Europäisches Patentamt
European Patent Office
Office européen des brevets

(19)

(11) Publication number: **0 155 418**
A2

(12)

EUROPEAN PATENT APPLICATION

(21) Application number: **84303715.1**

(51) Int. Cl.[4]: **G 21 F 9/04**

(22) Date of filing: **04.06.84**

(30) Priority: **06.12.83 JP 229091/83**

(43) Date of publication of application: **25.09.85 Bulletin 85/39**

(84) Designated Contracting States: **DE FR GB SE**

(71) Applicant: **MITSUBISHI JUKOGYO KABUSHIKI KAISHA**
5-1, Marunouchi 2-chome Chiyoda-ku
Tokyo(JP)

(72) Inventor: **Sato, Koei c/o Mitsubishi Jukogyo K. K.**
5-1 Marunouchi 2-chome
Chiyoda-ku Tokyo(JP)

(72) Inventor: **Yamauchi, Noriyuki c/o Mitsubishi Jukogyo K. K.**
5-1 Marunouchi 2-chome
Chiyoda-ku Tokyo(JP)

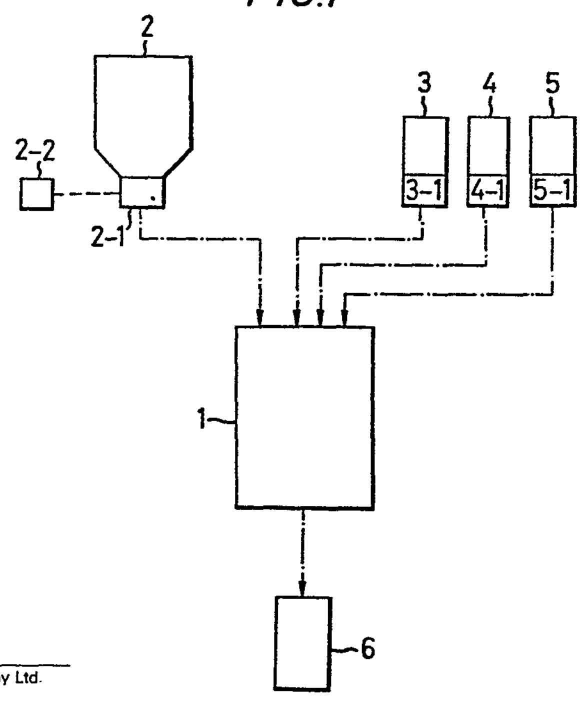
(72) Inventor: **Hirayama, Toshihiko c/o Mitsubishi Jukogyo K. K.**
5-1 Marunouchi 2-chome
Chiyoda-ku Tokyo(JP)

(74) Representative: **Sommerville, John Henry et al,**
SOMMERVILLE & RUSHTON 11 Holywell Hill
St. Albans Hertfordshire, AL1 1EZ(GB)

(54) **Method of volume-reducing disposal of radioactive wastes.**

(57) In a method of disposal of radioactive waste involving volume reduction, a product containing radioactive material obtained as a result of the treatment, such as by drying, calcination and or incineration, of radioactive waste liquid and or combustible solid, is mixed in an appropriate proportion with waste product comprising glass fibers having radioactive material adherent thereto, both wastes being generated from an atomic energy installation, and the mixture is melted by heating to be converted, on subsequent cooling, into an integrated stable solidified mass.

FIG.1



Croydon Printing Company Ltd.

0155418

A METHOD FOR DISPOSAL OF RADIOACTIVE WASTE INVOLVING VOLUME REDUCTION

This invention is concerned with a method, involving volume reduction, for disposal of radioactive waste such as is generated from a radioactive material handling installation, for example a nuclear power station.

In addition to radioactive exhaust gas, atomic energy installations, such as nuclear power stations, are known to generate radioactive waste liquid, combustible solid wastes (combustible miscellaneous solids such as clothes, paper and wood, active carbon or similar materials used in air-conditioning systems, used ion exchange resin and waste oil, and non-combustible solid wastes (for example, glass wool and metallic parts).

Medium-level or low-level radioactive wastes, after they have been subjected to volume-reduction such as by concentration, drying, calcination, incineration or compression and further subjected to various solidification treatments such as asphalt solidifiction and cement solidification according to their usable physical and chemical properties, are conventionally stored in the form of solidified masses.

For instance, in the case of radioactive waste liquid, reduction is achieved by converting it into a sparingly soluble material by chemical reaction and then it is separated by drying or concentration, volatile components being treated by calcination. The precise treatment of such liquids, as is well known, depends upon the precise nature of the liquids themselves. For example in a PWR power station the liquid is mostly liquid containing boric acid used for neutron suppression and in a BWR power station it is mostly whole liquid containing sodium sulphate. Combustible solid wastes such as wood and paper are generally reduced in volume by incineration to yield ash including as principle components silicon oxide and metal oxides. Non-combustible solid wastes are often reduced in volume by compression.

The choice of such treatment is made individually, depending upon the kind of waste. In all cases improvements in efficiency of volume-reduction and stability of the solidified masses are desirable.

However, the stable solidification of the products obtained from the volume reduction treatments mentioned above often requires the addition of various additives upon solidification which increases the volume. Furthermore waste, used glass-fibers from air filters of air-conditioning systems are non-combustible and the efficiency of their volume-reduction is insufficient using the compression treatments of the prior art. As a result the amount of waste to be stored increases year by year and requires early countermeasures.

As to high-level radioactive wastes, while volume-reduction and disposal by solidifying them with glass has been tried, glass frit of good quality is needed as a raw material for the solidifying glass. Furthermore, it is necessary to select, as glass frit, material which has been mixed in an optimum proportion depending on the nature of the high-level radioactive waste involved. There is also a problem in that, since glass frit of good quality is conventionally brought for use from outside the atomic energy installation, it is expensive. The use of this frit also obviously increases the volume of the waste.

We have now found that a product obtained as a result of drying, calcination and/or incineration, of waste liquid and combustible solid found in radioactive wastes (hereinafter sometimes called "product of incineration or the like") together with waste glass fibers contaminated by radioactivity when mixed together in an appropriate proportion, and then combined by melting together in a furnace, can form a stable solidified body for storage thus overcoming disadvantages as outlined above.

Accordingly there is provided a method of disposal of radioactive atomic energy installation waste products characterised in that a product containing radioactive material and obtained as a result of the treatment, such as by drying, calcination and/or incineration, of radio-active waste liquid and/or combustible solid, is mixed in an appropriate proportion with waste product comprising glass fibers having radioactive material adherent thereto, and the mixture is melted by heating to be converted, on subsequent cooling, into an integrated stable solidified mass.

The invention will now be described in more detail with reference to the accompanying Figure 1, which is a schematic flow diagram of one preferred embodiment of the method of the invention.

In Fig. 1, a melting furnace 1 is connected with a container 2 for a product of incineration or the like, through a cutting device 2-1 for the product of incineration or the like, which device is provided with a setter 2-2. The furnace 1 is also connected with containers 3, 4 and 5 for different kinds of waste glass fibers each of which containers is provided with its own cutting device, 3-1, 4-1 and 5-1 respectively, for the waste glass fibers therein. A device 6 for collecting melt discharged from the melting furnace 1 is also provided. The product of incineration or the like comprises ash-like powder, while the waste glass fibers are fiber-like solidified masses. When mixing these materials together in an appropriate proportion and throwing the mixture into the melting furnace 1, although the appropriate proportion can of course be attained by varying the amount of either material, we will describe the variation of proportions in terms of varying the amount of the product of incineration or the like with reference to the amount of the waste glass fibers.

It is to be noted that, since the compositions of waste glass fibers differ depending upon the kinds of different products used, a more stable solidified mass can be obtained by treating at the same time waste glass fibers having the same composition, which is why as shown in Fig. 1 the glass fibers are illustrated as being classified into three kinds. However, classification into any arbitrary number of kinds is desirable depending on the different compositions of glass fibers present.

We show below, in Table 1, as one example of components of a general product of incineration or the like, the compositions, in weight %, of a calcined product of a pressurized water reactor (PWR) and an incinerated product of a combustible miscellaneous solid body, respectively.

Table-1

Specimens \ Components	SiO_2	B_2O_3	Na_2O_3	Other Metal Oxides
A Calcinated Product of PWR	20.3	57.3	4.6	17.8
B Incinerated Product of Combustible Miscellaneous Solid Body	60 - 70	-	-	40 - 30

It will be noted that if a radioactive waste liquid is calcined for the purpose of volume reduction, it is converted into a composition principally comprising various metal oxides containing non-volatile radioactive materials. For instance, in the case of a PWR power station, boric acid B is a principal component, and in the case of a BWR power station, Na is a principal component.

As regards glass wool compositions, one example of a representative composition of E-glass is shown in Table-2.

Table-2

Components	SiO_2	B_2O_3	Al_2O_3	CaO	Others
% (weight)	53.5	9	14.5	18.5	4.5

For simplicity we will explain the Invention in terms of the representative container 3 for waste glass fibers in Figure 1. First, a certain amount of waste glass fibers is thrown into the melting furnace 1 by the waste glass fiber cutting device 3-1. Next, a product of incineration or the like in an optimum amount for the composition of the glass fibers in the waste glass fiber container 3 and the amount thereof fed to the furnance 1 is mixed into the waste glass fibers by manipulating the cutting setter 2-2.

The both materials, mixed together within the melting furnance 1, are heated to a temperature at which the waste glass fibers melt. In this case, among the various components adhering to or contained in the both materials, metals and metal oxides having higher melting points than that of the waste glass fibers may be present in the form of granular or other particulate solid material in the melt, but combustible materials are burnt, and thereby a glass-like product having a maximum overall rate of volume reduction can be produced. Furthermore, if heating is continued to a temperature at which the metals and metal oxides having higher melting points than that of the glass fibers also melt, then the glass fibers and these metals and metal oxides melt together and become coupled in molecular structures to give a more stable glass-like product.

When the melting has been finished, the melt is transferred from the melting furnace 1 to the melt collecting container 6, then it is cooled to yield a solidified mass in which radioactivity, metals, metal oxides and the like are sealed. Alternatively the melt can be cooled and solidified within the melting furnace 1 without being transferred to the melt collecting container 6, and the material for disposal can be taken out of the furnace 1 in the form of a solidified mass.

The invention will now be described for the purposes of illustration only, in the following Examples of some preferred embodiments.

Example 1

Using the above-described method, a preferred mixing proportion between the waste glass wool and the product of incineration or the like is, for example, 20% of A in Table-1 and 80% (wt %) of glass wool. These, are mixed together and heated to about 1,400°C to become molten and yield a product of the following composition:

SiO_2	B_2O_3	Al_2O_3	Na_2O_3	CaO	Other Components
46.9	18.7	11.6	0.9	14.8	7.1

This product on cooling is a solidified mass of the conventional borosilicate glass, and at room temperature it has strength, stablity, durability and exudation- resistance (as represented by the weight of component exuding through a unit area when forced stirring has been effected in distilled hot water at 100°C).

Example 2

Using another preferred mixing proportion, 20% of A, 25% of B and 55% (wt %) of glass wool are mixed together and heated at 1,500 - 1,600°C to become molten. The composition of the obtained product was as follows:

SiO_2	B_2O_3	Al_2O_3	CaO	Na_2O	Other Components
49.8	16.4	8.0	10.2	0.9	17.7

When this product was compared with the product of Example 1 product, they were nearly similar and no distinct difference was found therebetween in their properties.

Example 4

In another preferred disposal technique of the invention, the product of incineration or the like as detailed in Table 1 and waste glass fibers of the composition given in Table 2 were used under the conditions indicated in Table 3 below, which also details results obtained.

Table 3

Embodiments	Mixing Propotion	Fusing Temperature	Pressure	Time (H)	Properties of Stable Solidified Body Obtained
4a	A 20kg glass fibers 80kg	1,400°C ∫ 1,500°C	Atmospheric Pressure	Melting for about 1.5H The temperature is held for about 1 H.	Inorganic crystalline mass whose properties are almost similar to conventional borosilicate glass, in which most metals are molten, and which has strength, stablility, durability and exudation-resistance.
4b	A 20kg B 25kg glass fibers 55kg	1,500°C ∫ 1,600°C	Atmospheric Pressure	Melting for about 1.5H The temperature is held for about 1H.	Properties and the like are similar to 4a except that the mass is colored brown or the like due to impurities (Fe or the like). No material difference from Embodiment 4a was found.

Although the effects and advantages of the invention are apparent from the foregoing description alone, the invention can overcome difficulties which arise in connection with the product of incineration or the like which is a pulverized material which is liable to sputter, is hygroscopic, and needs countermeasures to be taken for maintenance and storage, and arising from the relative inefficiency of the known volume reduction by compression of the waste glass fibers. The invention further makes it possible to provide a stable solidified mass which has an extremely small exudation rate of radioactive material and which has appropriate strength and ageing properties. Moreover, in the invention, which uses glass fibers which have become radioactive solid wastes from their presence in air filters, and which are therefore necessarily produced in atomic energy installations, the raw material cost is zero and increase in the amount of waste is avoided.

It will be appreciated that the appropriate proportions of the various waste products in the practice of the present invention will vary depending upon the particular installation and the precise nature of the washes therefrom. However the determination of the proportions of Bi_2O_{31}, SiO_2, other metal oxides, sodium sulphate, glass etc. which should be used can be made by simple trial and error experimentation to ensure that the resultant molten and resolidified glass, that is borosilicate glass, has the required enclosing capability as well as mechanical strength.

Since glass is a noncrystalline (amorphous) material, it has a flexibility in that it can enclose foreign materials, such as metal oxides and achieve the required mechanical strength in a range of appropriate amounts rather than in amounts of fixed proportions.

Since the kinds and amounts of the wastes discharged from an atomic power installation are nearly determined for each installation, a first melting experiment may be carried out in a small crucible as above described and in practice the respective amounts of the wastes described above are suitably determined at such proportions that the entire amounts of such waste glass and treated liquids and solids can be molten together without leaving any excess nor running into shortage, that is, at such proportions that the amounts of the waste materials which are in short supply and hence may have to be brought in from outside of the atomic power installation can be minimized, preferably can be made zero. Such a trial-and-error experiment will determine the appropriate proportions for each instance.

As a matter of routine, it is of course necessary to test the molten and solidified mass obtained after the disposal to examine whether or not the disposal technique has been successful in that the radioactive waste material has been successfully enclosed in a mass of appropriate strength for storing.

CLAIMS

1. A method of disposal of radioactive atomic energy installation waste products, characterised in that a product containing radioactive material and obtained as a result of the treatment, such as by drying, calcination and/or incineration, of radioactive waste liquid and/or combustible solid, is mixed in an appropriate proportion with waste product comprising glass fibers having radio-active material adherent thereto, and the mixture is melted by heating to be converted, on subsequent cooling, into an integrated stable solidified mass.

2. A method as claimed in Claim 1, wherein the glass fiber waste products from the installation are classified into products of substantially the same composition for separate treatment of each classified product.

FIG.1