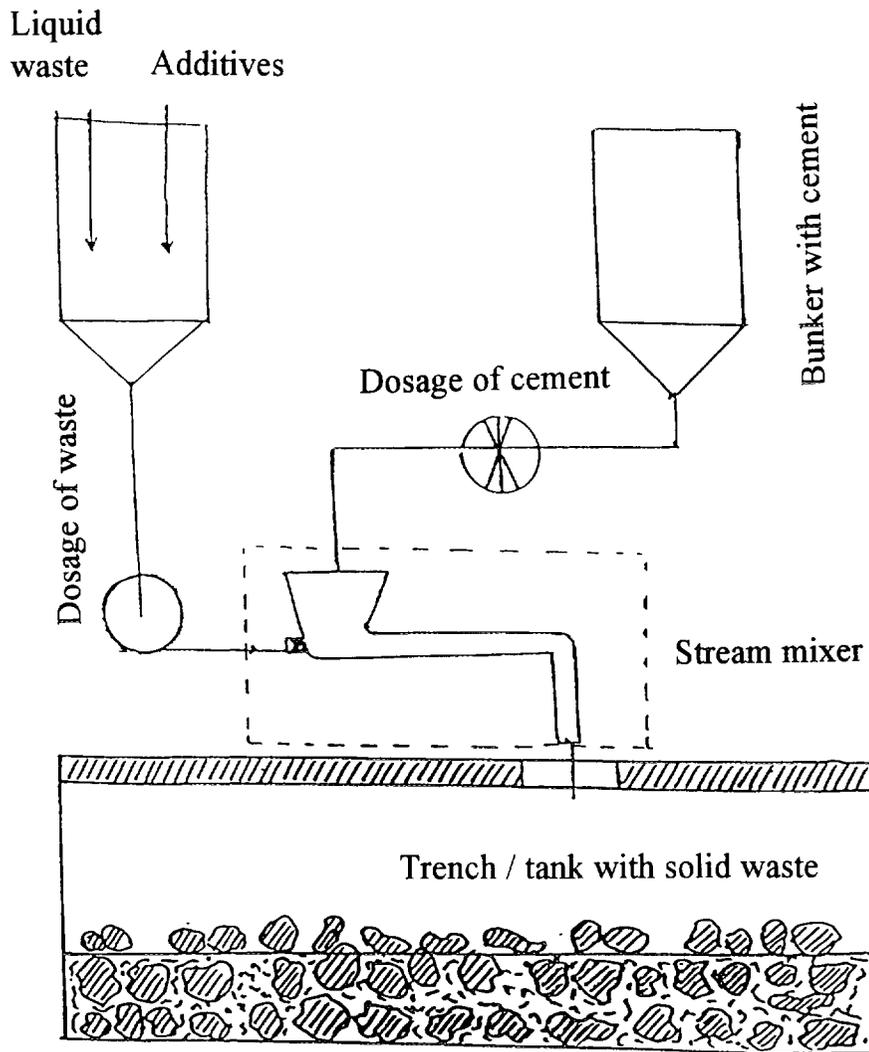
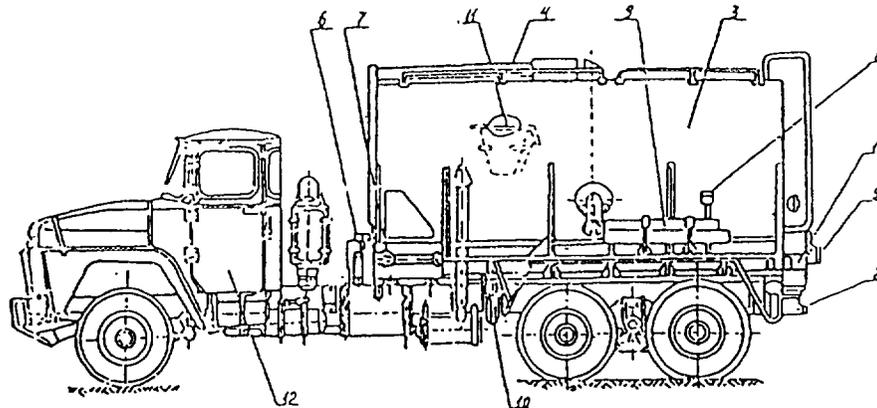


FIG. 15. Schematic layout of equipment for in situ immobilization of solid and liquid waste at Sergiev Posad, Russian Federation.



1 – receiving chamber; 2 – mixer; 3 – bunker; 4 – loading drive; 5 – metering screw; 6 – power take-off box; 7 – cardan shafts mounting; 8 – vibrator; 9 – loading screw; 10 – mounting of jacks; 11 – feed hopper; 12 – chassis

FIG. 16. Mobile unit for grout preparation and in situ immobilization of solid and liquid wastes in tanks and trenches at Sergiev Pasad, Russian Federation.

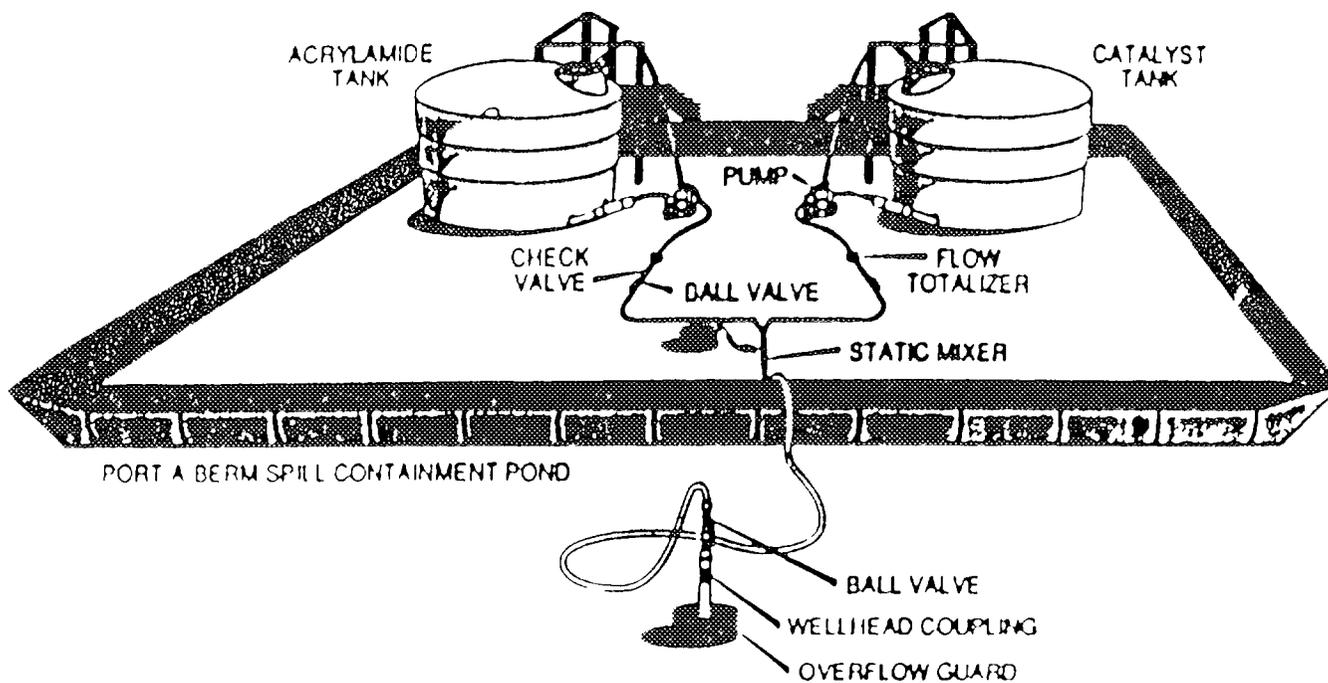


FIG 17 Layout of grouting and injection equipment used for in situ immobilization at the ORNL trenches USA

Demonstrations of in situ grouting with polyacrylamide were carried out on two undisturbed burial trenches and one dynamically compacted burial trench in the solid waste storage area at ORNL. The total volumes of grout delivered to four trenches are summarized in Table XIII. The injection of polyacrylamide was achieved quite easily for the two undisturbed burial trenches which were filled with grout at typical pumping rates of 95 L/min in several batches injected over several days.

The compacted burial trench failed to accept grout at more than 1.9 L/min even when pressure was applied. Thus, it appears that burial trenches stabilized by dynamic compaction have a permeability too low to be considered groutable. The water table beneath the burial trenches did not respond to grout injections, indicating a lack of hydrologic connection between fluid grout and the water table which would have been observed if the grout failed to set. Because grout set times were adjusted to less than 60 min, the lack of hydrologic connection was not surprising. Post-grouting penetration testing revealed that the stability of the burial trenches was increased from 26% to 79% that measured in the undisturbed soil surrounding the trenches. In situ permeation tests on the grouted trenches indicated a significant reduction in hydraulic

conductivity of the trench contents from a mean of 2.110^{-3} to 1.8510^{-6} cm/s. Field demonstrations indicated that grouting with polyacrylamide is a potential method for both improved stability and hydrologic isolation of radioactive waste and its incidental hazardous constituents.

TABLE XIII. IN SITU GROUTING AT ORNL, USA
(POLYACRYLAMIDE GROUT INJECTIONS)

Trench number	Grout delivered (m ³)	Trench area (m ²)	Trench volume (m ³)	Fraction grouted (%)
7	67.9	65.2	298.1	22.8
165	41.1	29.6	135.5	30.4
6 (compacted)	1.5	57.6	263.2	0.6
8 (partial fill)	23.0	52.5	239.9	9.6

3.1.1.4. In situ immobilization of sludge in a concrete tank at Trombay, India

In Trombay, India, an open topped circular concrete tank (about 20 m diameter) used for interim storage of liquid low level waste which was awaiting treatment/immobilization and disposal accumulated about 0.3 m of thick sediment in the bottom [2]. The liquid waste arose from a number of processes at the facility including the plant decontamination. Although some of the liquid waste streams contained suspended solids, much of the sediment in the tanks arose from wind blown sand. The sediment contained mostly Cs and Sr. For operational and radiological reasons in situ immobilization of the sediment in the tank was carried out. Figure 18 shows the layout of the tank. The in situ immobilization process consisted of the following steps:

- 1) Erection of a moveable gantry bridge over the tank carrying a grout injection and stirring device;
- 2) Emplacement of several hundred topless and bottomless drums (i.e. tubes) into the waste resting on the base of the tank;
- 3) Via the overhead gantry, cement grout was injected into sludge and stirred. The void space between neighboring drums was also mixed and grouted in the same way;
- 4) After in situ immobilization of the waste in each drum a concrete cap was emplaced over the tank to form a large monolithic repository.

3.1.2. Future developments

Very little appears to be done internationally to develop generic technologies for in situ treatment/immobilization of existing wastes in tanks or disposal trenches. One exception is in situ vitrification (ISV) where two further development studies are underway, both being managed by Geosafe Corporation based in Richland, Washington. The biggest of these studies is being carried

out at the Maralinga Test Range located in a remote area of the Great Victoria Desert in Southern Australia and is being funded by the Australian Commonwealth. The site has extensive radioactive contamination resulting from British nuclear weapons tests conducted in the 1950s and 1960s. Phase 2 of a four phase project is underway to immobilize plutonium contaminated mixed wastes buried in 21 pits at the Taranaki area of the test range. The 21 pits contain massive amounts of steel and other debris contaminated with plutonium, uranium and heavy metals such as barium, lead and beryllium. Organic based wastes are also buried in the pits [20].

In another study vitrification has been investigated as in situ solidification solution for immobilizing radioactive sludges stored in underground tanks. The concept involves installing a graphite gas release vent to the bottom of the tank, prefilling the head space in the tank with inert material (scour or soil), and vitrifying the tank contents. Several nonradioactive, pilot scale tests have been conducted with a good success, but a nonradioactive field test resulted in a pressurization vent that expelled molten soil onto the steel off-gas collection hood. Consequently, further developments of in situ vitrification for underground tanks has been suspended [21].

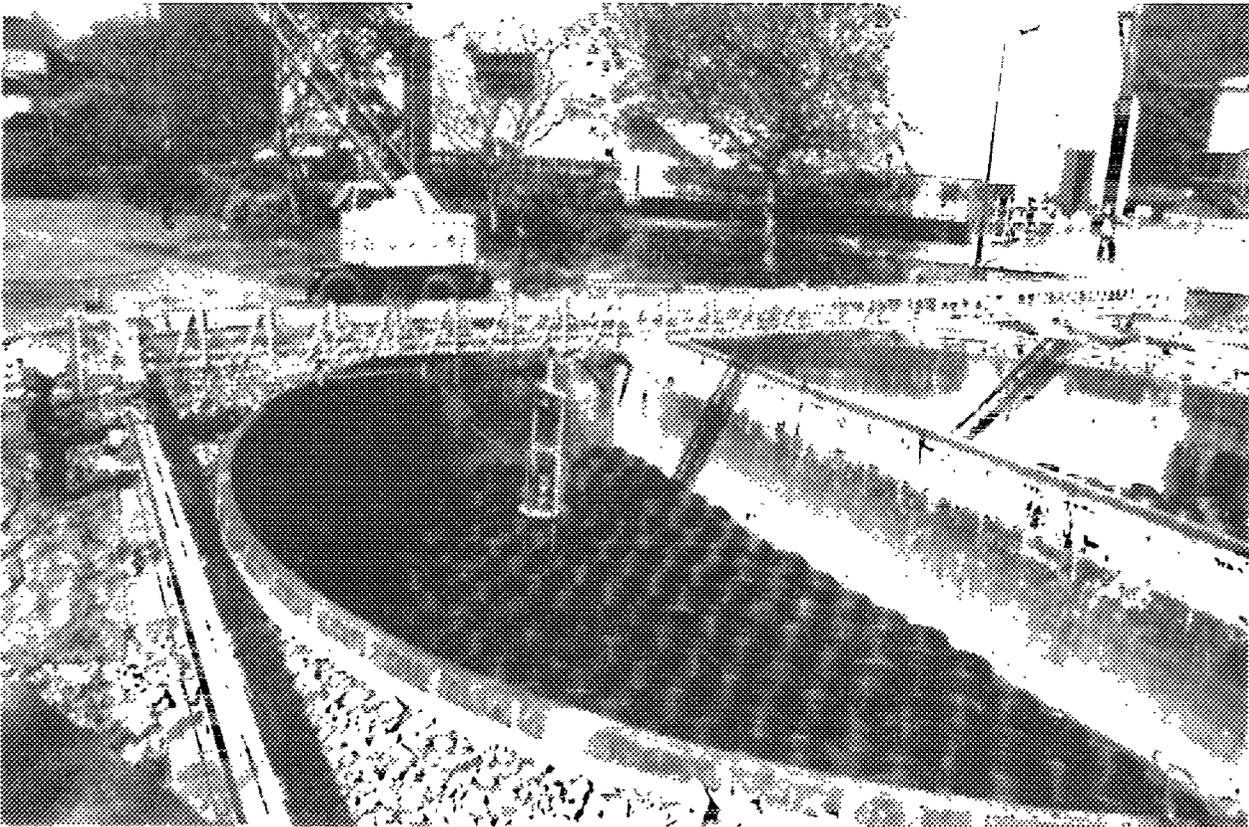


FIG. 18. In situ immobilization of sludge in a concrete tank at Trombay, India.

3.2. IN SITU IMMOBILIZATION OF GEOSPHERE CONTAMINATION

There are numerous locations with widespread radioactive soil contamination around the world. These may have arisen because of accidents or because of historical disposal practices which are not now performing adequately. Whatever the origin, the resultant geosphere contamination may be posing a radiological or environmental hazard and therefore requires remediation. If for economic or radiological reasons, or for reasons of practicality, it is decided not to exhume the ground contamination but to immobilize it in situ, then the technologies described in this section are applicable. In some cases where the ground contamination is widespread but contained at a shallow depth, such as may result from atmospheric deposition, then it may be beneficial to scrape the contaminated surface material into one location. This may be carried out, for example, using a bulldozer. The accumulated pile of waste may be tipped into an excavated hole for in situ immobilization. Alternatively, if the contamination is not distributed but is contained in one clearly delineated location, then in situ solidification/immobilization may take place without further disturbance of the waste. The key technology described in this section for immobilization of this type is in situ vitrification. In situ vitrification has largely been pioneered in the USA and has been tested on conventional chemically contaminated sites. The process is also undergoing further radioactive waste feasibility studies at the Oak Ridge National Laboratory.

One other technology is described in this section which again is being pioneered in the USA and is called Deep soil mixing. This involves direct mixing of the waste using a large auger device while at the same time injecting a grout formulation to form a homogenous admixture which subsequently hardens in situ.

3.2.1. Operating experience

3.2.1.1. *In situ grouting using deep soil mixing*

In situ deep soil mixing involves mixing soil with a grout formulation injected into the soil through a hollow stemmed auger [22–23]. The auger turns and mixes the soil with the grout slurry (see Fig. 19). The slurry continues to be injected as the auger is withdrawn to ensure thorough soil mixing. The resulting mixture is a cement-like matrix that immobilizes the contaminated soil in situ.

A recent demonstration of the deep soil mixing technique was performed in 1992 at the US Portsmouth Gaseous diffusion plant where the site is contaminated with volatile organics mixed with low concentrations of uranium and technetium from the diffusion enrichment process.

The soil boring/mixing tool uses either a single or dual bladed auger into the soil and provides lifting, mixing and injection points for grout, air or other fluid (see Fig. 20). At the Portsmouth demonstration, both 1.8 m and 3.0 m diameter augers were used. A bar transmits rotational torque to the soil boring/mixing tool and provides a path for the injected air or grout. A fiberglass shroud is used to seal off the area above the drilling. A negative pressure can be maintained inside the shroud which is connected to an off-gas treatment system. The parameters and criteria for deep soil mixing are shown in Table XIV.

With regard to in situ immobilization of the contaminated soil, full scale testing was completed using the following processes:

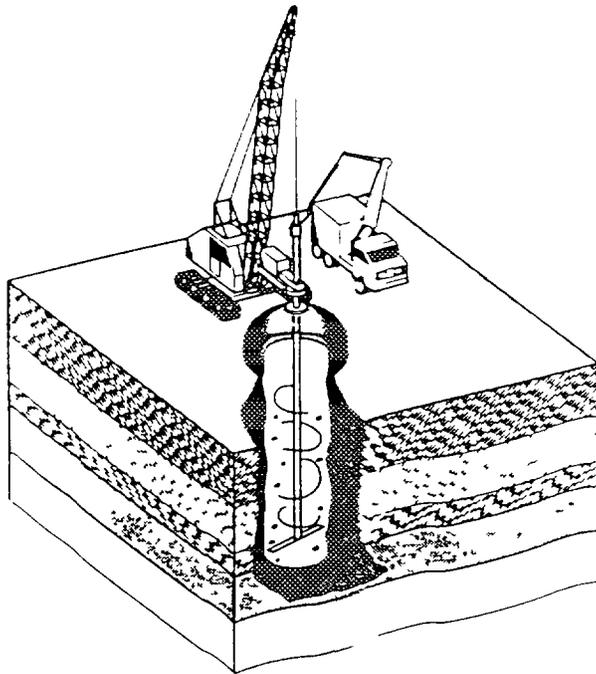


FIG 19 Deep soil mixing concept for in situ immobilization of soil contaminants

TABLE XIV PARAMETERS AND CRITERIA FOR DEEP SOIL MIXING

Parameter	Criteria
Soil treatment depth	Up to 5 m
Treated soil column diameter	Up to 9 m
Typical stabilizing agents	Portland cement/lime slurry
Water to slurry ratio	Varies with soil moisture content
Final closure	Treatment of hazardous contaminants may require installation of final cover to provide long term minimization of infiltration
Contaminant mobility	Possible migration of contamination away from the injection and mixing zone

Hot Air Injection. Hot air injection treatment was performed on three 4.6 meter deep test cells for approximately four hours each. One 6.7 meter test cell was also treated;

Ambient Air Injection. Ambient air injection treatment was performed on three 4.6 meter deep test cells for approximately four hours each;

Hydrogen Peroxide/Water Injection. Hydrogen peroxide water injection treatment was performed on three 4.6 meter deep test cells for approximately one hour and fifteen minutes each;

Helium Trace Injection. A helium tracer test was performed on one 4.6 meter deep test cell for approximately three hours each;

Grout Injection. The demonstration was performed on three 4.6 meters deep test cells. The grout was injected at a rate of 0.9 cm^3 per minute through the mixing system as the mixing blade slowly rotated at 10 rpm down through the soil. Mixing up and down continued for approximately one hour. A total of six samples were taken, at 1.5 m and 3 m depth

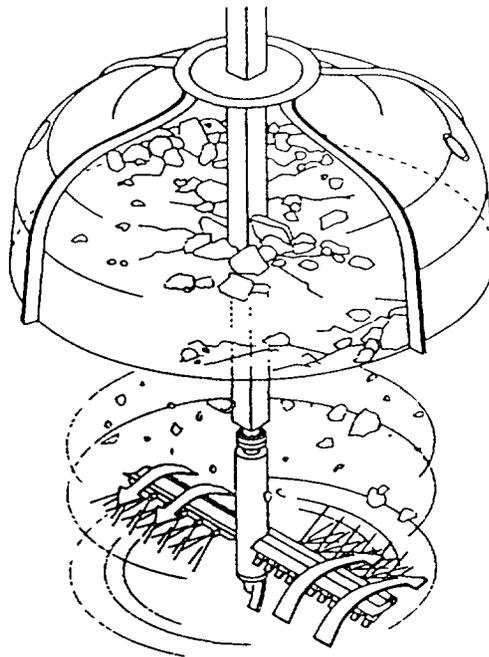


FIG 20 Deep soil mixing equipment with dual bladed auger and vapour control shroud.

3 2 1 2 *In situ vitrification*

a) *In situ vitrification without involving disturbance of the waste*

In situ vitrification (ISV) is a thermal immobilization process that converts radioactive and chemically contaminated soil into a stable vitrified product [24–29]. ISV has been demonstrated on actual radioactively contaminated soils on two occasions and is being considered for application at several more radioactive sites. The first demonstration was conducted on a transuranic contaminated soil site at Hanford in the USA known as the 216–Z 12 crib. This crib contained Pu and Am concentrations of up to 2000 nCi/g in the soil underlying a tile field at depth of 7 m. This demonstration was the first full scale application of ISV at an actual radioactive waste site. With the exception of achieving the complete process depth, the demonstration met all the demonstration goals of equipment performance, off-gas containment and waste form quality. However a subsequent demonstration was not as successful in vitrifying through the entire contamination depth and so development work on the technology is continuing to improve the process depth.

The second radioactive application of ISV known as the 116–B–6A crib took place in 1990 also at Hanford site. This site is contaminated with mixed fission products, caesium and strontium, and heavy metals, lead and chromium. This crib, during its 17 year operating history, accepted decontamination solutions for disposal in a subsurface, 3.7 m x 3.7 m x 2.5 m deep wooden timber crib. The crib and its content was vitrified by ISV to the 4.5 m depth. Except for achieving the target depth of 6.5 m, all objectives of the demonstration were met including obtaining > 99 wt% retention of contaminants in the glass, containing off-gases generated from the combustible timber crib, and producing a durable glass product. The inability to vitrify below the crib to the target depth was attributed to the presence of a cobble layer which has been found to be resistant to the vitrification process. This discovery has renewed efforts to enhance the processing depth of ISV. Since this demonstration, ISV has been successfully tested to 6.1 m by a commercial supplier of ISV services in the USA.

Present considerations for future radioactive applications of ISV include Oak Ridge National Laboratory seepage trenches, Hanford transuranic contaminated soil sites, uranium mill tailings and uranium contaminated soils (Japan). In addition to these sites and applications, ISV has been identified as the preferred remediation technology at 10 hazardous chemical sites in the USA. At the present time ISV is being applied on the first of these sites.

The technology of ISV involves four cylindrical graphite electrodes in a square array inserted vertically a few centimeters (approximately 30 cm) below the surface of the soil. A shallow and narrow trench (approximately 4 cm wide x 9 cm deep) is partially filled with a mixture of graphite flakes and glass frit to facilitate starting of the process. As alternating electrical voltage is applied to the four electrodes current begins to flow in the graphite starter path which heats the surrounding soil to approximately 1600^oC. As the soil melts it becomes electrically conductive itself thus being heated by the electrical current. The molten soil mass grows outward and downward as the process ensues until the desired depth is reached. The electrodes are lowered into the soil as the molten soil proceeds downward. Figure 21 illustrates the process. ISV is generally applicable to soils contaminated with either radioactive or chemical constituents or both. It is generally applicable across a broad range of site conditioning, such as soil type and composition, contaminant type, soil inclusions.

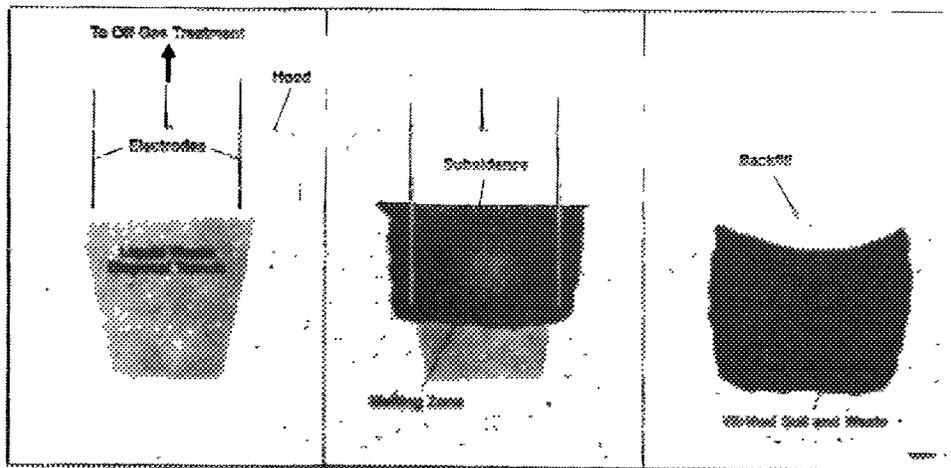


FIG. 21. Disposition of materials during ISV processing.

The off-gas and power systems of ISV are designed to accommodate soil moisture concentrations of up to 50 wt%, as long as groundwater recharge of the treatment zone is prevented during processing. With an energy consumption rate of 0.7 kW•h/kg and a power capability of 3500 kW for the process, the maximum processing rate is calculated at 5000 kg wet soil/h. At this processing rate for soil at 50 wt% moisture, 52 STD m³/min of water vapor would be generated. This is approximately half the maximum off-gas rate designed for the process. If the soil moisture is due to a high groundwater table, the groundwater would need to be temporarily lowered by pumping or diversion during processing to prevent recharge.

Vitrification of soil is largely independent of soil classification, whether it be sand, silt, clay or combination of these types. However, minimum concentrations of silica and alkaline element oxides are required to achieve a molten soil. ISV has been successfully tested with soils with minimum combined sodium and potassium oxide concentrations of 1.4 wt% in the soil. Most soils have sufficient concentrations of sodium and potassium oxides to be vitrified. However, some soils along the southeastern seaboard of the USA, for example, are weathered to the extent that they are not currently processable without the addition and mixing of alkaline oxide.

ISV has been tested and demonstrated on several types of radionuclides mixed with inorganic and organic contaminants. The process was originally developed for transuranic waste (> 3.7 kBq/g of transuranic elements such as Pu and Am) as an alternative to removal and repository disposal. It has since been demonstrated for mixed fission products, heavy metals and organics. Most of the radionuclides and inorganic contaminants are retained and immobilized in the glass and crystalline product. Table XV shows leach test results from one bench scale test of ISV. Organic and some inorganic contaminants are either destroyed by pyrolysis or removed by volatilization during the process. The certainty of removal and destruction of volatile organic contaminants (VOCs) (those with boiling points < 100°C) have not yet been fully demonstrated. When VOCs coexist with radioactive contaminants, treatability studies and special monitoring in the soil region outside the area being treated may be necessary to ensure VOCs are effectively treated by the process.

TABLE XV. LEACH TEST RESULTS FROM ONE BENCH SCALE TEST OF ISV

Contaminant	Initial concentration in soil (g/g)	TCLP results of ISV product, metal concentration (mg/L)	Allowable concentration (mg/L)
Arsenic	4400	<5	5
Barium	4400	<1	100
Cadmium	4400	<1	1
Chromium	270 to 4400	<0.2 to 2.7	5
Silver	4400	<0.1	5
Lead	50	<0.1	5
Mercury	46	<0.0001	0.2

ISV is limited by certain soil inclusions, primary sealed intact containers or tanks. Normally, water vapour and other gases generated by the process are vented through the dry soil zone surrounding the molten soil into the off-gas collection hood. However, when sealed containers are present, gases inside the container cannot escape outside the melt and are forced to vent through the molten soil. This behavior can result in occasional violent bubbling or expulsion of molten soil onto the off-gas hood surface. Consequently, the presence of sealed containers and buried wastes with intact sealed containers are to be avoided.

The ISV process is amenable to other types of radioactively contaminated soil inclusions such as solid metallic and combustible components which frequently co-exist with radioactively contaminated soils. The process has been successfully tested on soil with up to 25 wt% scrap metal. When metals are present, a secondary metallic waste form is formed at the bottom of the glass and crystalline product. Successful tests have included aluminum and iron based metals such as stainless steels. Tests with combustible components have included up to 0.3 wt% based on 2380 kg of timbers in 749 000 kg of soil. The process is designed to treat up to 7 wt% combustibles.

b) *In situ* vitrification involving disturbance of the waste

For situations where there is widespread shallow ground contamination a variation of ISV has been developed where the waste is scraped into one location (such as using a bulldozer) and then solidified in situ. This variation of on-site vitrification could be applied with low capital cost and high production rate for contaminated soil and buried wastes where direct in situ vitrification is not feasible or practical because of the aerial extent of the contamination. As shown in Fig. 22, vitrification is accomplished in a pit excavated in the earth with a high temperature refractory roof. Contaminated soil and waste material are consolidated at the waste site and fed to the surface of a molten glass pit through a port in the refractory roof. The molten pool is heated by electrodes fed through the roof. Combustion air is supplied above the molten glass pool to oxidize combustible wastes. Molten glass product is accumulated within the pit or drained through a discharge section into waste containers for immobilization in an on-site disposal pit. Metallic wastes melt and accumulate at the bottom of this vitrification pit, where they solidify

at the completion of vitrification operations. Several other types of vitrification processes exist for a wide variety of wastes. These include the following: cyclone furnace, microwave energy melter, low temperature vitrification, direct current (DC) arc furnace, plasma centrifugal furnace, stirred glass melter, entrained bed gasifier, ceramic lined melter, induction heated crucible.

Based on information currently available, these processes do not lend themselves to in situ immobilization or in situ disposal of radioactive wastes. Discussions, therefore, will be limited to the in situ vitrification variation.

The on-site vitrification process is claimed to be applicable to various forms of waste, including contaminated soil, combustible wastes, metallic components, sludges and slurries. Applicable contaminants include radioactive, heavy metal and inorganic contaminants. Gaseous elements, iodine and mercury are excluded from applicability because of their volatile nature from the process. For inorganic contaminants, elemental concentrations are limited by their solubility in most waste glasses. Table XVI provides general limits of contaminants and constituent concentrations.

Laboratory testing of this technology has been completed on high radium containing soils, municipal solid waste, plating sludge, 25 wt% sewage sludge, cardboard recycling sludge, asbestos, dry active waste (typical of those generated in nuclear power plants) and ion exchange resins. No published data on the performance of this technology on these waste types are currently available, however.

A 25 tonne/day contaminated soil melter has been constructed for potential use on radioactively contaminated soils at Hanford in the USA. Currently, however, the on-site vitrification process is not being implemented for any radioactive applications.

A great deal of literature has been published on the performance of waste glass forms. Much of this information is directly applicable to the vitrified product produced from the on-site vitrification process. However, no published information is currently available on the waste form characteristics or the specific glasses produced from this type of vitrification process for the types of wastes mentioned.

TABLE XVI. CONTAMINANT AND CONSTITUENT LIMITS FOR IN SITU VITRIFICATION

Solubility limit	Constituents
<0.1 wt%	Ru, Rh, Pd, Ag, Pt, Au
1-3 wt%	C, S, Cl, Cr, As, Se, Tc, Sn, Sb, Te
3-5 wt%	Ti, Mn, Co, Ni, Cu, Mo, Bi
5-15 wt%	F, La, Ce, Pr, Nd, Gd, Th, rare earths
15-25 wt%	Li, B, Na, Mg, Al, K, Ca, Fe, Zn, Rb, Sr, Cs, Ba, Fr, Ra, U
> 25 wt%	Si, P, Pb

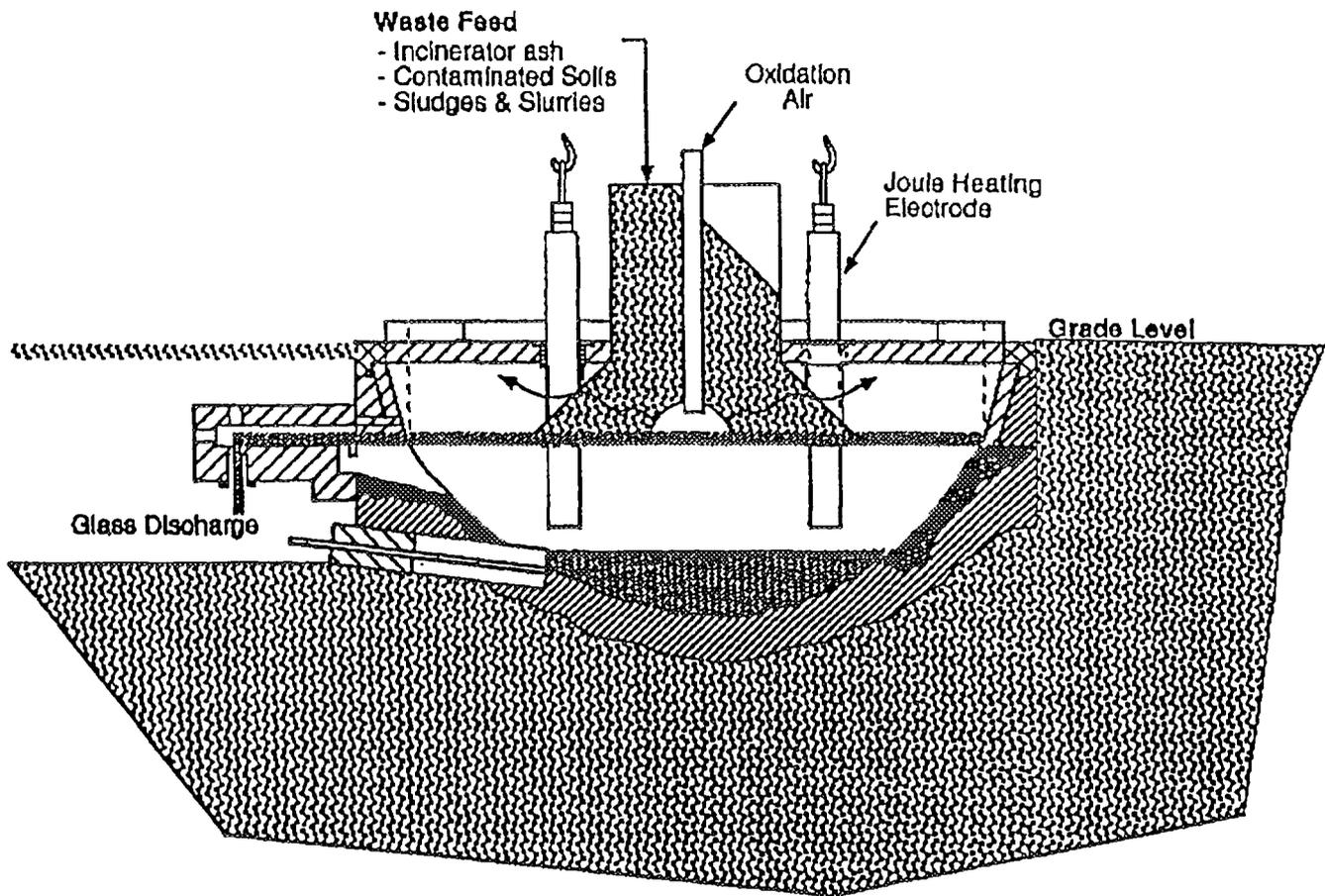


FIG. 22. Layout of in situ vitrification process involving disturbances of the waste.

The cost for a 100 tonne/day melter for contaminated soils using electricity at \$0.06/kW•h have been estimated at \$125/tonne. These estimates are claimed to include capital recovery costs for the melter, operating labor, energy and maintenance. Capital costs have been substantiated by the erection of the 25 tonne/day melter for less than \$500 000. Operating cost estimates have not been publicly substantiated through field operations.

In Japan, a 3 year engineering scale testing programme has been completed, in which the application of ISV for vitrifying a low level waste burial vault has been evaluated [28].

3.2.2. Future developments

Conventional ISV techniques currently use electrical ohmic heating. Alternative power sources are being investigated including plasma and chemical heating. The first of these, using

plasma arc technology, is being researched at under a project called PRISM (plasma remediation of in situ materials). This technique is being made viable by advances in plasma arc technology. Conceptually, a plasma arc torch can be lowered into a borehole to any depth and operated to melt contaminated materials into a type of magma or lava, which cools into a zone of vitrified material. Subsequently, the plasma torch is slowly raised and operated at progressively higher levels to thermally convert a mass of soil into a vertical column of vitrified and remediated material. By applying this technique over a systematic grid pattern, the process becomes a viable means of in situ thermal vitrification for burial pits containing contaminated materials. Three small scale laboratory tests have been performed with a 100 kW plasma arc torch to simulate in situ thermal vitrification of soils. It is anticipated that the PRISM concept would be applicable to the same situation currently tackled by conventional ISV techniques. Plasma arc torches applied to ISV operate at power levels exceeding 1 MW and would be expected to produce vitrified columns greater than 3 m [30].

4. IN SITU CONTAINMENT APPLIED TO WASTE DISPOSAL AND CONTAMINATED SITES

Containment of radioactive waste at disposal and contaminated sites is used primarily to prevent dissolution of contaminants or at least to reduce their migration in the geosphere. A second purpose of some containment technologies is to prevent intrusion into the site by humans, animals, plants, or other naturally occurring phenomena. Implicit in these purposes is the need to manage the flow and movement of water in and around the waste site.

A containment technology may be applied to a site for one of the following reasons:

- To provide temporary containment of reagents during certain in situ treatment technologies, such as soil flushing and some biotreatments;
- To provide interim containment of the waste pending future treatment action (for some sites, a viable treatment method may not yet exist); and
- To provide permanent containment of the site, effectively eliminating the need for future treatment action.

In situ containment technologies are those which are applied to existing waste disposal or contaminated sites without first removing the waste materials and which are applied in a manner that minimizes intrusion into the site during emplacement.

In situ containment technologies, or barriers, can be divided into three main categories:

- surface barriers (caps)
- vertical barriers (cut-off walls)
- sub-horizontal barriers (floors).

The following discussion is organized around these categories. In addition to these categories, some containment technologies exist involving chemical barriers that retard migration of selected contaminant species without impeding the groundwater flow.

Since, in most cases, resistance to the flow of water is the objective of containment technologies, an important measure of barrier performance is bulk hydraulic conductivity (permeability). Unfortunately, there are no international standards for performance of containment technologies applied to radioactive sites. The US Environmental Protection Agency (EPA) guidance for hydraulic barriers, including caps and liners, over and around disposal sites regulated under the Resource Conservation and Recovery Act (RCRA) of 1976, recommends achieving an average hydraulic conductivity of 10^{-7} cm/s. This value is generally accepted in the USA as a benchmark for evaluating other barriers, including the in situ containment technologies discussed in the following sections. In some cases, higher values may be acceptable in the USA depending on the specific application for the barrier. An excellent overview and discussion of containment technologies is provided in Ref. [31]. The authors provide an assessment related to all types of barrier application at US DOE (i.e. nuclear waste) sites; therefore the information is broadly applicable.

4.1. SURFACE BARRIERS

Surface barriers are placed over a waste site with the primary purpose of separating or isolating the subsurface waste from the surface environment. Therefore, the goals of the containment technology used as a surface barrier are:

- to control the infiltration of surface moisture (e.g., rain water, snow melt) which in turn minimizes dissolution of contaminants into the groundwater;
- to prevent direct contact with receptors (e.g., humans, animals, and plants); and
- to control the release of gases and vapors generated from the waste site [30].

Surface barriers can be constructed using single layer or multilayer designs. The method and materials of construction are generally based on the desired design life and performance requirements. Depending on their design and the nature of the underlying waste, surface barriers require periodic inspection and maintenance for subsidence (settling), climatic erosion, and invasion by deep rooted plants or burrowing animals during the period of institutional control. The frequency of inspection and maintenance of the cap will depend on the climate, flora, and fauna of the particular site.

4.1.1. Operating experience

Numerous efforts are being pursued worldwide regarding cap design, barrier materials, and construction methods, to improve and predict long term barrier performance. Unfortunately, much of this is unpublished. As a result, the information below largely refers to U.S. data which are readily obtainable from a number of sources.

4.1.1.1. Single layer caps

Single layer caps are generally used as an interim measure where short term containment of a site is desired, pending selection and implementation of a longer term solution. Materials used in a single layer cap may, for example, be soil, asphalt, concrete, or a synthetic material. Clay materials, while sometimes useful in the subsurface of a multilayer cap, are not typically a good material for single layer cap construction because weathering (i.e., exposure to heat/cold

and wet/dry cycles) can cause cracking of the clay, thus minimizing its effectiveness as a barrier material. The profile of single layer caps may be domed or have a less pronounced gradient depending on the hydraulic properties of the construction materials and the hydrological design criteria.

Since the materials for a single layer cap are well developed and the application of the single layer cap is fairly limited, there is minimal development work being done in this area.

4.1.1.2. Multilayer caps

Most surface barriers are engineered using multiple components [32–34]. In the USA barriers for hazardous waste applications are governed by RCRA and are designed to provide a useful life of 30 years or more. Since there are no official standards regulating barrier design and construction for radioactive waste sites, the RCRA standards are generally applied at radioactive and mixed waste sites in the USA for long term, though not permanent, containment, pending future remedial action.

In countries where wastes with longer half-lives are disposed of in near surface repositories, surface barriers may have to be engineered with a design life of hundreds, or even thousands, of years. In such cases a detailed understanding of cap performance criteria is required so that caps can be designed that have extended anti-erosion and long term hydrological properties. There are a number of internationally available numerical codes to model cap performance and therefore aid design, such as HELP from the EPA.

In general, the various layers typically consist of an upper layer, a drainage layer, and a low permeability layer; each of these general layers may be constructed using multiple components. For example, the EPA recommends two alternative multi layer surface barrier designs. The standard design incorporates a top layer (generally composed of about 600 mm of top soil with vegetation), a drainage layer (generally composed of about 300 mm of sand or fine gravel having a minimum hydraulic conductivity of 10^{-2} cm/s); and a low permeability final layer (generally consisting of a geomembrane such as PVC, LDPE, or HDPE, depending on design life and other concerns) at least 0.5 mm thick overlying a 600 mm thick layer of low permeability compacted soil with saturated hydraulic conductivity of 10^{-7} cm/s.

Use of clay-rich soil has been widely used as the low permeability compacted soil layer. Unfortunately there are several possible problems associated with such clay-rich soil liners that may cause them to be susceptible to damage, thus compromising their effectiveness. Specifically, they are difficult to effectively compact on a soft foundation (i.e., some waste materials). Future collapse of the underlying waste materials may cause differential settlement and result in cracking or shearing of the clay-rich soil layer. In addition, clay-rich soils are highly dependent on moisture content for their hydraulic conductivity properties; dehydration will therefore cause cracking unless adequately protected against moisture loss. Finally, compacted soil liners of any type are difficult to repair because of their proximity to the waste zone and the complexity of the overlying composite layers. However, special consideration and attention to these issues during design of the cap can help alleviate these potential problems.

Alternatives to clay-rich soils include soil/bentonite blends and geosynthetic clay liners (GCL). The former are not as susceptible to damage as clay-rich soil liners. The latter offer improved performance over soil/bentonite blends but are relatively new and have not yet been broadly applied. The GCL is constructed of thin 'blankets' of bentonite attached to one or more

geosynthetic materials (e.g. a geotextile or geomembrane). A detailed description of various GCLs is provided in Ref. [33]. Special consideration of the durability and design life of the cap must be given if synthetic materials are to be employed in place of natural materials.

The alternative surface barrier design offered by the EPA, in addition to the component layers of the standard design, includes the following options. Firstly, cobbles at the surface to provide enhanced protection from climatic erosion. Secondly, a layer of cobbles about 300 mm thick beneath the top soil layer to provide a biotic barrier, preventing intrusion of deep rooted plants and burrowing animals. Thirdly, a high permeability layer (similar to the drainage layer in the standard design) about 300 mm thick beneath the low permeability soil layer and above the waste, to provide a vent for controlled release of gases such as methane, hydrogen and radon generated by decomposition, corrosion or radioactive decay of the waste. In addition, a geomembrane or geosynthetic filter may be used at the top of either the biotic barrier and/or the gas vent layers. The need for the various additional layers is determined by assessing factors related to the waste characteristics, site characteristics, geology, hydrology and cap design life.

Although no construction standards exist for containment barriers for low and intermediate level waste, the design life of surface barriers in the USA is addressed by various regulatory agencies. For example, while the RCRA cap provides for at least 30 year design life, the US NRC (through 10 CFR 61) allows 100 years, the US DOE (through DOE Order 5820.2a, 1988) allows for 150 years. Other criteria for LLW site stability are given in 10 CFR requiring 500 years, while 40 CFR 193 requires 1000 years. In other countries, such as the UK, the regulatory environment is much less stipulative. The requirement is merely that any surface barriers have to be designed and demonstrated with confidence that they perform adequately throughout the time period that the waste remains a hazard. Thus, the requirements of surface containment are dictated by the results of a site performance assessment.

4.1.2. Future developments

4.1.2.1. Materials of construction

Barrier design and construction continues to improve as new geosynthetic materials are developed. The use of naturally occurring materials (e.g., soil, sand, clay and gravel) in multi-layer surface barrier construction will never be totally replaced. However, optimized geosynthetic materials in surface barriers may greatly decrease the risk of barrier failure, thus increasing the performance of the cap during its intended design life. Improved fundamental understanding of the physical processes governing performance will also greatly improve cap design. Advanced numerical and experimental modeling will undoubtedly have an increasing role to play where long term durability and performance is required.

4.1.2.2. 'Permanent' surface barriers

No surface barrier to date has been designed nor constructed with a design life of 1000 years or more. Such a barrier would be required to survive and retain its function and integrity beyond the end of institutional control, without routine inspections and maintenance/repair, and through potential climatic changes. However, development of such a barrier is underway, for example, at the Hanford Site in southeastern Washington, USA and other sites worldwide where long term containment of radioactive waste is required [33–35].

Long term isolation surface barriers have been proposed to protect disposed wastes from transport back to the environment. The Hanford Barrier concept uses engineered layers of natural materials (e.g., fine soil, sand, gravel, rip-rap, and asphalt) to create an integrated structure with redundant protective features. Natural materials have been selected to optimize barrier performance and longevity; no synthetic materials or clay-rich soils, therefore, are used. The Hanford Barrier Development Programme was initiated in 1985 as a collaborative programme among Hanford site scientists and engineers as well as outside contractors, universities and consultants. The programme consists of three stages to build up the necessary information and experience needed to design and assess the performance of long term barriers. These activities include field tests, experiments (including extensive lysimeter studies), computer models and natural analogues. Results from these activities have provided the necessary input for design and construction of a prototype barrier that was completed in 1994. Following construction, a minimum of 3 years of testing and monitoring is planned to evaluate its design performance. Since only a finite amount of time is available to test a prototype barrier that is designed for a minimum of 1000 years, an accelerated testing programme has been designed to stress the barrier to measure performance within a reasonable time frame.

An interesting issue associated with developing a barrier with a design life of greater than 1000 years is that of markers identifying the site as dangerous to human health and the environment and thus discouraging overt or inadvertent intrusion into the barrier. This and many other issues are being addressed in the US barrier development programme, expected to be completed in 1998. Similar issues have been considered in other countries, resulting in some cases in plans not to use markers to prevent archeological interest in the site in the far future, and to use only indigenous natural materials in the cap construction to prevent future scavenging activities.

4.2. VERTICAL BARRIERS (CUT-OFF WALLS)

Vertical barrier walls are placed around a site with the primary purpose of controlling migration of contaminants laterally away from the site. Vertical barriers can also serve to prevent lateral intrusion into the waste site by hydrological, biotic or human agents.

Vertical barrier walls are generally constructed using techniques related to excavation and backfill, injection or soil mixing. The objective is to generate a relatively 2-dimensional underground structure (i.e. long and deep, with relatively small thickness). Vertical barriers are generally constructed using cementitious materials so that they are easily handled and delivered during placement, but once in place the materials set and form a virtually permanent barrier structure. Depending on the placement technique, the operation can be conducted in either a batchwise or continuous fashion. Alternatives to the cementitious materials are barrier walls which are placed using thermal means, by either melting (vitrification) or freezing (cryogenics) the soil, and mechanically introduced sheet piling [32, 36–40].

4.2.1. Operating experience

4.2.1.1. Slurry walls

Slurry walls are vertical barriers that reduce groundwater flow in unconsolidated earth materials. Slurry wall construction involves excavating a narrow vertical trench through pervious soils and then backfilling the trench with an engineered material. The process is conducted entirely under the slurry in the excavation trench. The backfill material is usually a mixture of

soil and bentonite (typically, using the soil excavated from the trench) or cement and bentonite. The cement–bentonite slurry walls provide greater structural strength than soil-bentonite walls, but their hydraulic properties are less favorable. The soil–bentonite slurry wall is the more common of the two.

Slurry cut-off walls have been used for over 40 years to control groundwater flow. They are typically 60 to 120 mm wide and can be up to 50 m deep, they must be keyed into the confining layer beneath the aquifer to seal off the groundwater flow. If the pollutant to be controlled is an organic layer floating on the groundwater, then the slurry wall need not be keyed nor extend through the entire depth of the aquifer, this is called a hanging slurry wall.

In evaluating or predicting the performance of a slurry wall it is important to consider its physical/chemical properties relative to their compatibility with the site hydrology and specific characteristics of the waste being contained (for example pH). An important feature of slurry walls in addition to their physical containment is their ability to also act as chemical barriers. For example, the hydrological performance of slurry walls constructed using bentonite clay may markedly decline over a period of a few decades, but the residual material will retain important sorption properties that may help to chemically retard contaminant migration.

4.2.1.2 Grout curtains

Historically, grout technology has been used in the construction industry for stabilizing soil (for example in damaged earthen dams, to provide foundation support). More recently, application of the technology has been made to waste sites for the purpose of containment. However, a limiting factor in the use of grout curtains is that, compared to the slurry wall, it is a relatively slow, labor intensive process that may produce a barrier wall with inferior hydraulic properties. Nonetheless, its application is growing as a vertical containment technology. There are several methods for forming grout curtains, jet grouting, permeation grouting, deep soil mixing and vibrating beam injection. Each of these methods is briefly described below.

Jet grouting

This technology uses high pressure air (6000 psi) to mix water and Portland cement with a column of soil, in situ. The mixture is introduced to the soil through a grouting head that has been inserted into a hole drilled in the soil to the desired depth. The grouting head is lowered to the bottom of the hole and the materials are introduced through the head, which then rotates through 360° horizontally and incrementally creates a flat disc. The head is raised uniformly upwards, producing a column of grouted soil. The resulting column is typically 0.6–0.75 meters in diameter with an hydraulic conductivity of the order of 10^{-7} cm/s. Sequential applications of the process, placed such that columns are staggered with slight overlap, result in a barrier wall that is about 1.5 columns in thickness.

Permeation grouting

This technology is similar to jet grouting in that a grouting head is delivered to the bottom of a hole drilled into the soil. It differs in the fact that the grout compound is introduced at low pressure so that the interstices of the soil particles are filled but the soil is not displaced. Typically, the column is generated starting at the bottom of the hole as with the jet grouting process, although the grouting head is not raised uniformly and multiple grouted sections are needed to produce the column. In producing a barrier wall with permeation grouted columns, it

is necessary to apply multiple rows with much overlap to provide a broad point of contact between adjacent pillars, because the pillars do not coalesce into a single monolith as in jet grouting. Multiple injections of different grout compositions and viscosities are also often required to ensure complete filling of the soil void space.

Deep soil mixing

This technology has been described previously in section 3.2.2.1 as an in situ immobilization technology. As a containment technology it is used to merely deliver a grout mixture to the soil column through the tips of the augers during both penetration and retraction of the equipment. The grout becomes thoroughly mixed with the soil and sets in situ; multiple applications in an overlapping pattern are used to create a vertical wall.

Vibrating beam

With this technology, a beam is driven or vibrated vertically into the ground. As the beam is withdrawn, grout is injected through nozzles at the bottom of the beam into the void remaining, creating a single column. The beam is then inserted into an adjacent location overlapping the previous beam setting. Since the grout fills the beam void rather than mixing with soil particles, this technology offers more control over the barrier properties.

4.2.1.3. Sheet piling

Interlocking steel panels, as used in the construction industry to make dams for bridge construction, can be used to form a vertical cut-off wall when placed around a waste site. The piles can either be driven or vibrated into place. Potential high permeability at the interlocking joints can be reduced by injecting grout through ports provided in the panels for this purpose.

4.2.2. Future developments

There are a number of interesting trends concerning the development of vertical barrier walls. Some research is being devoted to developing new and improved grouts. Other research activities are directed toward thermal technologies such as soil freezing (cryogenics) and soil melting (vitrification). The former is proposed to provide a barrier wall that is completely temporary and can even be 'turned on or off' as desired. The latter is proposed to provide a permanent structure, maintaining its integrity not just for hundreds or perhaps thousands of years (as with the grout technologies) but for geologic time periods. Research is also being devoted to chemical additives to the soil that will provide a filtration or sorbent barrier to selectively capture or otherwise limit the migration of contaminants without impeding the groundwater flow. The following discussion provides a brief description of each of these developing technologies.

4.2.2.1. Grout improvements and alternatives

A number of additives to traditional grout mixtures as well as alternative materials are being developed.

Polymer additives

In the USA, research is being conducted on several different advanced polymer materials for use in subsurface barriers for radioactive material containment at US DOE sites. Polymer

binders such as polyester styrenes, vinylester styrenes and high molecular weight acrylics are being investigated for grout composites [36, 37]. These materials have been used in many commercial applications, for example in sewage and brine handling systems, and as a repair mechanism for dams, bridges and highways. Results of laboratory testing of these materials for wet/dry cycling, resistance to various chemicals and irradiation and hydraulic conductivity have been favorable. Field testing of some of these materials as a barrier prototype is expected in the future.

New flowable grouts

Two new grout materials are being developed for creating low permeability barriers in unconsolidated soils for application to radioactive material containment at US DOE sites. One grout material is a montan wax and bentonite mixture developed in Germany, the other is a glyoxal-modified sodium silicate chemical grout developed in France. Research has been conducted in the laboratory and in the field to evaluate the performance of the grouts over a range of soil and contaminant conditions. Field experiments have included single borehole injection tests and results have been promising for certain soil types.

4.2.2.2. *Thermal technologies*

Frozen soil cut-off walls

This technology, also known as cryobarriers, is an established technique used in the construction industry as a temporary containment technique to consolidate ground during excavation. Although application of frozen soil cut-off walls as vertical barriers to waste sites is a logical adaptation of the technology and has been proposed for many years, application to such sites has not yet been demonstrated. Several pilot demonstrations of the technology are under way at some US DOE sites; however, results of the testing are not expected to be available for at least another year or more.

Frozen soil cut-off walls are traditionally installed in a vertical orientation for construction applications. Frozen soil walls are created by circulating refrigerated brine or liquid nitrogen (which is more effective, but also more expensive) into a linear series of closely spaced wells. As the soil around the well pipes cools, the moisture in the soil freezes and expands, filling the voids in the soil and thus reducing the permeability. The nominal thickness of the frozen soil wall is on the order of the spacing of the wells, or about 0.75–1.5 m. The proper spacing of the wells depends somewhat on the soil properties, particularly moisture content, and is necessary to ensure sufficient overlap of the frozen soil columns. If there is insufficient moisture in the soil to produce a good barrier, water may be injected into the freezing area; however, this requirement may be counter productive toward the overall objective of maintaining containment of contaminants at a waste site. The principle advantages of frozen cut-off walls are that they are relatively easy to install and that they are temporary; once the barrier is 'deactivated,' the original properties of the soil are restored [38–39].

Vitrified soil cut-off walls

This technology is a proposed adaptation of in situ vitrification (ISV) developed by the Pacific Northwest Laboratory and now commercially available for hazardous waste remediation. Some modifications to the commercial process would be necessary to generate a vitrified soil cut-off wall; however, the general principles of ISV soil melting apply. Soil is vitrified by delivering

electrical power to an array of electrodes (in this case, two or more in a linear pattern) placed in the ground. Once melting is initiated between the electrodes at the soil surface, the pool of molten soil grows downward until the target depth is reached. This is currently limited to about 6 m in the commercial remediation arena, although much greater depths are projected, especially for the cut-off wall application. Continuous walls are produced by overlapping contiguous melt settings to form a single monolithic structure. The properties of vitrified soil are analogous to igneous rocks, particularly obsidian or basalt. The advantages of a vitrified soil cut-off wall are that it is extremely durable, highly impermeable to moisture, and virtually permanent in the environment. If the wall is damaged, it is easily repaired by remelting. A description of vitrified barriers and some laboratory results are given in Ref. [40].

4.3. UNDERGROUND HORIZONTAL BARRIERS

Horizontal barriers, or floors, are installed beneath a waste site as a containment measure to prevent or minimize downward migration of contaminants into the ground water, or conversely, to prevent percolation of the ground water upward into the waste site (Fig.23). They are used in conjunction with cut-off walls to provide containment during in situ remediation activities. If used in conjunction with walls and caps, they can provide for isolation of the contaminants from the environment. Barrier floors are necessary in situations when it is not possible to key a cut-off wall into an impermeable stratum; either the stratum does not exist, or its depth is impractical [41–43].

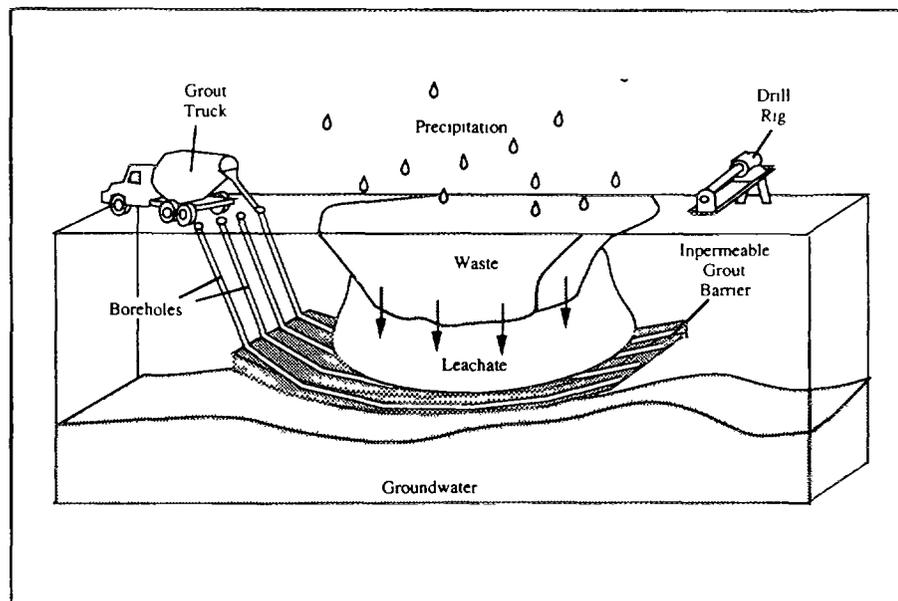


FIG. 23. Subsurface barrier application [43].

Barrier floors are installed using materials and techniques similar to those used in the vertical grout cut-off walls, such as permeation grouting and jet grouting. A floor can be installed as a separate horizontal barrier in the soil or as an angled barrier, shaped like an inverted (and perhaps laterally elongated) pyramid or cone, that serves as a combination floor/wall to limit both lateral and downward migration. In general, horizontal floors have not found widespread use in the area of waste confinement, particularly since the application usually requires drilling directly through the waste site. Alternative methods using directional drilling and boring techniques are being investigated for applying materials in a subsurface horizontal fashion while minimizing the direct encounter with the waste materials.

4.3.1. Operating experience

The application of subsurface horizontal formations for waste confinement is not the primary use of the following technologies. They are discussed here since the technologies are readily available for that application

4.3.1.1 Permeation grouting

Permeation grouting has been used to form a floor in dam construction where the permeability of the bedrock formation is unsuitable. The two main differences between using permeation grouting to install a cut-off wall, as described previously, and to install a floor are that only a narrow band is grouted at the predetermined depth, and the number of rows of injection wells is configured to place a floor in an expansive area as possible

4.3.1.2 Jet grouting

Jet grouting, also known as kerfing, can be used to place a subsurface floor-like structure. The technique is similar to installing grout curtains using jet grouting, except that the grouting takes place at one depth. Installation involves boring a hole to the desired depth and inserting the jet. The jet applies a grout mixture at high pressure to carve out a 1 to 3 m-diameter disk. Many overlapping settings are required to complete the floor. The technique has been commercially developed for mining ore bodies, since the cuttings generated in the process are removed to the surface.

4.3.2. Future developments

Traditional methods of constructing horizontal barriers beneath a waste site require multiple boreholes through the waste site. This can be a major drawback since it results in the generation of contaminated spoils at the soil surface. It can also be a major limitation if there are buildings or underground tanks or other structures with which to contend. Advanced drilling techniques are being investigated as an alternative to vertical boreholes to produce horizontal boreholes in the ground. A number of directional drilling technologies appear to be readily adaptable for the grouting methods described above. These come mainly from the oil and gas industries and the mining and mineral exploration industries. The utilities have also been using similar technologies for years to drill beneath roads, rivers, buildings, etc., to install electrical cables and telephone lines.

There are several other barrier techniques available or being pursued that do not involve emplacement of materials intended to be impermeable. The first involves gravel layers and curtains. These are often installed to divert or otherwise manage the flow of water in the ground.

in and around the waste site. Because the gravel layer or curtain generally has a permeability that is much higher than that of the soils in which it is placed, the water will drain into the gravel area and be directed to the desired location. This technique can be used to lower a water table in which a waste site is located or to direct the ground water flow from passing through the waste site. In this manner, migration of contaminants is reduced and often the conditions are improved for performing other in situ remedial actions.

The second type of non-impermeable barrier is a sorbent barrier. Sorbent barriers are designed to reduce or eliminate the migration of contaminants while allowing for the normal flow of moisture or ground water through the barrier. They are constructed by placing sorbent materials into the soil in order to adsorb or react with contaminants. They can be installed, depending on the chemical adsorbent, similar to the gravel layers and curtains or using a soil mixing technique. In principle, sorbent barriers operate on the same principle as an ion exchange or an activated carbon adsorption column used in the ex situ treatment of waste water. The selection of the adsorbent material will depend on the target contaminant(s). For example, zeolites have high surface area cation exchange properties and are used to remove a number of heavy metals and radionuclides in waste water treatment applications and calcite has been shown to coprecipitate strontium and plutonium with the phosphate ion in the water.

5. TRENDS AND DEVELOPMENTS ON IN SITU IMMOBILIZATION AND ISOLATION TECHNOLOGY

In situ immobilization of radioactive waste at the place of their origin, i.e. at NPPs, is a stable and well developed technology. Different in situ immobilization technologies described in the report are routinely applied at different nuclear fuel cycle facilities. However, more substantial developments on in situ immobilization and isolation in the recent years can be observed in the area of remediation of environmental contaminations. The activity on environmental remediation is growing in recent years and consequently new technological approaches are under development to increase efficiency of remediation work, safety of generated waste isolation, and cost of remediation actions. This relates both to remediation of old radioactive waste disposal sites (which have not appropriate engineered barriers to prevent migration of radionuclides [44]), and to rehabilitation of locally contaminated environment (mainly as a result of the past nuclear activity on different sites [45]).

After several years of testing and application of different treatment techniques for environment restoration and rehabilitation it has been recognized, that in many cases treatment technologies have certain limitations when applying for remediation of waste disposal sites or for cleaning/isolation of contaminated soil [46–50]. Hydrogeological, chemical and waste matrix complexities may far exceed the capabilities of current treatment based remedial technologies. The technology which could in some cases overcome these limitations may be In Situ Vitrification (ISV) technology. Over 200 laboratory tests and experiments plus almost 100 large scale field tests, demonstrations and commercial melts have demonstrated the broad applicability of this technology for a broad range of contaminated soil and waste/debris application, for the destruction/removal of organic contaminants and permanent immobilization of inorganic and radioactive contaminants within a high integrity vitrified product. This technology has successfully applied since 1993 on a broad range of contaminated soil and buried waste and debris applications at US Superfund sites and overseas sites [51]. However, this technology also has its economical and technical constraints.

Limitations of the treatment based remediation technologies have led to increasing usage of and reliance upon subsurface barrier walls and caps to prevent and control contaminants migration from isolated sites. This more common usage of isolation barriers technologies has been supported by considerable advancements in construction technologies. Barrier walls can now be constructed by many different techniques, each offering particular features. Many of these techniques also can attain much greater depth than earlier conventional techniques. New development in barrier wall techniques has been described and analysed in recent publications [52–56]. Different emerging barrier wall technologies are described, addressing their advantages, limitations, document track records, and cost. In particular, the following technologies are analysed and discussed [55]

- deep soil mixing barrier walls,
- jet grouting cutoff walls,
- waterloo sealable sheet piles,
- self-hardening slurries walls,
- permeation grouting,
- ground freezing,
- geomembrane cutoff walls,
- deep barrier walls,
- plastic concrete slurry trenches

Each of the above technologies also may have many permutations. There are many varieties of self-hardening slurries that can be tailored to specific site conditions and design objectives. In particular barrier technology consists of controlling ground water flow under or through a man-made structure. New compositions are under development for preparation of low permeable grouts to provide isolation of contaminants. Cement–bentonite grouts of different compositions are intensively used in many countries for this purpose. However, in many cases permeability of such materials is not satisfactory and investigations are carried out to develop more water proof materials with less permeability. Examples of a new compositions for low permeable grout preparation can be found in recent publications [56–57].

There are also a wide variety of permeable barrier walls under testing and development not to block, but to clean groundwater flow from different contaminants. This subject will be analysed and discussed in another Agency's publication which is now under preparation.

6. CONCLUSIONS

In the cases where it is either uneconomic or technically unfeasible to condition radioactive waste outside a disposal site, in situ immobilization of waste can be a solution. In situ immobilization processes aim at converting radioactive waste, sludge or contaminated soil into a durable leach resistant product which meets the established requirements for waste to be disposed of at certain conditions (in the near surface repositories or in the deep underground structures). These technologies do not generally require that solidified waste be excavated and transported for disposal. In situ solidification and immobilization processes can be potentially applied to on-site immobilization and disposal of radioactive waste, remediation of soil, buried sites, underground storage tanks, etc.

One generic solution does not exist since no single technology can be applied for all sites or even for a single contaminant. The operating experience reviewed in this report shows that in

situ solidification / immobilization in some cases include application of different technologies in sequence.

While assessing these technologies for a potential application, questions important to address are:

- Which technology is best suited for a particular type of waste or contaminants?
- What problems will be encountered in achieving regulatory compliance?
- Is there a need for further development and testing?

Some in situ immobilization technologies are still under development and the effectiveness of their applications for variable waste, combination of different contaminants, etc. is still the subject of further improvement. In many cases in situ immobilization technologies should be considered as an alternative for reducing environmental problems created in the past by not adequate waste management practices.

The performance assessment, safety analyse and environment impact analysis are important components of the practical application of the in situ solidification and immobilization technologies and therefore should be always considered when the decision is made to apply these technologies.

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