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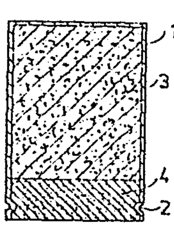
(54) **A method in the storage of nuclear waste of intermediate-level radioactivity, deriving E.G. from nuclear power plants and a waste unit produced hereby.**

(57) A mixture of waste and a liquid hydraulic binder is introduced through an opening in a container, the binder is allowed to solidify, the container opening is sealed, and the container is optionally moved a repository site. The container comprises an iron material and has gas impervious walls and bottom; the seal is formed from a material or a material combination which exhibits high resistance to outward diffusion of radioactive nuclides but is permeable to gas, and in the event of the container being moved to a repository site, the container is embedded in a concrete block in the repository site. A waste unit, comprising at least one substantially cylindrical container (1) which is provided with a bottom and which accommodates a mixture (3) of intermediate-level radioactive waste deriving e.g. from a nuclear power plant, and a binder and is provided with a seal or closure means (4).

Fig. 1



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A method in the storage of nuclear waste of intermediate-level radioactivity, deriving e.g. from nuclear power plants and a waste unit produced hereby

The present invention relates to a method in the storage of nuclear waste of intermediate-level radioactivity deriving e.g. from nuclear power plants, in which method a mixture of the waste and a liquid binding agent is charged to a container through an opening provided therein; the binder is allowed to solidify; the container opening is closed and sealed; and the container is optionally transported to a final or terminal storage site. The invention also relates to a waste unit produced when carrying out the method according to the invention.

In operation, nuclear power plants, plants for reprocessing of nuclear fuel, and research stations produce so-called intermediate-level radioactive waste. By this is meant waste with which the level of radioactive radiation is so low that the waste can be shielded with the aid of relatively simple means and does not give rise to any appreciable increase in temperature. Such waste should also contain mainly radioactive nuclides of moderate half-life. This means that the radiation emitted by the waste will decrease strongly during the first centuries.

The intermediate-level radioactive waste to a large extent comprises ion exchange masses used for cleansing various waterflows in the plant. Another typical constituent of such wastes is salt concentrates resulting from the vaporization of contaminated solutions. The normal method of handling this type of waste is to mix the waste with some substance or another, often cement, bitumen or a synthetic resin. Several benefits are sought in this respect; these benefits being

- improved handling possibilities and reduced risk of waste spreading to the surroundings in conjunction with storing the waste on the spot;
- facilitated transportation of waste to terminal-storage repository sites;

and

- decreased risk of escape from a repository site to the surroundings, and ultimatively to the biosphere.

The most common binder used, and one which affords valuable advantages, is cement. Cement is used in the following way; Waste in the form of a water-saturated ion exchange mass, or a concentrated salt solution, or in some cases a slurry, is mixed with a slurry of cement and water in suitable proportions. The mixture is poured into a container, which may have the form of a conventional 200-liter barrel or drum, or a prefabricated concrete vessel. When the mixture has solidified, the container is sealed with a lid of metal and cement, respectively.

The barrels, or concrete containers, are often stored for some years in locations on the site on which they were filled. The containers are then transported to a repository depot. This repository depot may be a rock cavity located some ten of meters beneath ground level. The containers are also in same instances stored in selected surface locations in an open trench, which is subsequently covered with soil or clay to a depth of some ten meters.

Extremely rigorous requirements are placed on the repository of such waste. Storage shall be such that no harmful activity is able to escape and reach human life over that period of time during which appreciable activity prevails in the stored waste. Those processes which can result in transportation of such harmful activity to humanity are diffusion and flow. Both of these processes take place with the aid of water as a transporting medium. Comprehensive and extremely expensive measures are taken to counteract these transportation possibilities.

- Choice of binder and optional additives thereto for increasing its retention properties;
- casting the cement-filled barrels in reinforced concrete bins;
- casting concrete around the packages subsequent to placing the barrels in the repository location;
- suitable selection of the repository site with respect to bedrock and surroundings;
- suitable choice of repository location with respect to recipient

conditions, i.e. the possibility of leaking activity being diluted.

Attention must be paid to all of these delaying effects when evaluating the safety of the repository site. Attention must also be paid to the slow processes taking place in the waste. The waste comprises a mixture of many different substances which undergo changes. Examples of such changes include:

- Chemical reactions, primarily corrosion of iron barrels, reinforcing rods, etc.;
- changes due to radioactive radiation;
- bacterial processes for which the organic content of the waste constitutes a nutrient.

All of these processes generate gas, some times in considerable quantities. These gases are able to depart when the various concrete barriers or surrounding rock has sufficient permeability. When the gases are unable to escape, there occurs a pressure build-up which is liable to cause cracks to form.

In the known storage method in which a mixture of waste and cement are cast in a container in the form of a simple metal drum or barrel having a roll-swaged bottom and therewith being permeable to liquid, ground water is able to enter the container (the metal drum) with no difficulty at all, and to come into contact with the concrete mass enclosing the waste. In order to delay penetration of the water into the concrete mass and the outward diffusion of the nuclides contained in the waste there is used a cement mixture which affords an extremely dense concrete (fine pores). In this case it is necessary to maintain a low quantitative ratio between waste and binder.

One object of the present invention is to provide a method in the storage of intermediate-level radioactive waste deriving e.g. from nuclear power plants and a waste unit produced in conjunction herewith, which enable a higher quantitative ratio to be used between waste and binder than that permitted with the known method, without increasing the safety risks.

Another object of the invention is to provide a method and a waste unit of the aforedescribed kind in which transportation of the nuclides of the waste by diffusion from the interior of the waste body (concrete mass) to its outer surface and out from the container to the surroundings is delayed and counter-acted to a greater extent than in the aforementioned known method and waste unit, at the same time as gas is able to leak from the container to an extent which prevents the build-up of harmful overpressures.

In accordance with the present invention these objects are achieved by a method of the kind described in the introduction which is characterized in

a) that the binder used is a hydraulic binder, preferably cement and that the container used is one made of an iron material and has gas-impervious walls and bottom;

b) that the container is sealed with a material or a combination of materials which exhibit high resistance to outward diffusion of radioactive nuclides but are gas permeable; and

c) that the container if moved to a repository site is cast in a concrete block in the said repository.

The term "hydraulic binder" is recognized within the building material industry to designate a binder capable of hardening in the presence of water. (Portland) cement is a prime example of conventional binders of this kind, although hydraulic lime and activated slags, such as, for instance, blast furnace slag activated with soda lye also fall under the designation "hydraulic binder".

In accordance with one embodiment of the method according to the invention the container is sealed by casting a concrete lid or cover in position in the container opening.

In accordance with one aspect of the aforesaid embodiment, a disk of highly porous concrete or a plate made of a metal or an alloy which can readily corrode in an alkaline environment can be placed nearest to the solidified mixture of waste and binder prior to casting the lid in place. This plate is provided with a multiple of apertures through which gas can pass.

In accordance with another aspect, the lid is cast from a cement mixture which affords an extremely high resistance to outward diffusion of radioactive nuclides, and a plurality of pre-manufactured concrete cylinders of substantial gas-permeability are cast in the lid or cover to provide gas through-flow channels.

When casting the container in a concrete block in the repository site, the container should be positioned so that the lid or cover faces downwards.

The waste unit according to the invention includes at least one substantially cylindrical container provided with a bottom and accommodating a mixture of intermediate-level radioactive waste derived e.g. from a nuclear power plant and a binder, and being provided with a seal or closure means, this waste unit being characterized in that

a) the container is manufactured from an iron material and has a gas-impermeable cylindrical wall and bottom ;

b) the binder is a hydraulic binder, preferably cement;

c) the seal comprises a material or a material combination which is highly resistive to outward diffusion of radioactive nuclides but permeable to gas.

The container is preferably provided with means for holding the seal or closure means in position.

In accordance with one embodiment of the waste unit according to the invention, the seal or closure means comprises a concrete lid or cover cast in position in the container opening.

In this embodiment the seal or closure means may also include a highly porous concrete slab or a plate made of metal or alloy capable of readily corroding in an alkaline environment, and arranged between the mixture of waste and binder and the concrete lid. The plate is provided with a plurality of apertures through which gas can pass.

In this embodiment of the waste unit according to the invention, the lid or cover may be cast from a cement mixture which affords an extremely high resistance to outward diffusion of radioactive nuclides, and may be provided with gas through-flow channels formed from a less impervious material.

The invention will now be described in more detail with reference to the accompanying drawing, which illustrates a number of examples of the waste unit according to the invention, without limiting the invention hereto.

In the drawing, Figures 1-4 are axial sectional views of a first, second, third and fourth embodiment, respectively, of a waste unit according to the invention.

The figures illustrate a cylindrical container 1 provided with an opening and manufactures from an iron material, for example unalloyed or plain steel , it having been ensured that the botton of the container and the container walls are gas tight. Located adjacent the container opening is a constriction 2.

The material from which the container 1 is formed has a thickness such that the container walls and bottom can be expected to remain impervious in prevailing environments for one thousand years. In view of the fact that the container is filled with a mixture containing hydraulic binder , preferably cement, and is intended to be cast into a concrete block in a repository the thickness of the material when using plain carbon steel is normally greater than 2 mm and less than 10 mm, preferably 3-6 mm.

In the illustrated example waste deriving e.g. from a nuclear power plant has been mixed with a slurry of cement and water, and the mixture has been poured into the conatiner 1 and then allowed to solidify to a hard mixture 3 of waste and cement. Unlike the case with previously known methods of storing this kind of waste, no special requirements have to be placed on the quality of the solidified mixture.

The volumetric ratio between waste and binder in the mixture varies with the type of waste concerned and the container measurements. Normally, however, a volumetric ratio of waste to binder in the range of 1:1 - 1:10 is applied.

The size of the container is chosen with a view to the ease of handling the same on one hand and the desire to accommodate as much waste as possible on the other. A diameter of 600 mm has been found a suitable compromise in this respect, together with a container height adapted to accommodate a waste-binder mixture to a height or depth of about 700 mm and the inclusion of a closure means or seal in accordance with the following:

In Figure 1 the seal has the form of a concrete lid 4, the concrete being such as to present a so-called effective diffusivity of about 10^{-13} m^2/s. A lid thickness of about 200 mm is suitable for a container of the aforesaid diameter and with a waste binder mixture of the aforesaid height or depth.

The waste unit according to this embodiment (Figure 1) presents considerable resistance to diffusion, but does not prevent the penetration of water other than when stored at shallow depths. The majority of nuclides, however, have time to decay totally before having passed through the lid or cover.

The constriction 2 serves as means for mechanically holding the lid 4 in the position in which it was cast.

In the embodiment illustrated in Figure 2 there is used a container which is about 100 mm taller than the container of the remaining embodiments. Subsequent to pouring the waste and cement mixture 3 into the container, a 200 mm thick slab 5 of highly porous concrete (aerated concrete) has been placed on top of the mixture 3. In addition hereto, a lid 4 has been cast in the manner described in Figure 1, the thickness of the lid, however, being 100 mm.

This construction prevents water from penetrating into and filling the pores of the mixture 3, even when the container is placed some tens of meters beneath ground level.

The waste unit illustrated in Figure 3 differs from the waste unit of Figure 1, insomuch as a perforated aluminium plate 6 having a plate thickness of 2 mm and a weight of 300 g has been placed over the waste and cement mixture 3 prior to casting the lid 4 in position.

When strongly alkaline cement water penetrates the container, the aluminium plate 6 corrodes relatively quickly to form hydrogen gas, which fills the pores in the mixture 3 and prevents water from penetrating into said pores.

It will be understood that instead of aluminium the plate 6 can be made of any other metal or alloy having approximately the same corrosion properties as aluminium in alkaline environments.

The waste unit illustrated in Figure 4 conforms with the waste unit illustrated in Figure 1, with the exception of the design of the lid. In the Figure 4 embodiment, a lid or cover 7 has been cast from a cement mixture which provides an extremely impervious concrete having an effective diffusivity of about 10^{-15} m^2/s. This means that the possibility of pressure build-up in the container cannot be excluded, and hence counter measures must be taken. Accordingly, this embodiment incorporates a plurality of pre-manufactured concrete cylinders 8 having a length of 210 mm and a diameter of 30 mm placed on the mixture (the waste body) 3, so as to be embedded in the lid 4, in the manner illustrated. The cement mixture from which the cylinders 8 are made is such as to provide an effective diffusivity of about 10^{-13} m^2/s., thereby enabling the cylinders 8 to function as gas through-flow channels. The number of cylinders 8 provided may vary in dependence on the type of waste to be stored. Normally, at least 5 cylinders are used, in an attempt to prevent an excessively wide gas through-flow surface becoming blocked simultaneously by chance should the waste unit be placed in the terminal-storage site in a less favourable manner.

The diffusivity is a measurement of the movement of ions in water under the influence of differences in concentration. By effective diffusivity is meant here movement of simple ions, such as chloride ions, through a porous body. It is defined by a law well known in physics, Fick's first law. The effective diffusivity is normally established experimentally. Two containers containing mutually different concentrations of, for example, chloride ions, are separated by a thin porous slab comprising, for example the cement mixture to be examined. The flow of chloride ions through the slab is then determined, and the effective diffusivity calculated from the measurements obtained.

When desiring a concrete which is highly resistive to outward diffusion of radioactive nuclides while, at the same time, permitting gas to pass therethrough to an extent such as to avoid the build-up of dangerously high gas pressures in the container, there is chosen a cement mixture which provides a concrete having a diffusivity in the region of 10^{-12} - 10^{-14} m^2/s. When separate gas through-flow channels are provided, such as with the embodiment illustrated in Figure 4, the major part of the lid may comprise a cement mixture which affords a more impervious concrete exhibiting a diffusivity of at least 10^{-14} m^2/s, and even more impervious concrete.

The diffusivity of the concrete may be regulated in a manner known per se, by selection of:

- cement quality
- mixing ratios, cement powder/water/filler
- filler

The filler may be sand of suitable particle size, together with additives or preferably comprises clays, such as bentonite, industrial silica, fly ash and other commercially available special products.

C L A I M S

1. A method in the storage of intermediate-level radioactive waste deriving e.g. from nuclear power plants in which a mixture of the waste and a liquid binder is charged to a container through an opening provided therein, the binder is permitted to harden, the container opening is sealed, and the container is optionally moved to a repository site, characterized in that

a) the binder used is a hydraulic binder, preferably cement, and the container is made of an iron material and has gas impervious walls and bottom;

b) the seal comprises a material or a material combination exhibiting high resistance to outward diffusion of radioactive nuclides but is permeable to gas, and

c) that the container if moved to said repository site is embodied in a concrete block in said terminal-storage site.

2. A method according to Claim 1, characterized in that the seal is effected by casting a concrete lid in position in the container opening.

3. A method according to Claim 2, characterized in that a highly porous concrete slab is placed close to the solidified mixture of waste and binder, prior to casting the lid in position.

4. A method according to Claim 2, characterized in that a plate made of metal or alloy which can readily corrode in an alkaline environment is placed nearest to the solidified mixture of waste and binder, prior to casting the lid in position.

5. A method according to any one of Claims 2-4, characterized in that the lid is cast from a cement mixture which affords an extremely high resistance to outward diffusion of radioactive nuclides, and in that a plurality of pre-manufactured concrete cylinders of substantial gas-permeability are cast in the lids of form gas through-flow channels.

6. A method according to any one of Claims 1-5, characterized in

that the container is positioned in the repository site with the lid facing downwards.

7. A waste unit comprising at least one substantially cylindrical container (1) provided with a bottom and accommodating a mixture (3) of intermediate-level radioactive waste deriving from e.g. a nuclear power plant and a binder, and presenting a seal or closure means, characterized in that

a) the container (1) is made from an iron material and has a gas impervious cylindrical container wall and bottom,
b) the binder is a hydraulic binder, preferably cement,
c) the seal (4-8) comprises a material or material combination which is highly resistive to outward diffusion of radioactive nuclides but gas permeable.

8. A waste unit according to Claim 7, characterized in that the container presents means (2) for holding the seal in position.

9. A waste unit according to Claim 7, characterized in that the seal is a concrete lid (4) cast in position in the container opening.

10. A waste unit according to Claim 9, characterized in that a highly porous concrete slab (5), or a plate (6) made of metal or alloy which will readily corrode in an alkaline environment, is arranged between the waste and binder mixture (3) and the concrete lid (4).

11. A waste unit according to Claim 9, characterized in that the lid (7) is cast from a cement mixture which affords extremely high resistance to outward diffusion of radioactive nuclides and is provided with gas through-flow channels (8) formed from a less impervious material.

Fig. 1

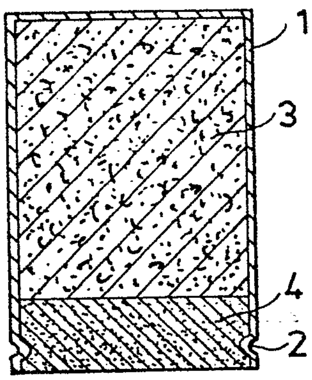


Fig. 2

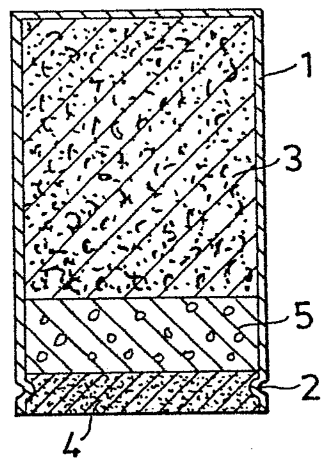


Fig. 3

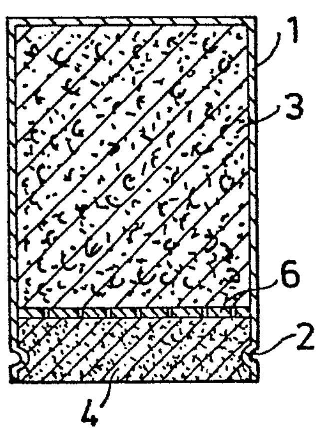
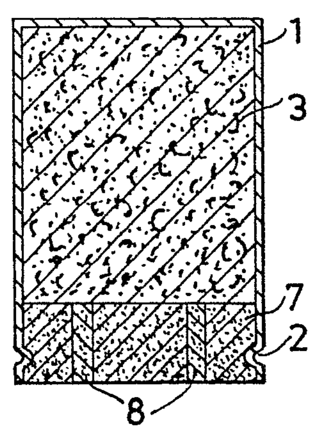


Fig. 4



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EUROPEAN SEARCH REPORT

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Application number

EP 85850353.5

Category	DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int Cl 4)
	DE - A1 - 2 748 774 (NUKEM)		G 21 F 9/36
X	* Fig. 1-3; page 5, lines 9-13; page 6, lines 16-32 *	7-10	G 21 F 1/04
			G 21 F 9/00
Y	* Fig. 1-3; pages 5,6 *	1,2	
A	* Fig. 1-3; pages 5,6 *	4,5,11	
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	GB - A - 2 009 486 (CENTRAL ELECTRICITY GENERATING BOARD)		
Y	* Fig. 7; page 3, lines 13-41, 42-53 *	1,2	
A	* Fig. 7; page 3; page 4, lines 46-100 *	5,7-10	
	--		
Y	DE - A1 - 3 410 370 (NATIONAL NUCLEAR)	1,2	TECHNICAL FIELDS SEARCHED (Int. Cl.4)
	* Fig. 1-3,6,7; abstract *		G 21 F 9/00

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
VIENNA	21-03-1986	KRAL

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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