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(54) METHOD OF TREATING RADIOACTIVE WASTE

(71) We, ASEA AKTIEBOLAG, a Swedish Company of Västerås, Sweden, 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:–

This invention relates to a method of surrounding a body containing radioactive waste from nuclear fuel with a casing of material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation.

In the method of reprocessing of radioactive waste from nuclear reactors which is being used today, the high-level waste is obtained in a strong nitric acid solution. The dominating radioactive substances in the waste during the first centuries are strontium-90 and cesium-137. The waste also contains, among other things, minor amounts of uranium and plutonium and transuranic elements, which have considerably greater half-lives than strontium-90 and cesium-137. Those skilled in the art are generally of the opinion that it is advantageous, after a suitable period of cooling, to convert the liquid high-level waste into a solid. A solid product of good chemical resistance is aimed at. It should be stable to leaching out of radioactive material, contained therein, by water. It should withstand heating produced by the fission products and it should also withstand stresses arising during management and transportation. Among materials that have been proposed for use for the containment may be mentioned glass, such as boron silicate glass and phosphate glass, quartz, titanium dioxide, certain zeolites and other minerals existing in nature, particularly those having the ability to retain gases.

In our U.K. Patent Specification No. 1 588 350 (Application No. 45379/77), there is disclosed in claim 1 a method of anchoring radioactive waste from nuclear fuel in a body resistant to leaching by water, the method being characterised in that a mass containing radioactive constituents and constituents which are resistant to leaching by water or which, when heated, form constituents resistant to leaching by water, is subjected to isostatic pressing enclosed in a casing at a pressure and temperature necessary for the formation of a coherent, dense body of the mass. However, in this known method, the said mass is not surrounded by a casing of a material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation.

In another known method of containing high-level waste in glass, the waste is evaporated and calcined and additives are added thereto which, when heated to 1000 to 1200°C, result in a glass melt. The melt is filled into tight steel containers which are transferred to a cooled and supervised storage plant. Calcination of high-level waste can take place at a temperature of the order of magnitude of from 300° to 500°C and results in the waste products being transformed into oxides.

To increase the corrosion resistance of the containers mentioned in the preceding paragraph after the final disposal of the high-level waste in rock, proposals have been made, among other things, to coat the containers with a thick layer of gold, for example a layer having a thickness of a few mm. It has been proposed to apply the gold electrolytically. Silver and copper have also been mentioned as materials for the layer. It is obvious that there are practical problems in applying thick corrosion-protecting coatings electrolytically on bodies of the size which would be used in this case. In addition, coatings of metals of said kind in thicknesses of the intended sizes can easily be mechanically damaged during transportation and other management.

If a layer of a corrosion-protecting material, such as a suitable ceramic material, is applied around a radioactive body and is pressed and sintered at high temperature in a conventional apparatus, there is a risk that gaseous constituents may escape from the radioactive body and spread to the surrounding environment. Further, it is impossible to achieve a finished layer of the required thickness, which has a uniformly high density and resistance to leaching by water.

The present invention aims to provide a method of surrounding a body containing radioactive waste from nuclear fuel with a corrosion-protecting layer which can be applied while maintaining complete control of the radioactive waste, thus eliminating any risk of gaseous constituents from the radioactive body spreading to the environment and, at the same time, counteracting the formation of gaseous constituents. Further, the invention aims to achieve under practical conditions corrosion-protecting layers of desired thicknesses and with high uniform density and resistance to leaching by water, which layers are difficult to penetrate by radioactive radiation.

According to the invention, a method of surrounding a body containing radioactive waste from nuclear fuel, i.e. high-level waste separated during reprocessing of nuclear fuel, with a casing of a material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation, is characterised in that the body, surrounded by the material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation and enclosed together with this material in a gas and liquid impervious capsule, is subjected to isostatic pressing at a pressure and temperature necessary for the formation of a coherent, dense casing of the material. The capsule is preferably evacuated prior to sealing.

The body containing radioactive waste may consist, among other things, of a solidified glass melt, described above, containing high-level waste. The body may further consist of a sintered body of high-level waste, possibly anchored in an ion exchanger and possibly mixed with a material resistant to leaching by water, such as one or more oxides of the kind normally included in glass of various kinds and in minerals, for example, SiO_2, B_2O_3, Al_2O_3, MgO, alkaline oxides, alkaline earth oxides, TiO_2, ZrO_2, Fe_2O_3 or minerals existing in nature and built up from silicates, aluminates, chromates or titanates. The body may also consist of a densely-pressed container of ceramic material in which radioactive waste has been arranged in spaces provided for this purpose. The body need not consist of one single coherent unit. It may also consist of several separate, minor bodies placed adjacent to each other, or of a particulate mass.

Suitably the casing, prior to being subjected to the isostatic pressure, is formed of a particulate, corrosion-resistant material.

Among suitable corrosion-resistant materials in the casing according to this embodiment may be mentioned various metallic materials such as tantalum, titanium, zirconium, tungsten, molybdenum and mixtures of at least two of these metals, copper and an alloy containing 70 *per cent* by weight copper and 30 *per cent* by weight nickel, various ceramic materials such as oxides of the kind normally comprised in glass of various kinds and in minerals, such as TiO_2, Al_2O_3, SiO_2, B_2O_3, MgO, ZrO_2 and Cr_2O_3, silicates, aluminates, chromates and titanates. As will become clear hereinafter, it may be suitable, particularly when using several layers of corrosion-resistant materials arranged outside each other, to make one of the layers of a noble metal. Among preferable materials may be mentioned tantalum, titanium and zirconium. The grain size of the corrosion-resistant materials is preferably below 1 mm. The thickness of the casing of the corrosion-resistant material in sintered state suitably amounts to at least 2 mm but can be made considerably thicker, for example several centimetres or several decimetres.

According to another embodiment of the method of the invention, the body containing radioactive waste is surrounded by elements of a metallic material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation, which elements – at least partly and preferably for the main part – are non-pulverous and are considerably larger than particles in pulverulent materials, the body then being enclosed together with the elements in a capsule and subjected to isostatic pressing at a pressure and temperature necessary for the formation of a coherent, dense casing of the elements. By using elements in this way, which at least for the main part are non-pulverous and thus considerably larger than particles in pulverulent materials, the shrinkage, that is the change of shape, can be considerably reduced during the pressing, which means that the equipment can be made correspondingly smaller. Another advantage is that evacuation of gases from the capsule prior to sealing is facilitated, since the elements are substantially free from voids.

The elements can at least for the main part consist of shaped elements, for example elements shaped as discs, rings, balls, cylinders or rods. In order to facilitate the compression, they are preferably symmetrically arranged with intermediate spaces around the body. It is, however, possible to use irregular elements, for example lumps. Particularly in the latter case, such elements are combined with powder particles to achieve the required filling. At least for the main part the elements may consist of sheet metal elements or of cast, forged or sintered bodies.

Among suitable materials for the casing in the last-mentioned embodiment, when particularly good corrosion-resistance to environments for long-term storage is aimed at, may be mentioned a plurality of metallic materials such as tantalum, tatanium, zirconium, tungsten, molybdenum and mixtures of at least two of these metals, copper, stainless steel, an alloy containing 70 *per cent* by weight of copper and 30 *per cent* by weight of nickel, and an alloy containing 94 *per cent* by weight of copper and 6 *per cent* by weight of aluminium. The exemplified materials, of course, also provide radiation protection, the efficiency of which is dependent on the nature of the material and the thickness of the casing.

Among suitable materials for the casing in the last-mentioned embodiment, when particularly good radiation protection is aimed at, may be mentioned steel, for example a roller or cast steel with a carbon content of from 0.1 to 0.5 *per cent* by weight and copper. Of course, such materials contribute to protect the enclosed body against corroding influences from the environment.

The thickness of the finished casing in the last-mentioned embodiment preferably amounts to at least 10 cm if a casing is aimed at which is sufficiently difficult to penetrate by radioactive radiation.

In certain cases it may be suitable in different embodiments of the invention to build up the casing of several layers of materials with different corrosion resistance to different environments, for example layers with different electrolytic potential or a layer which provides particularly good protection against acid waters over or under a layer which provides particularly good protection against basic waters.

Particularly when using several layers of corrosion-resistant materials in the casing, it may be suitable to make one layer of noble metal such as gold, platinum or platinum-iridium. The noble metal can suitably be applied electrolytically if it is applied as the outermost layer. If applied between two other layers of corrosion-resistant materials which are applied simultaneously on the body by isostatic pressing, it may be possible to place a thin foil of the noble metal between said two layers before pressing, for example a gold foil between a titanium layer located inside it and a titanium layer located outside it.

The use of several layers of different materials is suitable if the casing is built up to a considerable extent of a material which provides efficient radiation protection but unsatisfactory corrosion protection. It is then suitable to build into the casing one or more layers of a material with better corrosion resistance than the radiation protecting material. Among suitable corrosion-resistant materials for such layers may be mentioned the materials which have been mentioned before as being suitable for manufacture of casings for which a particularly good corrosion resistance is aimed at. The corrosion-resistant material can be applied in the form of a powder mass. The application may then take place at the same time as the radiation-protecting material is applied by isostatic pressing or in a separate pressing operation after the radiation-protecting material has been applied. The thickness of the layer of the corrosion-resistant material in sintered state then suitably amounts to at least 1 mm. It can also be applied otherwise, for example electrolytically.

The capsule in which the body with radioactive waste and the casing-forming material is contained consists, of course, of a material which is yielding under the pressing conditions. The capsule may, among other things, be made from one or more sheets of steel, tantalum, titanium, zirconium or alloys based on these metals, for example, "Zircaloy" (Trade Mark). In certain cases it is also possible to use a capsule of quartz.

The pressure during the isostatic pressing of the casing around the body containing radioactive waste suitably amounts to at least 100 bar and is preferably at least 500 bar. The range between 500 and 3000 bar is particularly suitable. The temperature is, of course, dependent on what casing-forming material(s) is/are used. However, for most practical casing-forming materials the temperature is at least 500°C. A suitable temperature for application of a radiation-protecting casing of steel without simultaneous application of a corrosion-protecting layer is 1000°C. A suitable temperature when using tantalum is about 1400°C, when using titanium about 950°C, when using zirconium about 1000°C, when using tungsten about 1500°C and when using molybdenum about 1350°C.

The invention will now be described, by way of example, with reference to the accompanying drawings, in which

Figure 1 is a schematic sectional view of a capsule containing a body of radioactive waste and a mass of corrosion-resistant material,

Figure 2 is a schematic sectional view of a body containing radioactive waste during the course of its production,

Figure 3 is a schematic sectional view of the body of Figure 2 in finished state and placed in a capsule in which it is surrounded by a material which is to form a radiation-protecting casing, and

Figure 4 is a partly sectioned side view of a high-pressure furnace in which the corrosion-resistant material according to Figure 1 and the radiation-protecting material according to Figure 3, respectively, are compressed and sintered to form a casing on the body of radioactive waste.

Figure 1 shows a cylinder 1 of a solidified melt of boron silicate glass containing high-level waste from a plant for reprocessing fuel used in nuclear reactors. The cylinder is surrounded by a tight container 2 of acid-proof steel. The radioactive waste has been contained using the known method for containment in glass described above. The cylinder may have a diameter of 30 cm and a length of 2 metres.

The container 2 with the cylinder 1 is then placed in a capsule in the form of a cylindrical container 3 of tantalum sheet having a thickness of 2 mm. At the bottom the container contains a layer 4 of tantalum powder with a grain or particle size of less than 200 microns. The space 5 between the envelope surfaces of containers 2 and 3 is filled with the same tantalum powder. A layer 6 of the tantalum powder is also placed on top of cylinder 2. The thickness of the layers of tantalum powder may be 10 mm. When the cylinder 3 has been packed in the manner described, a lid 7 of tantalum sheet is welded onto the cylinder. The lid is provided with a tube 8 which can be connected to a vacuum pump for evacuating the capsule. After evacuation, the capsule is sealed by heating and fusing the tube immediately above the upper surface of the lid.

In Figure 4, the numeral 32 designates a displaceable press stand. It is supported by wheels 33 running on rails 34 on a floor. The press stand is of the type which consists of an upper yoke 36, a lower yoke 37 and a pair of spacers 38 which are held together by a prestressed strip sheath 39. The press stand is movable between the position shown in Figure 4 and a position where the stand surrounds a high-pressure chamber 42. The high-pressure chamber 42 is supported by a column 49 and contains a high-pressure cylinder which is built up from an inner tube 50, a surrounding prestressed strip sheath 51 and end rings 52 which axially hold together the strip sheath and constitute a suspension device by which the high-pressure chamber is attached to the column 49. The chamber 42 has a lower end closure 53 projecting into the tube 50 of the high-pressure cylinder. In the end closure 53 there is a slot in which there are arranged a sealing ring 54, a channel 55 for the supply of pressure medium, suitably argon or helium, and a channel 56 for cables for feeding heating elements 57 for the heating of the furnace. The elements 57 are supported by a cylinder 58 resting on a thermally insulating bottom 59, which protrudes into a thermally insulating sheath 60. The chamber 42 has an upper end closure comprising an annular portion 61 with a sealing ring 62 which seals against the tube 50. The sheath 60 is suspended from portion 61 and connected in gas-tight manner thereto. The upper end closure also comprises a lid 63 for closing the opening in portion 61, which is usually permanently mounted in the high-pressure cylinder. The lid is provided with a sealing ring 64 sealing against the inner surface of portion 61 and with a thermally insulating lid 65 which, when the high-pressure chamber is closed, projects into cylinder 60 and constitutes part of the thermally insulating shell which surrounds the furnace space 66. The lid 63 is fastened to a bracket 67 which is carried by a raisable, lowerable and rotatable operating rod 68. Yokes 36 and 37 take up the compressive forces acting on the end closures 53 and the lid 63 when pressure is applied to the furnace space.

When the capsule 3 has been placed in the furnace space 66, the lid 63 first having been lifted up and then lowered for closing the furnace space, the pressure and the temperature are successively increased to around 1000 bar and about 1400°C, respectively, and are maintained at these values for about two hours, when the desired density and sintering are obtained in the tantalum layers 4, 5 and 6. The capsule with the enclosed material is then allowed to cool, whereafter the pressure is reduced to atmospheric pressure and the capsule is removed from the furnace. Normally, the capsule is allowed to remain on the pressed product when it is deposited for long-term storage.

According to another embodiment, the container 2 with the glass cylinder 1 can be placed in a container of titanium where it is embedded in two layers of titanium powder arranged one outside the other and a layer of gold in the form of a film arranged therebetween.

Figure 3 shows another example of a body 11 containing radioactive waste. It can be manufactured in a manner which is schematically shown in Figure 2. A prepressed cylinder 12 of a corrosion-resistant material, for example aluminium oxide, is provided with holes 13 and has surface-ground ends. The cylinder rests on a prepressed surface-ground bottom plate 14 and is provided with a similarly surface-ground lid 15 so as to cover the holes 13. The bottom plate and the lid are of the same material as the cylinder and are manufactured by isostatic pressing, for example in a device of the kind shown in Figure 4, at 1000 bar and 1350°C for a time of 1 hour. Spent fuel rods 16 from a nuclear reactor are placed in the holes 13. The cylinder with its lid and bottom is placed in a container 17 of steel sheet having a thickness of about 3 mm with a welded-on lid 18. The lid 18 is provided with a tube 19 which can be connected to a vacuum pump for evacuation of the container 17. The space between the inner walls of the container 17 and the cylinder 12 and between the lid 15 and the bottom 14 and the cylinder 12 is filled with powder 20 of the same corrosion-resistant material, of which the cylinder, the bottom and the lid are manufactured. After evacuating the container 17 it is sealed by fusing the tube 19. The container with its content is then subjected to an isostatic pressing at 1000 bar and 1350°C in the device shown in Figure 4. This results in a connection between the end surfaces of the cylinder 12, bottom 14 and the lid 15, a densification of the powder 20 and in the powder 20 being joined to the prepressed bodies 12, 14, and 15 into one single monolithic body 11 (Figure 3). The holes 13 in the body 11, even after the isostatic pressing, have greater dimensions than the outer dimensions of the fuel rods 16. In this way it is possible to avoid stress problems which are caused by the different coefficients of thermal expansion of the different materials included.

As will be clear from Figure 3, the body 11, possibly after removing the container 17, is placed in a capsule 21 comprising a cylindrical container 22 of 5 mm thick steel sheet. The container 22 contains in the bottom a plurality of steel discs 23. In the space between the inner envelope surface of the container 22 and the outer envelope surface of the body 11 there are wound a plurality of turns 24 of steel sheet. Above the body 11 the container 22 contains a plurality of steel discs 25. The steel in the parts 23, 24 and 25, which constitute elements of a material difficult to penetrate by radiation, is of a type having low carbon content and the thickness of the sheet may be 10 mm. The parts 23, 24 and 25, are surrounded in the container 22 by a 10 mm thick layer 27 of a material with better corrosion resistance than steel to environments during long-term storage, for example stainless steel, in the form of a powder with a maximum grain size of 0.7 mm. A lid 26 of steel sheet is mounted on the container 22. The lid is provided with a tube 28 which can be connected to a vacuum pump for evacuating the capsule. After evacuation, the capsule is sealed by fusing the tube 28.

After the capsule 21 has been placed in the furnace space 66 in the high-pressure furnace according to Figure 4, the lid 63 first having been raised and then lowered for sealing the furnace space, the pressure and the temperature are successively raised to about 1000 bar and about 1150°C, respectively, and are maintained at these values for about 2 hours, when the desired density and sintering of the items 23, 24, 25 and 27 are obtained. The capsule with the contained material is then allowed to cool, whereupon the pressure is reduced to atmospheric pressure and the capsule is removed from the furnace. The items 23, 24 and 25 together form a casing around the body 1, which provides an efficient protection against radioactive radiation. The casing may in the exemplified case have a thickness of about 25 cm. The casing also comprises the layer 27 which becomes well anchored to the other layers and provides a good protection against corrosion when the pressed body is deposited for long-term storage.

If the application of the radiation-protecting casing is done in the same pressing operation in which the body containing the radioactive waste is sealed, a suitable temperature is 1350°C.

The sheet steel turns 24 may, for example, be replaced by rings of steel sheet which have somewhat larger holes than the diameter of body 11 and which are stacked on top of each other to form a bundle. They may also be replaced by sector-shaped longitudinal rods which are arranged parallel to the axis of the body 11 and with intermediate gaps to facilitate an efficient anchoring thereof to the body during the pressing. The turns 24 may also be replaced by a cast cylinder of the radiation protecting material. Similarly, each bundle of discs 23 and 25 may be replaced by one or more cast and relatively thick discs. The parts 23, 24 and 25 may also be replaced by geometrically regular bodies, for example cylindrical or spherical bodies which are packed around the body 11. Particularly when using elements in the form of lumps of irregular shape it may be suitable, as mentioned before, to use simultaneously filling material in powdered state.

If the steel in the described example is replaced by, for example, a copper alloy of 70 *per cent* by weight of Cu and 30 *per cent* by weight of Ni or 94 *per cent* by weight of Cu and 6 *per cent* by weight of Al, a casing is obtained in which a good corrosion resistance is particularly aimed at. In this case no layer corresponding to layer 27 need be applied.

As will be clear from the preceding description, the body 11 containing radioactive waste in the cases described in the examples may be replaced by a body consisting of a glass melt containing radioactive waste or a body consisting of a sintered body of radioactive waste and possibly a corrosion-resistant material.

The method according to the invention is not only useful for containment of fuel used in nuclear reactors as described in the examples. It may, for example, also be used for containment of high-level waste from fuel reprocessing for the manufacture of plutonium for nuclear weapons.

WHAT WE CLAIM IS:–

1. A method of surrounding a body containing radioactive waste from nuclear fuel with a casing of a material which is

corrosion-resistant and/or difficult to penetrate by radioactive radiation, characterised in that the body surrounded by the material which is corrosion-resistant and/or difficult to penetrate by radioactive radiation and enclosed together with this material in a gas and liquid impervious capsule, is subjected to isostatic pressing at a pressure and temperature necessary for the formation of a coherent, dense casing of the material.

2. A method according to claim 1, in which the casing, prior to being subjected to the isostatic pressure, is formed of a particulate, corrosion-resistant material.

3. A method according to claim 1, in which the casing-forming material comprises non-pulverous elements of metallic material.

4. A method according to claim 3, in which the elements at least for the main part consist of shaped elements.

5. A method according to claim 4, in which the shaped elements are discs, rings, balls, cylinders or rods.

6. A method according to any of claims 3 to 5, in which the elements at least for the main part consist of sheet elements or cast, forged or sintered bodies.

7. A method according to any of claims 1 to 6, in which the body containing the radioactive waste comprises a dense body, sintered under pressure, of a corrosion-resistant material, in which body the radioactive waste is arranged.

8. A method according to any of claims 1 to 7, in which the casing-forming material consists of tantalum, titanium or zirconium.

9. A method according to any of claims 1 to 7, in which the casing-forming material consists of tungsten or molybdenum.

10. A method according to any of claims 3 to 6, in which the casing-forming material consists of steel.

11. A method according to any of claims 1 to 10, in which the capsule consists of tantalum, tatanium, zirconium or steel.

12. A method according to any of claims 1 to 11, in which the body is coated with several layers of different casing-forming materials arranged one outside the other.

13. A method of surrounding a body containing radioactive waste substantially as herein described with reference to Figures 1 and 4 or Figures 2 to 4 of the accompanying drawings.

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1590108 COMPLETE SPECIFICATION

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

Sheet 1

Fig. 1

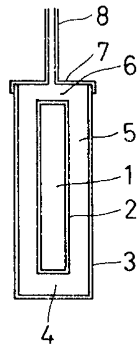
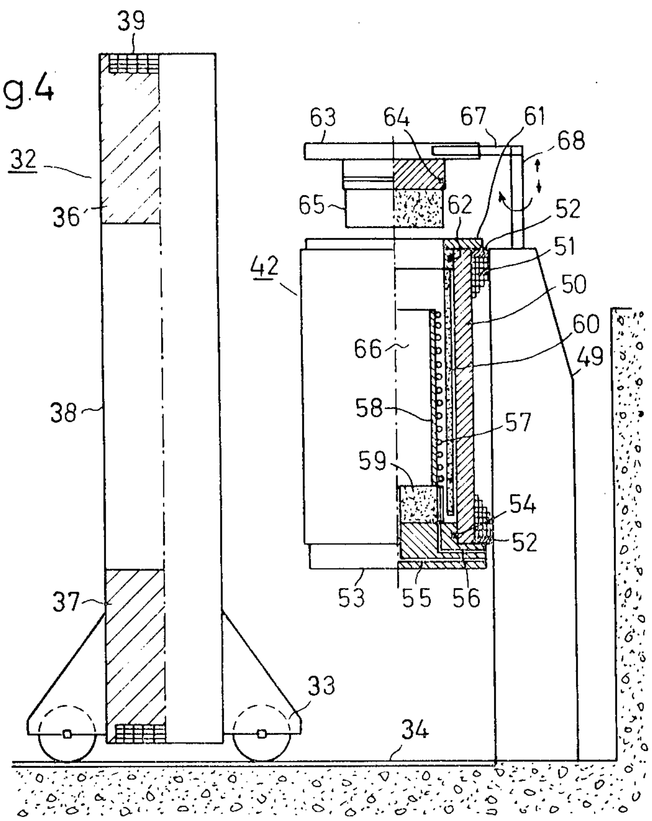


Fig.4



1590108 COMPLETE SPECIFICATION

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

Sheet 2

Fig. 2

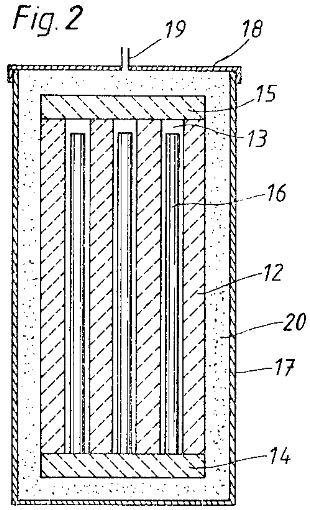


Fig. 3