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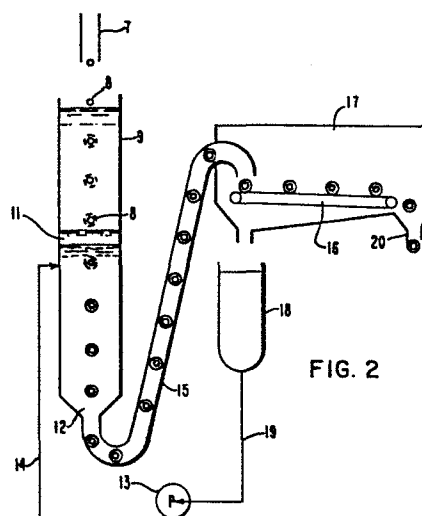
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84 **Method of encapsulating spheroids containing nuclear waste.**

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METHOD OF ENCAPSULATING SPHEROIDS CONTAINING
NUCLEAR WASTE

This invention relates to a method of encapsulating spheroids containing nuclear waste.

Approximately 25 million gallons of high-level nuclear waste has accumulated at the Savannah River Laboratory (SRL) of the Department of Energy (DOE) from the
5 production of defense materials during the past 25 years. One procedure under consideration for disposing of this nuclear waste is to encapsulate it in glass or ceramic spheroids. The spheroids are then coated to reduce the
10 leachability of the nuclear waste from the glass material. The coated glass or ceramic spheroid waste form concept is one having particular appeal because the spherical waste form is produced directly from a liquid in a dustless process that is especially amenable to remote operation.
15 However, the procedures planned for coating the spheroids pose some serious difficulties because three separate coatings are to be applied by chemical vapor deposition at temperatures greater than 1100°C. First, pyrolytic carbon will be deposited by thermal decomposition of acetylene in
20 a fluidized bed reactor. Then, two alumina layers will be applied by using gaseous aluminum tetrachloride and hydrogen in a rotating drum furnace. The use of highly combustible gases and high temperatures in combination with nuclear waste creates a danger of an explosion or fire
25 with the potential release of radionuclides. Also, com-

plex manipulations must be performed by remote operations, and hydrochloric acid by-product solutions are produced which are difficult to treat.

Accordingly a method of encapsulating glass or ceramic spheroids containing nuclear waste characterized by (A) coating said spheroids with a composition which comprises (1) from 30% to 85% by weight, calculated as SiO_2 , of a partially hydrolyzed silicon compound having the general formula $\text{SiR}_m(\text{OR}')_n\text{X}_p$ or $\text{Si}(\text{OSiR}_3)_4$, where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, each X is independently selected from chlorine and bromine, m is 0 to 3, n is 0 to 4, p is 0 to 1, and $m + n + p$ equals 4; (2) up to about 30% by weight, calculated as Al_2O_3 , of a partially hydrolyzed aluminum compound having the general formula $\text{AlR}'_q(\text{OR})_r\text{X}_s$ or $\text{Mg}(\text{Al}(\text{OR})_4)_2$, where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, q is 0 to 3, r is 0 to 3, s is 0 to 1, and $q + r + s$ equals 3, and (3) from 30 to 50 percent by weight of an alcohol; (B) preventing said coated spheroids from touching while hardening said coatings; and (C) curing the coatings on said spheroids.

We have discovered a method of applying an amorphous coating to glass or ceramic spheroids containing nuclear waste. The coating is of very low leachability and can be made to have about the same thermal expansion as the spheroids do, so that cracking does not occur with changes in temperature. The coating can be applied and cured at temperatures of from 400 to 500°C, but if desired, it can be heated at about 850°C to give a dense, amorphous coating of the same quality as coatings produced conventionally at much higher temperatures.

In order that the invention can be more clearly understood, convenient embodiments thereof will now be described, by way of example, with reference to the accompanying drawings in which:

Figure 1 is a diagrammatic side view, in section, illustrating a spraying process for coating spheroids, and Figure 2 is a diagrammatic side view, in section, showing a two-liquid immersion process for coating spheroids. Alternatively, this two-liquid immersion process could also be utilized in reverse provided the specific gravities of the constituent fluids were properly matched.

Referring to Figure 1, a sphere dispenser 1 drops spheroids 2 one at a time into chamber 3. At the top portion of the chamber, spray nozzles 4 spray a coating composition according to this invention onto spheroids 2. In the lower portion of chamber 3 a heating coil 5 heats the coatings 6 on the spheroids and hardens them.

In Figure 2, a sphere dispenser 7 drops spheroids 8 one at a time into trough 9. The trough 9 contains a hydrolyzed alkoxide coating composition 10 in its upper portion, an immiscible liquid layer 11 in its middle portion, and a hardening agent 12 in its lower portion.

The immiscible liquid layer, which separates the coating composition from the hardening agent, is necessary because otherwise the two fluids will mix and the entire coating composition will become hard. A silicone oil layer 1/8 to 1/4 inch thick has been found suitable for this purpose. It is preferable to heat the hardening fluid up to about 75°C to accelerate the hardening process. Higher temperatures, however, should be avoided as they may result in bubbles in the coating. A pump 13 circulates the hardening fluid through line 14 which forces the coated spheroids up tube 15 onto moving belt 16 into heated chamber 17. Excess hardening fluid is collected in vessel 18 where it passes through line 19 to the pump for recirculation. In heated chamber 17 the coated spheroids are calcined to glassify the coating. The spheroids then pass through opening 20 where they are collected for disposal.

In the two liquid-immersion process the hardening fluid may be either a setting ("electrolytic") agent or a dehydrating agent. Sodium hydroxide, ammonium hydroxide, hydrochloric acid, acetic acid, and hot water, are suitable setting agents. Sodium hydroxide is preferred because it promotes the most rapid hardening, although it does contaminate the coating somewhat with sodium ions. The purpose of the hardening agent is to make the coating sufficiently hard and non-tacky so that the spheroids will not stick together and the coating will not be damaged in handling. The amount of setting agent required to produce hardening of the coating may vary from 0.03 to 0.1 moles per mole of alkoxide glass forming constituent. Generally, equal volumes of alkoxide coating solution and setting agent are present in the system when sodium hydroxide is used as the setting agent. With the other setting agents which take a longer time for hardening, about 3 to 4 times as much as the coating solution is used to give an increased travel time.

The dehydrating agent works by removing water from the alkoxide composition, thus further cross-linking the silicon and aluminum oxides. The preferred dehydrating agent is trichloroethylene as it has been found to work well, although 2-ethyl-1-hexanol or octanol may also be used. When a dehydrating agent is used, a molecular sieve should be inserted into the recirculating line leading from the pump to remove the water captured by the dehydrating agent. It is preferable to use a dehydrating agent instead of a setting agent as dehydrating agents do not introduce contaminant species such as sodium into the coating, even though they promote less rapid hardening of the coating.

The final step in the process of this invention is to cure the coating which can be accomplished by heating the coated spheroids to 400 to 500°C in air for about one hour. At this temperature the cured coating contains closed porosity. Heating to about 800°C will completely

densify the coating. To avoid thermal shock it is desirable to expose the coated spheroids to a temperature of about 100°C before exposing them to the higher temperature.

5 The process of this invention may be repeated using the same spheroids in order to enhance the thickness of the coating to the desired level. Generally, a coating of about 0.5 mm thick is considered desirable for the disposal of nuclear waste. Each coating cycle, however,
10 should not add more than about 0.1 mm to the thickness of the coating in order to avoid cracks in the coating. Preferably, each cycle should add about 0.05 mm to the coating thickness. This normally means from 5 to 10 passes are required to produce a coating of a desired
15 thickness.

 The nuclear waste contained in the spheroids may take a variety of forms such as a sludge consisting of a mixture of complex hydroxides or hydrolyzed oxides of aluminum, iron, magnesium, manganese, silicon, calcium,
20 sodium, potassium, ruthenium, mercury, nickel, cesium, strontium, uranium, molybdenum, the transuranics, and other elements. Defense nuclear waste can also include up to about 10 percent by weight sulfate, phosphate, nitrate, or mixtures thereof, and up to about 95 percent by weight
25 water. The radioactive elements in nuclear waste may include uranium, thorium, cesium, ruthenium, iodine, and strontium. The nuclear waste in the spheroids may also be the result of fuel reprocessing which produces an aqueous nitrate solution of many of these elements.

30 The nuclear waste is contained in various types of glass or ceramic, usually of a borosilicate type by processes well known in the art. See, for example, the reference "Ceramics in Nuclear Waste Management," Proceedings of an International Symposium held in Cincinnati,
35 Ohio, April 30-May 2, 1979, sponsored by the American Ceramic Society and the U.S. Dept of Energy, pp. 73-122. There are several basic processes for containing the

nuclear waste in spheroids. One process is a gel precipitation process. In this process the nuclear waste is mixed with a gel-support additive such as polyvinyl chloride, methyl cellulose, and/or formamide. Drops of the mixture are then permitted to fall into a gelation agent, such as ammonium hydroxide, which hardens the drops into small spheroids. The spheroids are then collected and are heated to remove the organics and to densify them, resulting in ceramic spheroids about 0.5 to about 3 mm diameter.

Another process for containing the nuclear waste in spheroids is a marble process. In this process nuclear wastes are mixed with glass frit containing such constituents as sodium oxide, silicon dioxide, and boron oxide. The mixture is then melted and cast into molds. This process produces glass spheroids from 10 to 25 mm in diameter.

The spheroids should be cleaned before they are used in the process of this invention. Cleaning may be accomplished by immersion in trichloroethylene, ethyl alcohol, one molar (1 M) nitric acid, or other cleaning fluids.

The composition used to coat the glasses is a mixture of alcohol and partially hydrolyzed alkoxides. The first glass-forming component of the composition is prepared from a silicon compound having the general formula $\text{SiR}_m(\text{OR}')_n\text{X}_p$ or $\text{Si}(\text{OSiR}_3)_4$ where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, each X is independently selected from chlorine and bromine, m is 0 to 3, n is 0 to 4, p is 0 to 1, and $m + n + p$ equals 4. The $\text{SiR}_m(\text{OR}')_n\text{X}_p$ compounds are preferred due to their availability, stability and compatibility with the other glass-forming constituents. The R' group is preferably alkyl to C_4 with $n = 4$ because these alkoxides are the most suitable starting compounds.

Examples of appropriate compounds which fall within the scope of the general formula include:

	Trimethylethoxysilane	$(\text{CH}_3)_3\text{Si}(\text{OC}_2\text{H}_5)$
	Ethyltriethoxysilane	$\text{C}_2\text{H}_5\text{Si}(\text{OC}_2\text{H}_5)_3$
	Tetrapropoxysilane	$\text{Si}(\text{OC}_3\text{H}_7)_4$
	Tetraethylorthosilicate	$\text{Si}(\text{OC}_2\text{H}_5)_4$
5	Tetratriethoxysiloxysilane	$\text{Si}[\text{OSi}(\text{CH}_3)_2\text{C}_2\text{H}_5]_4$
	Triethylchlorosilane	$(\text{C}_2\text{H}_5)_3\text{SiCl}$
	Vinyltriphenoxysilane	$\text{CH}_2=\text{CHSi}(\text{OC}_6\text{H}_5)_3$

The preferred silicon compound is tetraethylorthosilicate because it is relatively inexpensive, readily available, stable, and easy to handle. Before the silicon compound is added to the composition, it is partially hydrolyzed because its rate of hydrolysis is slower than the other compounds, and preferential precipitation may result if the components are hydrolyzed after they have been combined. Partial hydrolyzation may be accomplished by the addition of water to the silicon compound, where either the water, the silicon compound, or both, have been diluted with alcohol. The molar ratio of a silicon compound to the alcohol can range from 0.2 to 2. The alcohol is preferably the same alcohol that is produced during hydrolyzation so that it is not necessary to separate two different alcohols. The mole ratio of the silicon compound to the water can range from 0.1 to 5. It is occasionally necessary to use up to about six drops of concentrated nitric acid per mole of water to aid in the hydrolyzation reaction.

The second component of the composition is an aluminum compound which has a general formula $\text{AlR}'_q(\text{OR})_r\text{X}_s$ or $\text{Mg}(\text{Al}(\text{OR})_4)_2$ where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, q is 0 to 3, r is 0 to 3, s is 0 to 1, and $q + r + s$ is 3. The $\text{AlR}'_q(\text{OR})_r\text{X}_s$ compounds, where r is 3 and R is alkyl to C_4 are preferred as they are the most stable and available and are easiest to handle. Examples of suitable aluminum compounds include:

	Trimethyl Aluminum	$\text{Al}(\text{CH}_3)_3$
	Triethyl Aluminum	$\text{Al}(\text{C}_2\text{H}_5)_3$
	Triethoxyaluminum	$\text{Al}(\text{OC}_2\text{H}_5)_3$
	Aluminum Isopropopate	$\text{Al}(\text{OC}_3\text{H}_7)_3$
5	Aluminum Secondary Butoxide	$\text{Al}(\text{OC}_4\text{H}_9)_3$
	Triphenyl Aluminum	$\text{Al}(\text{C}_6\text{H}_5)_3$
	Aluminum Magnesium Ethoxide	$\text{Mg}[\text{Al}(\text{OC}_2\text{H}_5)_4]_2$
	Diethylaluminum Chloride	$(\text{C}_2\text{H}_5)_2\text{AlCl}$

10 The preferred aluminum compound is aluminum
 secondary butoxide because it is stable, available, and
 does not require special handling. These compounds are
 hydrolyzed prior to addition to the composition to avoid
 inhomogeneities. Hydrolysis can be accomplished using a
 molar ratio of aluminum compound to water of from 0.0007
 15 to 0.03 and using from 0.03 to 0.1 mole of 1M HNO_3 per
 mole of $\text{AlO}(\text{OH})$ produced. The water is preferably heated
 to from 70 to 100°C .

20 In addition, the composition may contain alkox-
 ides of boron or sodium which may be needed to match the
 thermal expansion of the coating with the thermal expan-
 sion of the spheroids. However, preferably no boron or
 sodium compounds are present as they increase the leach-
 ability of the coating. Sodium compounds which could be
 used have a general formula NaOR or NaZR'_3 where each R is
 25 independently selected from alkyl to C_{10} and alkenyl to
 C_{10} , each R' is independently selected from R and aryl,
 and Z is carbon or boron. The NaOR compounds where R is
 alkyl to C_4 are preferred as they are more stable and
 compatible. The sodium compounds should be hydrolyzed
 30 prior to being mixed into the composition to avoid differ-
 ential hydrolyzation. A molar ratio of a sodium compound
 to water of from 0.003 to 0.1 may be used for hydrolyza-
 tion. Suitable sodium compounds which fall within the
 scope of the general formula include:

Sodium Methylate	NaOCH_3
Triphenylmethylnsodium	$\text{NaC}(\text{C}_6\text{H}_5)_3$
Triphenylborylnsodium	$\text{NaB}(\text{C}_6\text{H}_5)_3$

5 Sodium methylate is preferred as it is easier to handle and is readily available.

Boron compounds which can be used have a general formula $\text{BR}'_q(\text{OR})_r\text{X}_s$ where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, q is 0 to 3, r is 0 to 3, s is 0 to 1, and $q + r + s$ is 3. The compounds where R is alkyl C_4 and r is 3 are preferred as they are relatively available and well-matched with the other constituents. A molar ratio of a boron compound to water of from 0.1 to 1.0 may be used. Dilution in the same alcohol as that of the boron compound at a level of from 15 to 25 moles alcohol to boron compound is preferred for a more homogeneous hydrolyzation. Suitable boron compounds which fall within the scope of the general formula include:

Trimethyl Boron	$\text{B}(\text{CH}_3)_3$
20 Triethyl Boron	$\text{B}(\text{C}_2\text{H}_5)_3$
Trimethyl Borate	$\text{B}(\text{OCH}_3)_3$
Triethyl Borate	$\text{B}(\text{OC}_2\text{H}_5)_3$
Triisobutyl Borate	$\text{B}(\text{OC}_4\text{H}_9)_3$
Triisopropyl Borate	$\text{B}(\text{OC}_3\text{H}_7)_3$
25 Triisobutylborine	$\text{B}(\text{C}_4\text{H}_9)_3$
Dimethyloxyboron Chloride	$(\text{CH}_3\text{O})_2\text{BCl}$
Diphenyl Boric Acid	$(\text{C}_6\text{H}_5)_2\text{BOH}$

Trimethylborate and triethylborate are preferred as they are relatively available and are compatible and require very little special handling.

30 Finally, the composition preferably includes up to about 2 percent of a surfactant to increase the adhesion of the coating to the spheroid. A suitable sur-

factant is octylphenoxypolyethoxyethanol sold under the trade designation Triton X-102 by Rohm and Haas of Philadelphia, Pennsylvania.

The invention will now be illustrated with reference to the following Examples:

EXAMPLE 1

Aluminum Alkoxide Preparation - batch I

To 108 g of deionized water at 85°C was added 13.014 g $\text{Al}(\text{OC}_4\text{H}_9)_3$ while stirring. Subsequently, 7.8 ml of 1M HNO_3 was introduced, and the resulting solution (actually a colloidal dispersion) was aged for 12 hours at 85°C.

Aluminum Alkoxide Preparation - batch II

The same procedure as above was followed with the exception that the weight of $\text{Al}(\text{OC}_4\text{H}_9)_3$ was doubled.

Silicon Alkoxide Preparation

To 104g $\text{Si}(\text{OC}_2\text{H}_5)_4$ was added 90 g of absolute ethyl alcohol, followed by 9 g of deionized water and 1 drop of concentrated HNO_3 .

Combining this hydrolyzed silicon alkoxide batch with batch I of the hydrolyzed aluminum alkoxide corresponded to a 90 SiO_2 :10 Al_2O_3 ratio on an oxide basis. The use of batch II of the hydrolyzed aluminum gave a 80 SiO_2 :20 Al_2O_3 oxide ratio.

Generally, in most of the coating experiments better results were obtained with the addition of a wetting agent to the hydrolyzed alkoxide mixture to enhance the application of the coating on the simulated waste-glass spheroids (or other glass shapes). After considerable screening, Triton X-102, a Rohm and Haas product, was found to be most effective at a concentration of about 0.5 volume percent, although acceptable coatings were obtained with up to ~2 v/o (% by volume).

A system, represented by Figure 1, was assembled which allowed either of the silicon-aluminum alkoxide mixtures to be atomized onto the waste-glass spheroids, followed by heating. This heating to cure the coating was

performed as depicted in Figure 1, or alternatively, by pre-heating the spheroids to about 250°C before spray application of the coating. In this case a wetting agent (surfactant) was not required. Subsequent heating to 500°C removed all water of hydration and residual alkyls to yield an adherent, amorphous coating. Although the coating produced at 500°C was porous, the porosity was not joined and the coating served as a durable barrier against leaching. Heating to 800-850°C produced a totally dense, amorphous coating identical to that which would be obtained by melting at much higher temperatures using conventional processes.

The coating process was repeated a number of times to increase the thickness of the coating and to ensure against imperfections such as connected porosity or cracks. Even when the coating composition did not exactly match that of the spheroid, cracks or spalling of the coating due to differences in thermal expansion coefficients were not observed. Compositional analysis by EDAX across the spheroid-coating interface indicated some short-range "diffusion" of sodium in particular from the spheroid into the coating. Perhaps this is promoted by the reactivity, resembling chemical etching, of the alkoxides with respect to the glass spheroid (substrate).

EXAMPLE 2

The system illustrated in Figure 2 was built to enable the spheroids to be individually dispensed into the alkoxide coating fluid and, then, through a solution which caused curing of the coating into a stiff gel before the spheroids came into contact with one another. The two liquids were separated by an immiscible silicone oil manufactured by Dow Corning Co. Using the silicon-aluminum batch II alkoxide mixture containing Triton surfactant prepared in Example 1 in the top of the column (Figure 2), and sodium hydroxide of about 0.5 M in the bottom segment, waste glass spheroids properly cleaned in trichloroethylene with ultrasonic agitation were success-

fully coated. The sodium hydroxide produced rapid and uniform curing of the coating. Other agents such as ammonium hydroxide, hydrochloric acid, and acetic acid required somewhat longer times for curing and, thus, a longer travel time in them was needed before the spheroids could be collected. Final heating to 500 or 800°C produced the quality vitreous coatings described in Example 1.

EXAMPLE 3

An alternative to using a "setting" agent in the bottom stage of the column in Figure 2, was to use a fluid which would cause curing of the coating by means of dehydration. Such a liquid was trichloroethylene or octanol or 2 ethyl-1 hexanol. The solubility of water in these fluids was sufficient, particularly when they were warmed to ~75°C, to cause the coating to harden. Although the hardening did not occur as rapidly as with the sodium hydroxide setting agent, a high quality coating as described in Example 1 could nevertheless be obtained. In a continuous system, the water would be extracted by molecular sieve, for example, and circulated back to the column.

What we claim is:

1. A method of encapsulating glass or ceramic spheroids containing nuclear waste characterized by (A) coating said spheroids with a composition which comprises (1) from 30% to 85% by weight, calculated as SiO_2 , of a partially hydrolyzed silicon compound having the general formula $\text{SiR}_m(\text{OR}')_n\text{X}_p$ or $\text{Si}(\text{OSiR}_3)_4$, where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, each X is independently selected from chlorine and bromine, m is 0 to 3, n is 0 to 4, p is 0 to 1, and $m + n + p$ equals 4; (2) up to about 30% by weight, calculated as Al_2O_3 , of a partially hydrolyzed aluminum compound having the general formula $\text{AlR}'_q(\text{OR})_r\text{X}_s$ or $\text{Mg}(\text{Al}(\text{OR})_4)_2$, where each R is independently selected from alkyl to C_{10} and alkenyl to C_{10} , each R' is independently selected from R and aryl, q is 0 to 3, r is 0 to 3, s is 0 to 1, and $q + r + s$ equals 3, and (3) from 30 to 50 percent by weight of an alcohol; (B) preventing said coated spheroids from touching while hardening said coatings; and (C) curing the coatings on said spheroids.

2. A method according to claim 1, characterized in that the spheroids are coated by spraying said composition onto them as they fall.

3. A method according to claim 2, characterized in that the coatings on said spheroids are hardened by heating them as they fall.

4. A method according to claim 1, characterized in that the spheroids are first heated to ~250°C and are coated by spraying said compositions onto them.

5. A method according to claim 1, characterized in that the spheroids are coated by dropping them into the composition.

6. A method according to claim 1, characterized in that the coatings on said spheroids are hardened by immersion in a setting agent.

10 7. A method according to claim 6, characterized in that the setting agent is an aqueous solution of MOH where M is selected from alkali metals, ammonium, and mixtures thereof.

8. A method according to claim 6 or 7, characterized in that the agent is heated to about 75°C.

9. A method according to claim 1, characterized in that the coatings on the spheroids are hardened by immersion in a dehydrating agent.

10. A method according to claim 9, characterized in that the dehydrating agent is trichloroethylene.

11. A method according to claim 9 or 10, characterized in that the dehydrating agent is heated to about 75°C.

12. A method according to claim 1, characterized in that the composition is hardened by coating the spheroids with a setting agent prior to coating them with said composition.

13. A method according to any of preceding claims, characterized in that the spheroids are from 1/2 to 25 mm in diameter.

14. A method according to any of the preceding claims, characterized in that steps (A), (B) and (C) are repeated from 5 to 10 times.

15. A method according to claim 1, characterized in that the spheroids are coated and hardened by dropping them into a vessel containing said composition on top and a hardening fluid on the bottom, said composition

and said hardening fluid being separated by an immiscible liquid.

16. A method according to claim 15, characterized in that the immiscible liquid is a silicon oil.

FIG. 1

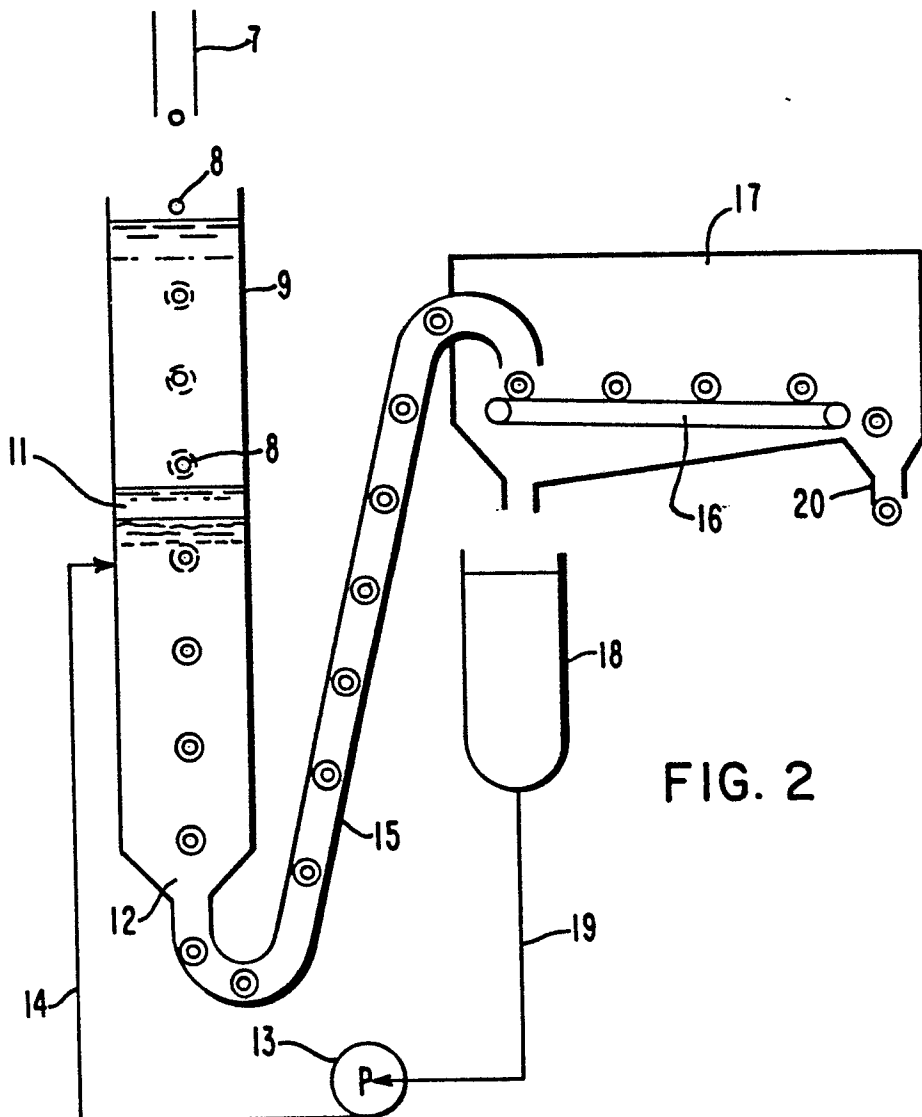
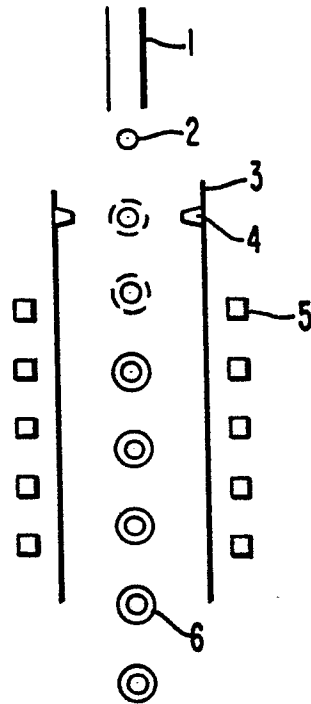


FIG. 2



European Patent
Office

EUROPEAN SEARCH REPORT

0073125

Application number

EP 82 30 4282

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
P,X	--- EP-A-0 042 770 (WESTINGHOUSE) *Claim 1*	1	G 21 F 9/34
A	--- CONF-801124--9, 1980, 8p, pages 1-8, Microfilm, Westinghouse Research & Development Center, Pittsburgh, Pennsylvania (USA); J.M.POPE et al.: "Advanced method for making vitreous waste forms". *The whole document*	1	
A	--- CONF-810528--16, 1981, 27p, Microfilm, ORNL, Oak Ridge, (USA); D.P.STINTON: "Coating crystalline nuclear-waste forms to improve inertness". *Page 15; abstract*	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl. 2)
			G 21 F
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 09-11-1982	Examiner NICOLAS H.J.F.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	