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(54) **Process for the Encapsulation of  
Radioactive Wastes**

(57) Radioactive waste material, particularly radioactive ion exchange resin in the wet condition, are encapsulated in a polyurethane by dispersing the waste in an aqueous emulsion of an organic polyol, a polyisocyanate and an hydraulic

cement and allowing the emulsion to set to form a monolithic block. If desired the emulsion may also contain additional filler e.g. sand or aggregate to increase the density of the final product. Preferred polyurethanes are those made from a polyester polyol and an organic diisocyanate, particularly hexamethylene diisocyanate.

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## SPECIFICATION

**Process for the Encapsulation of Radioactive Wastes**

This invention relates to the encapsulation of radioactive waste for transportation, storage or disposal.

5 Various proposals have been made for the encapsulation of radioactive waste into polymeric resins prior to transportation, storage or disposal. In U.K. Patent Specifications Nos. 1,353,121 and 1,418,277, for example, radioactive wastes, which have been converted to a dry powder are incorporated into an unsaturated resin, for example, an unsaturated polyester resin, which is cross-linkable at room temperature by copolymerisation with a cross-linking monomer, e.g. styrene. Such a system is, however, disadvantageous when it comes to handling liquid or wet radioactive wastes which first have to be dried before encapsulation. 10

In U.K. Patent Specification No. 1,433,419, radioactively contaminated phosphoric acid esters, used as extractants in nuclear fuel reprocessing, are formed into solid blocks by admixture with polyvinyl chloride chips, the mixture being allowed to fuse into a solid block at room temperature or with heating. 15

Radioactive waste ion exchange material may also be incorporated into the mixture with the phosphoric acid ester. In U.K. Patent Specification No. 1,478,628, solid wastes are treated in a similar way.

In U.K. Patent Specification No. 1,460,379 radioactive organic liquid wastes are encapsulated by first forming a homogeneous mixture with an organic binder, e.g. bitumen, or a plastics material such as polyvinylchloride or polyethylene, and distributing the mixture into an aqueous hydraulic cement mixture which sets to form a solid block. 20

In U.K. Patent Specification No. 1,482,780, liquid radioactive wastes are first of all evaporated to reduce the waste to a mass substantially anhydrous particles which are then mixed with a binder, e.g. a mixture of polyethylene and paraffin wax, or an unsaturated polyester resin/styrene mixture, and which sets to form a monolithic structure embedding the radioactive waste particles. 25

In U.K. Patent Specification No. 1,483,080, organic liquid wastes are encapsulated by admixture with a monomer charge containing both mono and polyvinyl monomers which are polymerised *in situ* to a solid polymer mass.

U.K. Patent Specifications Nos. 1,414,073 and 1,510,494 relate to methods of dispersing dried radioactive wastes in organic binders of a lower specific gravity than the waste, in particular bitumen. 30

Finally, U.K. Patent Specification No. 1,547,952 discloses a process in which a wet radioactive waste material is kneaded at elevated temperature with a thermoplastic binder, e.g. polyethylene, and drying the mixture by evaporation.

35 A proposal for the encapsulation of wet radioactive wastes, e.g. ion exchange resins and solids resulting from the filtration and/or flocculation of radioactive material, which does not require the elimination of the water content of the waste is disclosed in U.K. Patent Specification No. 1,479,150. According to this proposal the wet waste, in the form of a sludge is treated with a polymerisable resin, e.g. an unsaturated polyester resin or an epoxy resin, which is polymerised *in situ* to form a solid block containing the radioactive waste. 40

Such methods of handling water wet wastes are generally energy intensive, requiring as they do evaporation of the moisture content of the waste either before or during encapsulation.

In accordance with the present invention, a method of encapsulating radioactive waste material, particularly, but not exclusively, water wet wastes such as radioactively contaminated ion exchange resins, is provided which avoids energy-intensive drying of the waste, minimizes the number of handling operations required on the waste and avoids energy-intensive mixing techniques for mixing the radioactive waste with the encapsulating material. 45

In accordance with this invention solid radioactive waste material is encapsulated by dispersing the waste in an aqueous emulsion containing an organic polyol, a polyisocyanate and an hydraulic cement and allowing the emulsion to set, thereby to form a monolithic block comprising said waste material embedded in a cement-polyurethane matrix. If desired to increase the density of the monolithic block, additional filler, e.g. sand or aggregate, may be incorporated into the emulsion. Also, to assist dispersion of the solids therein, there may be added to the emulsion one or more wetting agents, e.g. a ligno sulphonate. 50

55 In the process of the present invention, the use of an aqueous emulsion as the encapsulating material provides ease of mixing for the radioactive waste which can be readily dispersed therein by stirring.

A variety of polyurethane forming reactant can be used, but preferred are polyester-polyols as the polyol component, and an organic diisocyanate, e.g. toluene diisocyanate or hexamethylene diisocyanate as the isocyanate reactant. A particularly suitable material is that sold under the trade name Ucrete which comprises an aqueous polyester-polyol emulsion as the first component, hexamethylene diisocyanate and a cement aggregate filler consisting of cement and crushed rock aggregate. 60

The aqueous emulsion used in the encapsulation process of the present invention may typically contain, percentages by weight:

5	aqueous organic polyol emulsion (15—40% by weight of water)	10—40%, preferably 25%	5
	organic diisocyanate	10—40%, preferably 25%	
	cement, or cement plus additional filler, if present	20—80%, preferably 50%	

Other optional ingredients which may be incorporated into the emulsion include wetting agents 0.1 to 5%, alkaline pH adjusters, e.g. calcium hydroxide 1 to 3% and fire retardants.

10 The hydraulic cement used in the process of this invention is preferably a Portland cement or Portland cement-aggregate, and serves to take up CO<sub>2</sub> liberated during the urethane reaction, thereby reducing the porosity of the final product and increasing its density. In general, the monolithic blocks of encapsulated radioactive waste produced in accordance with the invention will have densities in excess of 1.2 and therefore suitable for disposal at sea.

15 The amount of radioactive waste which may be encapsulated by the process of the invention may be up to 30% (dry weight) based on the weight of the emulsion and may contain up to 50% by weight of water. The radioactive waste is dispersed into the emulsion by stirring, for example, in a hydraulic torque mixer.

20 The monolithic blocks of polyurethane encapsulated radioactive waste produced in accordance with this invention show remarkable dimensional stability with no measurable expansion during setting.

25 The process of the invention is particularly applicable to the encapsulation of wet, radioactive ion exchange resins, but may also be applied to the encapsulation of other radioactive waste material, primarily in granular or sludge form, but also radio-actively contaminated articles such as ladles and other items of equipment either intact or broken up into pieces.

The process of this invention is illustrated by the following example.

#### Example

A viscous aqueous emulsion was prepared having the following composition:

30	aqueous polyester-polyol emulsion (% water)	39.0 kg	30
	hexamethylene diisocyanate (MDI)	34.3 kg	
	sand	26.5 kg	
	Portland cement	47.5 kg	
35	calcium hydroxide	5.2 kg	35
	sodium lignosulphonate	1.05 kg	
	Total	<u>153.55 kg</u>	

40 The emulsion was prepared by adding the MDI to the aqueous polyester-polyol emulsion in a hydraulic torque mixer and dispenser and mixing for 4 minutes. The filler components were then added to the mixer and mixing continued for a further 3 minutes, followed by the addition of the pH adjuster and wetting agent. A water wet, waste radioactive ion exchange material (107.8 kg) containing approximately 50% by weight of water was then added to the mixer, thereby providing an amount of about 35% dry weight of ion exchange resin, based on the total weight of the emulsion. Mixing was continued for a further short period of 4—5 minutes after which the mixture was cast into a drum as a single monolithic block. Setting took place over a period of 6 hours. The whole mixing operation was performed at room temperature but due to the exothermic reaction, the temperature of the reaction mix rose to 40°C at the time of casting with the peak exotherm (74°C) occurring 3.5 hours after casting. No visible change of dimension, i.e. expansion or contraction, took place in the material cast in the drum during the setting process. The cured block showed a compressive strength of about 20 MN/m<sup>2</sup> after curing for 6 hours. The resultant monolithic block was non-flammable, contained little or no free water, was substantially void free and had a specific gravity in excess of 1.2.

#### Claims

55 1. A method of encapsulating radioactive waste material, which comprises dispersing the waste in solid form in an aqueous emulsion of an organic polyol, a polyisocyanate and an hydraulic cement and allowing the emulsion to set, thereby to form a monolithic block containing said waste embedded in a cement-polyurethane matrix.

2. A method according to claim 1, wherein the emulsion contains a filler in addition to the hydraulic cement.
3. A method according to claim 2, wherein said filler is sand or aggregate.
4. A method according to claim 2 or 3, wherein said emulsion contains, on a weight basis:
- 5 10—40% of an aqueous organic polyol emulsion containing 15—40% water; 5  
10—40% organic diisocyanate;  
20—80% filler, including cement.
5. A method according to any one of claims 1—4, wherein the organic polyol is a polyester polyol and the polyisocyanate is hexamethylene diisocyanate.
- 10 6. A method according to any one of claims 1—5, wherein the hydraulic cement is Portland cement. 10
7. A method according to any one of claims 1—6, wherein the radioactive waste material is a radioactive ion exchange resin containing water in an amount up to 50% by weight, based on the weight of the wet resin.