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ORP/TAD-76-4

Available Methods of Solidification for Low-Level Radioactive Wastes in the United States

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AVAILABLE METHODS OF SOLIDIFICATION FOR LOW-LEVEL RADIOACTIVE WASTES IN THE UNITED STATES

by

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EPA Review Notice

PREFACE

The mission of the Technology Assessment Division (TAD) is to provide the primary assessments of the technologies currently utilized, or being proposed for use in our society in activities which have a potential radiation impact on man or his environment. TAD has attempted to fulfill its technology assessment mission by addressing the major functional responsibilities, as described in the authorities and responsibilities given by the reorganization plan under which EPA was created. One of the major responsibilities for EPA which directly involves TAD is to render technical assistance to the individual States within EPA's fields of expertise. This report was prepared in order to review the currently available and proposed methods of solidification for low-level radioactive wastes as requested by the National Conference on Radiation Control Program Directors Task Force on Radioactive Waste Management.

Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

David S. Smith
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Abstract

This paper reviews the numerous solidification systems and related matrix materials that are presently being offered, or proposed, to the commercial nuclear power industry. Included, where possible, is the nature of how these materials and/or systems are affected by the physical, chemical, and radiolytic characteristics of the treated radioactive waste materials. Key features of the equipment used in individual solidification processes are discussed in order to clarify the relative utility of these processes for either power plant or fuel cycle application. Finally, a discussion of current problems facing this phase of the nuclear industry is presented.

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AVAILABLE METHODS OF SOLIDIFICATION FOR LOW-LEVEL RADIOACTIVE WASTES IN THE UNITED STATES

I. INTRODUCTION

Radioactive wastes are customarily categorized as low- or high-level (see page 5 for definitions) depending upon the concentrations of radionuclides. However, the long-term hazard associated with each waste is not necessarily proportional to the level of radioactivity, but rather to the specific toxicity and decay rate of each radionuclide.

Due to restrictions placed upon the disposal of wastes at the commercial burial sites and the increasing amount of wastes being generated both in volume and activity by the nuclear industry, considerable interest has been shown during the last several years concerning various methods and systems for the solidfication of liquid and solid radioactive wastes from nuclear power plants and, to a varying degree, from other fuel cycle facilities.

The existing commercial low-level radioactive waste disposal sites in the the U.S. were licensed while implementing the concept of containment of radioactive wastes within site boundaries. The present U.S. method for the disposal of low-level radioactive wastes is by burial in shallow trenches dug in the earth's surface.

There are many varieties of solidification materials and techniques available. Solidification agents include portland cement, concrete, plaster of paris, asphalt (or bitumen), polymers, and a blend of absorbent material and cement or plaster. The method of solidification used should not be a reversible process which can return to the liquid form after placement of the solid in the disposal trench. One benefit of solidification is to remove the liquid and reduce the potential for movement through the soil of the radionuclides incorporated in the solidified waste. A second benefit of solidification of radioactive liquids and sludges is to produce an inert immobilized waste matrix which is safer to handle during transportation and receiving operations at the burial site.

It is the intent of this report to review the various solidification systems and matrix materials that are presently employed or proposed; and to include where possible, how these materials and methods are affected by the physical, chemical, and radiolytic characteristics of the waste materials. Presently, most of the solidification systems used in the United States utilize either cement or an organic polymer, such as urea formaldehyde, as the basic solidification matrix material.

Table I lists the various solidification agents and system vendors. Some of the individual solidification equipment features are included, along with some of the current problems anticipated in the operational phase of the alternate systems. This report is primarily a "state-of-the-art" survey of the various solidification systems and technology. It will not present a statement of systems preference.

The first commercially operated burial site began operation in 1962 and since that time, the industry has expanded to include three private companies operating six sites. The sites are located at Maxey Flats, Kentucky; Beatty, Nevada; Sheffield, Illinois; Barnwell, South Carolina; West Valley, New York; and Richland, Washington. Earlier reports have described these sites in some detail (1, 2, 3).

The burial facilities are managed by private industry, but, by regulation, are located upon Federal- or State-owned land (4). They are, in general, regulated by the State in which they are located, according to the provision of agreements between the individual States and the U.S. Nuclear Regulatory Commission (NRC). The one exception is the site located at Sheffield, Illinois, which is regulated by the NRC.

The increasing emphasis upon the environmental considerations relating to the nuclear industry has focused attention on radioactive waste management operations. Experience in such operations has raised questions concerning the environmental acceptability of current practices and methods used for the disposal of low-level waste.

EPA is responsible for providing assistance to State agencies on environmental radiation-related matters. This responsibility, contained in the Public Health Service Act, was given to EPA under Reorganization Plan No. 3, provides EPA with authority to assist States in their efforts to assess the safety of radioactive waste management activities carried out under State license (5).

The Office of Radiation Programs (ORP) of EPA, at the request of the National Conference on Radiation Control Program Directors' Task Force on Radioactive Waste Management, reviewed the various systems for solidifying wastes and prepared this summary report (6).

Estimation of the rate of leaching from a solidified matrix during burial is one of the important considerations in the assessment of a solidification method as it will strongly influence the amount of treatment, containment, and surveillance that will be needed. Low matrix solubility will improve the safety of waste management by reducing the likelihood of an unplanned release.

Table [illegible]

SOLIDIFICATION SYSTEMS VENDORS

TYPE	VENDOR
I. Cement System	
Portland cement and sodium silicate	United Nuclear Industries
Cement/Vermiculite	ATCOR
Portland Cement Type II	Stock Equipment Company
Portland Cement Type I with Activities	Oak Ridge National Lab a/
Cement with organic polymers	Brookhaven National Lab a/
Cement/vermiculite	Westinghouse Electric Corp.
Cement/shale/clay	Delaware Custom Materials
Cement	Aerojet Energy Conversion Co.
	Chem.-Nuclear Systems
	Hittman Nuclear & Development Corp
	General Electric Company
	Energy Inc.
II. Urea Formaldehyde Systems	United Nuclear Industries
	Chem.-Nuclear Systems
	Hittman Nuclear & Development Corp.
	Protective Packaging Inc.
	ANEFCO, Inc.
	Stock Equipment Company
	Aeroject Energy Conversion Co.
	Energy Inc.
III. Bitumen (Asphalt) Systems	Energy Inc
	Werner and Pfleiderer Corp.
	Aerojet Energy Conversion Co.
IV. Organic Polymer Systems	Food Research and Technical Division
	Dow Chemical Company

a/ Non-commercial applications

This becomes increasingly important when long-term disposal is considered.

Present regulations involving the solidification of reactor-generated wastes are currently not the result of burial considerations, but of transportation considerations. The transport of radioactive wastes is done in compliance with existing Department of Transportation (DOT) and Nuclear Regulatory Commission rules for the safe transport of radioactive materials. This will normally ensure that radioactive waste transportation will not result in an unacceptable radiation hazard to man and the environment.

II. GENERAL CONSIDERATIONS

Wastes from the nuclear fuel cycle and particularly from power plants can be solid, liquid or gaseous with varying chemical, physical, and radiological characteristics. Since most solidification systems process special liquids which produce wet solids such as evaporator concentrates, resins, etc., it is necessary to also categorize or classify those liquid wastes which produce the wet solids. The following terms which are used in this report are defined.

1. Absorb - to immobilize by a method in which the liquid is totally retained by physical means (e.g., by use of such processes as absorption or microcellular capture).

2. Immobilize - to treat the radioactive liquid wastes in such a manner as to eliminate characteristics of fluidity, dispersibility, or freedom of movement within the receptacle.

3. Low-Level Wastes - all those wastes from the fuel cycle other than the transuranium-contaminated and high-level wastes. They usually consist of contaminated paper, cloth, filters, clothing, filter material, demineralizer resins, evaporator concentrates, sludges, activated structural components, etc.

4. High-Level Wastes - (a) high-level liquid waste or (b) the products from solidification of high-level liquid wastes, or (c) irradiated fuel elements, if discarded without processing.

5. High-Level Liquid Waste - the aqueous waste resulting from the operation of the first - cycle extraction system, or equivalent concentrated wastes from subsequent extraction cycle, or equivalent wastes from a process not using solvent extraction, in a facility for processing irradiated reactor fuels.

6. Receptacle - the primary containment receptacle, into which the radioactive liquid waste and the immobilizant are placed for immobilization.

7. Solidified Radioactive Wastes - products resulting from the immobilization or chemical fixation of liquid, semi-liquid or solid radioactive wastes.

8. Transuranium-Contaminated Wastes - any wastes containing significant amounts of long-lived alpha emitters such as plutonium (this number has not been resolved, but is considered by some to be greater then 10 nanocuries per gram).

The radioactive liquid wastes from the power reactors, such as, primary system blowdown, equipment drains, resin sluicing water, evaporator condensates, decontamination solutions, demineralizer regenerative solutions, laundry and laboratory wastes, evaporator concentrates, spent ion exchange resins, filter precoat and cake materials (powdex and solka-floc), cartridge filter units, and diatomaceous earth are suitable for immobilization. These wastes contain the bulk of the volume and radioactivity of the solidified wastes sent to the commercial low-level burial facilities.

The processing of these wastes can be broken down into five steps (7): (a) waste collection; (b) solids pretreatment; (c) the solidification process; (d) mixing and packaging; and (e) final handling (Figure 1). Collection usually takes place in sumps or tanks; the contents are then processed on a batch or semi-continuous basis. The solids pretreatment operation consists of reducing the volume of the wet solids by using an evaporator or other volume reduction device. The solidification and mixing steps involve the use of an agent, such as cement or an organic polymer with additives or a catalyst, to produce an immobilized, monolithic, inert matrix. The container handling operations include inspection to ascertain that solidification took place, capping the container and adding appropriate shielding, decontamination, marking and labeling, container testing, and storage awaiting shipment to a burial facility.

There are several physical, chemical and radiological properties which should be considered as of primary importance in assessing the potential environmental impact from the solidified wastes on the shallow-land burial facilities. Some of these properties include: (a) leachability; (b) thermal conductivity; (c) chemical stability; (d) radiation resistance; (e) mechanical ruggedness; (f) noncorrosiveness of shipping container; (g) total solidified volume, (h) pH of solidified matrix, (i flammability; (j) density; etc.

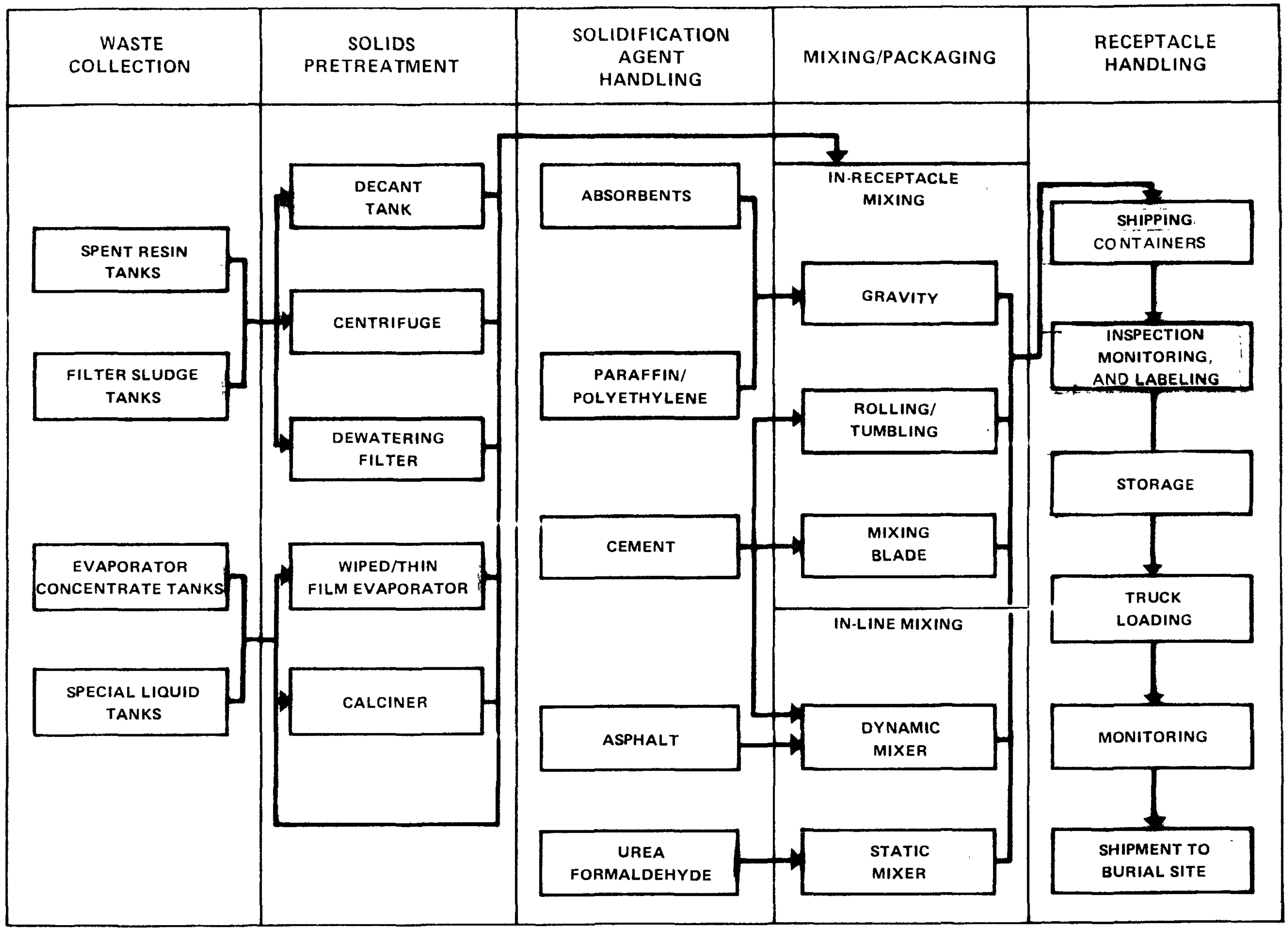

FIGURE 1
WASTE PROCESSING STEPS

III. SOLIDIFICATION OF RADIOACTIVE WASTES IN BITUMEN

The use of bitumen to solidify low-level radioactive wastes has been successfully applied on an industrial scale for many years in different countries (8-21). Bitumen or asphalt, a mixture of high-molecular weight hydrocarbons, is a by-product residue from the petroleum refining processes. Various grades of bitumen are commercially available with a wide range of physical properties. Bitumen processes generally operate in the range of 150 to 230 °C, at which temperature water originally present in the waste can be potentially volatilized.

Basically, all of the bitumen processes consist of mixing the waste solution, slurry, or solids with commercial emulsified asphalt or molten base-asphalt and raising the temperature to evaporate the waste fluid. The solids remain intimately dispersed in the asphalt and the product flows out of an evaporator into a receptacle. The process has been successfully demonstrated in both continuous and batch mixing operations (8, 9, 10). Four different processes have been developed for the bitumen-waste incorporation process: (a) stirred evaporation, (b) film evaporation, (c) the emulsified bitumen process, and (d) the screw extrusion.

Operational Experience

Several versions of the bitumen processes for incorporating radioactive wastes for disposal have been utilized on an industrial scale in Europe (8-15). The first plant scale bitumen process, using the stirred evaporator method was started in 1964 with an evaporation rate of 100 liters per hour at Mol, Belgium (9, 15). The initial operation was directed at the bitumen incorporation of radioactive chemical sludges. Subsequently, concentrated solutions, incinerator ash, vermiculite, ion-exchange material and sand have been processed. Difficulties were experienced with sand and ash owing to the abrasive nature of the material, phase separation occurring due to density differences between the ash and bitumen, and foam formation with organic ion-exchange material. However, these problems were solved using a slow-mixing device, particle separation, and resin incineration (9, 15). Certain oxidizing salts, particularly nitrates, produce an undesirable hardness in the final product; however, this difficulty can be solved by use of reducing agents or different bitumen mixes. Finally, boric acid solutions can be incorporated into bitumen if the solutions are first neutralized to prevent acid volatilization and water leaching of final product. The waste treatment plant at Harwell in the United Kingdom was designed on the same principles as that at Mol and was intended primarily for the incorporation of chemical sludges, but is presently not in routine use (9,11,15).

The emulsion bituminization process for radioactive sludges was pioneered at the Marcoule Centre, France (9,15). This process requires a much higher bitumen content where greater than 1 Ci/m^3 material is processed. Various surface-active agents, (a surface-active agent is a soluble compound that reduces the surface tension of liquids, or reduces interfacial tension between two liquids or a liquid and a solid), variable reaction times, and different bitumen varieties are used depending on the sludge to be treated. Experimental work on this process was also done at the Oak Ridge National Laboratory (ORNL), Tennessee (9). In this process the initial mixing of liquid waste and bitumen could be affected readily at any convenient temperature below the boiling point of the waste solution with the water and/or volatiles removed by heating. The liquid wastes of special interest to ORNL were the evaporator concentrates and solutions of sodium metaborate, nitrate and nitrite. No significant difficulities were experienced in the incorporation of 60 weight per cent (w/o) of solids in bitumen from evaporator concentrates, however, the best final products were produced at solids contents of 45-50 w/o. Boron compounds required higher temperatures, neutralization, and low sodium to boron ratios, especially with tetraborates as the bitumen hardens making stirring impossible. ORNL never established this method for commercial or routine use (9).

Studies were initiated for incorporation of concentrated solutions using a thin-film and screw extruder evaporators; and the screw extruder evaporator concept was developed at the Eurochemic Plant at Mol, Begium and the Karlsruhe Nuclear Research Center in West Germany with apparently good success at both facilities (10,19). The industrial bituminization plant for evaporator concentrates utilizing the screw-extruder-evaporator has been sucessfully operated and commercialized by Werner & Pfleiderer Corporation at an evaporation capacity of 140 kg of water per hour with the final product containing 50% salts (16, 17, 19). ORNL has also investigated the film evaporator process, along with Marcoule, for incorporation of industrial, urban and radioactive wastes in asphalt (8, 9, 15).

Additional research and operational work involving radioactive wastes in bitumen has been done by other countries and nuclear facilities; specifically, the USSR, Bulgaria, Japan, Hungary, and Austria (9,15). The Hungarian Mineral, Oil and Natural Gas Research Institute developed a technique for the incorporation of nitrate-containing wastes. The technique utilizes an inert gas being introduced during the process which eliminates the danger of fires and explosions; the gas also ensures adequate mixing which results in a more satisfactory dispersion of the salts in the bitumen. Workers in India and Japan (9) prefer incorporation of radioactive wastes in bitumen emulsions with the addition of suitable surface active agents. The ease of mixing, low water content in product and reduced leaching rates in sea water are main reasons for this preference. The Soviet Union has a great deal of interest in the solidification of radioactive concentrates by incorporation in bitumens; they have had a pilot plant in operation since 1969 to evaluate the conditions and feasibility of incorporating wastes enriched in sodium nitrate. Also, both the British and Russians have developed methods for immobilizing radioactive wastes attached to netural and synthetic sorbents such as vermiculites, zeolites, clinoptilolite, and ion exchange resins, into a bitumen matrix (9, 15).

The main reported advantages and disadvantages of using bitumen for the insolubilization of radioactive wastes are as follows (9, 12, 15):

Advantages

(a) The leach rate of the final product can be expected to be between 10^2 and 10^3 times lower than similar cement mixes.

(b) There are a large number of types of bitumen with a wide variety of properties; thus it is usually possible to obtain a suitable material for any waste.

(c) Bitumen has good coating characteristics and good adhesion to incorporated material.

(d) The solubility of bitumen in water is negligible.

(e) Bitumen possesses a degree of plasticity and elasticity which are of benefit during the incorporation process.

(f) Bitumen is resistant to attack by microorganisms.

(g) There is some evidence that bitumen is more suitable than cement for wastes which emit emanations.

Disadvantages

(a) There is always an inherent risk in working with organic material at elevated temperatures; however, there is no evidence that incorporation of inert material into bitumen increases the risk of fire or explosion.

(b) There is some evidence that the presences of nitrates or nitrites and other oxidizing agents can increase the fire risk.

(c) It is obvious that no substance should be added to bitumen which decomposes at the working temperature. Difficulties may be experienced with certain plastics and compounds such as sodium citrate.

(d) Heating of bitumen mixes can result in the releases of oils, fumes and mercaptans.

(e) It is necessary to work at high temperatures to obtain efficient mixing.

(f) To obtain the best final product it is necessary to remove as much as possible of the water present in the waste. Leachability increases significantly with increasing amounts of retained water.

(g) Strict temperature control is required in the bituminization process.

(h) Mixing tetraborates and iron and aluminium salts with bitumen causes hardening to an extent which can interfere with discharge of the final product from the equipment.

(i) Irradiation of bitumen modifies the chemical and physical properties. In some cases the effect is negligible and in others considerable. Irradiation up to an integrated dose of 10^{10} rad by an external source or up to 10^{8} rad due to the incorporation of radionuclides can be accepted provided a suitable type of bitumen is chosen.

(j) Phase separation in bitumen mixes is likely to occur more readily than in cement-waste products, particularly during accidental fires in transport or storage.

(k) Experiments at Marcoule and Karlsruhe have shown that swelling of certain bitumen products can occur in water.

IV. SOLIDIFICATION OF RADIOACTIVE WASTES IN CEMENT

The cement solidification process with and without additives has been in common practice at nuclear waste treatment facilities for many years in different countries on an industrial scale (', 12, 13, 14, 20). In 1968, the IAEA concluded that low-level sludge incorporation into cement is considered an adequate treatment (9). As recent as 1974, both bitumen and cement incorporation were considered by IAEA to be acceptable techniques (12).

Cement has been described as an adhesive substance, lime being the principal constituent, capable of uniting fragments of solid matter into a compact monolithic structure. The most commonly used cement for the incorporation of radioactive wastes is the "Portland" variety. It is obtained by intimately mixing silica-, alumina-and ferric oxide-bearing materials to the lime and burning these materials to a very hard brick and grinding the resulting brick. There are various types of portland cement depending on the fineness of the grinding and on the addition of certain additives or the amount of various constituents. Portland cement Type I has been most commonly used, but other Types, such as Type V which is resistant to sulfate salt deterioration, have also been employed (22, 23).

Basically all the cement solidification processes consist of mixing the cement with waste solution, slurry, or solids within the receptable. The actual kinetic process leading to the curing of cement is not known. However, the mechanism for the setting of cement, according to the literature (9, 22, 23), has been postulated to include a reaction between water and cement which formed solid particles and causes crystallization of the calcium hydroaluminate, hydroferrite, and hydrosilicate with the crystals giving the strength to the hardened cement. If the cement product is to be in satisfactory condition for transportation or burial it must have adequate compressive strength. It is common practice to neutralize the acid wastes before the cementation process and to control the salt content. Poorly cured cement will crack and spall, causing more surface area to be exposed to leaching conditions.

The mixing of the cement with the various radwaste forms, i.e., sludges, resin beads, etc., affects the properties of the product. The strength of the cement will be a function of the total salt content in the sludges, resins, etc., where there is a narrow range in the acceptable values for the ratio of basic and acidic oxides in the final product. In this regard, waste to cement ratios recommended for proper curing vary significantly among both foreign and domestic suppliers (see Table II) (9). USSR studies have shown that in order to produce cement of acceptable structural strength the concentration of sodium nitrate salt should preferably not exceed 130 g per kg of cement (9).

Some of the U.S. utilities employ a combination of vermiculite and cement to solidify their radwaste. The expanded vermiculite is porous, permitting the infiltration of dry cement into the vermiculite structure. This would act like a sponge absorbing the liquid and giving a better final product than when cement is used alone (14). Two cement solidification vendors, United Nuclear Industries (UNI) and Delaware Custom Materials, have developed a process which utilizes sodium silicate as an additive with portland cement (24, 25). However, the addition of sodium silicate to cement-waste mixtures increases the volume of waste per volume of solid formed.

In the non-commercial area of solidification ORNL blends their radioactive wastes with a dry mixture of cementitious materials and clays. The dry solids consist of a mixture of portland cement Type I, with a variety of clays including grundite which has a high retention capacity for cesium (14).

The presence of water, nitrates, sulfates, borates, and other unstable (in a radiation environment) compounds in the cement could give rise to gaseous radiolytic products (13). Gases also could result from volitalization of compounds by elevated temperature in the cement-waste mixture causing voids to form within the crystalline structure.

Brookhaven National Laboratory (BNL) found that the leachability properties of cement could be improved by developing a polymer impregnated concrete (PIC) matrix (26, 27). PIC composites containing tritiated aqueous waste, solid calcine, incinerator ash, aqueous and solid sodium nitrate, reactor waste, acidic and neutralized fuel reprocessing wastes, and ion-exchange and sludge materials have been produced. The PIC process utilizes a soak impregnation technique to fill the pores with a styrene monomer, which is then polymerized in situ by heating to 50-70°C (27). In addition, the mechanical properties of these cements are significantly improved as a result of the styrene impregnation. PIC also possesses improved resistance to chemical attack and radiation stability.

Table II

Cementation Practices at Various Establishment (9)

Establishment	Nature of Waste	Composition of Mixture
France	(a) Evaporator (400 g/liter)	250 liters sludge 300 kg cement 40 kg vermiculite
	(b) sludge	83 kg sludge 55 kg cement
F. R. Germany	Evaporator concentrate	100 to 110 liters 150 to 200 kg cement
USA	(a) Evaporator concentrate (20% solids)	Vermiculite (2.7 m^3) and Portland cement (0.68 m^3)
	(b) Neutralized concentrated	(i) 75 liters of concentrate, 128 kg cement, 4 kg vermiculite (ii) 20 to 35 liters/min concentrate, 60 to 65 kg/min of cement
	(c) Evaporator Resins	91 kg of cement 100 liters of waste
	(d) Evaporator Resins	3 to 1 ratio cement to waste with a sodium silicate additive
Czechoslovakia	(a) Sludge with solids content of 20 to 25%	35 liters of sludge 110 kg of cement
	(b) Evaporator concenrate neutralized to pH 6 to 8 (200 g/liter	10 kg of sludge 5 kg of evaporator concentrate 22 kg of cement
USSR	Evaporator concentrates (Max. 150 g/liter)	130 g of salt of the sodium nitrate-type per kg of cement

Operational Experience in the United States

As exhibited in Figure 2, the cement and radwaste could be mixed either within the shipping container or prior to loading the shipping container. For example, ATCOR performs all its mixing with an in-line dynamic or mechanically driven mixer (28). Cement and the liquid radwaste are driven into one end of the mixer and a homogeneous mix is discharged into the shipping container (which has vermiculite added) where in-container solidification occurs. Often the cement will pre-harden causing the mixer to jam.

Batch mixers have also been employed. The components are introduced into a mixer; a mixing blade blends the constituents and the mixture is drained into a receptacle. Earlier plants employed roller and tumbler mixers. When the cement is initially loaded in a drum with a definite mixing weight, a measured quantity of waste is injected into the drum, and the drum is physically rolled and/or tumbled (9, 11, 12).

Another system that can be characterized as an in-drum mixing process has been developed by Stock Equipment Company (S-E Co.) (29). Since transport of fresh cement has historically presented difficulties due to the premature hardening and resultant incomplete curing of the waste, S-E Co has developed a process to overcome this difficulty, by having mixing take place in the final storage drum at the rate of 50 to 200 kg of waste per hour. S-E Co has concluded that the quantity of cement and/or additive in each 208 liter (55 gal) drum averages about 91 kg (200 lbs) and the amount of radwaste averages about 106 liters (28 gal). For this system, cap removal, filling, cap replacement, and mixing is an automatic operation. Therefore, the operator does not have to estimate the correct prescription for solidification.

The UNI and Delaware Custom Material (which uses the Chem-Fix process) systems for solidifying radwaste use an in-line batch mixer for waste and cement which is then mixed with sodium silicate in the shipping container. The UNI and DCM systems provide: (a) proportional pumps for metering waste feed; (b) in-line mixer to assure homogeneity; (c) single fill port for wastes; and (d) in-container solidification (24, 25, 30, 31).

The main reported advantages and disadvantages of using cement for insolubilization are as follows (9, 11, 12, 13):

Advantages

(a) No complex equipment; it is often possible to carry out the incorporation in the disposal receptacle.

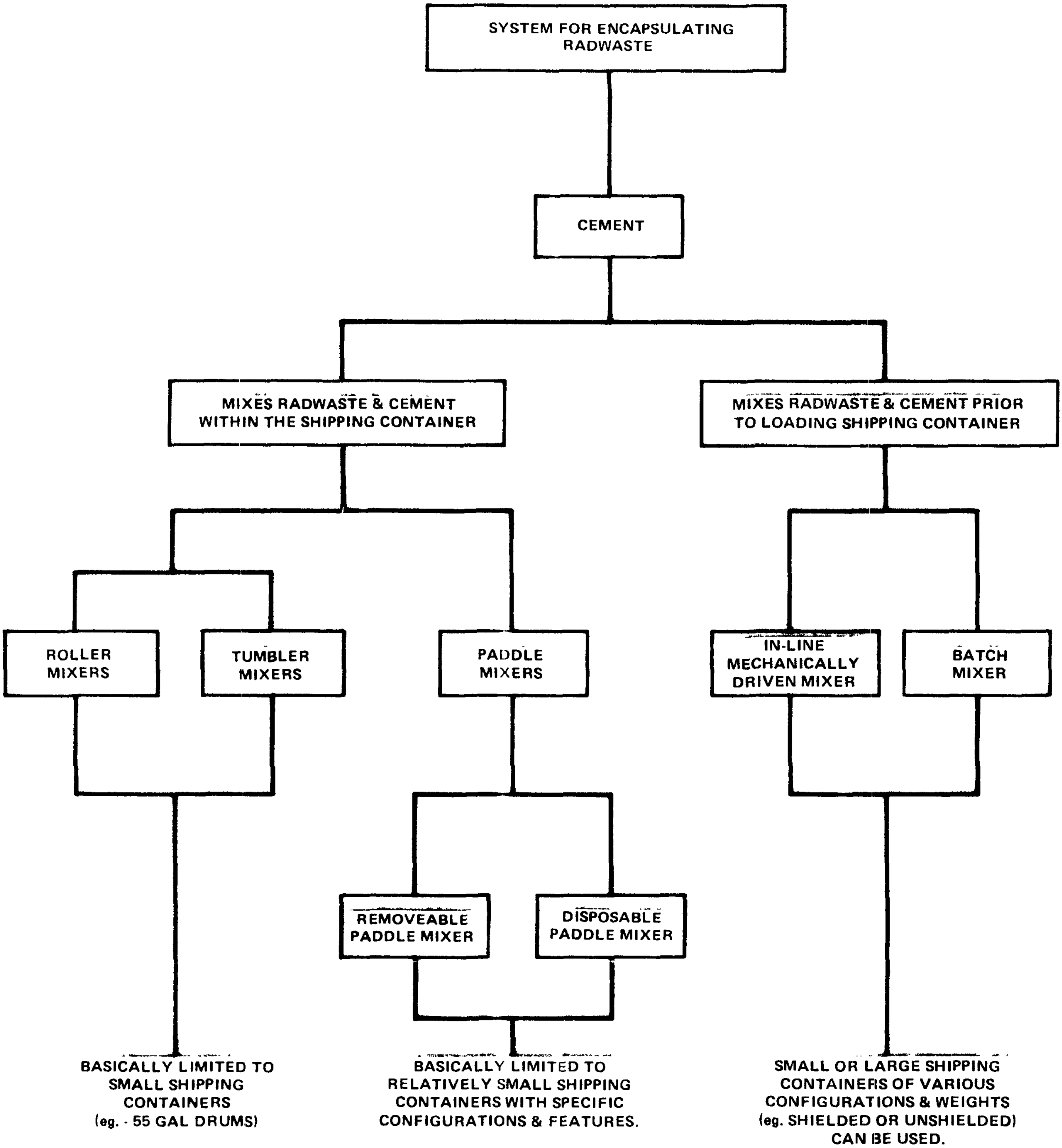

FIGURE 2
FLOW DIAGRAM FOR
CEMENT INCORPORATION PROCESSES

(b) Low capital investment and low running costs; power requirements minimal.

(c) No applied heat required; low operating temperature means no fire risk and eliminates difficulties with off-gas purification.

(d) Most systems fully automatic; and therefore, operators can be trained easily.

(e) Waste-cement mixes are not grossly affected by pH.

(f) Cement is relatively cheap, but this is often off-set by the greater quantity required.

(g) Chemical and physical properties of cement well known.

(h) Cement imparts good shielding properties.

(i) Natural alkalinity of cement is useful in helping to neutralize acidity in waste solutions.

(j) Little reported trouble with phase separation in the mix.

(k) Water is required for setting the mix so there is no need for extensive dewatering provided a satisfactory water/cement ratio is maintained.

(l) Presence of nitrates and nitrites and other oxidizing agents do not have the same detrimental affects as they can have when mixed with an organic material such as bitumen.

(m) Less subject to irradiation damage than bitumen.

Disadvantages

(a) The concentration of certain salts, such as borates, may cause the cement-waste matrix some difficulty in curing and causing deterioration over time thus leaching at an abnormally high rate.

(b) The weight and volume of the final product will normally be about twice that for other corresponding solidification processes. Experience at Fez, Czechoslovakia (a government laboratory), is that 1.5 m^3 of filtered sludge, containing 20 to 25% of solids, would result in a volume of 4.8 m^3 after incorporation into cement. The weight and volume increase is mainly due to the amount of cement which must be added to react with the residual water in the waste.

(c) If mixing equipment experiences operational trouble and frequently breaks down, this could require frequent cleaning of the equipment, particularly the blades.

(d) Nonautomated systems require several manual operations during the solidification process.

(e) Most studies have shown that when buried and after the container rusts away the cement will leach if in contact with ground water.

(f) Cement is relatively cheap, but this is often off-set by the greater quantity required.

(g) Chemical and physical properties of cement well known.

(h) Cement imparts good shielding properties.

(i) Natural alkalinity of cement is useful in helping to neutralize acidity in waste solutions.

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V. ABSORBENTS

Absorbents are used to eliminate free standing liquids by virtue of their ability to hold water molecules within their pores. The absorbent is, however, not chemically bound to the waste nor does it represent a free standing monolithic solid; therefore, the absorbents should not be considered as solidification agents. Further, they do not provide or enhance resistance to leaching, if water comes in contact with the absorbed radioactive materials. The absorbents are stored in a dry environment and are placed in the shipping container prior to adding radioactive liquids. Some commonly used absorbents are vermiculite, clays, silica gel, plaster of paris, microcell and/or diatomaceous earth filter aid (13, 20). The prime use of absorbents is at older plants that do not have installed solidification systems. Not all the burial facilities will accept wastes shipped with an absorbent, because of its unacceptable properties, such as leachability after burial (see Table III).

Vermiculite, dehydrated clay granules, and diatomite absorbents have been routinely used for liquid wastes, with perhaps vermiculite the most widely used. The absorbent method, when properly applied, will physically entrap the waste liquid so that no appreciable free liquid will leak out if the container is breached. With most of the absorbents, the liquids are physically entrapped and can be displaced readily by the addition of water.

In preparation the receptable is filled with vermiculite and liquid waste equivalent to about 1/3 to 1/2 of the volume. For some materials such as the diatomaceous earth, physical mixing of the liquid and absorbent may be necessary. Care must be taken with all absorbent to avoid supersaturation.

Table III

BURIAL REQUIREMENTS AT THE SIX COMMERCIAL BURIAL SITES

Requirements	N.Y. *	KY.	ILL.	NEV.	WASH.	S.C.
A. ACCEPTANCE OF RADWASTE TYPES						
1. DEWATERED RESINS	NO	YES	NO	YES	YES	YES
2. DEWATERED POWDEX	NO	YES	?	YES	YES	NO
3. DIATOMACEOUS EARTH	?	YES	?	?	YES	NO
4. DEWATERED SLUDGES	NO	?	NO	YES	YES	NO
5. FREE LIQUIDS	NO	NO	NO	YES	YES	YES
B. SOLIDIFICATION AGENTS						
1. ALL TYPES OF CEMENT	NO	YES	YES	YES	YES	YES
2. UF SYSTEMS	NO	?	YES	YES	YES	YES
3. OTHER ORG. POLYMERS	NO	?	?	?	?	NO
4. ASPHALT	NO	?	?	?	?	?
C. SELECTED REQUIREMENTS						
1. RETRIEVABILITY	NO	NO	NO	NO	NO	YES
2. Pu LIMITATIONS	YES	YES	YES	YES	NO	YES

* PRIOR TO ITS CLOSING ON 3/19/75.

? UNCERTAIN ABOUT ACCEPTANCE

VI. POLYMERIC SOLIDIFICATION PROCESSES

Incorporation of radioactive wastes into polymeric fixation agents is a relatively new solidification process when compared to incorporation in cement or bitumen (13, 14). The solidification process can take place either at ambient temperatures or with hot evaporator concentrates (up to 60°C). Presently, several U.S. companies sell urea-formaldehyde (UF) solidification systems (as shown in Table I). All the organic processes are essentially batch processes where a catalyst is generally mixed with the wastes and polymer either in a premixer vessel or in the receptacle itself (14, 32, 33). The polymeric processes do not really solidify the wastes; the long chain molecules of the organic polymer are linked together to form a multi-voided sponge that "traps" the waste. Not all U.S. burial sites at this time, however, will accept radwaste solidified with an organic polymer (see Table III).

Paraffin and polyethylene based solidification agents can also be used to solidify wastes. These agents must be liquified by heating prior to mixing with the wastes (20, 21, 34).

The only industrial experience with polymeric solidification systems to date has been with the UF process. The process description, advantages and disadvantages are based on systems using UF, whereas there are other organic polymer processes, such as the Dow Chemical and the Todd Research processes (35, 36), which are either not operational or have not been in operation long enough to provide operational information comparisons.

Urea Formaldehyde

The physical method of organic polymeric mixing depends upon the type of solidification agent and receptacle used. In general, there are three types of UF mixing:

1. In-container disposable paddle mixer
2. In-line static mixer
3. In-line mechanically driven mixer

The in-container mixer is generally used for cement (see Chapter IV) but is also used for mixing resin beads with UF. The in-line dynamic mixer is used by UNI to mix liquid waste and UF prior to discharging into a receptacle. UF systems generally employ an in-line static mixer which contains stationary helical vanes to mix the fluids as they flow through the mixer. Just as the mixed polymer and waste are injected into a container, the acidic catalyst is added to initiate solidification. Figure 3 is a flow diagram for UF indicating the incorporation steps.

The UF solidification process system developed by Protective Packaging, Inc., (PPI) is called Tiger Lock. Tiger Lock is a registered trade mark for a proprietary augmented agent of urea formaldehyde resin that is manufactured to strict physical and chemical specifications. The PPI system currently being sold includes Tiger Lock and catalyst (usually sodium bisulphate-$Na_2 SO_4$), associated processing equipment, and container liners for use in the transportation and burial of the solidified radwaste.

PPI suggests that the desired ratio of Tiger Lock to radwaste is 2:1 by volume. For this system the operator has the task of estimating the correct amount of catalyst for solidification which is highly dependent upon the quantity and type of radwaste that would be solidified by the PPI system.

Gel time of the product can be adjusted from minutes to hours by the catalyst concentration (normally about 1 to 3%). The UF catalyst is used at a pH of 3 (32). If UF is used after its shelf life has been exceeded or at low temperatures or low viscosity, the "cottage cheese" effect will occur, i.e., little solidifying and essentially a settling of materials of different density within the container.

The main reported advantages and disadvantages of using urea-formaldehyde for insolubilization are as follows (13, 14):

Advantages

(a) The amount of waste capable of incorporation in a receptacle with UF is about 30% by volume more than with cement.

(b) For shipping not requiring radiation shields, shipping cost with UF or polymerics is less than for cement and bitumen due to the ability to put more waste in a given container and a lower density for the mixture.

(c) Mixture of UF and radwaste are not combustible. Further, no detectable exothermic reaction occurs with UF.

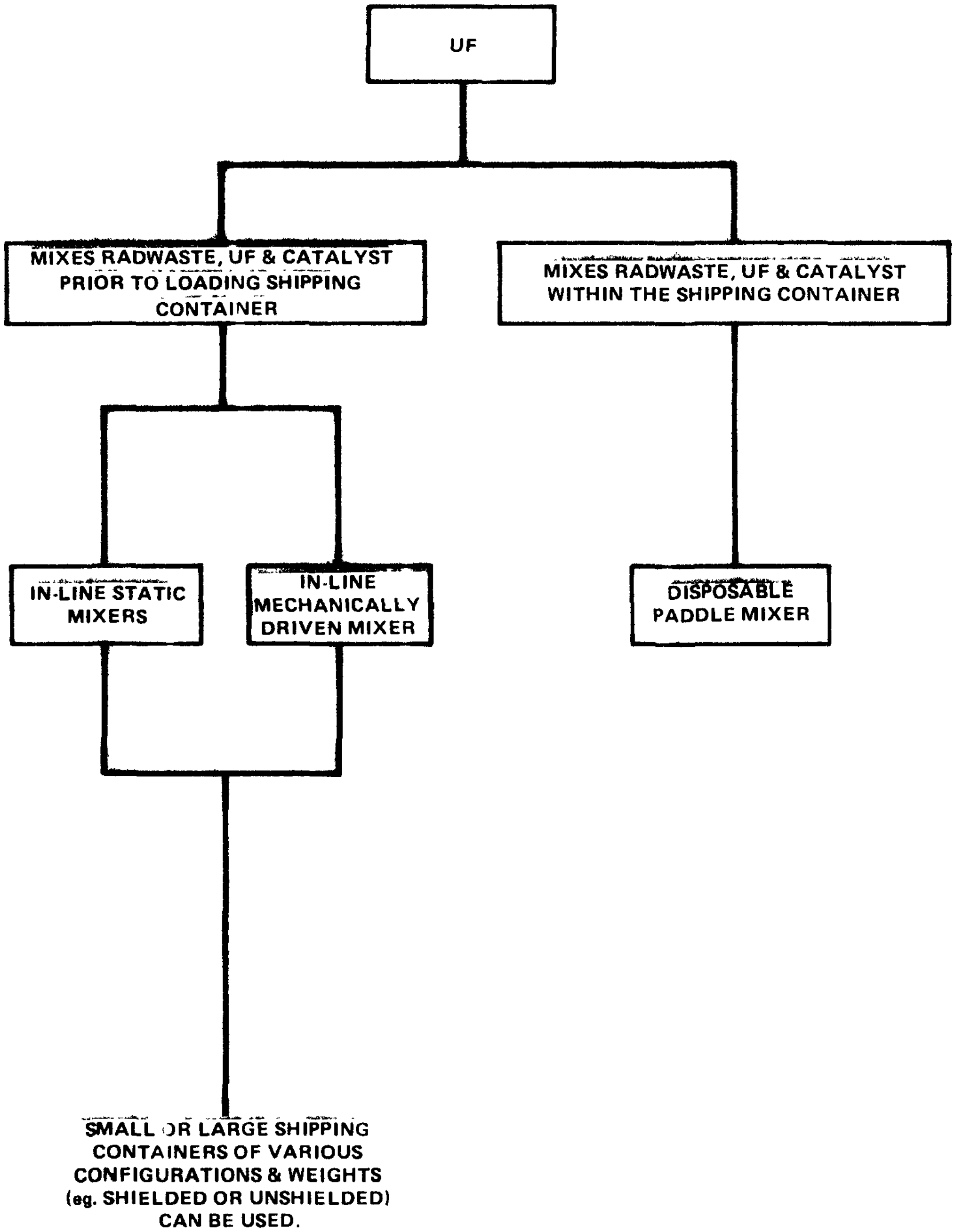

FIGURE 3
FLOW DIAGRAM FOR UF
INCORPORATION PROCESSES

Disadvantages

(a) For shipments requiring radiation shields, UF or polymeric solidified materials, due to its lower bulk density and higher activity, requires more shielding than materials solidified with cement.

(b) It appears that routine attainment of the complete elimination of free standing water is a problem with encapsulated UF radwaste, particularly those having lower concentrations of the polymer (ratios of 1 to 3 or less) (37).

(c) According to the utility operators, it is difficult to work with UF because of the relatively low viscosity of the mixture, which permits settling or floating (segregation) of materials of different densities.

(d) Solidification time is affected by both the pH of the mixture, which is regulated by the amount of catalyst, and the temperature of the mixture. Operating experience at the utilities indicates that the optimum conditions for solidifying are 29°C (85 °F) and a pH of 3 (37, 38).

(e) The UF shelf life is limited and is dependent upon storage conditions.

(f) Equipment must be designed to eliminate fume problems with UF; the odor is disagreeable even in small concentrations.

(g) Some manufacturers of UF have stated that this product is biodegradable; also the catalyst is corrosive to most metals.

(h) During the solidification process when the UF-radwaste mixture is exposed to air, water vapor evaporates from the mass, but if the matrix remains in an air-tight container, the mixture will remain semi-liquid.

Polyethylene Process

Polyethylene agents are not used commercially in the U.S. Polyethylene is a superior solidifying agent for most organic liquid wastes. The waste is combined with molten polyethylene inside a heated chamber in which the water and other volatile constituents are evaporated. The mixed and dehydrated liquid product is discharged to a container where it solidifies upon cooling. The final product is a solid plaster block, which is relatively inert at room temperature and is insoluble in water. It has good freeze-thaw characteristics and a storage life of several years. Polyethylene is completely combustible and can be incinerated. It is flammable with a flash point of 250°C (20, 21, 34).

VII. LISTING OF WASTE SOLIDIFICATION AND PACKAGING SYSTEMS SUPPLIERS

The previous sections discussed the various solidification methods while briefly mentioning some of the companies offering the process. This section is intended to provide a listing of those companies which supply partial or complete waste solidification and packaging systems. These systems may or may not include options for using either cement or polymerics and in many cases these systems do not have actual on-line processing experience in the United States nuclear industry. The coverage of each company in this listing varies in length and detail, but in most cases is representative of their process information brochures. The reason for this is the lack of actual use of the systems, and in many cases the process ingredients are proprietary.

1. Werner & Pfleiderer Corporation (10, 16, 17, 19, 28, 58)

Werner & Pfleiderer Corporation, (WPC) offers a waste solidification system that includes a volume reduction system. With asphalt as the solidification agent, WPC indicates the end product is stable, particularly against leaching. Although it has only been recently introduced in North and South America, it has been operating routinely in Europe since 1965. The WPC radwaste solidification process yields a liquid-free (0.5%) solid using a continuous, fully automatic process with a multi-screw compounding extruder. The extruder-evaporator simultaneously provides homogenous mixing (including reagent additives), liquid evaporation and solidification in one machine. The extruder evaporator normally discharges the asphalt/salts mix into standard DOT 208 liter (55 gal) drums at a rate from 1 to 114 liters (1/4 to 30 gal) per hour, depending on the the size and speed of the extruder and the concentration of the feed stream. The entire process, complete with interlocks, can be controlled remotely.

The WPC extruders, originally developed for the plastics industry, are designed and built to operate a full year without maintenance. The operating record established in Europe bears this out and is quite impressive (over 134,000 hours operation at Marcoule without mechanical failure).

2. ATCOR, Inc. (28)

ATCOR has developed a process system to mix liquid and solid wastes with a cement or a cement and vermiculite mixture to produce a solidified product within a disposable receptacle. Basically, the system mixes separate feeds of moist radioactive waste or evaporator concentrates and dry cement in a small volume continuous mixer. Solid waste materials are preconditioned within the radioactive waste feed tank to provide sufficient moisture when mixed with the dry cement to achieve an acceptable cement mixture. The system not only solidifies resins, sludges and evaporator concentrates, but it can also be used to fix spent filter cartridges within a solidified matrix. In this case drums or large volume liners containing spent filter cartridges could be filled with a cement mixture that contains radioactive wastes.

The cemented waste mixture can be loaded directly into standard 208 liter (55 gal) drums or larger receptacle. Where waste is to be packaged in drums, drum capping and decontamination can also be provided. There is no preparation of drums required prior to filling.

3. Stock Equipment Company (28, 29)

The Stock Equipment Company (S-E-Co.) Solid Radwaste System is designed and manufactured as a completely integrated system utilizing components which are designed specifically for the service expected rather than attemping to modify standard equipment. The S-E-Co. System is furnished complete for placement into the radwaste building and interfacing with liquid system piping, utilities, etc.

The S-E-Co. design uses cement as a solidification agent and packages the solid radwaste into standard DOT, 208 liter (55 gal) drums. The S-E-Co. system is also easily adaptable to other types of solidification agents such as urea formaldehyde. The S-E-Co. waste system consists of: a cement storage hopper; a storage tank to hold liquid wastes that contain concentrated solutions of dissolved solids; a decant tank for filter media, resins, and/or the solid waste slurry from the storage tanks; a drum processing unit which is fully automatic for uncapping the drums, filling the drums with cement, filling from the decant tank, reinsertion of the cap, and for the mixing/tumbling operation.

4. United Nuclear Industries, Inc. (24, 28, 30, 31)

United Nuclear Industries (UNI) offers radwaste solidification systems utilizing as the solidification agent either urea formaldehyde (UF) or Portland cement with sodium silicate as an additive.

The use of the UF material permits the use of in-line static mixers with no moving parts. Solidification of the waste - UF mixture is accomplished using either a sodium bisulfate catalyst (pH range of 3 to 7.5) or a phosphoric acid catalyst (pH range of 3 to 10).

5. Aerojet Energy Conversion Company (28, 39-43)

The Aerojet Energy Conversion Company (AECC) has marketed a VR-20 Radioactive Waste Management System which reduces the volume and encapsulates the waste. The volume reduction is achieved by conversion of all liquid wastes into anhydrous calcined solids and drying of dewatered spent resins and sludges. These solid wastes can then be encapsulated in cement, U-F or bitumen as the solidification agent for subsequent shipment and burial. Using the VR-20 process, the volume of liquid waste could be reduced by a factor of 10-20, while the volume of liquid spent resins and sludges could be reduced by a factor of 2-4 when compared with other solidification methods.

The main feature of this system is a fluidized bed calciner which receives the radioactive liquid waste feed containing the dissolved chemical solids and processes these aqueous solutions into free-flowing anhydrous particles. Concentrated radioactive liquid waste (evaporator bottoms, etc.) is pumped from the concentrated liquid waste storage tank to a heated fluidized bed calciner concentrator. The volatiles exist with the water vapor at the top of the fluidized bed concentrator, leaving behind the dissolved solids. The granular solid produced can then be encapsulated.

6. Chem-Nuclear Systems (28, 44)

Chem-Nuclear Systems Inc., offers either portions of or a complete waste system design, component selection, procurement, fabrication, construction, installation and operation of solidification systems using either cement or urea formaldehyde as the solidification agent. Chem-Nuclear also has available a mobile solidification unit using the UF system.

7. Protective Packaging, Inc. (28, 32, 33)

Protective Packaging, Inc. (PPI), a wholly owned subsidiary of Nuclear Engineering Co., developed and was the first company to design and sell, a system using a chemical solidification agent other than Portland Cement. Since then, they have filed several patent applications on the system and trademarked the name "TIGER-LOCK". Their patent applications cover both the use of the liquid solidification agent (TIGER-LOCK, a type of urea formaldehyde polymer), and also all the related hardware that makes up a TIGER-LOCK Radwaste Solidification System. This includes the process equipment, control panel, power panel, and associated material handling equipment. The key aspects of the PPI design are: (a) three separate tanks, pumps, different size liners for radwaste, TIGER LOCK, and catalyst respectively; (b) a premixer for the radwaste and UF to homogenize the slurry prior to contact with the catalyst; (c) a manual decoupling device to seal the liners that contain the cured waste; and (d) an automatic level detector to indicate filling to 90% volume.

8. ANEFCO, Inc. (28)

ANEFCO, Inc., offers a waste solidification system using urea formaldehyde and sulfuric acid, or an equivalent catalyst, as a solidifying agent. Their process system uses a 3785 liter (1000 gal) batch tank, a static mixer and a disposable polyethylene liner in the disposal container.

9. Hittman Nuclear & Development Corporation (28, 45)

Hittman Nuclear & Development Corporation (HNDC) offers radioactive waste solidification systems using cement or a polymer such as urea formaldehyde as the solidifying agent. Chemical additives are used with both agents to enhance the efficiency, i.e., volume of waste per unit volume of solidified product. The disposable containers used to package radwaste vary in size from a standard 208 liter (55 gal) drum up to 5.7 m^3 (200 ft^3) capacity.

10. General Electric Company (28, 46)

The present General Electric Solid Radwaste Systems use cement as the solidification agent with a disposable mixer and large disposal containers.

11. Westinghouse Electric Corp. (47)

The Westinghouse Waste Encapsulation System is basically a vacuum packaging process in which spent radioactive resins and waste evaporator bottoms are encapsulated using a cement vermiculite mixture in standard DOT 17 H drums.

12. Delaware Custom Materials (25)

The Delaware Custom Materials company utilizes the Chemfix process which offers a complete service of equipment and chemicals for solidification of radioactive wastes, including a variety of inorganic and organic sludges. The process uses a combination of cements, shales and clay as the solidification agent.

13. Dow Chemical Company (35)

Dow Chemical Company has developed a radwaste solidification system that produces radwaste free of liquid, reasonably hard, and free standing. The solidification system is usable for all anticipated chemical decontamination solvents and regular wastes from nuclear power stations.

To date, Dow has solidified the following simulated wastes in the laboratory and in 208 liter (55 gal) drums, containing no detectable free liquid: (a) spent decontamination solutions at pH's of 3 to 5 and 9 to 10 with 40% solids; (b) filter aid and slurries, 90/10 by volume; (c) ion exhange resins, 90/10 by volume; (d) PWR evaporator bottoms with a pH of 2.5 and 7% solids; and (e) BWR evaporator bottoms with a pH of 10.6 and 6% solids. After casting, the drums solidified within one hour. The radwaste to agent ratio is at least 1.25 to 1 and as high as 2.5 to 1. A field demonstration was carried out by successfully solidifying 3400 liter (900 gal) of radioactive decontamination solvent at a nuclear power plant.

To simulate disposal conditions, Dow evaluated the solidification product for the following: (a) compressive strength; (b) temperature cycling; (c) radiation stability; (d) leachability; (e) impact testing; (f) heat exposure; and (g) free liquid.

14. Todd Research and Technical Division (36)

Todd Research and Technical Division is marketing a solidification agent called SAFE-T-SET, which is a long chain linkage organic polymer. The agent can be used with concentrated low-level liquid radioactive wastes from filtration, precipitation, ion-exchange or evaporation. The set-up time varies from one minute to several hours depending on the amount used in proportion to the volume of waste. One-half kilogram (1 lb) of SAFE-T-SET will solidify 3.8 liters (1 gal) of liquid material. The agent can be tailored to any particular system or circumstances including pumping the waste and SAFE-T-SET mixture and can be adapted to molds of any type. The solidified matrix remains stable under conditions of freezing, high temperature and leaching.

15. United Technologies (48)

United Technologies - Chemical Systems Divison offers the Inert Carrier Process which handles ingredients in an inert liquid. The operating concept is based on dispersal of the reactants in an inert carrier to provide maximum surface area for solid - liquid reaction mechanism. In addition the process provides for a clean separation of the reaction product from the inert carrier. The waste materials are low viscosity dispersions in an inert carrier. The inert carrier is a fluid selected so that neither the starting materials nor the products are soluble in it or chemically reactive with it. The process has particular advantages in operations which require (a) preparation of compositions which are too viscous to mix by ordinary methods; (b) extremely intimate mixing of solids with small quantities of liquids; (c) safe control of highly exothermic chemical reactions; or (d) a closed system and/or remote controlled processing of hazardous, toxic, or explosive materials.

16. Energy Incorporated (49)

Energy Incorporated and Newport News Industrial Corporation has developed a Radioactive Waste Reduction (RWR) system to convert all low and medium level liquid and solid combustible radioactive wastes to solids by a fluidized bed calcining process. The system produces a granular, anhydrous solid which may be placed directly in burial containers or incorporated into matrices such as concrete, urea formaldehyde or bitumen for burial.

VIII. CONCLUSIONS AND RECOMMENDATIONS

In the foregoing sections brief descriptions have been presented of several established and proposed processes for the solidification of low-level radioactive wastes. Each of the processes as well as each of the solidified waste products (cement, bitumen, UF, etc.) have a number of advantages and disadvantages. Table I listed the solidification agents and/or systems with each supplier. In general, the following conclusions can be drawn concerning the three major systems:

- Bitumen - some question concerning thermal stability, particularly above ambient temperature, but possesses good mechanical ruggedness and radiation resistance.

- Cement - good thermal stability, mechanical ruggedness, and radiation resistance but questionable ability to properly fix certain salts with the sludges or resins; also some system operation problems.

- Urea Formaldehyde - questionable radiation resistance, thermal stability, and biodegradability properties but the best waste to agent ratio; also some process operation problems.

- Other Organic Polymers - not enough actual on-line experience to provide an adequate conclusion.

The most likely mechanism of radionuclide release to the surroundings is by solution in the water existing in the environs of the burial site. Therefore, measurements are usually attempted to indicate the rate at which radionuclides are leached from the solidified products. The leachability properties of a radwaste solidification matrix will strongly influence the amount of treatment, containment and surveillance that will be required.

A review of the various leach rate tests conducted by industry, (50-57) indicates that the results were not developed under standardized laboratory conditions and were not correlated to simulate current burial conditions at the various commercial shallow-land sites. Studies currently underway which are investigating the properties of solidified wastes include: (a) a study by Brookhaven National Laboratory (under contract with NRC) to evaluate various solidified wastes generated by commercial nuclear power plants and establish a standardized leach test for solidified matrixes (57); (b) a study by the Army Corps of Engineers (under contract to EPA-SWRL-Cinn) to evaluate solidification agents for solidifying hazardous waste sludges (52); and (c) a study of leach rates for various solidified wastes by the Oak Ridge National Laboratory (50, 51, 54).

The studies are intended to provide information on the leach resistance of various solidified waste products. Leach rates for alkali and alkaline earth, rare earth, and actinide elements from various waste matrices are compared in Table IV (20). A comparison shows: (a) cement has wide ranging leach rates; (b) calcines are extremely leachable; that for a given waste matrix the leach rates for rare-earth and actinide elements are about a factor of 1,000 less than those for alkali and alkaline earth elements; and (d) the leach rates for rare-earth and actinide elements from cements and grouts are as low as those from glasses. Additional leachability work needs to be accomplished in the near future to permit a more complete understanding of the environmental impact of solidified radioactive waste.

Table III indicated some of the burial requirements at the six commercial sites. The requirements have been arranged into three categories: (a) the type of radwaste shipped by the utilities that are being accepted; (b) the solidification systems that are being accepted; and (c) other requirements. The question marks indicate uncertainties the burial sites have concerning certain requirements. Several key aspects which can be pointed out are: (a) there are inconsistencies concerning what each burial site will accept; and (b) there is a lack of information concerning the merits of the various solidification agents, so that the licensing agencies can pass judgement on what matrix media would be acceptable for burial.

The quality assurance requirements for the various solidification processes should also be improved at the nuclear plants. It is important that each waste shipping container have a consistent composition of solidified matrix. This condition would improve the reliability of the leachability measurements.

In conclusion, this summary has drawn together in a list the various vendors of solidification processes and systems. There is definitely a need for specific standards for leach testing the solidified matrices and developing standardized burial requirements for the sites

Table IV

COMPARISONS OF LEACH RATES FOR VARIOUS SOLIDIFIED WASTE PRODUCTS (20)

LEACH RATES GRAMS/CM^2-DAY

	Alkali and Alkaline-Earth	Rare Earth	Actinide
Calcines	10^{-1}-1	10^{-4}-10^{-3}	--
Ceramics			
Phosphate	10^{-5}-10^{-2}	10^{-9}-10^{-6}	--
Devitrified Phosphate glass	10^{-4}-10^{-2}	--	--
Glasses			
Borosilicate	10^{-7}-10^{-5}	10^{-9}-10^{-7}	--
Phosphate	10^{-8}-10^{-5}	10^{-9}-10^{-6}	--
Aluminosilicate	10^{-8}-10^{-7}	--	--
Bitumens	10^{-7}-10^{-4}	--	10^{-8}-10^{-7}
Cements	10^{-6}-10^{-1}	--	10^{-9}-10^{-7}
Grouts	10^{-7}-10^{-4}	--	10^{-7}

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