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(54) **PROCESS FOR PREPARING A TARGET FOR THE GENERATION OF RADIOACTIVE ISOTOPE**

(57) The present invention relates to a process for preparing a two-layer or three-layer target for the generation of radioactive isotopes (radionuclides) by bombardment with accelerated particles; said process comprises the coupling of the layers through the passage of an electric current (pulsed, alternating or direct) and a simultaneous application of pressure. The present invention also

relates to said two-layer or three-layer target comprising a first layer and a support layer with thermal conductivity or a first layer, an intermediate layer and a support layer with thermal conductivity. Said first layer comprises an element/isotope/chemical compound and has a thickness/diameter ratio ≤ 0.25 .

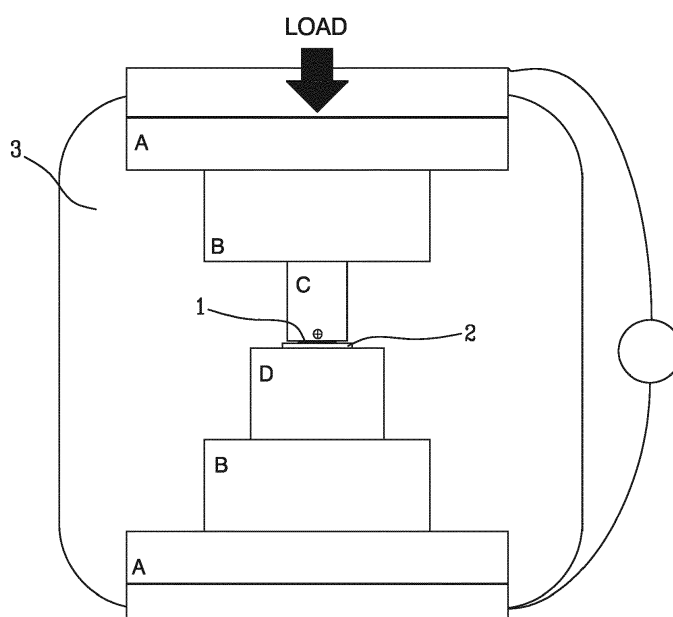


Fig.2

Description

FIELD OF THE INVENTION

[0001] The present invention relates to a process for the manufacture of targets suitable for the production of radioactive isotopes, in particular ^{100}Mo targets for the production of the radioactive isotope $^{99\text{m}}\text{Tc}$ by means of a cyclotron.

BACKGROUND OF THE INVENTION

[0002] Radioactive isotopes are widely used in medicine for diagnostic procedures, for example in the field of oncology, cardiology, endocrinology, etc.

[0003] In the diagnostic field, a radionuclide is used as a tracer in order to have an emission of radiations from the inside of the human body and be able to evaluate the functional aspect of an organ or a drug, which are thus visualised by means of the PET (Positron Emission Tomography) or SPECT (Single Photon Emission Computed Tomography) techniques. The most used radionuclide is $^{99\text{m}}\text{Tc}$, with a half-life of 6 h: every day about 60,000 patients around the world undergo diagnostic procedures with this isotope, which represents about 80% of the total images obtained with techniques of nuclear medicine.

[0004] The production of the radionuclide $^{99\text{m}}\text{Tc}$ mostly takes place from the precursor ^{99}Mo . ^{99}Mo is an unstable isotope with a half-life of 66 h: it is mostly produced in several nuclear plants and subsequently quickly sent to nuclear medicine units where the radioactive decay is exploited for the formation of $^{99\text{m}}\text{Tc}$.

[0005] As a result of the planned closure of some nuclear plants (Chalk River in Canada and Petten in the Netherlands), and thus of the decrease in the availability of the raw material ^{99}Mo , it becomes necessary to use other methods for the production of $^{99\text{m}}\text{Tc}$. One of the most interesting methods provides for the bombardment of a solid ^{100}Mo target with protons accelerated by a cyclotron. The rate of production of $^{99\text{m}}\text{Tc}$ by means of the $^{100}\text{Mo}(p,2n)^{99\text{m}}\text{Tc}$ nuclear reaction is maximum when the proton acceleration energy is equal to 15 MeV: this energy value is easily obtainable with the normal cyclotrons already present in many hospital units, whose present task is to produce other isotopes such as ^{18}F , ^{11}C , ^{15}O , ^{13}N etc.

[0006] At the same time ^{100}Mo is a rather stable isotope and can be easily obtained with an enrichment greater than 99% starting from natural isotopes of molybdenum: it is usually supplied in the material form of an angular powder with a particle size of around 100 μm . Once the target composed of ^{100}Mo has been irradiated and thus contains a minimum amount of $^{99\text{m}}\text{Tc}$ it is quickly dissolved by chemical attack and the different isotopes/elements can be separated and used. Only a minimal part of the isotope ^{100}Mo is converted into $^{99\text{m}}\text{Tc}$, but it is sufficient to make the process economically and environ-

mentally sustainable. It is therefore possible to produce $^{99\text{m}}\text{Tc}$ directly from ^{100}Mo in every hospital unit equipped with a cyclotron suited to the purpose.

[0007] Other radioactive isotopes used for diagnostic purposes are: ^{57}Co obtained from ^{58}Ni , ^{44}Ti obtained from ^{45}Sc , ^{88}Y obtained from ^{88}Sr and still others are continually discovered and used.

[0008] The obtainment of radioactive isotopes requires the preparation of a target, which is definable as the physical object against which the appropriately accelerated physical particles (protons, electrons, alpha particles, beta particles, light ions), also called an accelerated beam, collide.

[0009] The target is usually composed of two disks or layers placed one on top of the other and joined together: a first disk (diameter D_1 and thickness t_1) composed of the chemical element from which the radioactive isotope is obtained by bombardment/irradiation of accelerated particles - for example ^{100}Mo bombarded by protons, from which $^{99\text{m}}\text{Tc}$ is obtained - and a second disk (diameter D_2 and thickness t_2) that acts as a mechanical support, whose function is to dissipate the thermal power generated by the particle beam incident upon the first disk.

[0010] An example of a target is illustrated in Figure 1, in which the number 1 indicates the first layer consisting of the chemical element from which the radioactive isotope is generated by bombardment of particles and the number 2 indicates the second layer that acts as a mechanical support, whose function is to dissipate the thermal power generated by the particle beam incident upon the first disk.

[0011] The incident beam of accelerated particles is schematically represented with the word "beam" and the arrow, whilst the target cooling system is schematically represented with "cooling water" and the respective arrow. An ideal target should meet several requirements:

- the first layer, composed of the suitably chosen chemical element (or the isotopes thereof), for example ^{100}Mo , must be easily predeterminable in terms of size and weight;
- the first layer must be made up of a solid material, sufficiently resistant to thermomechanical stresses and movements/vibrations;
- the second layer must be a solid with high thermal conductivity so as to dissipate the heat produced by the collision of the particles against the first layer;
- the second layer must be compact and cannot be too porous, since it must be able to maintain the vacuum towards the side of the first layer (irradiation zone), whereas on the other side it is subjected to the passage of a fluid that enhances its cooling;
- the second layer must withstand the chemical attack methods necessary for dissolving the first layer after irradiation;
- the second layer must not produce strongly radioactive isotopes in the event that part of the incident ray

of accelerated particles is not shielded by the first layer;

- the second layer must resist creep (i.e. slow deformation due to thermomechanical stress);
- the first and the second layers must have a good continuity, adherence and junction so as to ensure mechanical strength and avoid thermal resistances;
- the layers can also have different shapes, rectangular, ovoidal, ellipsoidal etc.

[0012] The process for preparing the target, on the other hand, should:

- ensure a rate of use of the chemical element/isotope/compound that is close to 100%, allowing the geometry of the first layer to be produced without losses given the high cost of the enriched isotopes;
- be clean, i.e. avoid pollution by spurious elements that could cause problems of radioactivity during and after irradiation or which could change the chemical reactivity during the chemical dissolution phase;
- be flexible in terms of the geometries that may be produced, as far as both the overall size of the target and the mass/thickness of the first layer are concerned;
- be able to process different elements (Mo, Cu, Al, Cr, Ag, Ni, Au, etc.), alloys thereof, compounds thereof, such as carbides, oxides, nitrides, borides and/or phases (metals, ceramics, metal-ceramic composites);
- be sufficiently flexible so as to allow the introduction of new intermediate layers to render the junction between the first and the second layers mechanically stable or impart resistance to corrosion during the subsequent step of chemical dissolution.

[0013] The production of the target must take account of the fact that the starting material from which the first layer (the active part of the object where the nuclear reaction takes place) will be formed is usually, albeit not exclusively, a powder. In some rare cases the starting material can be a metal foil with a controlled composition.

[0014] Over the years, different techniques have been used to produce targets: pressing and sintering in an oven, plasma spray, electrodeposition, brazing, sputtering etc., each with limitations, above all as regards the low rate of use (plasma spray, sputtering), mechanical stability (electrodeposition, pressing and sintering in an oven) and contamination (brazing).

[0015] The methods of pressing and sintering in an oven do not ensure a sufficient mechanical strength: the prolonged sintering times result in the growth of intrinsically fragile intermetal layers which severely impair the solidity of the layer.

[0016] In contrast, the methods which make use of plasma spray, sputtering or electrodeposition have a low coefficient of use: a large part of the enriched isotope is lost in the solution (electrodeposition) or is not deposited

solely on the target (plasma spray, sputtering).

[0017] Finally, brazing could be the best method in that it has a rate of use of the element/isotope that is close to 100% and the junctions are mechanically very resistant: unfortunately, however, brazing alloys are composed of elements with a high atomic number (Z) (silver, platinum, palladium, tin, zinc, cadmium, lead), which, following the irradiation phase, can transmute into highly radioactive isotopes that are difficult to control.

[0018] Thus, in the field there remains a need to provide a process for the production of a target suitable for bombardment with particles, in particular protons generated by a cyclotron, which can overcome the above-illustrated problems typical of the known techniques in the field.

SUMMARY OF THE INVENTION

[0019] The present invention relates to a process for preparing a two-layer or three-layer target for the generation of radioactive isotopes (radionuclides) by bombardment with accelerated particles, in particular protons generated by a cyclotron. The radioactive isotopes obtained starting from the target prepared with the process according to the invention are used in medicine for diagnostic investigations. The process of the invention envisages the preparation of a layer (called "first layer"), characterised by a thickness/diameter ratio ≤ 0.25 , comprising a chemical element or an isotope of a starting chemical element, i.e. a chemical element or an isotope of a chemical element capable of generating a known radionuclide when subjected to bombardment with accelerated particles. The starting chemical element or isotope of a chemical element can also be in the form of a chemical compound, such as, for example, a carbide, oxide, nitride, boride, or silicide. The coupling between this first layer and a support layer, characterised by thermal conductivity, takes place by means of a passage of electric current (pulsed, alternating or direct) through the two layers (the first layer and the support layer) and a simultaneous application of pressure.

[0020] In one embodiment of the invention, between the first layer and the support layer there is an intermediate layer made of metal material whose function is to prevent contact of the support layer with the chemical reagents used for the dissolution and recovery of the radionuclide from the first layer after irradiation with the accelerated particles.

[0021] In one embodiment, the coupling between the first layer, the support layer and an intermediate layer, if present, takes place through the use of the Spark Plasma Sintering (SPS) technique, without the use of a containing mould. This technique can be defined as a technique of sintering in the presence of electric current and pressure.

[0022] The invention also relates to a target for the generation of radionuclides by bombardment with protons which comprises a first layer comprising a chemical ele-

ment or isotope, for example ^{100}Mo , in an amount $\geq 99\%$ by weight, and a support layer of copper, preferably containing from 0.1% to 0.2% by weight of Al_2O_3 . The isotope ^{100}Mo can also be in the form of a chemical compound such as a carbide $^{100}\text{MoC}/^{100}\text{Mo}_2\text{C}$, oxide $^{100}\text{MoO}_2$, boride ^{100}MoB , nitride $^{100}\text{Mo}_2\text{N}$ or silicide $^{100}\text{MoSi}_2$. In such a case the compound itself will be present in an amount greater than 98%. Alternatively, the support layer is a layer made of diamond or a composite material comprising from 60% to 80% by volume of synthetic diamond powder and 20% to 40% by volume of silver alloy powder. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

[0023] In one embodiment, the target also comprises an intermediate layer, which can be made of nickel, gold, tantalum, niobium, silver, zirconium, titanium, chrome, yttrium, vanadium, tungsten, manganese, cobalt, platinum, zinc, aluminium, tin, said intermediate layer being positioned between the first layer and the support layer.

BRIEF DESCRIPTION OF THE FIGURES

[0024]

Figure 1 illustrates a two-layer target according to the invention comprising a first layer 1 consisting of the chemical element from which the radioactive isotope is produced by bombardment with accelerated particles and a second support layer 2;

Figure 2 illustrates an embodiment of the process of the invention for preparing a two-layer target;

Figure 3 schematically represents an enlargement of the $^{100}\text{Mo}/\text{Glidcop AL-15}^\circ$ interface from which it is possible to note the mechanical gripping between the two layers;

Figure 4 illustrates an embodiment of the process of the invention for sintering the first layer;

Figure 5 shows a three-layer configuration of the target of the invention, wherein the number 1 indicates the first layer, the number 2 the support layer and the number 4 the intermediate layer;

Figure 6 illustrates an embodiment of the process of the invention wherein the three-layer target is obtained in a single process cycle;

Figure 7 illustrates an embodiment of the process of the invention wherein a plurality of two-layer targets is prepared simultaneously with a single process cycle.

DETAILED DESCRIPTION OF THE INVENTION

[0025] "First layer" means a layer comprising a starting chemical element or isotope of a chemical element from which the radionuclide is obtained by bombardment with accelerated particles. The starting chemical element or isotope of a chemical element can also be in the form of a chemical compound, such as, for example, a carbide, oxide, nitride, boride or silicide.

"Green layer" means a first layer that is not sintered according to the sintering step a1).

"Sintered green layer" means a first layer subjected to sintering according to step a1).

5 "First sintered layer" means the first layer of the two- or three-layer target after step d) of the process of the invention.

"Support layer" means a layer on which the first layer is applied. "Intermediate layer" means a layer that is positioned between the first layer and the support layer.

10 "Angular powder" means a non-spherical powder or, in other words, a powder of an irregular shape.

"Target" means the physical object against which the accelerated particles collide and which for the purposes of the invention can be a two-layer or a three-layer one.

15 "Intermetal phase" means a chemically and crystallographically established phase defined by a metallic bond and an ordered lattice, characterised by low tenacity.

"Protective atmosphere" means a non-oxidising atmosphere, for example a nitrogen, argon or hydrogen gas.

20 For the purposes of the present invention, the current density and pressure measurements always refer to the dimensions of the first layer.

[0026] The present invention relates to a process for preparing a two-layer or three-layer target for the generation of radioactive isotopes (radionuclides) by bombardment with accelerated particles, comprising the steps of:

25 a) providing a first layer comprising an amount $\geq 98\%$ by weight of a chemical element, an isotope of a chemical element or of an isotope of a chemical element suitable for generating a radionuclide following bombardment with accelerated particles, wherein the first layer is characterised by a thickness/diameter ratio ≤ 0.25 ;

30 b) providing a support layer characterised by thermal conductivity;

35 c) positioning the first layer on the support layer, inserting it into a vacuum chamber and applying the vacuum and/or a protective atmosphere;

40 d) applying an electric current density comprised between 0.5 and 25 A/mm² simultaneously subjecting the first layer and the support layer to a pressure comprised between 0.1 and 100 MPa.

[0027] In step a) the first layer is a green layer prepared from a powder of a chemical element or of an isotope of a chemical element or of a compound of a chemical element or of a compound of an isotope of a chemical element.

50 The powder contains an amount of the element/isotope/chemical compound $\geq 98\%$ by weight, preferably $\geq 99\%$ by weight. In particular, in the green layer there are no binding agents or other additives that are typically used in the known processes for improving the sintering and thus the mechanical strength of the layer.

55 The powder is preferably a powder isotopically enriched

with an element/isotope/chemical compound, for example it is a ^{100}Mo or $^{100}\text{MoC}/^{100}\text{MoC}_2$ powder enriched to values greater than 99% by weight. The powder is introduced inside a metal mould and subjected to a pressure comprised between 100 and 2000 MPa, preferably between 500 and 900 MPa.

The metal mould can have cavities of different shape, for example it can be rectangular, cylinder-shaped or of an ellipsoidal shape so as to provide a corresponding green layer of the desired shape.

[0028] An ellipsoidal shape of the first layer is particularly useful in the event of use of the target in high-power cyclotrons. In this case the first ellipse-shaped layer is coupled with a support layer of a rectangular shape for maximum performance and inclined by a certain angle relative to the direction of the incident ray of bombardment so as to increase the area of incidence.

The green layer has a relative density comprised between 60% and 90%, according to the compressibility of the powder; this means that the porosity of the green layer is comprised between 10% and 40%. A greater or lesser porosity of the green layer determines a greater or lesser reactivity of the layer during the chemical attack phase following the bombardment with accelerated particles, which can thus be carried out with variable times. The mechanical strength of the green layer is determined by the mechanical gripping due to the rearrangement and plastic deformation of the powder particles, in particular the surface roughness. Therefore, the mechanical strength of the green layer depends on the morphology of the initial powder, which is preferably an angular powder. Spherical powders are less preferred, since they provide green layers with poor mechanical strength and which are thus difficult to handle.

The amount of powder to be introduced into the mould depends on the diameter of the mould itself and the thickness of the green layer it is desired to obtain. It is important to maintain a thickness/diameter ratio of the green layer ≤ 0.25 , preferably < 0.15 , since ratios higher than the limit indicated bring about a possibility of deformation of the first layer during the subsequent pressing and current passage step d), ultimately arriving, in the worst cases, at the loss of the original shape.

With this method of preparing the green layer, the percentage of use of the powder of the element/isotope/chemical compound is $\geq 95\%$ by weight, preferably $\geq 98\%$ by weight. This result represents an undoubted advantage compared to the known processes in which the percentage of use of the powder is about 50%. For example, since the cost of ^{100}Mo enriched powder is very high, the process of the invention presents itself as an absolutely advantageous process, compared to the known ones, also from an economic viewpoint.

The chemical element or the isotope of a chemical element which is used, in powder form, to form the green layer can be selected from: ^{100}Mo , ^{58}Ni , ^{45}Sc , ^{88}Sr , ^{54}Cu , ^{57}Cu , ^{51}Cr , ^{59}Fe , ^{89}Y , ^{68}Zn , ^{112}Cd , ^{64}Ni , ^{48}Ti , ^{55}Mn , ^{50}Cr , ^{52}Cr , ^{44}Ca , ^{54}Fe , ^{56}Fe , ^{61}Ni , ^{59}Co , ^{63}Cu , ^{60}Ni , ^{66}Zn , ^{65}Cu ,

^{94}Mo or from the natural isotopes or chemical compounds thereof, such as, for example, carbides, oxides, nitrides, borides, silicides.

The isotope is preferably ^{100}Mo , which gives rise to the radionuclide $^{99\text{m}}\text{Tc}$ when subjected to proton bombardment.

Other radionuclides obtained by proton bombardment are: ^{57}Co obtained from ^{58}Ni , ^{44}Ti obtained from ^{45}Sc and ^{88}Y obtained from ^{88}Sr .

One example of a first layer is illustrated in figure 1 with the number 1.

In one embodiment of the invention, the first layer can be a preformed foil of an element/isotope/chemical compound, in which case no pressing step is necessary.

[0029] The support layer of **step b)** can have a rectangular or disk shape and dimensions greater than those of the first layer, as schematically illustrated with the number 2 in figure 1.

The support layer must be characterised by a good ability to dissipate heat and thus by a thermal conductivity that is as high as possible.

The support layer is made of a metal material selected from: copper, preferably containing from 0.1% to 0.2% by weight of Al_2O_3 , aluminium, gold, silver and alloys thereof. The thermal conductivity of these metals is the following: silver $\kappa=429 \text{ W}/(\text{m}\cdot\text{K})$, copper $\kappa=390 \text{ W}/(\text{m}\cdot\text{K})$, gold $\kappa=317 \text{ W}/(\text{m}\cdot\text{K})$, aluminium $\kappa=237 \text{ W}/(\text{m}\cdot\text{K})$.

Alternatively, the support layer can be made of synthetic diamond which has a κ of up to $2000 \text{ W}/(\text{m}\cdot\text{K})$ and is highly resistant to chemical agents, even if fragile and costly, or of electrically conductive diamond obtained by chemical vapor deposition (CVD) using boron as the doping agent.

It is possible to make the support layer from a composite material obtained by mixing 65-80% by volume of synthetic diamond powder and 20-35% by volume of silver alloy powder. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

The support layer in this case is produced by means of the Spark Plasma Sintering (SPS) technique or with other techniques, for example infiltration, classic pressing & sintering, or metal injection moulding.

Alternatively, the support layer can be made of a conductive or semi-conductive ceramic material, for example TiB_2 or SiC , or electrically insulating ceramic materials, for example Al_2O_3 , rendered electrically conductive by using carbon nanotubes or graphene.

The support layer can also be made of isotropic graphite, C/SiC (carbon fibres infiltrated with Si), C/C (carbon reinforced with carbon fibres), SiSiC (silicon carbide infiltrated with silicon) or graphite/SiC, commercially known as CarbocellTM, composed of spherical graphite particles bound together by SiC. CarbocellTM is electrically conductive, has good thermal conductivity ($\kappa=200 \text{ W}/(\text{m}\cdot\text{K})$) and resists chemical attack by acids; furthermore, unlike common isotropic graphite, it is impermeable to gases. In some cases, the support layer can be made of metals with a modest thermal conductivity, such as niobium, tan-

talum, tungsten, titanium, zirconium or vanadium, in order to meet other needs, in particular resistance to specific thermochemical treatments for dissolving the first layer.

Copper material containing from 0.1% to 0.5% by weight, preferably from 0.2% to 0.4% by weight, of Al_2O_3 , known, for example, by the name Glidcop AL-15®, is particularly preferred. This material, besides having excellent thermal conductivity, shows good resistance to deformation at high temperatures and also a high capacity to maintain good mechanical strength at high temperatures.

[0030] The support layer is obtained by machining a bar or sheet of one of the materials listed above or else, if it is made of a composite diamond/silver alloy material, by using one of the techniques listed above.

[0031] In **step c)**, the first layer is placed on top of and aligned with the support layer and then the two-layer target thus obtained is inserted into a vacuum chamber. Preferred combinations of the first layer and support layer are: first layer of ^{100}Mo and support layer of copper or copper alloy, for example copper containing from 0.1% to 0.5% by weight, preferably from 0.2% to 0.4% by weight, of Al_2O_3 ; first layer of ^{100}Mo and support layer of CarboCell™; first layer of ^{100}Mo and support layer of synthetic diamond or a composite material obtained by mixing 65-80% by volume of synthetic diamond powder and 20-35% by volume of silver alloy powder. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

[0032] In **step d)**, after the insertion of the two-layer target into a vacuum chamber and setting of a vacuum or a protective atmosphere, an electric current density comprised between 0.5 and 25 A/mm^2 , preferably between 3 and 16 A/mm^2 , is applied, while the two-layer target is simultaneously subjected to a pressure comprised between 0.1 and 100 MPa, preferably between 2 and 30 MPa.

The electric current applied can be pulsed, alternating or direct.

This step is schematically illustrated in figure 2, in which the number (1) indicates the first layer, the number (2) the support layer and the number (3) the vacuum chamber.

The graphite blocks (also called "spacers") are indicated with the letters A, B, C and D. The sizes of said graphite blocks depend on the available equipment, the sizes of the target and the type of graphite they are made from.

[0033] Given that both the first layer (1) and the support layer (2) are electrically conductive, the circuit is closed and it is possible to apply a pulsed, alternating or direct electric current. Since the two layers are electrically conductive, it is not necessary to apply a conductive or semi-conductive lateral containing mould, for example made of graphite, to close the electrical circuit, since the electric current passes totally through the first layer and the support layer, thus determining an excellent junction between the layers and a consolidation/sintering of the first layer.

Since the electric current passes totally through the first layer and the support layer, a high local heating, and thus a high temperature and interdiffusion, is caused in the points of contact between the powder particles and between the powder particles and the support layer as a result of the Joule effect ($P = R \cdot I^2$). Overall, this determines a rapid sintering (i.e. a bond between the layers) even when the layers are made up of very different materials.

During the application of the electric current, the instantaneous contact temperature between the particles can be very high, even higher than the melting temperature, whereas the average temperature of the target is maintained at acceptable values, well below the melting values.

The graphite of the spacers, being a semiconductor, has a much higher resistivity than the materials of the target: therefore, a large part of the heating power generated as a result of the Joule effect is produced in the spacers and not in the target, which in fact remains at a lower temperature.

Therefore, by making geometrically different graphite blocks it is possible to generate and control temperature gradients during the whole of step d). It is for this reason that the spacers C and D in figure 2 have been made with different dimensions: the spacer C, having a smaller cross section, has a higher resistance than the spacer D and therefore, given that the current in transit is the same, heats up more quickly and reaches a higher temperature.

The choice of the dimensions of the graphite blocks means that the spacer C at a higher temperature is in direct contact with the first layer to be sintered, whilst the spacer D is in contact with the support layer: the first layer is thus positively subjected to a temperature that is higher than that of the support layer. This fact is advantageous because the material of the first layer requires high temperatures in order to be sintered, much higher than the melting temperature of the support layer. Since no lateral containing mould is present, there arises the problem of measuring the temperature during step d). In order to remedy this problem, the temperature of the spacer C can be measured immediately near the first layer and also that of the spacer D near the support layer by means of a pyrometer or thermocouple. This measurement represents a fairly accurate estimate of the temperature of the first layer and of the support layer.

The mechanical pressure applied throughout the cycle (2-30 MPa) tends to be low in order to avoid the deformation and compression of the layers, but sufficient to ensure electrical contact.

As regards the application of the current density, it can take place in two ways: by applying a certain fixed value, for example 5 A/mm^2 , and allowing heating to take place freely up to a certain temperature, or setting a thermal cycle in terms of heating rate, maximum temperature and time of stasis. Depending on the geometry, the size and the materials the target is composed of, one or the other way can be chosen.

For example, if the target is formed by a first layer of ^{100}Mo and a support layer of copper or copper alloy, for example copper containing from 0.1% to 0.5% by weight, preferably from 0.2% to 0.4% by weight, of Al_2O_3 , the temperature measured at the graphite blocks during step d) will be comprised between 700 and 1000°C.

[0034] Once step d) has ended, the target is cooled and shows excellent solidity: it is necessary to apply a considerable shear stress between the first layer and the support layer in order to be able to detach them. Not even the use of a scalpel or cutter enables the two layers to be detached.

The analysis of the separated layers has made it possible to verify that the first layer has a slightly smaller mass than the initial one (thus a rate of use $\geq 98\%$) and the same density as at the start, but the mechanical strength is higher compared to the initial green layer. Therefore, an actual sintering process with a constant relative density has taken place: the pores have not been reduced, but the link between the different powder particles is no longer due only to the plastic deformation and rearrangement obtained after pressing, but rather sintering necks have formed thanks to the high local temperature.

From a metallurgical viewpoint, the junction between two very different materials, such as, for example, molybdenum and copper, should be poorly resistant: this is due to the fact that the two materials are mutually insoluble. The reason for the considerable resistance of the junction that occurs between the first layer and the support layer after step d) is of a mechanical type. At the temperature that is generated in step d), the first layer, for example of molybdenum, is scarcely deformable whereas the support layer, for example made of Glidcop AL-15®, deforms sufficiently and penetrates into the pores of the first layer, thus creating a mechanical gripping as shown, for example, in figure 3. Figure 3 shows an enlargement of the ^{100}Mo /Glidcop AL-15® interface after the application of step d).

The treatment with an electric current and application of pressure in step d) takes place in a short time, for example comprised between 10 and 600 seconds, preferably between 60 and 300 seconds. The duration of the entire process can be longer because of the time necessary for reaching a vacuum or introducing a protective atmosphere.

The rapidity of heating and the absence of stasis at a high temperature, besides ensuring high productivity, limits the growth of intermetal phases between the layers in contact, which are thus joined by mechanical gripping. The shorter the time for which the temperature is maintained, the smaller the amount of intermetal phases that will be formed: this is an undoubted advantage of the process of the invention, since intermetal phases are usually very fragile and impair the mechanical solidity of the junction.

[0035] In a preferred embodiment, the first layer (or green layer) is subjected to sintering after step a) and before the subsequent steps c) and d) so as to increase

its mechanical strength.

As explained above, in step a) the powder of an element/isotope/chemical compound is inserted into a metal mould and subjected to a pressure comprised between 100 and 2000 MPa, preferably between 500 and 900 MPa, so as to form the green layer. This operation imparts to the layer a certain mechanical strength and manageability which, however, can be insufficient during phases of irradiation with high-energy accelerated particles.

In this case it is possible to subject the green layer on its own to a sintering step (**step a1**) which precedes the subsequent steps c) and d) of coupling to the support layer.

In order to sinter the green layer on its own, the same method as described for step d), and schematically illustrated in figure 4, can be used.

Given the small thickness/diameter ratio of the green layer, it is possible to use the same method as provided for step d) of the process, i.e. the total passage of current inside the layer to be sintered. This ensures a high productivity and short processing times. It should be noted that in this case the graphite blocks are of the same size, because no temperature gradient is necessary, as only one layer is to be sintered.

[0036] The applied electric current density has values comprised between 1 and 25 A/mm², preferably between 7 and 16 A/mm². The applied pressure is comprised between 5 and 100 MPa.

Although the applied pressure and also the sintering temperature obtained are higher than those in step d), the density of the green layer (pre- and post-sintering) remains practically unchanged: the sintered green layer remains considerably porous and thus maintains the benefits with respect to chemical reactivity and adhesion with the subsequent support layer. However, the mechanical strength is considerably greater: it is no longer possible to break the sintered green layer with one's hands.

[0037] In a preferred embodiment of the invention, inserted between the first layer and the support layer there is an intermediate layer which has the purpose of preventing contact between the support layer and the solution used for the chemical attack of the first sintered layer and the recovery of the radionuclides generated by the irradiation with particles.

The solutions commonly used for the chemical attack of the first layer are acidic or basic. They normally also enter into contact with the support layer linked to the first layer. However, the support layer may also not be sufficiently resistant to such reactions and thus partially degrade, thereby contaminating the phase of recovery of the radionuclides.

In order to avoid this problem, it is possible to provide for the insertion of an intermediate layer, between the first layer and the support layer, manufactured from a material that is resistant to chemical attacks. The chemical solution will thus be confined to the zone of the first layer and of the intermediate layer by using an O-ring or an object of an equivalent shape.

One example of a three-layer target according to this embodiment is illustrated in figure 5, in which the intermediate layer is identified by the number (4).

[0038] The intermediate layer can be a metal layer of nickel, gold, tantalum, niobium, silver, zirconium, titanium, chrome, yttrium, vanadium, tungsten, manganese, cobalt, platinum, zinc, aluminium, tin, lead, cadmium or iron.

The lateral dimensions of the intermediate layer are greater than those of the first layer and equal to or less than those of the support layer, as schematically illustrated in figure 5.

In order to obtain the three-layer target it is possible to apply step d) of the process two times: a first cycle to form the junction between the intermediate layer and the support layer and a second cycle to form the junction between the green layer, or the green layer sintered according to step a1), and the intermediate layer anchored to the support layer. The beneficial effects of the passage of current between the various interfaces enable solid, resistant junctions to be obtained. The intermediate layer is as thin as possible so as to influence the thermal conductivity as little as possible, but at the same ensure the separation between the first layer and the support layer. The diameter of the intermediate layer, on the other hand, depends on the system of chemical dissolution.

[0039] The preferred combinations for the three-layer target are: support layer of copper or copper alloy, for example copper containing from 0.1% to 0.5% by weight, preferably from 0.2% to 0.4% by weight, of Al_2O_3 , intermediate layer of nickel and first layer comprising ^{100}Mo ; support layer of copper or copper alloy, for example copper containing from 0.1% to 0.5% by weight, preferably from 0.2% to 0.4% by weight, of Al_2O_3 , intermediate layer of gold and first layer comprising ^{100}Mo .

[0040] One of the disadvantages of the process just described for the production of the three-layer target is the need for a double cycle of step d), which lowers productivity and thus increases production costs.

An alternative to this production method is a process wherein the intermediate layer is applied and anchored to the support layer by means of a conventional method selected from: welding, electrochemical deposition, physical vapor deposition (PVD), chemical vapor deposition (CVD) and the like; the first layer can subsequently be positioned and anchored to the two-layer target comprising the support layer and the intermediate layer according to step d) of the process of the invention.

[0041] A further alternative to the two processes illustrated above is a process wherein the three-layer target is obtained with a single cycle of step d) of the invention by suitably increasing the pressure during step d).

In this case, therefore, the green layer or the green layer sintered according to step a1) is positioned on the support layer together with the intermediate layer according to the configuration in figure 5. After this, the assembled target is inserted into a vacuum chamber according to step c) and the vacuum is applied or a protective atmos-

phere is introduced.

The configuration of the graphite blocks A, B, C and D, is equal to the one illustrated, for example, in figure 2, the spacer C being smaller than the spacer D.

5 After the vacuum has been applied, or a protective atmosphere introduced, an electric current is made to circulate at the values previously indicated for step d); a constant current is preferably applied. Simultaneously, an initial pressure comprised between 5 and 20 MPa is applied. When a sintering temperature comprised between 900°C and 1100°C (measured on the graphite block C as represented in figure 2) is reached, the pressure is increased to a value comprised between 20 and 80 MPa.

10 The increase in pressure causes the intermediate layer and the first layer to penetrate inside the support layer until the graphite block C is in contact with the support layer. See, by way of example, the configuration in figure 6.

15 In this manner one obtains a target with the interposition of the intermediate layer in a single step; however, the support layer will have dimensions larger than the initial ones because of the interpenetration between the first layer and the intermediate layer. The adhesion of the first layer is good and poses no problems of detachment from the other two layers.

[0042] In one embodiment of the invention, since the thickness of every target is very limited, it is possible to stack various targets, as illustrated, for example, in figure 7 and then proceed to a single sintering cycle so as to obtain a high productivity and low costs. In this embodiment, the targets to be sintered are stacked in such a way that the graphite blocks C are facing towards the first layer and the graphite blocks D are facing towards the support layer of each target. Therefore, each target is stacked in a manner opposite to that of the next target, as illustrated in figure 7.

This embodiment of the process of the invention can also be applied to the three-layer targets that comprise an intermediate layer between the first layer and the support layer, both in the case where the intermediate layer has already been anchored to the support layer with other techniques, and in the case where the 3 layers undergo simultaneous sintering according to the configuration in figure 6.

[0043] In a preferred embodiment of the invention, the process of the invention can be definable as a process in which the known sintering technique aided by mechanical pressure and electric current, called Spark Plasma Sintering (SPS), suitably adapted, is applied without the use of a mould for containing the powder to be sintered. The SPS technique exploits the heating resulting from the Joule effect thanks to the passage of current through the layers and thus enables many materials to be easily sintered in a short time and at lower temperatures compared to the usual sintering times and temperatures.

The SPS technique is also called FAST (Field Assisted Sintering Technique), PECS (Pulsed Electric Current

Sintering), EFAS (Electric Field Assisted Sintering), DHP (Direct Hot Pressing) or DCS (Direct Current Sintering). A variant of the SPS technique is the ESF (Electro Sinter Forging) technique, which uses a bank of capacitors capable of accumulating a large amount of electric charge, which is then discharged onto the sample to be sintered. Another variant is the technique that uses electrical induction to generate an electromagnetic field and hence an electric current inside the sample to be sintered. Many variants of the SPS process exist, therefore: in the case of the process of the present invention, what matters is that there be the passage of an electric current (whether or not pulsed, alternating or direct) inside the target to be sintered, without a lateral containing mould, and a simultaneous application of a mechanical pressure. Therefore, in one embodiment, the process of the invention is an SPS process for preparing a target for the generation of radioactive isotopes by bombardment with accelerated particles, comprising the steps of:

- a) providing a first layer comprising an amount $\geq 98\%$ by weight of a chemical element, or of an isotope of a chemical element, or of a compound of a chemical element or of an isotope of a chemical element suitable for generating a radionuclide following bombardment with accelerated particles, wherein the first layer is characterised by a thickness/diameter ratio ≤ 0.25 , preferably ≤ 0.15 ;
- b) providing a support layer characterised by thermal conductivity;
- c) positioning the first layer on the support layer and inserting them in a vacuum chamber of an SPS system;
- d) applying an electric current density comprised between 0.5 and 25 A/mm^2 , simultaneously subjecting the first layer and the support layer to a pressure comprised between 0.1 and 100 MPa .

Preferably, the method of use of the SPS technique adopted here does not envisage the use of a lateral mould for containing the target.

[0044] Once the two-layer or three-layer target sintered with the process according to the invention has been obtained, it is used as a source of radionuclides when subjected to irradiation with accelerated particles which strike the first layer comprising a chemical element or an isotope of a chemical element or a compound of a chemical element or a compound of an isotope of a chemical element.

Given the high cost of the powder of the chemical element or of the isotope of the chemical element or of the compound of the chemical element or of the isotope of the chemical element, which is preferably an isotopically enriched powder, the non-transmuted part (i.e. the part not converted into the radionuclide) can be recovered after the chemical attack, for example with hydrogen peroxide solutions or basic solutions, converted into the powder of the starting element/isotope/chemical compound (for

example ^{100}Mo) and again used for the production of further targets.

[0045] The invention also relates to a two-layer target comprising a first layer comprising a chemical element or an isotope of a chemical element or a compound of a chemical element or of an isotope of a chemical element in an amount $\geq 98\%$ by weight, preferably $\geq 99\%$ by weight, wherein the first layer has a thickness/diameter ratio ≤ 0.25 , preferably < 0.15 in contact with a support layer with a high thermal conductivity, preferably comprised between $\kappa=50 \text{ W/(m}\cdot\text{K)}$ and $\kappa=2000 \text{ W/(m}\cdot\text{K)}$, more preferably between $\kappa=200 \text{ W/(m}\cdot\text{K)}$ and $\kappa=950 \text{ W/(m}\cdot\text{K)}$. The first layer is anchored to the support layer by sintering according to the process of the invention.

The anchorage between the first layer and the support layer is a gripping of a mechanical type, wherein the support layer has penetrated into the pores of the first layer. In general, in the case of materials/elements that do not exhibit solubility the anchorage/junction will mainly be due to a mechanical action, whereas in the case of materials/elements that exhibit solubility the anchorage/junction will be mainly due to diffusion/interdiffusion. The presence of intermetal phases between the two layers in contact is preferably nearly or completely nil.

[0046] The chemical element or the isotope of a chemical element that is used, in powder form, to form the green layer can be selected from: ^{100}Mo , ^{58}Ni , ^{45}Sc , ^{88}Sr , ^{54}Cu , ^{57}Cu , ^{51}Cr , ^{59}Fe , ^{89}Y , ^{68}Zn , ^{112}Cd , ^{64}Ni , ^{48}Ti , ^{55}Mn , ^{50}Cr , ^{52}Cr , ^{44}Ca , ^{54}Fe , ^{56}Fe , ^{61}Ni , ^{59}Co , ^{63}Cu , ^{60}Ni , ^{66}Zn , ^{65}Cu , ^{94}Mo or from the natural isotopes or chemical compounds thereof, such as, for example, carbides, oxides, nitrides, borides, silicides. The isotope is preferably ^{100}Mo , which gives rise to the radionuclide $^{99\text{m}}\text{Tc}$ when subjected to proton bombardment.

The support layer is made of a material selected from: copper, preferably containing from 0.1% to 0.2% by weight of Al_2O_3 (for example Glidcop AL-15®), aluminium, gold, silver and alloys thereof, diamond, a composite material obtained by mixing $65\text{--}80\%$ by volume of synthetic diamond powder and $20\text{--}35\%$ by volume of silver alloy powder (preferably a silver alloy containing 3% by weight of silicon), TiB_2 , SiC , Al_2O_3 rendered conductive by using carbon nanotubes or graphene, isotropic graphite, C/SiC (carbon fibres infiltrated with Si), C/C (carbon reinforced with carbon fibres), SiSiC (silicon carbide infiltrated with silicon) or graphite/ SiC , commercially known as Carbocell™, composed of spherical graphite particles bound together by SiC .

The two-layer target of the invention preferably comprises the first layer of ^{100}Mo and the support layer made of copper containing from 0.1% to 0.2% by weight of Al_2O_3 (for example Glidcop AL-15®) or of diamond or a composite obtained by mixing $65\text{--}80\%$ by volume of synthetic diamond powder and $20\text{--}35\%$ by volume of silver alloy. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

[0047] The invention also relates to a three-layer target comprising a first layer comprising a chemical element

or an isotope of a chemical element or a compound of a chemical element or a compound of an isotope of a chemical element in an amount $\geq 98\%$ by weight, preferably $\geq 99\%$ by weight, wherein the first layer has a thickness/diameter ratio ≤ 0.25 , preferably <0.15 , an intermediate metal layer with high corrosion resistance in contact with the first layer and a support layer in contact with the intermediate layer, which has high thermal conductivity, preferably comprised between $\kappa=50 \text{ W/(m}\cdot\text{K)}$ and $\kappa=2000 \text{ W/(m}\cdot\text{K)}$, more preferably between $\kappa=200 \text{ W/(m}\cdot\text{K)}$ and $\kappa=950 \text{ W/(m}\cdot\text{K)}$. The three layers are anchored to one another by sintering according to the process of the invention.

The anchorage between the first layer and the intermediate layer is a gripping of a mechanical type wherein the intermediate metal layer has penetrated into the pores of the first layer, whereas between the intermediate layer and the support there is an interdiffusion between the elements such as to form a metallurgical junction. In general, in the case of materials/elements that do not exhibit solubility the anchorage/junction will mainly be due to a mechanical action, whereas in the case of materials/elements that exhibit solubility the anchorage/junction will mainly be due to diffusion/interdiffusion.

The presence of intermetal phases between the three layers in contact is preferably nearly or completely nil. The first layer and the support layer are as defined above. The intermediate metal layer is a layer of nickel or gold, tantalum, niobium, silver, zirconium, titanium, chrome, yttrium, vanadium, tungsten, manganese, cobalt, platinum, zinc, aluminium, tin.

In a preferred embodiment, the three-layer target comprises the first layer of ^{100}Mo , the intermediate layer of gold and the support layer of copper containing from 0.1% to 0.2% by weight of Al_2O_3 (for example Glidcop AL-15®) or of synthetic diamond or a composite obtained by mixing 65-80% by volume of synthetic diamond powder and 20-35% by volume of silver alloy. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

In a preferred embodiment, the three-layer target comprises the first layer of ^{100}Mo , the intermediate layer of nickel and the support layer of copper containing from 0.1% to 0.2% by weight of Al_2O_3 (for example Glidcop AL-15®) or of diamond or a composite obtained by mixing 65-80% by volume of synthetic diamond powder and 20-35% by volume of silver alloy. Said alloy is preferably a silver alloy containing 3% by weight of silicon.

[0048] In the case of both embodiments, two-layer or three-layer, the first layer has a relative density comprised between 60% and 90% depending on the compressibility of the starting powder; this means that the porosity of the first layer is comprised between 10% and 40%.

In the case of both embodiments, two-layer or three-layer, the first layer can be a green layer or a sintered green layer.

[0049] The invention also relates to the use of the two-layer or three-layer target according to the invention for

the production of radionuclides by bombardment with protons produced by a cyclotron. The radionuclides are used in diagnostic techniques such as PET (Positron Emission Tomography) or SPECT (Single Photon Emission Computed Tomography).

EXAMPLE 1

Example of production of a $\text{Cu}/^{100}\text{Mo}$ target with interposition of a gold foil suitable for the production of $^{99\text{m}}\text{Tc}$ by means of a cyclotron.

[0050] 0.5 g of ^{100}Mo powder enriched to at least 99.04% (Isoflex, San Francisco, USA) was poured into a metal mould with a 15.5 mm diameter cylindrical cavity. A pressure of 800 MPa was then applied in order to obtain a green layer with a thickness of 0.34 mm and thus a density $\rho=7.64 \text{ g/cm}^3$ ($\rho_{\text{rel}}=74.3\%$).

[0051] The green layer is solid and resistant and weighs 0.495 g: the rate of use of this first step is thus around 99%.

[0052] A disk with a diameter of 32 mm and thickness of 1.5 mm was obtained from a bar of Oxygen-Free High Conductivity (OFHC) copper; a small foil with a diameter of 25 mm was punched out from a pure gold foil with a thickness of 0.1 mm. The copper disk and small gold foil were positioned in an SPS apparatus with a spacer C with a diameter of 30 mm and height of 30 mm and a spacer D with a diameter of 45 mm and height of 35 mm. After the chamber was evacuated and vacuum of 10 Pa was produced, a constant load of 3 kN (corresponding to a pressure of 15.9 MPa) and a current of 1600 A (corresponding to a current density of 8.5 A/mm^2) were applied until a temperature of 850°C was reached in the spacer C: the time necessary to reach this temperature was 160 s.

[0053] A second SPS cycle was then carried out using the green ^{100}Mo disk and the Cu/Au support just mentioned: the same graphite block configuration as in the first SPS cycle was used.

[0054] The chamber was again evacuated and a constant load of 2 kN (corresponding to a pressure of 10.6 MPa) and a current of 1400 A (corresponding to a current density of 7.4 A/mm^2) were applied until a temperature of 850°C was reached in the spacer C: the time necessary to reach that temperature was 140 s.

[0055] Summing together the weight of the disk (1), the small gold foil and the disk (2) relative to the final target, a rate of use of the ^{100}Mo powder equal to 98% was obtained: therefore, the overall rate of use of ^{100}Mo powder is 97%.

[0056] The target thus obtained was used in a cyclotron and bombarded with protons with a specific power of up to 1 kW/cm^2 for 6 hours: no detachment of the disk (1) from the small gold foil was noted. The chemical dissolution test also gave excellent results.

EXAMPLE 2

Example of production of a two-layer ^{58}Ni /Carbocell™ target

[0057] A ^{58}Ni target for the production of ^{57}Co was produced. The ^{58}Ni isotope was supplied in the form of a 13 x 13 x 0.1 mm small foil.

[0058] In order to remedy the problem of corrosion of the support during the phase of chemical dissolution by an acid, use was made of the Graphite/SiC composite commercially known as Carbocell™ (Toyo Tanso, Osaka, Japan): it is composed of spherical graphite particles bound together by SiC. Carbocell™ is electrically conductive, has good thermal conductivity $\kappa=200 \text{ W/(m}\cdot\text{K)}$ and easily resists chemical attack by acids; moreover, unlike common isotropic graphite, it is impermeable to gases. The Carbocell™ disks used had a diameter of 25.4 mm and thickness of 2 mm: the object obtained easily passed the proton bombardment and chemical resistance test during the dissolution step.

[0059] The same target could have been produced using a copper support with the interposition of a gold foil, but the use of Carbocell™ showed two large advantages:

- a single SPS cycle for the production of the target;
- the lack of any use of gold and the low cost of Carbocell™ render the target a "disposable" object after the dissolution step, with undoubted advantages from a nuclear viewpoint (long decay times and thus a long wait before reuse of the support).

EXAMPLE 3

Example of sintering of a green layer of ^{100}Mo in disk form ($D_1=15 \text{ mm}$, $t_1=0.35 \text{ mm}$).

[0060] For sintering use was made of a constant current cycle at 1700 A (corresponding to a current density of 9.6 A/mm^2) and a constant load of 11 kN (corresponding to an applied pressure of 62 MPa): after 90 s the pyrometer read a temperature of 1250°C and the cycle was stopped. The initial density of the green disk remained practically unchanged, that is, around 79%: the sintered green disk is still considerably porous and thus maintains the benefits with respect to chemical reactivity and adhesion with the subsequent support disk. However, the mechanical strength after sintering is considerably greater: it is no longer possible to break the disk with one's hands. For the formation of the target, it is possible to proceed according to the method of figure 2, obtaining a perfect junction between the sintered disk and the support made of Glidcop Al-15.

EXAMPLE 4

Example of preparation of a three-layer target in a single cycle according to the geometries of example 1.

[0061] The configuration of the process is the one shown in figure 2, but with the assembly of the target as per figure 5: the disk (2) made of Glidcop Al-15, a nickel foil (4), and the disk (1) (green disk or sintered green disk, according to circumstances) are positioned in the SPS chamber with the small-sized spacer C and the large-sized spacer D.

[0062] A constant current (for example 1700 A, corresponding to a current density of 9 A/mm^2) and a low initial load (3 kN, corresponding to an applied pressure of 15.9 MPa) are imposed until a maximum sintering temperature (1050°C) is reached: at this point, the load is increased to 11 kN (corresponding to an applied pressure of 58 MPa). The increase in the load causes the nickel foil (4) and the disk (1) to penetrate into the disk (2) of Glidcop Al-15 until the spacer C is in contact with the disk (2) (figure 6). In this manner, one obtains a flat target with the interposition of the nickel foil in a single step: the only drawback is that the disk (2) of Glidcop Al-15 will have larger dimensions than initially due to the interpenetration of the foil and the disk (1). The adhesion of the disk (1) is good and poses no problems of detachment.

EXAMPLE 5

Example of preparation of a two-layer ^{100}Mo /Glidcop AL-15® target according to the configuration in figure 2 and geometries of example 1.

[0063] The SPS cycle provides for a constant load of 3 kN (corresponding to an applied pressure of 15.9 MPa) and the application of a constant current of 1800 A (corresponding to a current density of 9.5 A/mm^2): after about 60 s the pyrometer read a temperature of 1050°C in the spacer C and the SPS cycle was interrupted. It should be noted that this is not the temperature value either of the disk (1) or the disk (2): it is not possible to read the temperature of the two disks because they are too thin. What can be measured is the temperature of the spacer D, which was equal to 700°C , a good 350°C less than the spacer C. It may be presumed that the local temperature of contact between the particles of ^{100}Mo is much greater. After cooling, the target has excellent solidity: it is necessary to apply a considerable shear stress between the disks (1) and (2) in order to be able to detach them. Not even the use of a scalpel or cutter enables the two disks to be detached.

[0064] The separation of the disks made it possible to examine the disk (1) more thoroughly after the SPS process: it has a slightly smaller mass (rate of use 98%) and the same density as at the start, around 79%, but its mechanical strength is much higher than that of the green disk. An actual sintering process with a constant relative

density thus took place: the pores have not been reduced, but the link between the various particles of ^{100}Mo is no longer due only to plastic deformation and rearrangement, but rather sintering necks have formed thanks to the high local temperature. From a metallurgical viewpoint, the junction between molybdenum and copper should be poorly resistant: this is due to the fact that these two elements are mutually insoluble.

[0065] The reason for the considerable resistance of the junction between the disk (1) and the disk (2) is of a mechanical type. At the temperature of the SPS process, molybdenum is scarcely deformable whereas copper, though it is Glidcop AL-15® (well known to be resistant to deformation) deforms and penetrates into the pores of the disk (1), bringing about a mechanical gripping as shown in figure 3. As proof of this fact, when the same SPS process was repeated with a rolled foil of ^{nat}Mo of the same size as the disk (1) a considerable reduction in the mechanical strength of the target was noted compared to that achieved with the powder disk 1: this is due to the fact that the ^{nat}Mo foil is not porous, and therefore the copper cannot penetrate inside it.

Claims

1. A process for preparing a target for the generation of radioactive isotopes by bombardment with accelerated particles, preferably carried out by means of the Spark Plasma Sintering (SPS) technique or Electro Sinter Forging (EFS), comprising the steps of:

a) providing a first layer comprising an amount $\geq 98\%$ by weight of a chemical element, or of an isotope of a chemical element, or of a compound of a chemical element, or of a compound of an isotope of a chemical element, suitable for generating a radionuclide following bombardment with accelerated particles, wherein the first layer is **characterised by** a thickness/diameter ratio ≤ 0.25 , preferably ≤ 0.15 ;

b) providing a support layer **characterised by** thermal conductivity;

c) positioning the first layer on the support layer, inserting it into a chamber and applying a vacuum and/or protective atmosphere;

d) applying an electric current density comprised between 0.5 and 25 A/mm^2 , preferably between 3 and 16 A/mm^2 , simultaneously subjecting the first layer and the support layer to a pressure comprised between 0.1 and 100 MPa , preferably between 2 and 30 MPa , preferably said electric current density and pressure are applied for a time comprised between 10 and 600 seconds, preferably between 60 and 300 seconds.

2. The process according to claim 1, wherein, in step c), an intermediate metal layer is positioned between

the support layer and the first layer, preferably said intermediate metal layer being a layer of nickel, gold, tantalum, niobium, silver, zirconium, titanium, chrome, yttrium, vanadium, tungsten, manganese, cobalt, platinum, zinc, aluminium, tin, lead, cadmium, iron.

3. The process according to claim 1 or 2, wherein said chemical element or isotope of a chemical element is selected from: ^{100}Mo , ^{58}Ni , ^{45}Sc , ^{88}Sr , ^{54}Cu , ^{57}Cu , ^{51}Cr , ^{59}Fe , ^{89}Y , ^{68}Zn , ^{112}Cd , ^{64}Ni , ^{48}Ti , ^{55}Mn , ^{50}Cr , ^{52}Cr , ^{44}Ca , ^{54}Fe , ^{56}Fe , ^{61}Ni , ^{59}Co , ^{63}Cu , ^{60}Ni , ^{66}Zn , ^{65}Cu , ^{94}Mo or from the natural isotopes or chemical compounds thereof, such as, for example, carbides, oxides, nitrides, borides or silicides.

4. The process according to any one of claims 1 to 3, wherein said first layer is obtained from a powder of a chemical element or of an isotope of a chemical element, or of a compound of a chemical element or of a compound of an isotope of a chemical element, subjected to a pressure comprised between 100 and 2000 MPa , preferably between 500 and 900 MPa .

5. The process according to any one of preceding claims, wherein the first layer is subjected to sintering (step a1) after step a) and before the subsequent steps c) and d).

6. The process according to any one of claims 1 to 5, wherein the support layer is made of a material selected from: copper, preferably containing from 0.1% to 0.2% by weight of Al_2O_3 , aluminium, gold, silver and alloys thereof, diamond, a composite material obtained by mixing $65\text{--}80\%$ by volume of synthetic diamond power and $20\text{--}35\%$ by volume of silver alloy, TiB_2 , SiC , Al_2O_3 rendered conductive using nanotubes of carbon or graphene, isotropic graphite, C/SiC (Si-infiltrated carbon fibre), C/C (carbon fibre-reinforced carbon), SiSiC (silicon-infiltrated silicon carbide), or graphite/SiC.

7. The process according to any one of claims 2 to 6, wherein if an intermediate layer is present between the first layer and the support layer, the target is prepared by applying step d) of the process of claim 1 at least two times: a first time to form the junction between the intermediate layer and the support layer and a second time to form the junction between the first layer, optionally sintered according to step a1) of claim 5, and the intermediate layer.

8. The process according to any one of claims 2 to 6, wherein if an intermediate layer is present between the first layer and the support layer, the target is prepared by subjecting said three layers in contact with one another to a current density, preferably constant, comprised between 3 and 16 A/mm^2 and simultane-

ously applying an initial pressure comprised between 5 and 20 MPa and subsequently increasing the pressure to values comprised between 20 and 80 MPa when the sintering temperature is comprised between 900 and 1100°C.

9. A process for preparing a target for the generation of radioactive isotopes by bombardment with accelerated particles, carried out by means of the Spark Plasma Sintering (SPS) technique, preferably wherein a lateral mould for containing the target is not present, comprising the steps of:
 - a) providing a first layer comprising an amount $\geq 98\%$ by weight of a chemical element, or of an isotope of a chemical element, or of a compound of a chemical element, or of a compound of an isotope of a chemical element suitable for the generation of a radionuclide following bombardment with accelerated particles, wherein the first layer is **characterised by** a thickness/diameter ratio ≤ 0.25 , preferably ≤ 0.15 ;
 - b) providing a support layer **characterised by** thermal conductivity;
 - c) positioning the first layer on the support layer and inserting it into a vacuum chamber of an SPS system;
 - d) applying an electric current density comprised between 0.5 and 25 A/mm², simultaneously subjecting the first layer and the support layer to a pressure comprised between 0.1 and 100 MPa.
10. The process according to claim 9, wherein the target is a three-layer target in which the first layer comprises ¹⁰⁰Mo, the intermediate layer is a layer of gold and the support layer is copper or copper containing from 0.1% to 0.2% by weight of Al₂O₃.
11. A target for the generation of radioactive isotopes by bombardment with accelerated particles, comprising a first layer comprising a chemical element or an isotope of a chemical element or a compound of a chemical element or of an isotope of a chemical element in an amount $\geq 98\%$ by weight, preferably $\geq 99\%$ by weight, wherein the first layer has a thickness/diameter ratio ≤ 0.25 , preferably < 0.15 , wherein said first layer is anchored to a support layer with high thermal conductivity, preferably comprised between $\kappa=50$ W/(m·K) and $\kappa=2000$ W/(m·K), by means of the process according to any one of claims 1 to 9.
12. The target according to claim 11, wherein an intermediate layer is present between the first layer and the support layer and the intermediate layer is anchored both to the first layer and the support layer by means of the process according to any one of

claims 1 to 9.

13. The target according to claim 11 or 12, wherein the first layer, the support layer and the intermediate layer, if present, are anchored by means of mechanical gripping and/or diffusion/interdiffusion.
14. The target according to any one of claims 11 to 13, wherein the presence of intermetal phases between the first layer, the intermediate layer, if present, and the support layer is nearly or completely nil.
15. The target according to any one of claims 11 to 14, wherein the chemical element or isotope of the first layer is ¹⁰⁰Mo, the intermediate layer is a layer of nickel or gold and the support layer is copper or copper containing from 0.1% to 0.2% by weight of Al₂O₃ or diamond or a composite obtained by mixing 65-80% by volume of synthetic diamond powder and 20-35% by volume of silver alloy.

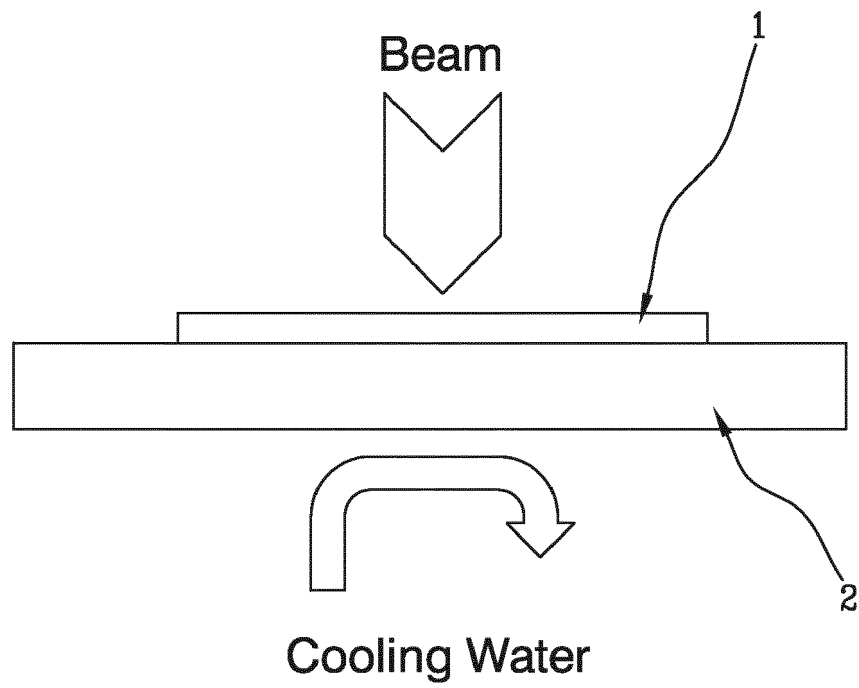


Fig.1

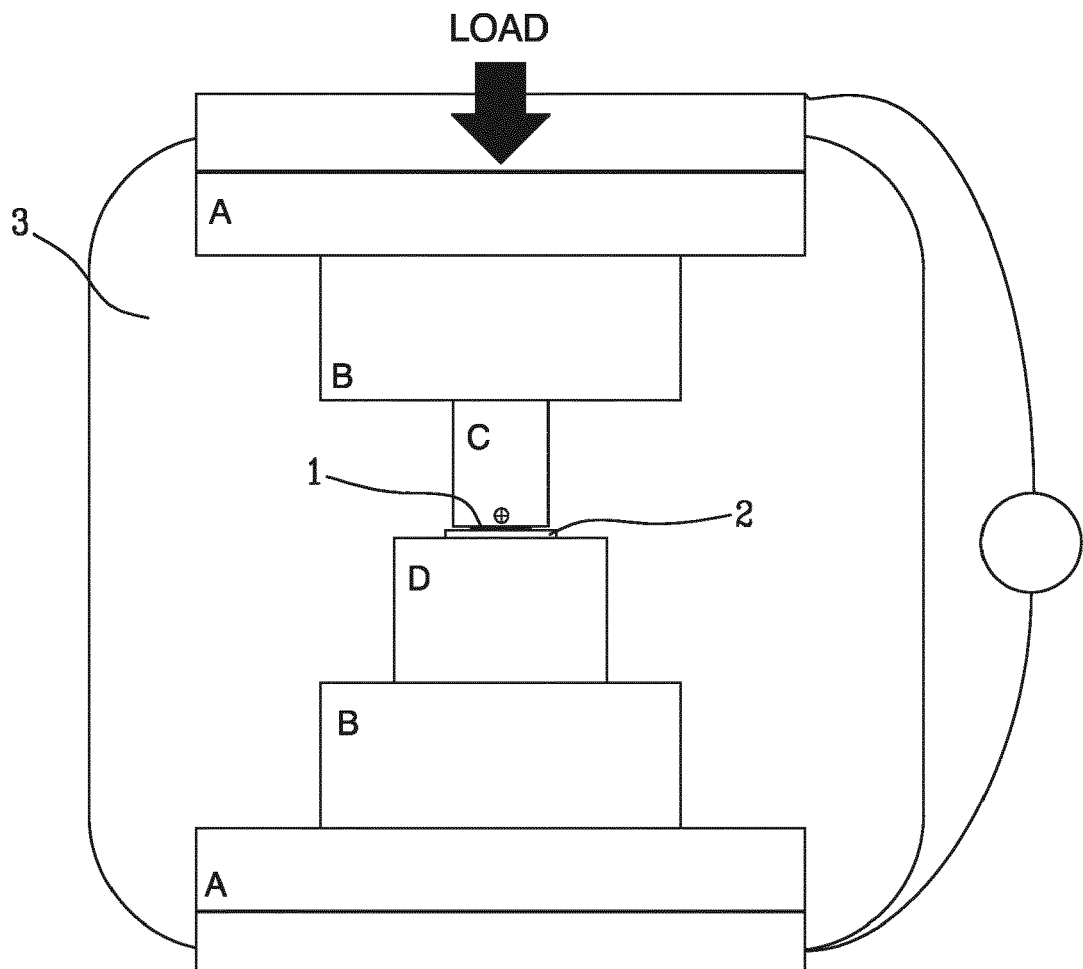


Fig.2

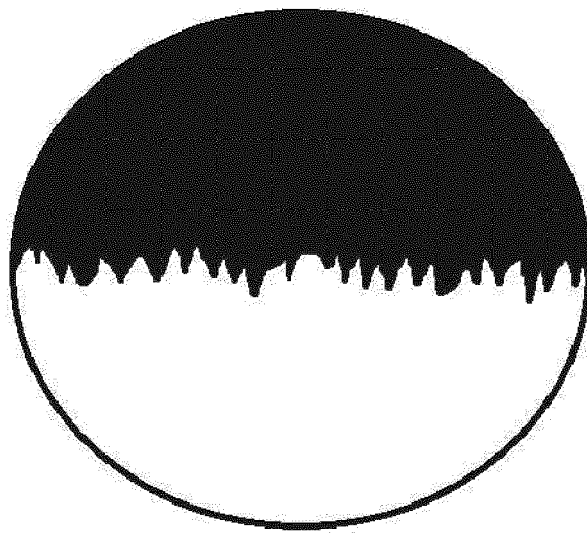


Fig.3

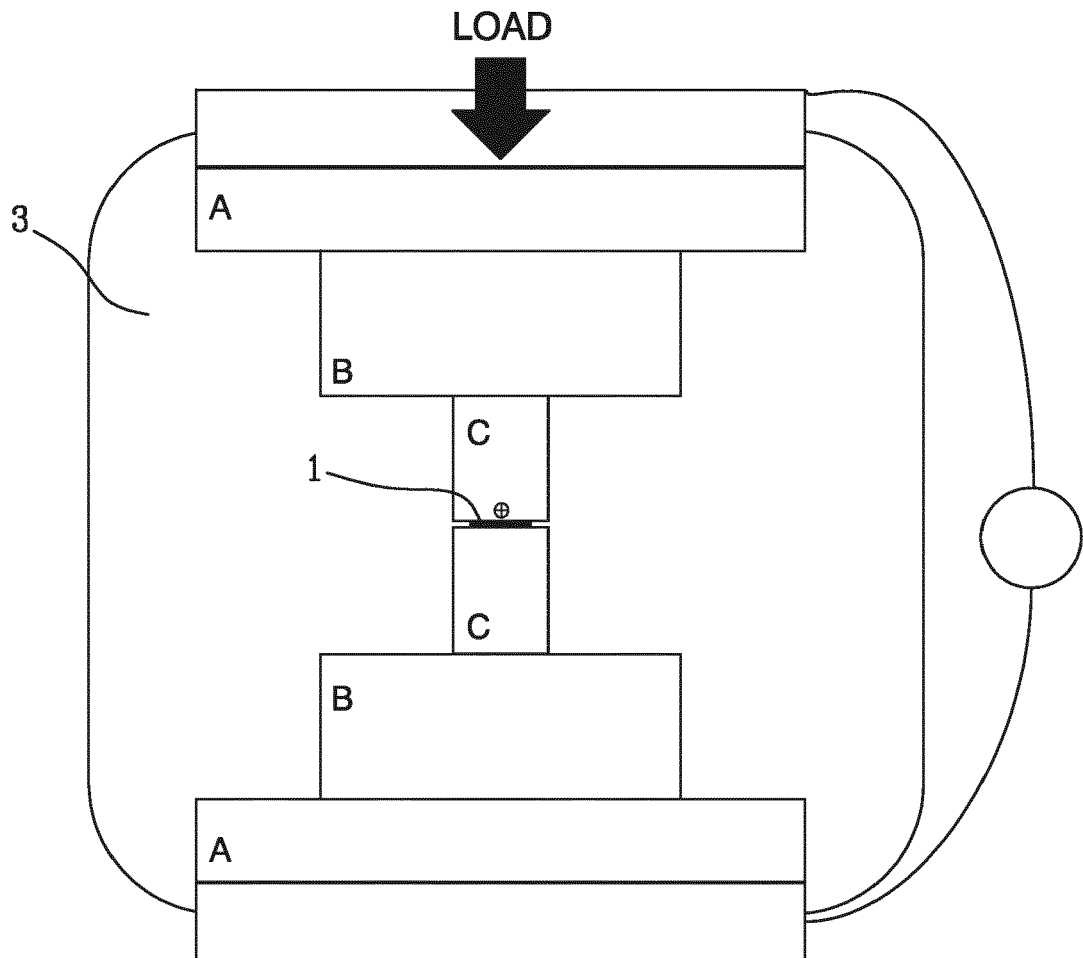


Fig.4

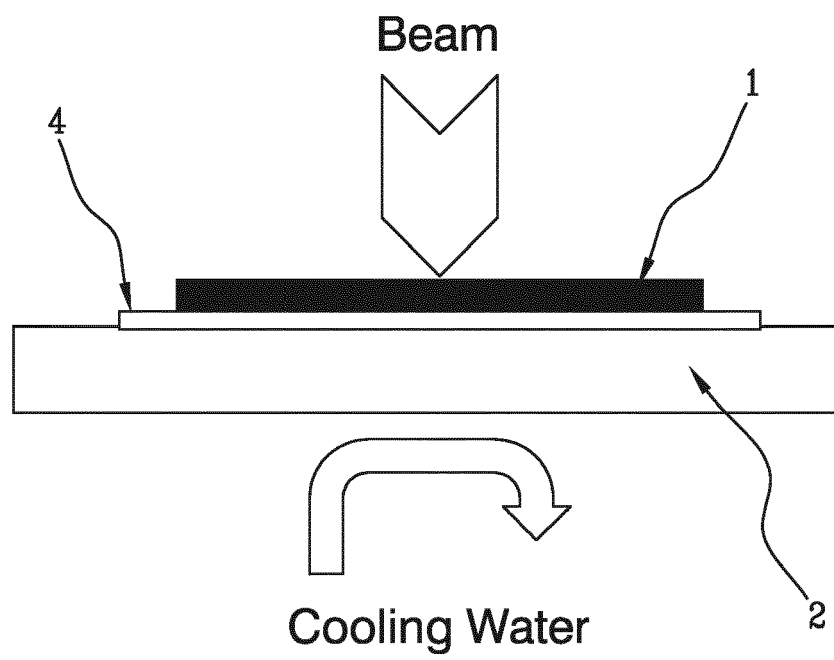


Fig.5

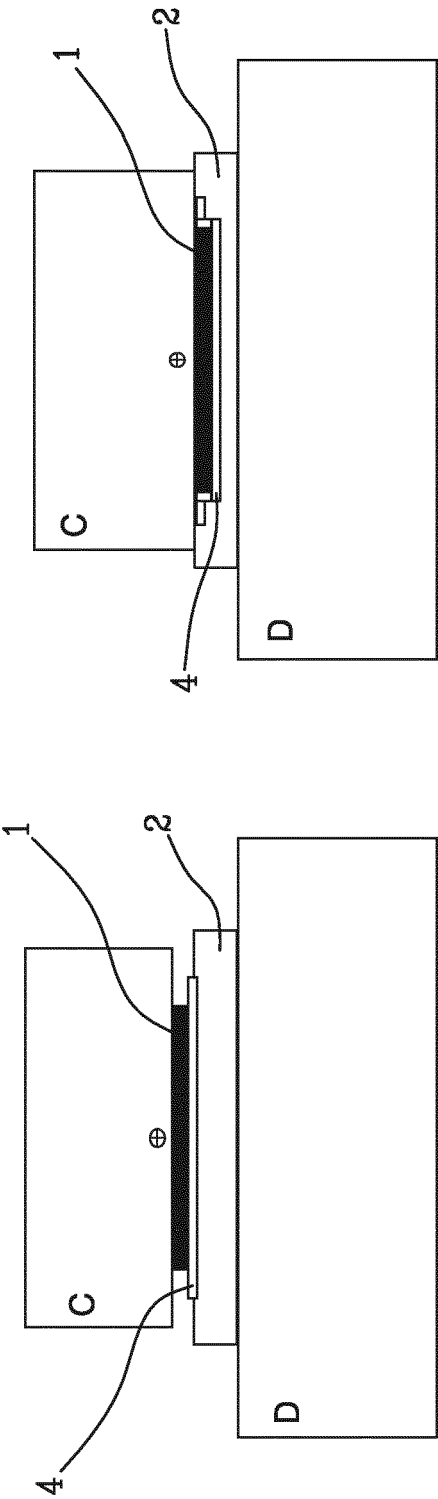


Fig.6

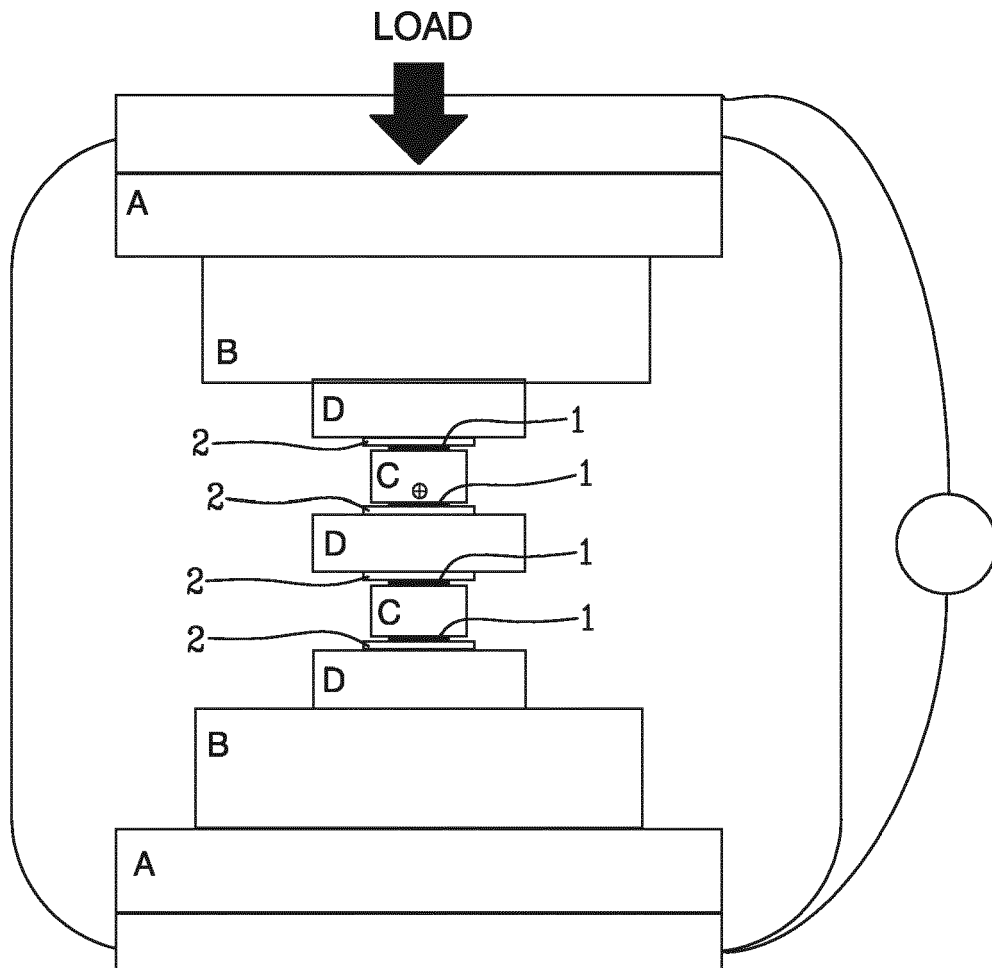


Fig.7



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