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Description

The present invention relates to the containment of waste material, and more particularly is concerned with arrangements for containing waste material which requires very reliable, very long term storage.

Extremely long term safe storage of nuclear wastes is a major problem for the nuclear industry and various proposals have been made for dealing with this problem. One proposal concerns immobilising the waste in a suitable borosilicate glass which can then be deposited in a suitable geological formation. However, doubts concerning possible devitrification of the glass and consequent leaching of radioactive elements have founded criticism of the safety of this technique.

Another recent proposal involves the formation of a synthetic rock in which the nuclear reactor waste is immobilised, details of this method being described by A. E. Ringwood et al in Nature March 1979. According to the disclosure, a selected synthetic rock is formed with the radioactive elements in solid solution. The constituent minerals of the rock or close structural analogues have survived in a wide range of geochemical environments for millions of years and are considered highly resistant to leaching by water.

The nuclear reactor waste is incorporated into the crystal lattices of the synthetic rock in the form of a dilute solid solution and therefore should be safely immobilised. A dense, compact, mechanically strong block of the synthetic rock incorporating the nuclear waste is produced by pressure and heat in a densification process and the block may then be safely disposed of in a suitable geological formation.

The following patent applications have been filed by the Australian National University based on the work by A. E. Ringwood et al:—

Australian Patent Application 48708/79 (now Patent AU—A—523,472) entitled "Safe Immobilisation of High Level Nuclear Reactor Waste"; and

United States Patent Application 124953 (now Patent US—A—4,329,248) entitled "A Proces for the Treatment of High Level Nuclear Wastes".

A further development in the field is disclosed in Australian Patent Specification 65176/80 (now Patent AU—A—531250) which is concerned with apparatus and method for immobilising waste material and is directed to selected methods for providing a containment arrangement which will provide a very high degree of safeguarding of the synthetic rock incorporating radioactive waste while nevertheless being produced in a process which is operable within the confines and limitations of a "hot cell". High level radioactive waste must be handled in a hot cell in which all operations are conducted automatically or by an operator using manipulators, and since the apparatus used will inevitably become contaminated itself, the apparatus should be of a form which facilitates servicing within the hot cell and ultimately disposal when its useful working life has come to an end.

Yet further developments in this field are disclosed in European Patent Application 0,044,692 in which additional apparatus and methods for immobilizing waste material such as high level radioactive waste in synthetic rock is disclosed. Among the configurations described is a method in which a powder comprising the synthetic rock materials intimately mixed with the radioactive waste is filled into a container having a bellows-like wall structure and after the container is closed, it is located inside an outer canister around which an induction heating coil is placed. A downwardly acting ram then applies pressure for sufficient time to cause densification of the contents of the bellows container. A series of bellows containers are adapted to be stacked in the outer canister which, when full, is then sealed and removed to a safe storage location.

Despite these various proposals, it is still considered that there is a need to conceive of a more effective, practical and, most importantly, reliable arrangement capable of convenient use in a hot cell.

Broadly, the present invention provides a method of containing and densifying particulate supply material comprising radioactive waste and synthetic rock precursor material, the method comprising pouring the supply material into a bellows container of generally cylindrical form with a side wall including a bellows-like formation and of heat and decay resistant material, closing the bellows container with a lid, placing the bellows container on an upwardly displaceable ram having a heat resistant surface portion, maintaining substantially axial pressure on the bellows container, applying heat and maintaining a sufficiently elevated temperature in the bellows container for a sufficient length of time to cause densification of said particulate supply material in the bellows container and axial compression of the bellows container, the arrangement being such that deformation of the bellows container occurs in its axial direction, and removing the bellows container after completion of the densification step characterized in that said pressure is applied substantially axially to the bellows container solely by the action of an upwardly acting ram which presses the container against a fixed abutment with the bellows-like formation free of surrounding support.

The invention thus can provide a most effective and reliable process for use with supply material in the form of high level radioactive waste and capable of long term operation in a hot cell with ease of operation of the process and maintenance of equipment. Furthermore, the method characterised by the upward pressing technique permits considerable economy of capital equipment and the hot cell space required by virtue of the use of a single ram with a fixed abutment. Since typically operating temperatures in the region of 1100°C will be used, a substantial refractory facing can readily be provided for the fixed abutment and also for the refractory ram. The upward pressing method facilitates, in a preferred embodiment, a most easily serviced apparatus since the refractory facing for the ram can simply be a disc-like pad located by simple locating means such as a spigot and socket with the pad essentially remaining in position under the force of gravity. Thus, using manipulators in a hot cell a worn refractory pad can readily be removed for disposal and a replacement pad fitted.

Furthermore, a major improvement in the process can occur when the broad method described above is used in combination with a preheating step substantially without the application of pressure.

The high working temperatures for the densification step are best achieved by the use of induction heating and therefore typically it takes many hours for the contents of the bellows container to come to a uniform working temperature. Therefore preheating of the bellows container to bring the contents up to a uniform temperature suitable for the densification step is a major advantage. Not only can the production rate for given capital cost be maximized but furthermore a substantial further advantage is that bringing the contents of the bellows container to the uniform densification temperature aids reliable and uniform densification thereby ensuring reliable axial compression of the bellows container which facilitates its later handling and storage. It is to be noted that the bellows container is typically of a heat resistant steel and preferably a stainless steel. Inevitably the mechanical strength of the steel is reduced at the high densification temperatures in the region of 1100 to 1200°C.

Furthermore, the broad method may be utilized in a surprisingly effective and synergistic combination of steps in which the bellows container is subjected to the densification step whilst in a cylindrical canister in which the bellows container becomes a tight fit after the pressing operation thereby providing a most effective and convenient extra containment for long term safe storage of the waste material. Such a method may be defined as consisting in a method for the containment of particulate waste material, the method comprising pouring the waste material into bellows containers of generally cylindrical form with a side wall including a bellows-like formation and of heat and decay resistant material, closing each bellows container with a lid, preheating in series the bellows containers to bring the contents thereof to a substantially uniform elevated temperature, placing each bellows container in turn on an upwardly displaceable ram and displacing the ram upwardly to insert the bellows container into a cylindrical canister and applying pressure and maintaining a sufficiently elevated temperature for sufficient time to cause densification of the contents of the bellows container with axial compression of the bellows container and relatively slight outward expansion thereof to cause the bellows container to grip the interior wall of the cylindrical canister, and when the canister has been filled with a series of such bellows containers, sealing the canister and removing the canister for storage.

Whilst the invention is particularly useful in relation to the incorporation of high level radioactive waste in synthetic rock of the type described by A. E. Ringwood (and referred to above), the invention can also be applied to other synthetic rock arrangements and furthermore can also be applicable to other materials which require storage and are capable of compaction under heat and pressure. One example of such other material would be shredded waste zirconium alloy nuclear fuel rod tubes and similar waste components.

It will be appreciated that the invention consists in a combination of steps which cooperate together in an advantageous relationship which permits efficient, economic, and convenient operations in a hot cell. The apparatus used can be relatively simple, and this can contribute greatly to the reliability and acceptability of the system due to simplicity of servicing and intrinsic reliability.

In a commercial scale operation, it is envisaged that the cylindrical canister will be of the order of 30 cm diameter and 2 metres long and each bellows container will be compressible from an initial height of about 40 cm to a final height of about 10 cm. The use of induction heating coils is the preferred method of both preheating and maintaining the necessary elevated temperature during the densification step, and due to the fact that heating of the 5 particulate material is due to conduction from the bellows container (which is heated by the induction heating coils) a considerable time is required for the preheating step. In a preferred embodiment of the invention, preheating for several hours can be effective whereas the final densification step will be much shorter, e.g. about one hour.

Most preferably, the bellows container is given a preliminary axial pressing which can be at ambient temperature or with advantage can be at a bellows container temperature of up to about 800°C to anneal the material of the bellows container. Since the bellows container will have a high degree of strength at ambient temperature and also at temperatures up to about 800°C, good control can be achieved in this preliminary axial pressing and, surprisingly, during the densification step at high temperature (typically 1200°C) excellent control of the axial pressing can be achieved thereby essentially minimizing the risk of the bellows container compressing with sideways shear rather than true axial compression.

Preferably, the pressure in the preliminary pressing is of at least (3000 lbs/sq. inch) $20{,}68 \cdot 10^6$ Pa.

Particularly when synthetic rock is to be formed, the material is preferably provided in the form of well graded fine particles up to about 2 mm maximum dimension whereby a readily pourable material is provided which can be easily densified in the process.

Preferably synthetic rock is used to incorporate radioactive waste, a mixture of synthetic rock precursor and high level waste being sprayed into a rotary kiln to produce the intimately mixed materials. In order to reduce what would otherwise be loss from the solid material of potentially volatile radioactive components, the temperature at the region in which the material is introduced into the rotary kiln is preferably controlled in the range of about 400—600°C, and the maximum temperature in the kiln is about 700—800°C with the exit from the rotary kiln being at ambient temperature.

A preferred embodiment of the invention can also provide further means for safeguarding the cylindrical canister from outward deformation under the pressure of expanding bellows containers within the canister. This is achievable by the use of a block of refractory material having a slightly tapered bore which at its narrowest diameter just fits over the canister, the refractory block being adapted to be moved downwardly in a series of steps corresponding to bellows container locations, the slightly tapered bores permitting release of the block even if some outward deformation of the canister has taken place in a step of densification and compression of the bellows container.

Most preferably, the refractory block is formed so as to embrace the induction heating coil for surrounding the canister.

Most preferably, the refractory block comprises a series of interlocking refractory segments arranged to be mounted inside a cylindrical containment shroud which absorbs any expansion forces applied from the canister.

According to a second aspect of the invention, there is provided apparatus for encapsulating particulate supply material in bellows containers within a cylindrical canister, the apparatus comprising means for pouring the particulate material into a bellows container, means for sealing the bellows container with a lid, means for moving bellows containers in sequence to a pressing station at which the container is heated and axial pressure is applied to the container causing the axial compression of the container, characterized in that said pressing station comprising an upwardly displaceable ram for receiving a bellows container, means for mounting a cylindrical canister with an open end directed downwardly towards said ram, means for upwardly pressing a bellows container supported on the ram into the canister, upper refractory support means to act as a fixed abutment for the canister and the bellows container pressed upwardly therein by said ram, heating means for maintaining an elevated temperature in said bellows container whilst said pressure is applied to cause densification of said supply material in the bellows container and to expand slightly the bellows container to cause it to jam in the canister, and the heating means being adapted to provide heating in a series of zones within the container corresponding to a series of bellows containers inserted one below the other in series therein, the canister being adapted by being removed and sealed when a series of bellows containers have been densified and secured therein.

Preferably, the apparatus includes a preheating station adapted to bring the contents of the bellows containers to a substantially uniform elevated temperature, and means for transferring a preheated bellows container to said pressing station.

For illustrative purposes only, the invention will now be exemplified by reference to the accompanying drawings wherein:—

Figure 1 is a representation of the preliminary portion of a process embodying the invention;

Figure 2 is a schematic representation of the final steps of the process initiated in Figure 1;

Figure 3 is an axial sectional elevation illustrating a preferred embodiment of apparatus for effecting the densification step of the waste material;

Figure 4 is an axial sectional elevation on an enlarged scale of a preferred form of refractory block configuration shown generally in slightly exploded view in Figure 3; and

Figure 5 is an isometric view from above of the refractory block and induction heating collar arrangement shown in Figure 4.

Referring first to Figures 1 and 2, the process has a preliminary mixing stage 21 in which synthetic rock precursor from supply 20 is formed into a slurry with high level radioactive waste from waste supply 19 which is in the form of a nitrate solution, and the slurry is passed along line 22 to be sprayed into the elevated temperature end of a rotary kiln, at which a maximum temperature in the range 700—800° is maintained. The spraying step immediately vaporises the water content of the slurry sprayed into the rotary kiln and causes chemical decomposition of the radioactive nitrates and will cause the mineral components of the synthetic rock to start to form with the radioactive elements starting to go into mineral phases. A chemical reducing control gas (such as argon-hydrogen, nitrogen-hydrogen, or CO—CO_2) is passed through the rotary kiln. The process could be operated so that synthetic rock particles incorporating the radioactive waste are completely formed in the rotary kiln but this is not essential. The rotary kiln produces cold particulate material of well graded particle size up to about 2 mm maximum dimension, whereas gases produced by the rotary kiln are fed back through a filter F to the preliminary mixing stage 21 since these gases will include some radioactive components.

In order to provide the necessary oxygen potential for the radioactive waste so that it is in the appropriate valency state to be incorporated into the synthetic rock, the particulate material produced by the rotary kiln is fed into a titanium mixing stage 24 which receives metallic titanium powder from a hopper 25 whereby the mixture poured into a bellows container 20 has about 2% titanium metal powder by weight.

An example of a suitable synthetic rock composition is indicated in the table set out below and is produced using tetraisopropyl titanate and tetrabutyl zirconate as ultimate sources of TiO_2 and ZrO_2. The components are mixed with nitrate solutions of the other components, co-precipitated by addition of sodium hydroxide and then washed to produce synthetic rock precursor.

Typical compositions of synthetic rock (Synroc) and constituent phases

	"Hollandite" 40%	Zirconolite 35%	Perovskite 25%	Bulk SYNROC composition
TiO_2	71.0	50.3	57.8	60.3
ZrO_2	0.2	30.5	0.2	10.8
Al_2O_3	12.9	2.5	1.2	6.3
CaO	0.4	16.8	40.6	16.2
BaO	16.0	—	—	6.4
Total	100.5	100.1	99.8	100.1

The precursor material is a product which possesses a very high surface area and functions as an effective ion exchange medium, which is mixed with additives containing Ca, Ba, and Al in solution and mixed in a hot cell with high level nuclear waste (HLW) in the form of nitrate solution to form a thick homogeneous slurry at mixing stage 21. Typically up to about 20% by weight of the solid content of the slurry may comprise the high level wastes.

The bellows container 20 is of a heat resisting steel such as an austenitic stainless steel, for example Sandvik grade 253MA which retains reasonable mechanical strength even at the elevated temperatures used in the process, although at these temperatures the container is relatively ductile. In the illustrated embodiment, a thin perforated metal liner 26 is located within the bellows container and the space between the liner and bellows wall is filled with zirconium oxide powder 27.

A stainless steel cap 29 is used to seal the bellows container which is then placed between a pair of pistons 30 for a cold pressing operation which can increase the density of particulate material from about 25% of the theoretical maximum density to about 36%.

The next stage is illustrated in Figure 2 in which the cold pressed bellows containers 20 are fed in sequence into a vertical induction furnace 31, each bellows container being supported on a refractory disk 32, the lowermost refractory disk being supported by a retractable latch 33. Over a period of several hours the temperature gradually increases up to about 1200°C.

A first water cooled ram 34 having a top spigot on which a refractory plate 35 is located is adapted to support and lower one at a time the bellows containers from the furnace for horizontal movement across a support table 36 to a pressing station having a second water cooled ram 37 of similar form. Figure 2 shows the ram 37 both in the lower receiving position and also in the upwardly displaced pressing position inside a metal canister 38 mounted on a support 39 and having its top sealed and in abutment against a fixed refractory block 40, vertically displaceable induction heating coils 41 being provided outside the canister 38.

In the lower position, the left hand side of the section of a bellows container 20*a* is shown in its configuration *before* hot pressing and the right hand side of the section shows a bellows container 20*b* as it would be *after* pressing. However, during the hot pressing, the bellows container slightly expands to become an interference fit within the canister 38 as shown by bellows 20*c* at the top of the canister 38.

The refractory plate 32 upon which each of the bellows containers is supported is removed after the pressing stage, the plate 32 being lowered on the water cooled ram 37 and then pushed onto a receiving table from which the plate can be recycled for further use.

Refractory plates will wear in use and must be replaced and an important advantage of the design illustrated in Figure 2 is a very simple and easily serviced arrangement made possible by the use of an upward pressing technique; this permits the replaceable refractory top plate 35 simply to sit on the head of each water cooled ram. Just a simple spigot and socket engagement is provided so that manipulators can readily remove a worn refractory plate and insert a new one.

Referring now to Figure 3, a practical embodiment of hot pressing apparatus is illustrated, the parts corresponding to the elements in Figure 2 being given the same reference numerals.

The apparatus further includes a base plate 42 with a set of upstanding tubular guides 43 on which sliding mounts for the support 39 and the induction furnace unit 41 are slidably mounted but adapted to be clamped at any selected position. The canister 38 is urged upwardly against the refractory block 40 which is supported by a top cap 44 adapted to be bolted to a top plate 45. Figure 3 shows the parts in slightly exploded view for clarity. The induction heating coil 41 is shown embedded within a refractory block 46 having a tapered bore, the drawing showing a greatly exaggerated taper and clearance between the bore and the container 38. The object of the tapered bore of the refractory block 46 is that any small expansion of the canister 38 causes the canister to be supported against further outward deformation by the refractory block but by virtue of the taper the refractory block can be released by downward motion to the next location for the succeeding bellows container.

In the enlarged view of Figure 4, like parts are given like reference numerals, and the parts are shown in the assembled condition just prior to pressing.

In this embodiment the refractory block is assembled from refractory segments comprising outer refractory segments 46a of cylindrical profile and inner refractory elements 46b having an inner profile adapted to cooperate to form a tapered bore with circumferentially extending grooves for accommodating the turns of the induction coil 41. The refractory elements are contained within a steel outer support cylinder 47 which absorbs the forces of any outward expansion applied by the canister 38.

Figure 5 shows in isometric view the refractory blocks 46a and 46b each having a semi-circular rib 46c on one side thereof and a corresponding cavity 46d on the other side for interengagement purposes.

Claims

1. A method of containing and densifying particulate supply material comprising radioactive waste and synthetic rock precursor material, the method comprising pouring the supply material (28) into a bellows container (20) of generally cylindrical form with a side wall (27) including a bellows-like formation and of heat and decay resistant material, closing the bellows container with a lid (29), placing the bellows container on an upwardly displaceable ram (37) having a heat resistant surface portion (35), maintaining substantially axial pressure on the bellows container, applying heat and maintaining a sufficiently elevated temperature in the bellows container for a sufficient length of time to cause densification of said particulate supply material in the bellows container and axial compression of the bellows container, the arrangement being such that deformation of the bellows container occurs in its axial direction, and removing the bellows container after completion of the densification step, characterized in that said pressure is applied substantially axially to the bellows container (20) solely by the action of an upwardly acting ram (37) which presses the container against a fixed abutment (40) with the bellows-like formation free of surrounding support.

2. A method as claimed in Claim 1 and wherein after closing the bellows container (20) with the lid (29), a preliminary pre-heating thereof is effected substantially without the application of axial pressure to the bellows container.

3. A method as claimed in Claim 2, wherein the bellows container is of metal and said pre-heating is by induction heating (41) for a period of several hours to bring the bellows container (20) and its contents substantially to a uniform temperature which is substantially elevated but sufficiently below the temperature to be achieved in the subsequent hot pressing step such that the bellows container has significantly greater strength at the pre-heating temperature compared with the hot pressing temperature.

4. A method as claimed in any one of the preceding claims wherein said hot pressing step is conducted with the temperature of the bellows container and its contents brought to about 1200°C.

5. A method as claimed in any one of the preceding claims and wherein immediately after placing the lid (29) on the bellows container (20) an axial compression is applied to the bellows container, the temperature not exceeding 800°C.

6. A method as claimed in claim 5, and wherein the axial compression applied in the pressing step of claim 5 is at least $20{,}68 \cdot 10^6$ Pa (3000 lbs/sq. inch).

7. A method as claimed in any one of the preceding claims, and wherein the supply material has particle size not greater than 2 mm and is readily pourable, the supply material being produced by spraying a slurry into a rotary kiln (23).

8. A method as claimed in any one of the preceding claims, wherein said axial compression of the bellows container at elevated temperature is carried out by inserting the bellows container (20) on the displaceable ram (37) into a cylindrical canister (38) having a downwardly projecting open end and in which the bellows container is a loose fit before the hot compression step, the bellows container undergoing a small radial expansion during compression so as to be pressed into an interference fit with the interior wall of the canister.

9. A method as claimed in claim 8, wherein the cylindrical canister (38) is elongated and a series of bellows containers (20) are upwardly pressed one at a time into the cylindrical canister, and when the canister has been substantially filled, the canister is sealed and removed for storage.

10. A method as claimed in Claim 8 or Claim 9, and including positioning a block of refractory material (46) having a slightly tapered bore over the canister (38) during hot pressing of a bellows container (20) therein, the tapered bore, at its narrowest diameter, being at most a sliding fit over the canister, whereby any tendency for outward deformation of the canister is resisted by the refractory block, the refractory block (46) being moved downwardly relative to the canister (38) after hot pressing of a bellows container.

11. A method as claimed in Claim 10, and wherein the refractory block (46) has an induction heating coil (41) extending therethrough.

12. A method as claimed in any of the preceding Claims, and including using a cylindrical partition (26) within the bellows container (20) and confining said particulate material to the zone within said cylindrical partition, an alternative supply material being located between said partition and the interior wall of the

bellows container whereby said supply material is excluded from the convolutions of the wall of the bellows container (20).

13. Apparatus for encapsulating particulate supply material comprising radioactive waste and synthetic rock precursor in bellows containers (20) within a cylindrical canister (38), the apparatus comprising means (24) for pouring the particulate material into a bellows container (20), means for sealing the bellows container with a lid (29), means (36) for moving bellows containers in sequence to a pressing station at which the container is heated and axial pressure is applied to the container causing the axial compression of the container, characterized in that said pressing station comprising an upwardly displaceable ram (37) for receiving a bellows container (20), means for mounting a cylindrical canister (38) with an open end directed downwardly towards said ram, means (37) for upwardly pressing a bellows container supported on the ram into the canister, upper refractory support means (40) to act as a fixed abutment, heating means (41) for maintaining an elevated temperature in said bellows container (20) whilst said pressure is applied to cause densification of said supply material in the bellows container and to expand slightly the bellows container to cause it to jam in the canister, and the heating means (41) being adapted to provide heating in a series of zones within the container corresponding to a series of bellows containers inserted one below the other in series therein, the canister being adapted by being removed and sealed when a series of bellows containers have been densified and secured therein.

14. Apparatus as claimed in claim 13, and wherein the apparatus includes a preheating station (31) adapted to bring the contents of the bellows containers (20) to a substantially uniform elevated temperature, and means (36) for transferring a preheated bellows container to said pressing station (37).

15. Apparatus as claimed in claim 14, and wherein said preheating station (31) comprises an upwardly extending cylindrical induction heated zone having refractory support means (32, 33) for holding a stack of bellows containers in said zone, and means for handling the bellows containers whereby the bellows containers (20) are inserted cold into the top of the cylindrical zone and are removed after preheating at the bottom of the zone, said transferring means operating in a horizontal direction to transfer the preheated bellows container to said displaceable ram.

16. Apparatus claimed in any one of claims 13 to 15, wherein said displaceable ram (37) has a refractory facing (35) located on the head of the ram by a spigot.

17. Apparatus as claimed in any one of claims 13 to 16 and further comprising compression means (30) for axially compressing substantially at ambient temperature each bellows container (20) after the bellows container has been sealed with the lid (29).

18. Apparatus as claimed in any one of claims 13 to 17 and further comprising a vertically displaceable block (46) of refractory material arranged to surround said canister (38), and the refractory block having a slightly tapered bore which at its narrowest is no more than a sliding fit over the canister, the refractory block being adapted to support the canister against radially outward expansion at the location at which a bellows container is being compressed, the refractory block subsequently being downwardly displaceable.

19. Apparatus as claimed in claim 18, wherein said refractory block incorporates turns of an induction heating coil (41).

20. Apparatus as claimed in claim 19, wherein said refractory block is formed from a series of interlocking refractory segments (46a, 46b) located within an outer cylindrical shell (47).

Patentansprüche

1. Verfahren zum Einschließen und Verdichten von körnigem Ausgangsmaterial, das radioaktiven Abfall und Zwischenmaterial aus synthetischem Gestein enthält, mit folgenden Verfahrensschritten: Zwischenmaterial (28) in einen Balgenbehälter (20) schütten, der eine im wesentlichen zylindrische Form und eine Seitenwand (27) mit balgenartiger Ausbildung hat und aus einem wärme- und verwitterungsbeständigen Material besteht, den Balgenbehälter mit einem Deckel (29) verschließen, den Balgenbehälter auf einen aufwärts verschiebbaren Stempel (37) bringen, der mit einem hitzebeständigen Oberflächenabschnitt (35) versehen ist, einen im wesentlichen axialen Druck auf den Balgenbehälter aufrecht erhalten, Wärme zuführen und eine ausreichend hohe Temperatur im Balgenbehälter für eine ausreichend lange Zeit aufrecht erhalten, um das körnige Ausgangsmaterial im Balgenbehälter zu verdichten und den Balgenbehälter axial zu komprimieren, so daß der Balgenbehälter in seiner Axialrichtigung deformiert wird, und den Balgenbehälter nach dem Abschluß der Verdichtungsstufe entfernen, dadurch gekennzeichnet, daß Druck im wesentlichen axial auf den Balgenbehälter (20) ausgeübt wird, und zwar allein durch die Wirkung eines aufwärts wirkenden Stempels (37), der den Behälter gegen einen festen Anschlag (40) drückt, wobei die balgenartige Ausbildung frei ist von einer sie umgebenden Abstützung.

2. Verfahren nach Anspruch 1, wobei der Balgenbehälter (20), nachdem der mit dem Deckel (29) verschlossen ist, einleitend vor-erhitzt wird, im wesentlichen ohne Ausübung von axialem Druck auf den Balgenbehälter.

3. Verfahren nach Anspruch 2, wobei der Balgenbehälter aus Metall besteht und das Vor-Erhitzen durch Induktionsheizung (41) für eine Dauer von mehreren Stunden durchgeführt wird, um den Balgenbehälter (20) und seinen Inhalt im wesentlichen auf eine gleichmäßige Temperatur zu und seinen Inhalt im wesentlichen auf eine gleichmäßige Temperatur zu bringen, die erheblich erhöht ist, aber

ausreichend unter derjenigen Temperatur liegt, die in der folgenden Warmpreßstufe erzielt wird, so daß der Balgenbehälter eine wesentlich größere Festigkeit bei der Vorheiztemperatur hat, verglichen mit der Warmpreßtemperatur.

4. Verfahren nach einem der vorangehenden Ansprüche, wobei die Warmpreßstufe durchgeführt wird, wenn die Temperatur des Balgenbehälters und seines Inhalts auf etwa 1200°C gebracht worden ist.

5. Verfahren nach einem der vorangehenden Ansprüche, wobei sofort nach dem Aufsetzen des Deckels (29) auf den Balgenbehälter (20) eine axiale Kompression auf den Balgenbehälter ausgeübt wird, während die Temperatur 800°C nicht übersteigt.

6. Verfahren nach Anspruch 5, wobei die in der Preßstufe nach Anspruch 5 ausgeübte axiale Kompression mindestens (3000 lbs/sq. inch) $20{,}68 \cdot 10^6$ Pa beträgt.

7. Verfahren nach einem der vorangehenden Ansprüche, wobei das Ausgangsmaterial eine Korngröße von nicht mehr als 2 mm hat und gut schüttbar ist und das Ausgangsmaterial durch Sprühen eines Breies in einem Drehofen (23) hergestellt wird.

8. Verfahren nach einem der vorangehenden Ansprüche, wobei die axiale Kompression des Balgenbehälters bei erhöhter Temperatur durch Einführen des Balgenbehälters (20) auf dem verschiebbaren Stempel (37) in einen zylindrischen Kanister (38) durchgeführt wird, dessen unteres Ende offen ist und in den der Balgenbehälter vor der Warmkompressionsstufe mit Spiel paßt, wobei der Balgenhälter während der Kompression eine kleine radiale Erweiterung erfährt, so daß er in einen Preßsitz mit der Innenwand des Kanisters gepreßt wird.

9. Verfahren nach Anspruch 8, wobei der zylindrische Kanister (38) langgestreckt ist und eine Anzahl von Balgenbehältern (20), einer nach dem anderen, nach oben in den zylindrischen Kanister gepreßt werden, und wobei der Kanister, wenn er im wesentlichen gefüllt ist, verschlossen und zum Lagern entfernt wird.

10. Verfahren nach Anspruch 8 oder 9, wobei ein Block (46) aus Feuerfestmaterial mit einem leicht konischen Hohlraum, während ein Balgenbehälter (20) darin warmgepreßt wird, um den Kanister (38) gesetzt wird, wobei die konische Bohrung bei ihrem geringsten Durchmesser höchstens mit Gleitsitz über den Kanister paßt, wodurch jedem Bestreben einer Auswärts-Deformation des Kanisters durch den Block aus Feuerfestmaterial Widerstand geleistet wird, und wobei der Block (46) aus Feuerfestmaterial nach dem Warmpressen eines Balgenbehälters gegenüber dem Kanister (38) abwärts bewegt wird.

11. Verfahren nach Anspruch 10, wobei sich eine Induktionsheizwicklung (41) durch den Block (46) aus Feuerfestmaterial erstreckt.

12. Verfahren nach einem der vorangehenden Ansprüche, wobei innerhalb des Balgenbehälters (20) eine zylindrische Trennwand (26) verwendet wird und das körnige Material auf den Bereich innerhalb der zylindrischen Trennwand beschränkt wird, wobei ein anderes Ausgangsmaterial zwischen der Trennwand und der Innenwand des Balgenbehälters untergebracht ist, wodurch das Ausgangsmaterial von den Wülsten der Wand des Balgenbehälters (20) ferngehalten wird.

13. Gerät zum Einschließen von körnigem Ausgangsmaterial, das radioaktiven Abfall und synthetisches Gesteins-zwischenmaterial enthält, in Balgenbehälter (20) innerhalb eines zylindrischen Kanisters (38), wobei das Gerät Mittel (24) zum Einschütten des körnigen Materials in den Balgenbehälter (20) aufweist, Mittel zum Verschließen des Balgenbehälters mit einem Deckel (29), Mittel (36) zum Befördern von Balgenbehältern nacheinander zu einer Preßstation, in der der Behälter erhitzt wird und zum axialen Komprimieren des Behälters axialer Druck auf ihn ausgeübt wird, dadurch gekennzeichnet, daß die Preßstation einen aufwärts verschiebbaren Stempel (37) zum Aufnehmen eines Balgenbehälters (20) aufweist, Mittel zum Anbringen eines zylindrischen Kanisters (38), so daß sein offenes Ende abwärts gegen den Stempel gerichtet ist, Mittel (37) zum Pressen eines auf dem Stempel sitzenden Balgenbehälters in den Kanister, obere Widerlagermittel (40) aus Feuerfestmaterial, die einen festen Anschlag bilden, Heizmittel (41) um eine erhöhte Temperatur in dem Balgenbehälter (20) aufrecht zu erhalten, während der Druck angewandt wird, um das Ausgangsmaterial in dem Balgenbehälter zu verdichten und um den Balgenbehälter etwas zu erweitern, um ihn im Kanister zu verklemmen, und daß die Heizmittel (41) geeignet sind, eine Erhitzung in einer Reihe von Zonen innerhalb des Behälters hervorzurufen, entsprechend einer Reihe von Balgenbehältern, die, einer unterhalb des anderen, in ihn eingesetzt sind, und daß der Kanister geeignet ist, entfernt und geschlossen zu werden, wenn eine Reihe von Balgenbehältern verdichtet und darin gesichert worden sind.

14. Gerät nach Anspruch 13, wobei das Gerät eine Vorwärmstation (31) aufweist, die geeignet ist, den Inhalt der Balgenbehälter (20) auf eine im wesentlich gleichmäßige höhere Temperatur zu bringen, und Mittel (36) zum Überführen eines vorgewärmten Balgenbehälters zur Preßstation (37).

15. Gerät nach Anspruch 14, wobei die Vorwärmstation (31) eine sich aufwärts erstreckende zylindrische Induktionsheizzone aufweist, die Tragteile (32, 33) aus Feuerfestmaterial besitzt, um einen Stapel Balgenbehälter in dieser Zone zu halten, und Mittel zum Handhaben der Balgenbehälter (20), wodurch diese kalt in den oberen Teil der zylindrischen Zone eingeführt werden und nach dem Vorwärmen am Unterende der Zone entfernt werden, wobei die Überführungsmittel in waagerechter Richtung wirken, um den vorgewärmten Balgenbehälter auf den verschiebbaren Stempel zu überführen.

16. Gerät nach einem der Ansprüche 13 bis 15, wobei der verschiebbare Stempel (37) eine Deckschicht (35) aus Feuerfestmaterial aufweist, die auf dem Kopf des Stempels duch einen Zapfen festgelegt ist.

17. Gerät nach einem der Ansprüche 13 bis 16 mit Kompressionsmitteln (30) zum axialen

Komprimieren jedes Balgenbehälters, im wesentlichen bei Umgebungstemperatur, nachdem der Balgenbehälter (20) mit dem Deckel (29) verschlossen worden ist.

18. Gerät nach einem der Ansprüche 13 bis 17 mit einem vertikal verschiebbaren feuerfesten Block (46), der so angeordnet ist, daß er den Kanister (38) umgibt, wobei der feuerfeste Block einen leicht konischen Hohlraum hat, der an seiner engsten Stelle mit nicht mehr als Gleitsitz Über den Kanister paßt, wobei der feuerfeste Block geeignet ist, den Kanister dort, wo ein Balgenbehälter zusammengedrückt wird, gegen radiale Erweiterung zu stützen, und wobei der feuerfeste Block danach abwärts verschiebbar ist.

19. Gerät nach Anspruch 18, wobei der feuerfeste Block Windungen einer Induktionsheizspule (41) enthält.

20. Gerät nach Anspruch 19, wobei der feuerfeste Block aus einer Reihe von ineinandergreifenden feuerfesten Segmenten (46a, 46b) aufgebaut ist, die innerhalb einer äußeren zylindrischen Scale (47) untergebracht sind.

Revendications

1. Procédé de confinement et de densification d'un matériau particulaire contenant des déchets radioactifs et d'un matériau précurseur d'une roche synthétique, le procédé comprenant le déversement du matériau particulaire (28) dans un récipient (20) en forme de soufflet ayant une forme générale cylindrique et une paroi latérale (27) comprenant une partie en forme de sourfflet et constituée d'un matériau résistant à la chaleur et à la radioactivité, la fermeture du récipient en forme de soufflet par un couvercle (29), la disposition du récipient en forme de soufflet sur un poston (37) mobile vers le haut et ayant une partie réfractaire de surface (35), le maintien d'une pression sensiblement axiale sur le récipient en forme de soufflet, l'application de chaleur et le maintien d'une température suffisamment élevée dans le récipient en forme de soufflet pendant un temps suffisant pour que le matériau particulaire soit densifié dans le récipient en forme de soufflet et que ce récipient subisse une compression axiale, l'arrangement étant tel que la déformation du récipient en forme de soufflet s'effectue en direction axiale, et l'enlèvement du récipient en forme de soufflet après la fin de l'étape de densification, caractérisé en ce que la pression est appliquée en direction sensiblement axiale au récipient (20) uniquement par l'action d'un piston ascendent (37) qui repousse le récipient contre une butée fixe (40), la partie en forme de soufflet n'ayant pas de support qui l'entoure.

2. Procédé selon la revendication 1, caractérisé en ce que, après la fermeture du récipient en forme de soufflet (20) par le couvercle (29), un préchauffage préliminaire du récipient est réalisé pratiquement sans application d'une pression axiale au récipient en forme de soufflet.

3. Procédé selon la revendication 2, dans lequel le récipient en forme de soufflet est constitué d'un métal et le préchauffage est réalisé par chauffage par induction (41) pendant une période de quelques heures afin que le récipient en forme de soufflet (20) et son contenu soient pratiquement à une température uniforme qui est assez élevée mais suffisamment inférieure à la température qui doit être obtenue pendant l'étape ultérieure de compression à chaud pour que le récipient en forme de soufflet ait, à la température de préchauffage, une résistance mécanique nettement supérieure à celle qu'il a à la température de compression à chaud.

4. Procédé selon l'une quelconque des revendications précédentes, dans lequel l'étape de compression à chaud est réalisée alors que la température du récipient en forme de soufflet et de son contenu a été portée à 1200°C environ.

5. Procédé selon l'une quelconque des revendications précédentes, dans lequel, juste après disposition du couvercle (29) sur le récipient en forme de soufflet (20), une compression axiale est appliquée au récipient en forme de soufflet, la température ne dépassant pas 800°C.

6. Procédé selon la revendication 5, dans lequel la compression axiale exercée pendant l'étape de compression de la revendication 5 est d'au moins $20{,}68 \cdot 10^6$ Pa.

7. Procédé selon l'une quelconque des revendications précédentes, dans lequel le matériau particulaire a une dimension particulaire qui ne dépasse pas 2 mm et peut être facilement versé, le matériau particulaire étant préparé par pulvérisation d'une suspension dans un four rotatif (23).

8. Procédé selon l'une quelconque des revendications précédentes, dans lequel la compression axiale du récipient en forme de soufflet à température élevée est réalisée par introduction du récipient en forme de soufflet (20) sur le piston mobile (37) dans un boîtier cylindrique (38) ayant une extrémité ouverte dépassant vers le bas, et dans lequel le récipient en forme de soufflet est introduit avec du jeu avant l'étape de compression à chaud, le récipient en forme de soufflet subissant une petite dilatation radiale pendant la compression si bien qu'il est comprimé contre le paroi interne du boîtier dans laquelle il est emmanché à force.

9. Procédé selon la revendication 8, dans lequel le boîtier cylindrique (38) est allongé, et une série de récipients en forme de soufflet (20) est comprimée vers le haut à raison d'un récipient à la fois dans le boîtier cylindrique, et, lorsque le boîtier a été pratiquement rempli, il est fermé de manière étanche et retiré avant stockage.

10. Procédé selon l'une des revendications 8 et 9, comprenant la disposition d'un block d'un matériau réfractaire (46) ayant un trou légèrement évasé au-dessus du boîtier (38) pendant la compression à chaud d'un récipient en forme de soufflet (20) placé à l'intérieur, le trou évasé, dans sa partie de plus petit

diamètre, permettant au plus un ajustement avec coulissement sur le boîtier si bien que toute tendance du boîtier à se déformer vers l'extérieur est empêchée par le bloc réfractaire, le bloc réfractaire (46) étant déplacé vers le bas par rapport au boîtier (38) après la compression à chaud d'un récipient en forme de soufflet.

11. Procédé selon la revendication 10, dans lequel le bloc réfractaire (46) a un enroulement (41) de chauffage par induction qui le traverse.

12. Procédé selon l'une quelconque des revendications précédentes, comprenant l'utilisation d'une cloison cylindrique (26) à l'intérieur du récipient en forme de soufflet (20), et le confinement du matériau particulaire dans la zone se trouvant à l'intérieur par rapport à la séparation cylindrique, une autre quantité du matériau particulaire étant placée entre la séparation et la paroi interne du récipient en forme de soufflet si bien que ce matériau est exclu des ondulations de la paroi du récipient en forme de soufflet (20).

13. Appareil d'enrobage d'un matériau particulaire contenant des déchets radioactifs et un matériau précurseur d'une roche synthétique, dans des récipients en forme de soufflet (20) placés à l'intérieur d'un boîtier cylindrique (38), l'appareil comprenant un dispositif (24) de déversement du matériau particulaire dans un récipient en forme de soufflet (20), un dispositif de fermeture étanche du récipient en forme de soufflet avec un couvercle (29), un dispositif (36) destiné à déplacer les récipients en forme de soufflet successivement vers un poste de compression auquel le récipient est chauffé et une pression axiale est appliquée au récipient afin que celui-ci subisse une compression axiale, caractérisé en ce que le poste de compression comporte un piston (37) mobile vers le haut et destiné à supporter un récipient en forme de soufflet (20), un dispositif de montage d'un boîtier cylindrique (38) avec une extrémité ouverte dirigée vers le bas, vers le piston, un dispositif (37) destiné à comprimer vers le haut un récipient en forme de soufflet supporté sur le piston, à l'intérieur du boîtier, un support réfractaire supérieur (40) destiné à jouer le rôle d'une butée fixe, un dispositif (41) de chauffage destiné à maintenir une température élevée dans le récipient en forme de soufflet (20) alors qu'une pression est appliquée afin qu'elle provoque une densification de la matière particulaire dans le récipient en forme de soufflet et une dilatation légère du récipient en forme de soufflet afin que celui-ci se coince dans le boîtier, le dispositif de chauffage (41) étant destiné à assurer le chauffage dans une série de zones placées à l'intérieur du récipient et correspondant à une série de récipients en forme de soufflet introduite en série à raison d'un récipient au-dessous de l'autre, le boîtier étant destiné à être retiré et fermé de manière étanche lorsqu'une série de récipients en forme de soufflet a été densifiée et fixée à l'intérieur.

14. Appareil selon la revendication 13, dans lequel l'appareil comporte un poste de préchauffage (31) destiné à mettre le contenu des récipients en forme de soufflet (20) à une température élevée sensiblement uniforme, et un dispositif (36) de transfert d'un récipient préchauffé en forme soufflet vers le poste de compression (37).

15. Appareil selon la revendication 14, dans lequel le poste de préchauffage (31) comporte une zone chauffée par induction, cylindrique et tournée vers le haut, ayant un support réfractaire (32, 33) destiné à porter une pile de récipients en forme de soufflet dans ladite zone, et un dispositif de manutention des récipients en forme de soufflet de manière que ces récipients (20) soient introduits à froid à partie supérieure de la zone cylindrique et soient retirés après préchauffage à la partie inférieure de la zone, le dispositif de transfert travaillant en direction horizontale et assurant le transfert du récipient préchauffé en forme de soufflet vers le piston mobile.

16. Appareil selon l'une quelconque des revendications 13 à 15, dans lequel le piston mobile (37) a un revêtement réfractaire (35) positionné sur la tête du piston par une saillie.

17. Appareil selon l'une quelconque des revendications 13 à 16, comprenant en outre un dispositif (30) de compression axiale pratiquement à température ambiante de chaque récipient en forme de soufflet (20) après que celui-ci a été fermé de manière étanche par le couvercle (29).

18. Appareil selon l'une quelconque des revendications 13 à 17, comprenant en outre un block (46) mobile verticalement, formé d'un matériau réfractaire et disposé afin qu'il entoure le boîtier (38), le bloc réfractaire ayant un trou légèrement évasé dont la partie la plus étroite ne permet pas plus qu'un emmanchement coulissant sur le boîtier, le bloc réfractaire étant destiné à supporter le boîtier en l'empêchant de se dilater radialement vers l'extérieur à l'emplacement auquel un récipient en forme de soufflet est en cours de compression, le bloc réfractaire étant ensuite déplaçable vers le bas.

19. Appareil selon la revendication 18, dans lequel le bloc réfractaire contient des spires d'un enroulement de chauffage par induction (41).

20. Appareil selon la revendication 19, dans lequel le bloc réfractaire est formé d'une série de segments réfractaires emboîtés (46a, 46b) disposés dans une enveloppe cylindrique externe (47).

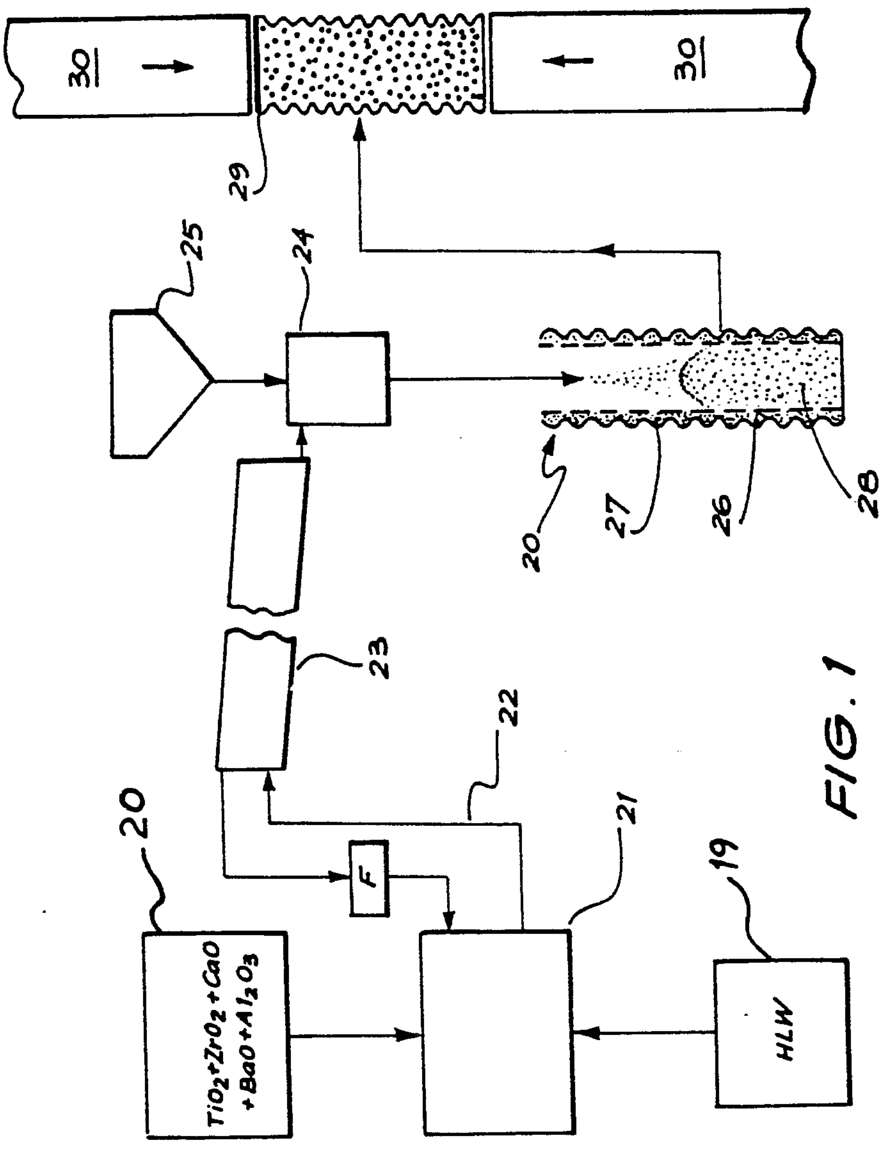
30
30
29
25
24
20
27
26
28
23
22
21
19
20
TiO_2+ZrO_2+CaO
$+BaO+Al_2O_3$
F
HLW

FIG. 1

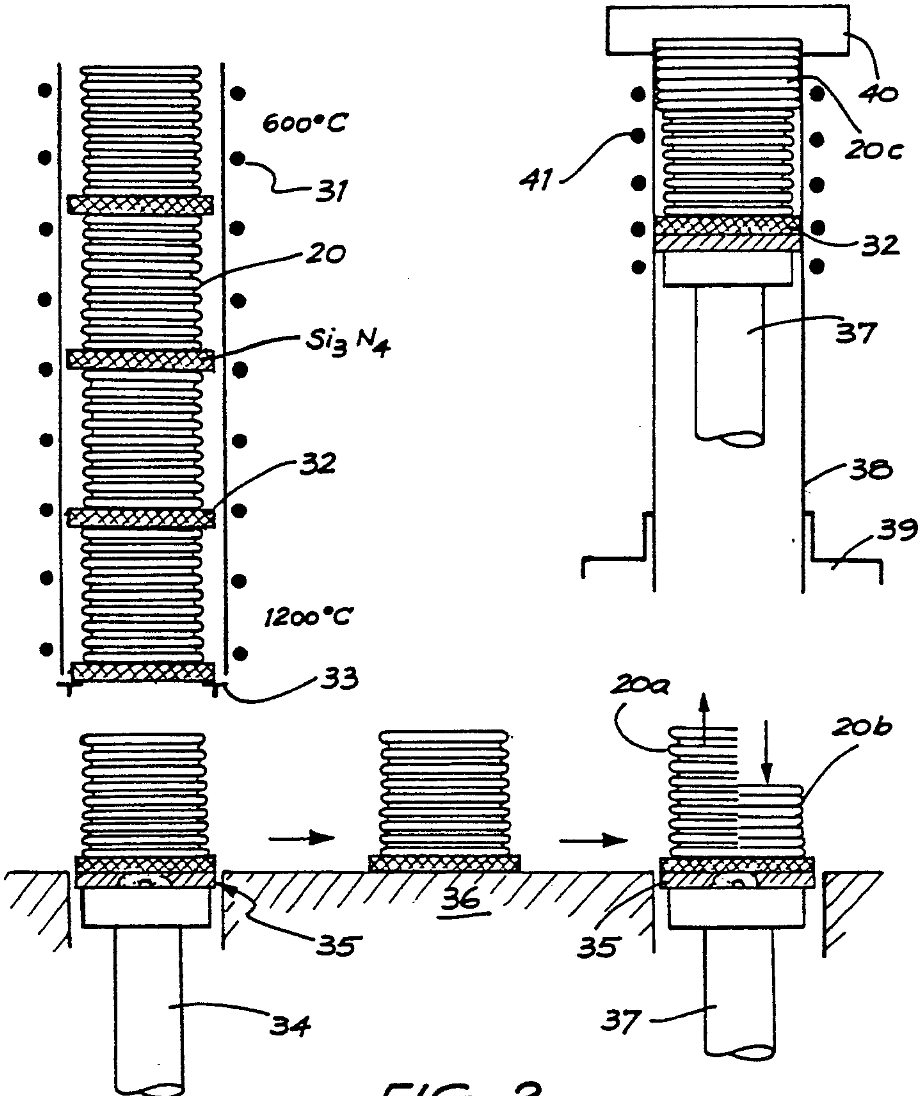
600°C
31
20
Si_3N_4
32
1200°C
33
40
20c
41
32
37
38
39
20a
20b
35
36
35
34
37

FIG. 2

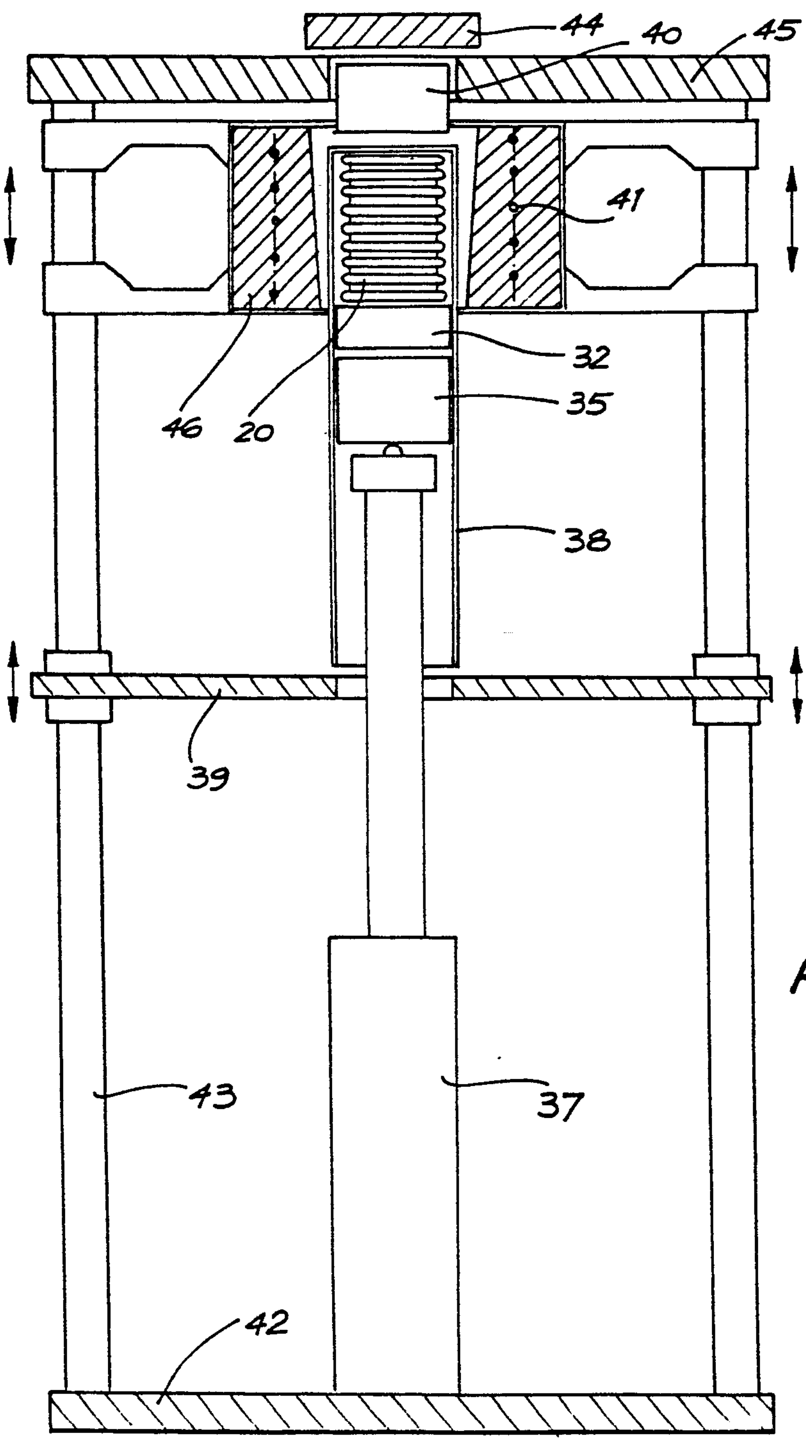

FIG. 3

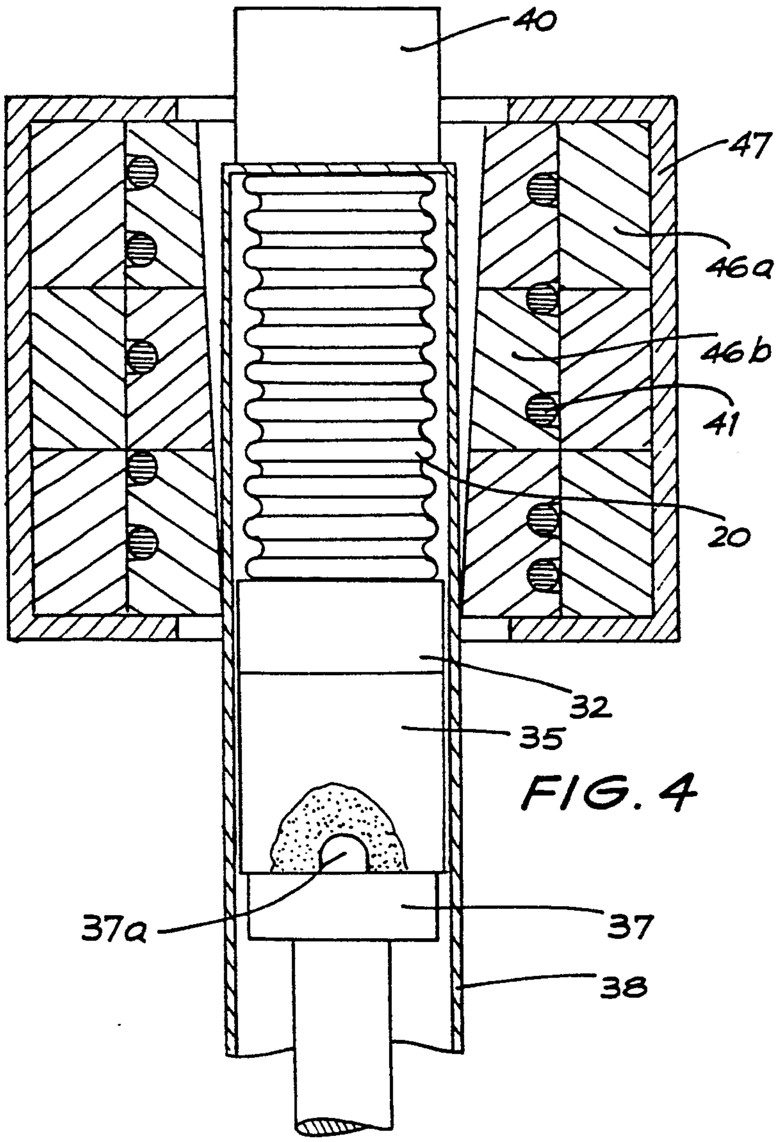

FIG. 4

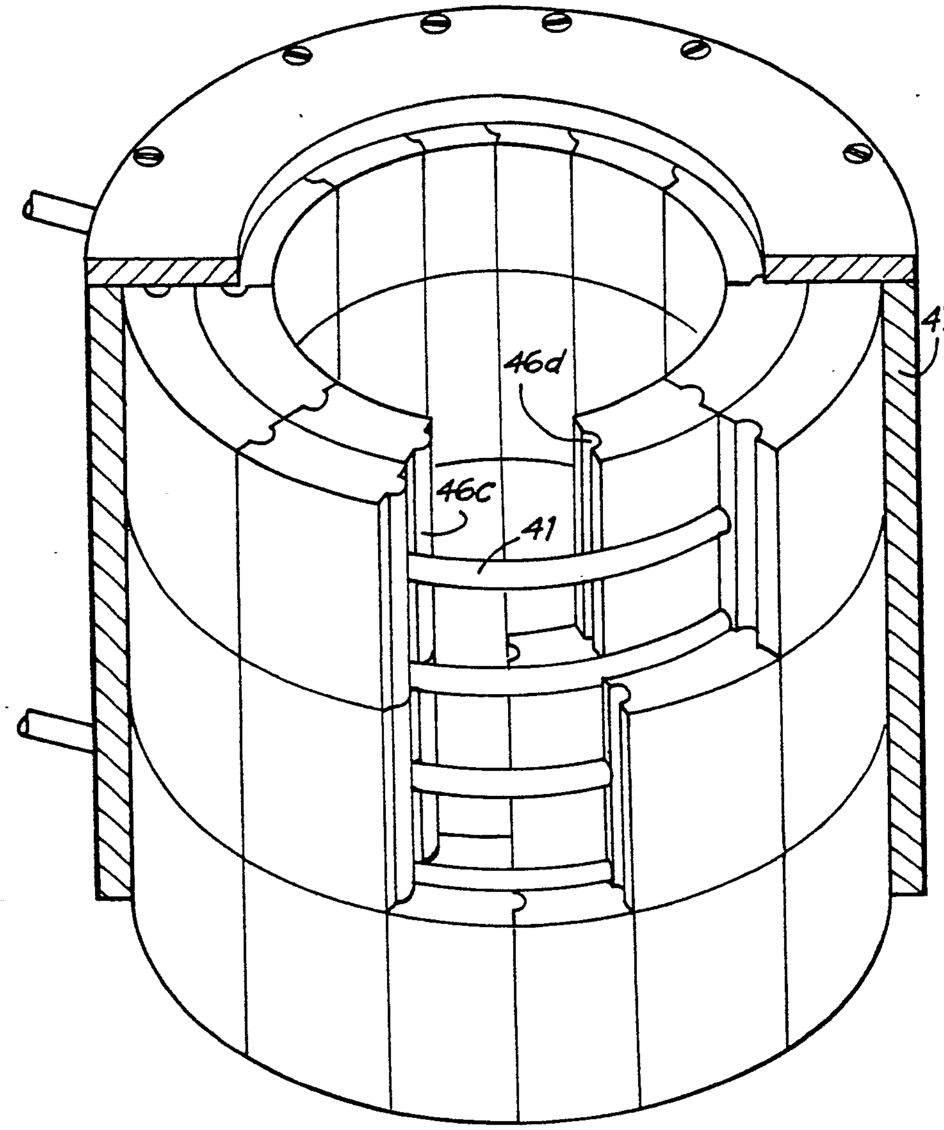

FIG. 5