PATENT SPECIFICATION

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(54) A METHOD OF TREATING RADIOACTIVE WASTE FOR STORAGE

(71) We, SOCIÉTÉ ANONYME DITE NOVATOME formerly NOVATOME INDUSTRIES, a French Body Corporate, of, 20, Avenue Edouard Herriot, 92350 Le Plessis Robinson, France, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a method of treating finely divided solid radioactive nuclear waste for storage.

When storing radioactive waste materials in this solid state it is known to reduce these materials into powders with a more or less fine grain size and then to incorporate and disperse the powdered materials in various storage media such as glass, concrete, plastics materials or asphalt or even water.

It is known that this incorporation of radioactive wastes in such media gives rise to numerous problems, in particular in the case of wastes which are still very active.

Indeed, in media e.g. glass or asphalt, it is necessary to avoid too high a local concentration of active products so as to avoid an abnormal rise in local temperatures.

Further, when the materials to be stored are still dangerously radioactive, they must be treated for storage in a protected "hot" chamber, so that expensive and complicated operations become necessary.

Lastly, incorporation in storage media must satisfy two apparently contradictory conditions: firstly, waste must be concentrated so as to make it occupy the smallest space possible in order to facilitate storage but secondly the waste must never be so concentrated that there is any danger of excessive local concentration of radioactive material. Unfortunately, it is not always easy to obtain a regular and uniform distribution of radioactive waste in media such as e.g. glass or asphalt. For security reasons, it will therefore be necessary to take into account the highest possible local concentration when defining the overall volume to be used. This volume will therefore always be higher than it would have been if the distribution of the concentration of the waste could have been determined beforehand with precision.

Proper application of the method of treating radioactive waste for storage provided by the present invention makes it possible precisely to define the concentration of the waste and to distribute the product with precision so that it will allow the storage volume to be reduced appreciably.

The present invention provides a method of treating powdered radioactive waste material for storage, the method comprising at least the following three operations:

– the powdered material is inserted in an elongate tubular metal casing;

– the powdered material is compacted in said metal casing; and

– the product thus obtained is embedded in a storage medium.

An advantage is that the direct incorporation of some radioactive wastes into media such as glass or asphalt for example entails difficulties if the wastes are put into direct contact with these media since the wastes may damage them. Thus the casing provides protection for the medium in addition to controlling the distribution of the waste.

A further advantage of the method of treating radioactive waste for storage according to the invention is that it should make it possible to avoid long and expensive operations and involve a series of operations which can be carried out automatically with reliable strongly built devices which can be used without drawback in a "hot" chamber.

During the first two operations, the nuclear waste in powder form will be greatly compacted. Measurements made on some waste materials have made it possible in particular to observe that the density of the compacted waste was almost the same as that of the solid waste and close to the density obtained by sintering a powder of the same product under pressure at a high temperature. Thus, the nuclear waste in powder form can be very greatly compressed while it is being covered with a cylindrical metal casing.

During the compacting operation, the density of the covered waste material is further increased by passing the metal casing and its enclosed powder product through one or several reducing dies. An analogous result is obtained by flattening said metal casing so that the final product to be incorporated in a continuous medium is in the form of a tape.

These operations can all be carried out with machines of known and reliable type and can be effected in completely automatic conditions; therefore the whole covering operation can be carried out in a "hot" chamber prohibited to human access.

The nuclear product thus obtained (i.e. covered compacted waste material) will have a density close to that of the solid material and will have an accurately predetermined radioactivity. Since the position of the radioactive material can now be accurately known when the material is inserted in a storage medium e.g. in the form of parallel strands or of a coil in which the spacing apart of the turns is well-defined, it is easy to calculate beforehand the amount of heating obtained in the storage medium and thus fix the spacing to be imparted to the various strands accordingly. Moreover, since the operation carried out is accurately repeatable, data supplied by calculation can be easily checked by experiment and, if need be, modified accordingly.

Covering and compacting will be distinct operations when the covered active product is again compressed during operations such as extrusion or flattening for example. Covering and compaction can be successive operations without any interruption between them when the radioactive waste material is inserted by vibration into a previously existing tube. Indeed, when the vibrations are continued after the phase during which the tube is filled, the radioactive waste material is compressed in the tube, the effect of this being equivalent to compaction but requiring in contrast a very much longer time which can last several hours.

Lastly, storage calls for a few remarks. Storage was initially effected using concrete as the storage medium, but experiment showed that with time, concrete is liable to disintegrate and to crack under the combined effects of outside agents and of internal corpuscular and thermal radiation. Hence, this storage method is satisfactory only for storage of products having low or medium radioactivity.

Glass has also been considered as a storage medium. However, in this case too, there are numerous difficulties.

Firstly, the heat conductivity of the glasses which can be used is particularly low, this considerably limiting the mass of high alpha to beta radioactivity which can be incorporated in a body of glass since the radioactivity of these products causes a great evolution of heat "in situ" which is liable to melt the glass.

Further, glass is a metastable chemical compound and ageing has frequently been observed in the presence of highly ionizing radiations. It can therefore be feared that with ageing speeded up by nuclear reactions, long term storage will lead to uncontrolled release of highly radioactive substances from which the environment needs protection.

Lastly, circumstances may lead to a subsequent attempt at recovering some of the most ionizing of these products with a view to some use which is at present unforseeable. Such recovery of dispersed products in glass appears hardly feasible because of the viscosity of the glass, which requires it to be brought to a high temperature.

The result of long experiments seems to show that for long storage periods it is now possible due to the invention to store the most active wastes in a denser form providing however that a storage medium is used which is a much better conductor. This condition is satisfied by using a metal as the storage medium.

The method according to the invention and the ensuing explanations are illustrated hereinbelow by the description of a few examples with reference to the accompanying drawings in which:

– Figure 1 shows schematically the covering operations when the cylinder containing the active material is shaped into the form of a wire;

– Figures 2 and 3 show two methods of storage of radioactive wastes; and

– Figure 4 shows schematically another method of covering.

Figure 1 shows a hopper 1 containing the product 2 to be stored in powder form. A metal tape (e.g. steel or stainless steel) whose thickness is 5 tenths of a millimetre is driven by a system of rollers 3, 4 and 5 as well as by their counterparts 3′, 4′ and 5′ which are symmetrical in relation to the axis of the metal tape. This system of rollers curves the metal tape 6 into a closed roll 7 which then contains the radioactive waste material already partially compacted by the action of the roller 5 and of its counterpart 5′. A welding torch (not shown) longitudinally welds the roll at 9 in order to seal it completely. After sealing has been checked the roll 7 is then pushed at 8′ towards a compaction die 8. At the output of this compaction die, rollers 11, 11′ drive the product 7′ towards a second die 12 from which the product emerges in the form of a wire 13. This wire has an outside diameter of about 4 millimetres; the thickness of the metal wall is 5 tenths of a millimetre so that the inside of the wire has a useful diameter of about 3 millimetres. It is therefore seen that during extrusion, only the inside powder has been greatly compressed and compacted.

In the case of temporary storage of nuclear wastes which are still very radioactive, water in a water-filled pit is used as the storage medium. The pit may be subdivided into a plurality of different cells by neutron absorbing means. The material to be stored is also enclosed in a stainless steel metal tape. The water allows the heat given off by the radioactive waste to be extracted.

Figures 2 and 3 show schematically two means for storing radioactive wastes. In Figure 2 the material is stored in a drum 31 in the form of parallel wires 32; the material to be stored is still highly radioactive but somewhat less than that stored temporarily in water. The diameter which can be imparted to the compacted product, and the distance at which the parallel elements 32 must be disposed for the rise in temperature to be compatible with the quantity of heat which can be dissipated are then determined.

Alternatively the product stored could be a low or medium radioactivity active ion exchanger resin.

The diameter of each strand is 5 millimetres. The tape is made of ordinary steel.

The average distance between the strands is kept to 10 millimetres.

The assembly is stored in the standard type drums: the storage medium chosen is concrete.

In figure 3, the radioactive product is less radioactive than that of figure 2 and is therefore stocked in the denser form of a coil of wire 33. The product is stored in standard type drums 31 and is potted in a thermosetting resin chosen because of the low radioactivity.

In the example described with reference to Figure 1, the tape was welded longitudinally. But more generally the tape can be closed round the material to be covered more simply by crimping according to the degree of sealing required; the tape can also be closed by cementing or brazing.

Lastly, the lengths of covered material are closed in the axial direction in the same manner as they are closed in the longitudinal direction.

In the examples described with reference to Figures 1, 2, and 3, water was used as the storage medium for temporary storage of very radioactive waste while concrete or thermosetting resin was used for medium or low radioactivity. For longterm storage of very radioactive wastes, the storage medium must be a metal.

Indeed, metals have the required qualities: their heat conductivity is satisfactory and is higher by several orders of magnitude than that of glass, resins or concrete. The stability of the metal with respect to physical or even chemical attacks is clearly greater in the majority of cases than that of other continuous media proposed.

EXPERIMENTS

The following experiments have been carried out

In a first experiment, materials simulating very radioactive fission products were covered by a steel tape with a thickness of 1 mm, then extruded. The wire obtained was wound in the form of a coil and placed in a cylindrical mould which kept the coil at an equal distance of 3 cm from the shell. The mould was filled with molten lead by low-pressure casting. After cooling, the block thus obtained was removed from the mould. A cylindrical block shaped like a 50-litre drum which can easily be handled was thus obtained. Very satisfactory covering of the coil with lead was observed. The cylinder has also undergone various long physical treatments, such as repeated shocks, and being kept at a given temperature. Up till now, no trace of even the smallest breakage has been observed in this block.

In a second experiment, an identical coil was covered with aluminium, again by low-pressure casting in a cylindrical mould provided with a central core; a hollow cylindrical block in the shape of a 200-litre drum was thus obtained; the block was easier to handle than the previous one and underwent various very severe thermal and mechanical tests and chemical tests under the same conditions as for the lead drum and with the same success (i.e. no trace of breakage). Lastly, it was immersed in salt water for a long period without undergoing any alteration or showing signs of ageing. Gamma radiation was very much less attenuated than in the case of a block of lead but in contrast the block can easily be stored for a long time e.g. in a fuel pit.

In a third experiment, the material to be stored was disposed in a cylindrical mould in the form of parallel strands of wire kept at an equal distance apart. The mould was filled with an aluminium alloy and a 100-litre cylindrical block was obtained after removal from the mould. This alloy block underwent the same tests as the hollow block containing a coil. The results obtained are comparable.

In the three above experiments, the coil of radioactive materials or the parallel wires are disposed in the cast metal block in such a way that they remain equidistant from each of the walls of the cylindrical metal block.

Although differently constituted, all the blocks thus formed seemed particularly strong and protected the products against sabotage. Also, they can be melted if necessary and the radioactive products can then be recovered without great difficulty because of the remarkable fluidity of molten aluminium. These storage experiments made with metals of very different kinds seem to show that the use of pure metals or alloys can be considered as very advantageous. Further, the metals used do not undergo any degradation due to the prolonged action of radioactivity so that their physical properties are not altered; this is not the case with continuous media such as thermosetting resins or concrete which seem better adapted for covering products of low radioactivity.

The metal matrix covering the parallel strands of wire also constitutes a second sealed barrier which is extremely stable and strong for retaining the radioactive wastes contained while providing a degree of protection against the radiation emitted. Conversely the wastes included in this block are protected against outside attack. Heat is satisfactorily dissipated from the inside of the block towards its surface. Easy storage in a water-filled fuel pit has already been mentioned, but the blocks thus constituted can also be dry stored and cooled by ventilation. Lastly, the transport thereof from the works to the storage zone does not raise any problems and reduces any danger of contamination.

Figure 4 relates to a device for covering and compaction of radioactive powders which differs somewhat from the device described with reference to figure 1 and provides various advantages: it is easier to implement in a hot laboratory and the sealing of the casing can be checked before its insertion in the hot laboratory.

The device of figure 4 uses a metal tube instead of a tape. Hence, sealing can be checked before the insertion in the chamber and there is no longitudinal welding operation.

In figure 4, a tube 44 is installed in the hot chamber on a vibration table 41, preferably mounted on a cable drum (not shown).

The input orifice 46 of the tube 44 is placed in contact with a funnel 43 of a filling hopper and remains stationary and is bound in a sealed manner to the funnel 43; the other end of the tube 44, i.e. an opening 47, is connected to a filling checking means 45 such as a photoeleceric cell for example. The sealing provided in the connection between the input orifice of the tube and the funnel 43 allows the tube to be filled without dispersion of the dust formed by the powder products in the hot laboratory. This cleanliness of filling simplifies subsequent handling of the tube 44 on its cable drum.

The table 41 is vibrated. The powder material will descend slowly down the tube and rise towards the opening 47 in the direction of the arrow 48. According to the diameter of the tube, this operation can take several hours, but it is completely automatic and is stopped by a signal from the filling checking means 45.

The powder product is then no longer allowed to come in through the funnel 43. However the compacting operation is continued for another hour. As the supply has been stopped, the vibrations of the vibrating table 1 clear the two orifices 47 and 46 of the tube 44 and bring the powder product back towards the centre of the tube 44. The tube is closed at both ends by crushing the ends and then welding them. It seemed particularly advantageous after the orifices 46 and 47 were cleared by the movement of the powder product towards the centre of the tube to fill these two ends before welding with a non-radioactive powder product which will be compressed in its turn by the further vibration of the tube 44. This product then forms a plug at the ends of the tubes thus providing in itself a degree of sealing at both its ends. After the two ends have been crushed it can be advantageous to weld these two ends either separately or together. Lastly, in some cases, the powder product at its ends was a metallic alloy which partially melts at the time of welding and further contributes to completing the sealing of the tube at its ends. After the tube was welded, compaction was further continued as follows: In a first version, the tube thus produced is extruded, this reducing the diameter between the initial tube and the extruded tube by a half or a third, lengthening the extruded tube by 5 to 10 times.

This operation gives good results with tubes having a diameter of the order of 10 millimetres which are relatively thin, i.e. of the order of 1 millimetre.

While a single layer coil 44 is described with reference to figure 4 it seemed particularly advantageous to use a coil with several *layers* of turns of hollow wire previously mounted outside the hot laboratory on a metallic spacing and support structure which is itself constituted by rigid metal wires defining precisely the geometrical portion of each layer of turns of the coil. After the filling operation, on the vibrating table 41, the coil with several *layers* of turns is still held on the vibrating table 41 to proceed with long compaction by vibration. Then the assembly constituted by the coil and its spacing structure is embodied in a metal body preferably made of aluminium or aluminium alloy. In another case, the coil 44 and the cable drum were replaced by a straight tube with thicker walls. After filling on the vibrating table 41, the tube was stopped at both its ends and was forged and hot drawn. The tube was thus lengthened by about 100 times with a reduction in diameter of the order of 1/10th and very high degree of compaction.

In another variant, a straight tube was flattened between two rolls after filling on the vibrating table without attempting to lengthen the tube. Since a flattened section has a smaller cross-section than a circular section excellent quality compaction was thus obtained, with the initial tube being given the form of a tape. After the two ends had been closed, this tape was wound in the form of a spiral spring and mounted on a spacing and support structure. Several elements thus formed were stacked on one another and embedded by casting in a metal such as aluminium.

Instead of being wound on a structure, the flattened tube can be wound in the form of a watch spring simultaneously with a solid metal wire which will serve as a spacer wire for it. As previously, several identical elements will be stacked on one another before casting. A metal (aluminium) cylinder with standard dimensions which is hence easy to handle will finally be obtained, allowing long and safe storage of radioactive materials.

In a final variant the straight tube was held in place on the vibrating table after filling on this table and its contents compacted by vibration. Several straight tubes were assembled by a light metal frame and embedded by casting in a metal such as aluminium. A generally cylindrical aluminium block was then obtained, the straight tubes being maintained parallel inside the block in a way analogous to that of figure 2.

For the storage of highly radioactive products, the method described with reference to figure 4 seems the most advantageous either using the variant having a coil with several layers of turns embedded in a cast aluminium block or the variant using straight rods compacted by vibration and subsequently embedded in cylindrical aluminium blocks analogous to those of figure 2.

Products with less radioactivity can be stored in a thermosetting resin or in concrete.

However, there are cases in which both solutions seem feasible. The man in the art will then stock radioactive products in a continuous metal medium, in particular in an aluminium block if safety is the most essential requirement and in a concrete block if the most essential requirement is the reduction of the cost of storage.

However, the main idea of the present invention remains the insertion of radioactive products in a sealed casing which is then placed in a storage medium in a known geometrical position in order to be able to determine beforehand the rise in temperature resulting from storage and hence to store without danger with optimum concentration and distribution of the radioactive products.

WHAT WE CLAIM IS:

1. A method of treating powdered radioactive waste material for storage, the method comprising at least the following three operations:

– the powdered material is inserted in an elongate tubular metal casing;

– the powdered material is compacted in said metal casing; and

– the product thus obtained is embedded in a storage medium.

2. A method according to claim 1, wherein at least the first two operations are effected automatically in a chamber separated from the outside environment.

3. A method according to claim 2, wherein the powdered materials are inserted in the casing by continuous deposition onto a metal strip which is then curved round the powdered materials and closed.

4. A method according to claim 2, wherein said powdered material is inserted in the casing by vibration in finished lengths of tubes with sealed side walls.

5. A method according to claim 3 or 4, wherein the powdered material is compacted in its casing by extrusion of said casing.

6. A method according to claim 4, wherein the powdered material is compacted in its casing by continuing to vibrate the casing after the material has been inserted therein.

7. A method according to claim 3 or 4, wherein the powdered material is compacted in its casing by flattening said casing.

8. A method according to claim 3 or 4, wherein the casing is thick and the powdered material is compacted by forging and hot drawing.

9. A method according to claim 5, wherein the extruded product is wound onto a metal spacing and support frame.

10. A method according to claim 5, 6, 7, 8 or 9, wherein the extruded product is wound in a plurality of layers of turns on a metal spacing and support frame constituted by rigid metal wires.

11. A method according to claim 7, wherein the flattened tube is wound in the form of a watch spring simultaneously with a

metal wire which serves as a spacer wire.

12. A method according to claim 5 or 6, wherein strands of wire of same length are assembled parallel so as to constitute a straight cylinder.

13. A method according to claim 9, 10, 11 or 12, wherein the covered and compacted radioactive material is embedded in a storage medium externally limited by a contour of a straight cylinder.

14. A method according to claim 13, wherein the storage medium is formed by a thermosetting resin when the radioactivity of the powdered material is low.

15. A method according to claim 13, wherein the storage medium is constituted by concrete when the radioactivity of the powdered material is medium.

16. A method according to claim 13, wherein the radioactive products are embedded in a metal storage medium.

17. A method according to claim 16, wherein the metal storage medium is formed by aluminium.

18. A method according to claim 16, wherein the metal storage medium is lead.

19. A method according to claim 13, wherein the block thus moulded assumes the form of a cylindrical drum in which the winding of radioactive materials is disposed at an equal distance from each of the outside walls.

20. A method according to claim 16, wherein the moulded block assumes the form of a hollow cylinder in which the winding of radioactive materials is disposed at an equal distance from each of the walls of the cylindrical metal block.

21. A method according to claim 16, wherein the metal storage medium is constituted by a metal alloy.

22. A method according to claim 4, wherein each finished length of the tube is sealed at both ends by crushing and welding.

23. A method according to claim 22, wherein both ends of each finished length of tube are stopped with a non-radioactive powder before they are sealed.

24. A method according to claim 23, wherein the non-radioactive powder stopping both ends of the tube is a metal alloy.

25. A method of treating powdered radioactive waste material for storage substantially as herein described with reference to Figures 1, 2 or 3 of the accompanying drawings.

26. A method of treating powdered radioactive waste material for storage substantially as herein described with reference to Figure 4 of the accompanying drawings.

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1580976 COMPLETE SPECIFICATION

3 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 1

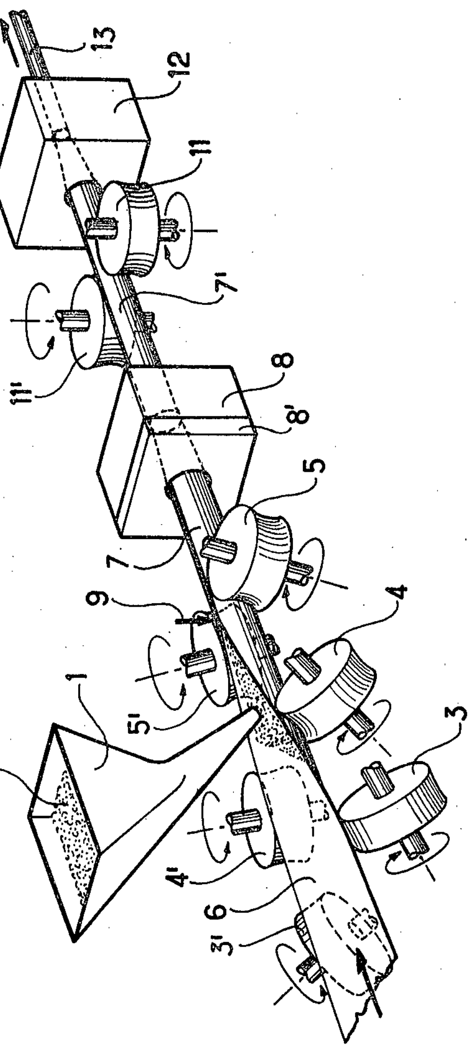

FIG.1

FIG.3

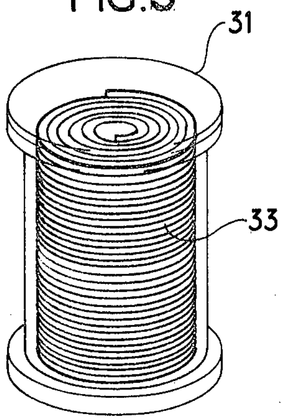

FIG.2

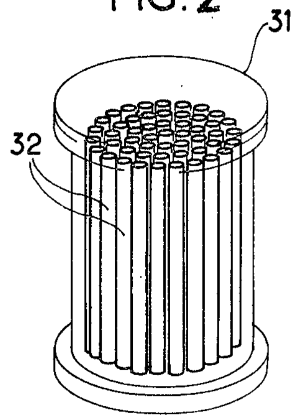

FIG. 4