(19) Octrooicentrum Nederland

(11) 2011415

(12) C OCTROOI

(21) Aanvraagnummer: **2011415**

(22) Aanvraag ingediend: **10.09.2013**

(51) Int.Cl.:
***C22C 1/02*** (2006.01) ***C22C 43/00*** (2006.01)
***G21G 1/08*** (2006.01) ***F27D 11/08*** (2006.01)

(43) Aanvraag gepubliceerd:
-

(47) Octrooi verleend:
**12.03.2015**

(45) Octrooischrift uitgegeven:
**18.03.2015**

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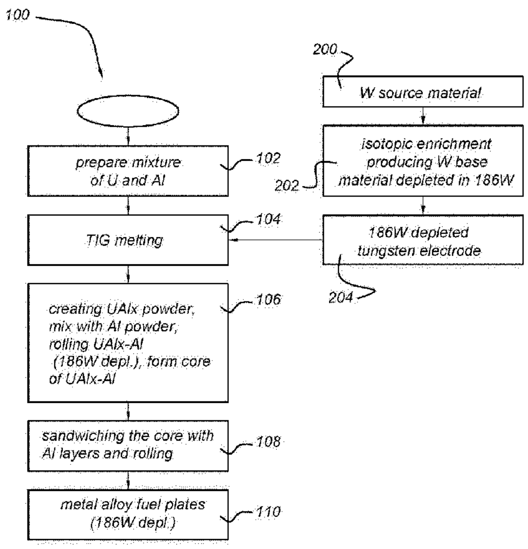
(54) **Manufacturing of a fissionable element metal alloy target.**

(57) A method for manufacturing a fissionable element metal alloy target includes a core and an enclosure, wherein the core is enveloped by the enclosure. The core material is a composite including an alloy of a fissionable element and one or more second elements, and a further element. The method includes collecting the material of the fissionable element and the material of the second element in a volume; melting the collected materials by applying heat supplied by an electric arc from a tungsten electrode to form and solidify an alloy of the fissionable element and the one or more second elements. The tungsten electrode consists of $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.




NL C 2011415

Dit octrooi is verleend ongeacht het bijgevoegde resultaat van het onderzoek naar de stand van de techniek en schriftelijke opinie. Het octrooischrift komt overeen met de oorspronkelijk ingediende stukken.

**Manufacturing of a fissionable element metal alloy target.**

Technical field

The present invention relates to a method for manufacturing a fissionable metal alloy target comprising a core and an enclosure, a use of a tungsten electrode in the manufacturing of such a target and a tungsten electrode.

Prior art

The most common technique for the production of the fission product $^{99}$Mo (molybdenum 99) is the neutron irradiation of metallic targets (e.g. $UAl_x$-Al), sometimes embodied as fuel plates, in a nuclear reactor. After the neutron irradiation the targets are dissolved within approximately 24 hours, after which the $^{99}$Mo is removed and purified from the dissolved material. $^{99}$Mo has a half-life of 66 hours. The $^{99}$Mo is a source that decays to $^{99}$Tc (Technetium 99), which is mainly used for medical applications, must be sufficiently pure in order to have an appropriate quality for medical injection. One of the chemical elements that is rather difficult to be removed from the $^{99}$Mo is tungsten. Tungsten is not a fission product but typically can be induced as impurity in the $^{99}$Mo, since tungsten impurity can be present in the target already at the start of the irradiation. Tungsten has several stable isotopes. The isotope $^{186}$W can absorb a neutron and becomes $^{187}$W, which has a half-life of 24 hours. $^{186}$W has a rather high abundance in natural tungsten (about 28 atomic%) and a rather high neutron absorption cross section for thermal neutrons (about 37 barn). This makes that natural tungsten impurities induce a significant amount of $^{187}$W during neutron irradiation. Also other radioactive tungsten radioisotopes can be formed due to neutron absorption of natural tungsten impurities, but these radioisotopes have a much smaller abundance and thereby a smaller radiological impact.

It is therefore an object of the present invention to provide a method that overcomes one or more of the disadvantages from the prior art.

Summary of the invention

The object is achieved by a method as defined in claim 1, for manufacturing a fissionable element metal alloy target comprising a core and an enclosure, wherein the core is enveloped by the enclosure;

the core material being a composite comprising an alloy of a fissionable element and at least one second element, and a further element;

the method comprising:

collecting the material of the fissionable element and the material of the second element in a volume;

melting the collected materials by applying heat supplied by an electric arc from a tungsten electrode to form and solidify an alloy of the fissionable element and the at least one second element ,

wherein the tungsten electrode consists of $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.

The tungsten in the metal alloy in the target originates from either impurities present in the fissionable and second element material from which the metal alloy target is formed or from impurities that enter into the metal alloy target from the manufacturing process. The material batches that are used for the fissionable and second element material are chosen such as to have a low amount of impurities, especially tungsten impurities. In this respect the melting and alloying of the fissionable and second element materials by exposing to a material that contains tungsten results in contamination of the materials by tungsten.

In particular, supplying heat from an  electron arc created by a tungsten electrode, e.g. a tungsten inert gas electrode (TIG),  to the metallic materials is a source for tungsten impurities in the alloy. The exposure of the tungsten electrode to the electric field, the electron arc and the associated discharge plasma causes a release of tungsten particles or vapor that can enter into the metal alloy target during the melting of the constituent materials. Typically, the release of tungsten is substantially independent on the isotope, which has the result that impurities in the metal alloy will have substantially the same isotope composition as the isotope composition of the tungsten electrode.

By reducing the relative abundance of the detrimental $^{186}W$ in the tungsten electrode, the amount of the $^{186}W$ in the tungsten impurity in the metal alloy and the metal alloy fuel plate will accordingly be reduced. As an advantage, during neutron irradiation, the creation of $^{187}W$ as impurity will be less due to the reduction of the relative abundance of $^{186}W$ in the tungsten impurity in the metal alloy target as caused by the TIG melting process.

Also, exposure of the materials to other tungsten-based tools for example in the form of a tungsten crucible during the manufacturing process can add tungsten impurities. According to the invention, such tungsten-based tools can also be made of $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.

The method can be used for uranium or any other fissionable element when the method involves melting of the collected materials by a tungsten based electrode.

According to an aspect, the present invention provides a method as described above that further comprises the step of forming an alloy powder from the solidified alloy of the fissionable element and the second element ;
mixing the alloy powder with powder of the further element to form to a powder mixture of the alloy and the further element;
and pressing the powder mixture to form the core.

According to an aspect, the present invention provides a method as described above that further comprises:
providing a first and a second plate of enclosure material;
creating a layered stack of the first plate, the second plate and the core, with the core being arranged between the first and second plates;
rolling the layered stack to form a fissionable element metal alloy target.

According to an aspect, the present invention provides a method as described above wherein
the second element is one selected from aluminum and silicon,
the further element is aluminum, and
the enclosure material is one selected from a group comprising aluminum, aluminum alloy, zirconium and Zr alloy.

Thus, in case the fissionable element is uranium, the alloy of the fissionable element and the second element maybe an uranium –aluminium compound (denoted $UAl_x$) or an uranium-silicon alloy (denoted here as USi). Such an alloy is manufactured by a melting process that involves a tungsten inert gas electric arc heating and possibly exposure to other tungsten based tools such as a crucible. Said alloy is then mixed with pure Al as further element to form an alloy compound – Al material (for example $UAl_x$-Al or USi-Al).

The enclosure material can be a pure aluminum but alternatively an aluminum alloy with low content of alloying elements as for instance Mg and/or Si can be used. Alternatively, the enclosure material can be zirconium or a zirconium alloy.

According to an aspect, the present invention provides a method as described above, wherein the compound material comprises a trace amount of tungsten, the trace amount of tungsten originating from the tungsten electrode being depleted in $^{186}W$ relative to the natural abundance of $^{186}W$ in tungsten.

According to an aspect, the present invention provides a method as described above, comprising:

the step of creating $^{186}W$ depleted tungsten by means of an isotope enrichment process that reduces the amount of $^{186}W$ relative to its natural abundance in tungsten.

According to an aspect, the present invention provides a method as described above, wherein the solid tungsten material of the electrode depleted in $^{186}W$ isotope has an abundance of elemental $^{186}W$ in tungsten being less than 20% at%.

According to an aspect, the present invention provides a method as described above, wherein the solid tungsten material of the electrode depleted in $^{186}W$ isotope has an abundance of elemental $^{186}W$ in tungsten being less than 10% at%.

According to an aspect, the present invention provides a method as described above, wherein the solid tungsten material of the electrode depleted in $^{186}W$ isotope has an abundance of elemental $^{186}W$ in tungsten being less than 5% at%.

According to an aspect, the present invention provides a method as described above, wherein the trace amount of tungsten in the compound material has an abundance of elemental $^{186}W$ in tungsten being less than 20 at%, preferably less than 10 at%, more preferably less than 5% at%.

According to an aspect, the present invention provides a method as described above, that further comprises creating a tungsten electrode depleted in $^{186}W$ from the $^{186}W$ depleted tungsten solid material.

According to an aspect, the present invention provides a method as described above, that further comprises the step of adding one or more high temperature oxides to the $^{186}W$ depleted tungsten before creating the tungsten electrode.

Additionally, the present invention relates to a use of a tungsten electrode in the manufacturing of a fissionable element metal alloy target as described above, with the tungsten electrode being arranged for melting the fissionable element and a second element to form an alloy of the fissionable element and the second element by tungsten inert gas electric arc heating, wherein the tungsten electrode consists of $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.

Also, the present invention relates to a fissionable element-based metal (uranium or other fissionable element) target comprising a layered stack of a first layer of a enclosure material, a second layer of the enclosure material, and a fissionable element-based core compound of an alloy of the fissionable element and at least one second element with a further element, the fissionable element-based core compound material being arranged between the first and second layers of the enclosure material, wherein the core material comprises a trace amount of tungsten, the trace amount of tungsten being depleted in $^{186}W$ relative to the natural abundance of $^{186}W$ in tungsten.

Moreover, the present invention relates to a tungsten electrode for an electric arc heating device, the electrode material comprising $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.

Furthermore, the present invention relates to a tungsten based tool for use in the manufacturing of a fissionable element-based core compound of an alloy of the fissionable element and a second element with a further element, wherein the tungsten-based tool comprises $^{186}W$ depleted tungsten, the depletion being relative to the natural abundance of $^{186}W$ in tungsten.

The tool can be selected from a group comprising an electric arc electrode and a crucible.

Advantageous embodiments are further defined by the dependent claims.

Brief description of drawings

The invention will be explained in more detail below with reference to drawings in which illustrative embodiments of the invention are shown.

The drawings are intended for illustration purposes only without limitation of the scope of protection, which is defined by the subject matter of the appended claims.

Figure 1 shows a process flow for manufacturing a fissionable element metal alloy target according to an embodiment of the invention.

Description of embodiments

Figure 1 shows a process flow for manufacturing a fissionable element metal alloy target according to an embodiment of the invention. A fissionable element metal alloy target is typically used in nuclear reactors to form fission products by irradiation with neutrons.

Such fission products are radio-isotopes of which some can be useful, i.e., radioactive isotopes that decay to stable isotopes and that can be used for radiation based applications, such as radiography (imaging), radiotherapy and radiopharmaceutical applications.

An example of a radiopharmaceutical application is the creation of $^{99}$Mo that decays to $^{99m}$Tc for application in a living being. The $^{99}$Mo isotope is created as a fission product by irradiating a fissionable element (e.g. uranium) metal alloy target (e.g., $UAl_x$-Al) by neutrons.

As mentioned in the introductory part, if the uranium target contains tungsten then also radioisotopes of tungsten will be created. Chemically separating molybdenum from tungsten is known to be difficult. Separating tungsten from molybdenum may require an additional processing step in the $^{99}$Mo production process, causing additional costs and delay. As a result, when $^{99}$Mo is chemically released from the metal alloy fuel plate, the obtained $^{99}$Mo product will contain also tungsten impurities that may be radioactive isotopes. In particular, $^{187}$W may have a detrimental impact on radiological application of $^{99}$Mo and it's daughter products.

In the process flow 100 of figure 1, the preparation of a metal alloy fuel target is described in which the contamination of the $^{99}$Mo product by $^{187}$W is significantly reduced.

Process flow 100 comprises after some initial steps a first step 102 of preparation of a mixture of a fissionable element and a second element. In an embodiment the fissionable element is uranium or an other fissionable element, the second element is aluminum or silicon. As mentioned above, silicon can be chosen as the second element. Optionally, the second element could be a mixture of Al and Si.

The fissionable element and the second element(s) are collected in a volume, such as a crucible.

Next in step 104, the mixture of the fissionable element and the second element(s) is melted using a tungsten electrode as heat source in a tungsten inert gas electric arc. Between the tungsten electrode and the mixture of the metals a high electric potential is generated, which creates an electric discharge (electric arc) between the electrode and the mixture of the metals. The tungsten electrode is surrounded by flowing inert gas. The electric discharge is accompanied by a release of heat that causes the mixture of metals to melt and form a compound material or alloy of the fissionable element and the second element(s).

In case of uranium as fissionable element and aluminum as second element a $UAl_x$ alloy is formed.

An alternative compound may comprise uranium and silicon for forming an USi alloy.

Subsequently the alloy material is then solidified. The alloy material is thereafter crushed, ground and sieved, thereby obtaining $UAl_x$ powder. The powder is inspected and thereafter mixed with aluminium powder. This mixture is homogenized and inspected. Thereafter core pressing or rolling of the $UAl_x$ -Al mixture takes place to create the fuel core.

In subsequent step 106, the core material is then machined into a shape of a plate.

In next step 108, the core material plate is arranged into an metallic enclosure by e.g. sandwiching between two plates of a third element for example, aluminum or Al alloy or zirconium or zirconium alloy. The stack of the plates is rolled (hot rolled) to form a uranium metal alloy target e.g., a target comprising a core of $UAl_x$-Al enclosed within two Al (alloy) plates or Zr (alloy) plates.

In case of uranium as first metal, aluminum as second metal and aluminium as a further metal, the metal alloy target comprises a stack of a first outer layer of aluminum, an intermediate layer of $UAl_x$-Al core and a second outer layer of aluminum, if aluminum is used as enclosing material.

If zirconium (alloy) is used as layer material, the metal alloy fuel plate comprises a stack of a first outer layer of zirconium (alloy), an intermediate layer of $UAl_x$-Al and a second outer layer of zirconium (alloy).

The metal alloy target can be further processed as will be described in more detail below.

With reference to the step 104 of melting the mixture of first and second metals it is noted that the tungsten electrode comprises tungsten that is depleted in $^{186}W$ relative to the natural abundance of $^{186}W$ in tungsten.

During the tungsten inert gas electric arc heating, some tungsten particles or tungsten vapor will be released from the tungsten electrode and enter the melted compound material as an impurity or trace amount. Thus the compound material will comprise a trace amount of tungsten. Since the release of tungsten during operation of the tungsten electrode is substantially independent of the isotope number, the trace amount of tungsten that is added to the uranium compound material during the tungsten inert gas heating will have a substantially same isotope composition as the tungsten electrode. Thus, if the electrode comprises tungsten that is $^{186}W$ depleted relative to the natural abundance of $^{186}W$ in tungsten, then the trace amount of W originating from the tungsten electrode in the compound material will show a $^{186}W$ depleted isotope composition.

The $^{186}W$ depleted tungsten electrode can be manufactured according to the steps 200 – 206.

In step 200, a tungsten source material is provided with a given abundance of $^{186}W$. Typically, the tungsten source material will be a tungsten source with natural abundance of $^{186}W$.

In step 202, the tungsten source material is converted in an isotopic enrichment process into a raw $^{186}W$ depleted tungsten material. The isotopic enrichment process could be based on an isotopic separation process for a gaseous tungsten compound such as tungsten-hexafluoride $WF_6$.

In step 204, from the raw $^{186}W$ depleted tungsten material a $^{186}W$ depleted tungsten electrode is fabricated using a method known in the art.

As known to a person skilled in the art, the $^{186}W$ depleted tungsten electrode may comprise one or more additives such as a high temperature oxide (e.g., a rare-earth oxide such as Lanthanum oxide) that improves the mechanical stability of the $^{186}W$ depleted tungsten electrode during high temperature operation.

The natural abundance of $^{186}W$ in tungsten is about 28.4 at.%. According to the invention, the abundance of $^{186}W$ in tungsten of the $^{186}W$ depleted tungsten electrode is about 20 at% or less, preferably about 10 at.% or less, more preferably the abundance of $^{186}W$ in tungsten of the $^{186}W$ depleted tungsten electrode is about 5 at.% or less.

The metal alloy target manufactured according to the invention, and that comprises the trace amount of tungsten depleted in $^{186}W$ can be used in a nuclear reactor (not shown) for creation of fission product from uranium by neutron irradiation.

The tungsten impurity in the metal alloy can form tungsten radioisotopes during the neutron irradiation. However, due to the $^{186}W$ depletion in comparison to the natural abundance, the amount of $^{187}W$ in the irradiated uranium metal alloy will be proportionally reduced.

After irradiation, the metal alloy target can be dissolved and radioisotopes such as $^{99}Mo$ can be isolated chemically. Since separation of Mo from W is difficult, the problem of contamination of $^{99}Mo$ by $^{187}W$ is reduced if the tungsten impurity in the metal alloy was depleted in $^{186}W$.

In particular for radiopharmaceutical applications of $^{99}Mo$, the use of $^{186}W$ depleted tungsten as electrode in the melting process of the fissionable element and the second element(s) is beneficial.

The skilled in the art will appreciate that the release of tungsten into the uranium-metal alloy may also originate from other tungsten-based tools such as a crucible. In accordance with an embodiment of the invention, such tungsten based tools can be manufactured from $^{186}W$ depleted tungsten material in similar manner as described above for a $^{186}W$ depleted tungsten electrode.

The invention has been described with reference to the preferred embodiment. Obvious modifications and alterations will occur to others upon reading and understanding the preceding detailed description. It is intended that the invention be construed as including all such modifications and alterations insofar as they come within the scope of the appended claims.

Conclusies

1. Werkwijze voor het vervaardigen van een metaallegering target van een splijtbaar element, het target omvattend een kern en een omhulsel, waarbij de kern wordt omgeven door het omhulsel;
het materiaal van de kern is samengesteld, omvattend een legering van een splijtbaar element en ten minste een tweede element, en een verdere element;
waarbij de werkwijze omvat:
verzamelen van het materiaal van het splijtbaar element en het materiaal van het tweede element in een volume;
smelten van de verzamelde materialen door toevoeren van warmte die geleverd wordt door een elektrische boog vanuit een wolfraam elektrode teneinde een legering van het splijtbaar element en het ten minste ene tweede element te vormen en vervolgens het stollen van de gesmolten legering,
waarbij de wolfraam elektrode bestaat uit $^{186}W$ verarmd wolfraam, waarbij de verarming is ten opzichte van het natuurlijk gehalte van $^{186}W$ in wolfraam.

2. Werkwijze volgens conclusie 1, verder omvattend de stap van het vormen van een legeringspoeder vanuit de gestolde legering van het splijtbaar element en het ten minste ene tweede element;
mengen van het legeringspoeder met poeder van het verdere element teneinde een poedermengsel te vormen van de legering en het verdere element;
en samenpersen van het poedermengsel teneinde de kern te vormen.

3. Werkwijze volgens conclusie 1 or 2, verder omvattend:
verschaffen van een eerste en tweede plaat van een omhulsel materiaal;
het vormen van een laagvormige stapeling van de eerste plaat, de tweede plaat en de kern, waarbij de kern is gerangschikt tussen de eerste en de tweede plaat van het omhulsel materiaal;
walsen van de laagvormige stapeling teneinde het target van de metaallegering van een splijtbaar element te vormen.

4. Werkwijze volgens willekeurig welke van conclusies 1 – 3, waarbij het ten minste ene tweede element er een of meer omvat die zijn gekozen uit

aluminium en silicium,
het verdere element aluminium is, en
het omhulsel materiaal er een is die gekozen wordt uit een groep omvattend aluminium, een aluminium legering, zirkonium en een zirkonium legering.

5. Werkwijze volgens willekeurig welke van conclusies 1- 4, waarbij het samengesteld materiaal van de kern een spoorhoeveelheid wolfraam bevat, waarbij de spoorhoeveelheid wolfraam die afkomstig is van de wolfraam elektrode verarmd in $^{186}W$ is ten opzichte van het natuurlijk gehalte van $^{186}W$ in wolfraam.

6. Werkwijze volgens willekeurig welke van conclusies 1 – 5, omvattend: de stap van het vormen van $^{186}W$ verarmd wolfraam door middel van een isotoopverrijkingsproces dat de hoeveelheid van $^{186}W$ ten opzichte van haar natuurlijk gehalte in wolfraam reduceert.

7. Werkwijze volgens conclusie 6, waarbij het vaste wolfraam materiaal van de elektrode dat verarmd in $^{186}W$ isotoop is, een gehalte van elementair $^{186}W$ in wolfraam heeft dat minder dan 20% at% is.

8. Werkwijze volgens conclusie 6, waarbij het vaste wolfraam materiaal van de elektrode dat verarmd in $^{186}W$ isotoop is, een gehalte van elementair $^{186}W$ in wolfraam heeft dat minder dan 10% at% is.

9. Werkwijze volgens conclusie 6, waarbij het vaste wolfraam materiaal van de elektrode dat verarmd in $^{186}W$ isotoop is, een gehalte van elementair $^{186}W$ in wolfraam heeft dat minder dan 5% at% is.

10. Werkwijze volgens conclusie 5, waarbij de spoorhoeveelheid van wolfraam in het samengesteld kernmateriaal een gehalte van elementair $^{186}W$ in wolfraam heeft dat minder dan 20at%, met voorkeur minder dan 10 at%, met de meeste voorkeur minder dan 5% at% is.

11. Werkwijze volgens willekeurig welke van conclusies 6 – 10, verder omvattend het vormen van een wolfraam elektrode die verarmd is in $^{186}W$ vanuit het vaste $^{186}W$ verarmd wolfraam materiaal.

12. Werkwijze volgens conclusie 11, verder omvattend de stap van het toevoegen van een of meer hoge temperatuur oxides aan het $^{186}W$ verarmd wolfraam voorafgaand aan het vormen van de wolfraam elektrode.

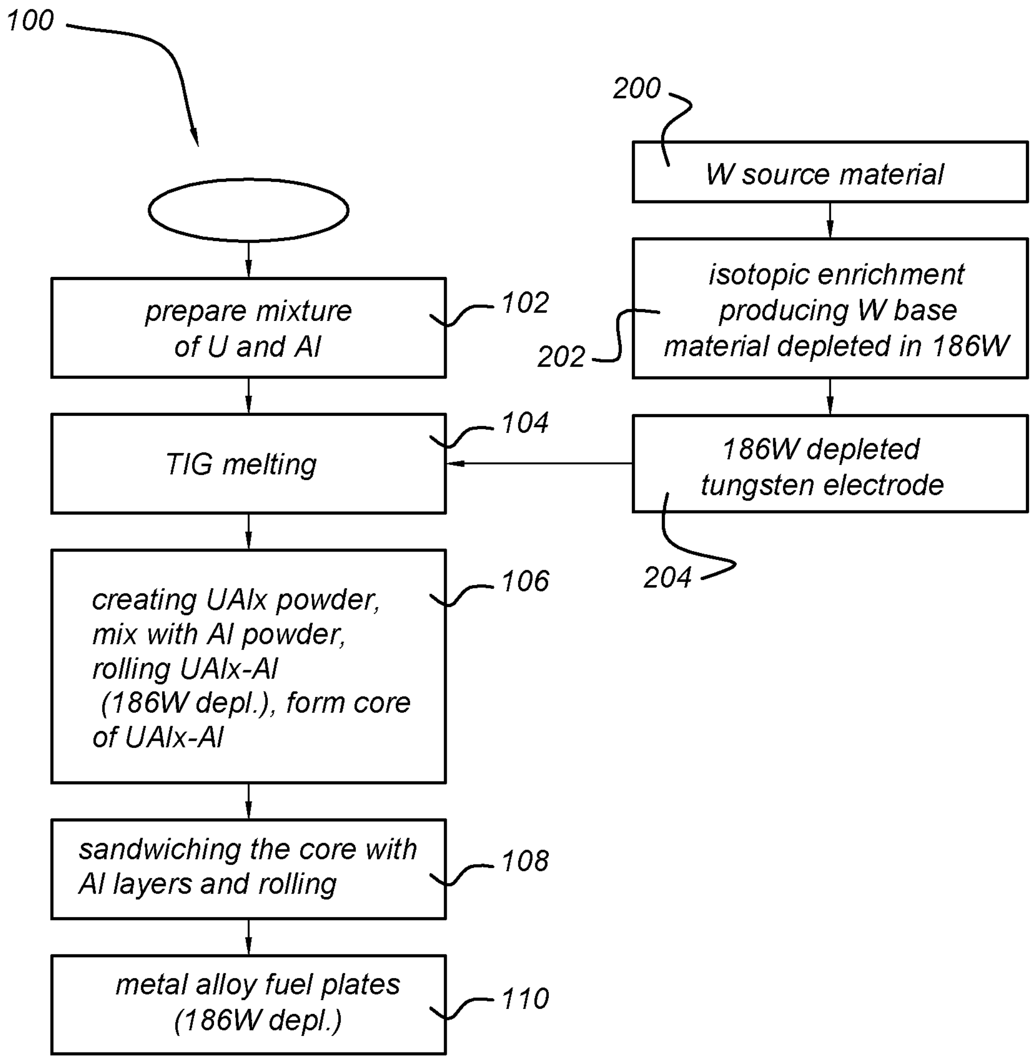
13. Gebruik van een wolfraam elektrode bij de vervaardiging van een metaallegering target van een splijtbaar element volgens willekeurig welke van conclusies 1 – 12, waarbij de wolfraam elektrode is ingericht voor het smelten van het splijtbaar element en ten minste een tweede element teneinde een legering van het splijtbaar element en het ten minste ene tweede element te vormen door verhitting middels een door de wolfraam elektrode gevormde elektrische boog, waarbij de wolfraam elektrode bestaat uit $^{186}W$ verarmd wolfraam, waarbij de verarming is ten opzichte van het natuurlijk gehalte van $^{186}W$ in wolfraam.

14. Metaaltarget op basis van een splijtbaar element omvattend een laagvormige stapeling van een eerste laag van een omhulsel materiaal, een tweede laag van het omhulsel materiaal, en een samengestelde kern op basis van het splijtbaar element van een legering van het splijtbaar element en ten minste een tweede element met een verder element, waarbij het op het splijtbaar element gebaseerd samengesteld kernmateriaal gerangschikt is tussen de eerste en tweede lagen van het omhulsel materiaal, waarbij het samengesteld materiaal van de kern een spoorhoeveelheid wolfraam omvat, waarbij de spoorhoeveelheid wolfraam verarmd is in $^{186}W$ ten opzichte het natuurlijk gehalte van $^{186}W$ in wolfraam.

15. Wolfraam elektrode voor een elektrische-boog-verhittingsinrichting, waarbij het elektrode materiaal $^{186}W$ verarmd wolfraam omvat, waarbij de verarming is ten opzichte van het natuurlijk gehalte van $^{186}W$ in wolfraam.

16. Op wolfraam gebaseerd gereedschap voor gebruik bij een vervaardiging een samengestelde kern op basis van een splijtbaar element-based omvattend het splijtbaar element en ten minste een tweede element, waarbij het op wolfraam gebaseerd gereedschap $^{186}W$ verarmd wolfraam omvat, waarbij de verarming is ten opzichte van het natuurlijk gehalte van $^{186}W$ in wolfraam.

17. Op wolfraam gebaseerd gereedschap volgens conclusie 16, waarbij het gereedschap er een is die gekozen wordt uit een groep omvattend een elektrode voor elektrische boog en smeltkroes.


100
200
W source material
isotopic enrichment producing W base material depleted in 186W
202
prepare mixture of U and Al
102
TIG melting
104
186W depleted tungsten electrode
204
creating UAlx powder, mix with Al powder, rolling UAlx-Al (186W depl.), form core of UAlx-Al
106
sandwiching the core with Al layers and rolling
108
metal alloy fuel plates (186W depl.)
110

# SAMENWERKINGSVERDRAG (PCT)

## RAPPORT BETREFFENDE NIEUWHEIDSONDERZOEK VAN INTERNATIONAAL TYPE

| IDENTIFICATIE VAN DE NATIONALE AANVRAGE | KENMERK VAN DE AANVRAGER OF VAN DE GEMACHTIGDE |
|---|---|
| | **P6046418NL** |
| Nederlands aanvraag nr.<br>**2011415** | Indieningsdatum<br>**10-09-2013** |
| | Ingeroepen voorrangsdatum |
| Aanvrager (Naam)<br>**Nuclear Research and Consultancy Group V.O.F.** | |
| Datum van het verzoek voor een onderzoek van internationaal type<br>**30-11-2013** | Door de Instantie voor Internationaal Onderzoek aan het verzoek voor een onderzoek van internationaal type toegekend nr.<br>**SN 61113** |

| **I. CLASSIFICATIE VAN HET ONDERWERP** (bij toepassing van verschillende classificaties, alle classificatiesymbolen opgeven) | | | |
|---|---|---|---|
| Volgens de internationale classificatie (IPC) | | | |
| | **C22C1/02** | **C22C43/00** | **G21G1/08** |
| | **F27D11/08** | | |

| **II. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK** | | | |
|---|---|---|---|
| Onderzochte minimumdocumentatie | | | |
| Classificatiesysteem | Classificatiesymbolen | | |
| **IPC** | **C22C** | **G21G** | **F27D** |
| Onderzochte andere documentatie dan de minimum documentatie, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen | | | |
| | | | |

| | | | |
|---|---|---|---|
| **III.** | ☐ | **GEEN ONDERZOEK MOGELIJK VOOR BEPAALDE CONCLUSIES** | (opmerkingen op aanvullingsblad) |
| **IV.** | ☐ | **GEBREK AAN EENHEID VAN UITVINDING** | (opmerkingen op aanvullingsblad) |

## ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2011415

A. CLASSIFICATIE VAN HET ONDERWERP
INV. C22C1/02 C22C43/00 G21G1/08 F27D11/08
ADD.

Volgens de Internationale Classificatie van octrooien (IPC) of zowel volgens de nationale classificatie als volgens de IPC.

B. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK

Onderzochte miminum documentatie (classificatie gevolgd door classificatiesymbolen)
C22C G21G F27D

Onderzochte andere documentatie dan de mimimum documentatie, voor dergelijke documenten, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen

Tijdens het onderzoek geraadpleegde elektronische gegevensbestanden (naam van de gegevensbestanden en, waar uitvoerbaar, gebruikte trefwoorden)

EPO-Internal, COMPENDEX

C. VAN BELANG GEACHTE DOCUMENTEN

| Categorie ° | Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages | Van belang voor conclusie nr. |
|---|---|---|
| A | KANWAR LIAQAT ALI ET AL: "Development of low enriched uranium target plates by thermo-mechanical processing of UAl2-Al matrix for production of 99Mo in Pakistan", NUCLEAR ENGINEERING AND DESIGN, deel 255, 1 februari 2013 (2013-02-01), bladzijden 77-85, XP055119329, ISSN: 0029-5493, DOI: 10.1016/j.nucengdes.2012.10.014 * het gehele document * ----- | 1-17 |

-/--

[X] Verdere documenten worden vermeld in het vervolg van vak C.

[ ] Leden van dezelfde octrooifamilie zijn vermeld in een bijlage

° Speciale categorieën van aangehaalde documenten

"A" niet tot de categorie X of Y behorende literatuur die de stand van de techniek beschrijft
"D" in de octrooiaanvrage vermeld
"E" eerdere octrooi(aanvrage), gepubliceerd op of na de indieningsdatum, waarin dezelfde uitvinding wordt beschreven
"L" om andere redenen vermelde literatuur
"O" niet-schriftelijke stand van de techniek
"P" tussen de voorrangsdatum en de indieningsdatum gepubliceerde literatuur

"T" na de indieningsdatum of de voorrangsdatum gepubliceerde literatuur die niet bezwarend is voor de octrooiaanvrage, maar wordt vermeld ter verheldering van de theorie of het principe dat ten grondslag ligt aan de uitvinding
"X" de conclusie wordt als niet nieuw of niet inventief beschouwd ten opzichte van deze literatuur
"Y" de conclusie wordt als niet inventief beschouwd ten opzichte van de combinatie van deze literatuur met andere geciteerde literatuur van dezelfde categorie, waarbij de combinatie voor de vakman voor de hand liggend wordt geacht
"&" lid van dezelfde octrooifamilie of overeenkomstige octrooipublicatie

| Datum waarop het onderzoek naar de stand van de techniek van internationaal type werd voltooid | Verzenddatum van het rapport van het onderzoek naar de stand van de techniek van internationaal type |
|---|---|
| 21 mei 2014 | |

| Naam en adres van de instantie | De bevoegde ambtenaar |
|---|---|
| European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. (+31-70) 340-2040,<br>Fax: (+31-70) 340-3016 | Mauger, Jeremy |

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## ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2011415

C.(Vervolg). VAN BELANG GEACHTE DOCUMENTEN

| Categorie ° | Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages | Van belang voor conclusie nr. |
|---|---|---|
| A | "Production technologies for molybdenum-99 and technetium-99m",<br>IAEA TECDOC 1065,<br>28 februari 1999 (1999-02-28), bladzijden 1-158, XP055119320,<br>Gevonden op het Internet:<br>URL:http://www-pub.iaea.org/MTCD/Publications/PDF/te_1065_prn.pdf<br>[gevonden op 2014-05-21]<br>* bladzijde 19 - bladzijde 24 *<br>----- | 1-17 |

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Formulier PCT/ISA/201 (vervolg tweede blad) (Januari 2004)

# WRITTEN OPINION

| File No. | Filing date *(day/month/year)* | Priority date *(day/month/year)* | Application No. |
|---|---|---|---|
| SN61113 | 10.09.2013 | | NL2011415 |
| International Patent Classification (IPC)<br>INV. C22C1/02 C22C43/00 G21G1/08 F27D11/08 | | | |
| Applicant<br>Nuclear Research and Consultancy Group V.O.F. | | | |

This opinion contains indications relating to the following items:

- ☒ Box No. I — Basis of the opinion
- ☐ Box No. II — Priority
- ☐ Box No. III — Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV — Lack of unity of invention
- ☒ Box No. V — Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI — Certain documents cited
- ☒ Box No. VII — Certain defects in the application
- ☒ Box No. VIII — Certain observations on the application

| | Examiner |
|---|---|
| | Mauger, Jeremy |

# WRITTEN OPINION

Application number
NL2011415

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**Box No. I Basis of this opinion**

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application and necessary to the claimed invention, this opinion has been established on the basis of:

   a. type of material:

   ☐ a sequence listing

   ☐ table(s) related to the sequence listing

   b. format of material:

   ☐ on paper

   ☐ in electronic form

   c. time of filing/furnishing:

   ☐ contained in the application as filed.

   ☐ filed together with the application in electronic form.

   ☐ furnished subsequently for the purposes of search.

3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

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**Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

| | | | |
|---|---|---|---|
| Novelty | Yes: | Claims | 1-17 |
| | No: | Claims | |
| Inventive step | Yes: | Claims | 1-17 |
| | No: | Claims | |
| Industrial applicability | Yes: | Claims | 1-17 |
| | No: | Claims | |

2. Citations and explanations

   **see separate sheet**

Application number

# WRITTEN OPINION

NL2011415

**Box No. VII Certain defects in the application**

see separate sheet

**Box No. VIII Certain observations on the application**

see separate sheet

## Re Item V

**Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1 Reference is made to the following documents:

D1 KANWAR LIAQAT ALI ET AL: "Development of low enriched uranium target plates by thermo-mechanical processing of UAl2-Al matrix for production of 99Mo in Pakistan",
NUCLEAR ENGINEERING AND DESIGN,
deel 255, 1 februari 2013 (2013-02-01), bladzijden 77-85, XP055119329,
ISSN: 0029-5493, DOI: 10.1016/j.nucengdes.2012.10.014

D2 "Production technologies for molybdenum-99 and technetium-99m",
IAEA TECDOC 1065, 28 februari 1999 (1999-02-28), bladzijden 1-158,
XP055119320,
Gevonden op het Internet:
URL:http://www-pub.iaea.org/MTCD/Publications/PDF/te_1065_prn.pdf
[gevonden op 2014-05-21]

2 Document D1 (see whole document) represents the closest prior art for the process of claim 1, the use of claim 13, the target of claim 14 and the tungsten tools/electrodes of claims 15 and 16 and discloses processes for manufacturing $UAl_2$-Al targets useful for the production of the medically useful isotope $^{99}$Mo. The process notably involved making the alloy for the target by arc melting (see sections 2.1 and 3). The document does not describe the electrode used in the arc melting.

2.1 The claimed subject-matter is novel because document D1 does not disclose tungsten electrodes depleted in respect of $^{186}$W, methods using or uses of such electrodes or targets made using such electrodes (the tungsten in such targets will also be depleted).

2.2 The technical effect of the use of such electrodes is that the target has a lower concentration of $^{186}$W, which forms $^{187}$W upon irradiation, a significant and difficult to remove radioisotope in $^{99}$Mo. The problem addressed by all the claims is thus providing means useful when seeking simpler methods to obtain pure $^{99}$Mo.

2.3 The solution to the problem is provided by $^{186}$W depleted electrodes.

2.4 The solution to this problem is not obvious. The problem of tungsten contamination is recognised in document D2 (see pages 19-24). The source of the tungsten was not identified in document D2 and the solution proposed in document D2 (chemical purification of the molybdenum oxide target used in this document) is completely different. Thus the claimed method, use, target and electrodes all involve an inventive step.

**Item VII**

**Certain defects in the application**

1 The relevant background art disclosed in documents D1 and D2 is not mentioned in the description, nor are these documents identified therein.

**Re Item VIII**

**Certain observations on the application**

1 Although claims 15 and 16 have been drafted as separate independent claims, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought and/or in respect of the terminology used for the features of that subject-matter. The aforementioned claims therefore lack conciseness.