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(54) **Fabrication process of uranium foil having fine grains solidified rapidly from melt using roll, and the fabrication apparatus**

Herstellungsverfahren von feinkörniger Uranfolie aus schnell abgekühlter Schmelze durch Kühlwalzen und Herstellungsgerät

Procédé de fabrication d'un ruban en uranium à grains fins solidifié rapidement par un cylindre refroidisseur et appareil de fabrication

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- **PATENT ABSTRACTS OF JAPAN vol. 018, no. 072 (M-1555), 7 February 1994 (1994-02-07) & JP 05 287307 A (SEIKO EPSON CORP), 2 November 1993 (1993-11-02)**
- **DATABASE CHEMICAL ABSTRACTS [Online] J. Metals (1954), 6 AIME Trans. 200, p.999-1003, 1954 W. SEYMOUR: "Preferred orientation of cold-rolled uranium foil" Database accession no. 48:63915 XP002190725**

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Description

BACKGROUND OF THE INVENTION

1. Field of the Invention

[0001] The present invention relates generally to a fabrication process for uranium foil having fine crystalline grains solidified rapidly from a melt using cooling rolls, and foil fabrication apparatus. In particular, the present invention relates to low or high enrichment uranium and uranium alloy foil, the fabrication apparatus, and the fabrication process therefore wherein uranium (U) and uranium alloy [U-(A)Q-(B)X-(C)Y (Q: Al, Fe, Ni, Si, Cr, Zr element, X: Al, Fe, Ni, Si, Cr, Zr element, Y: Al, Fe, Ni, Si, Cr, Zr element, $Q \neq X \neq Y$, $(A) \leq 1$ wt%, $(B) \leq 1$ wt%, $(C) \leq 1$ wt%)] foils are directly cast from a melt, not through a vacuum induction melting & casting, ingot-cutting, hot-rolling and heat-treatment process, but through a twin-roll-casting process.

2. Description of the Prior Art

[0002] Generally, the conventional fabrication method for uranium foil has the disadvantages of complicated processes as follows: casting the uranium or the uranium alloy after holding at about 1300 °C in a vacuum induction furnace; cutting the resulting rod-type ingot to suitable size for hot rolling at 600 °C or higher; rolling through many passes a thick piece of the ingot to gradually thin it to fabricate a uranium foil of 100-200 µm thickness; and finally heat-treatment at 800 °C and quenching the fabricated uranium foil to produce the required gain size and orientation.

[0003] In the conventional method, the uranium must be heated and rolled under vacuum or in an inert atmosphere because it is a reactive material. The hot rolling is repeated several times to obtain a suitable thickness of the uranium foil. As the hot-rolling process takes long time, productivity is relatively low. A washing/drying process must be done to remove surface impurities after hot rolling. In order to obtain the fine grain structure which has a more stable behavior during irradiation, heat-treatment and quenching must be performed. The high hardness and low ductility of uranium or uranium alloy make it difficult to roll the foil. The foil is liable to crack owing to residual stress during the process, resulting in a low yield. The present invention is expected to improve the economy of producing uranium foil, due to a higher productivity and yield than the conventional method.

[0004] Meanwhile, a uranium foil having excessive residual stress from hot rolling may be deformed or damaged during thermal cycling during irradiation. Furthermore, deformed areas or cracks generated during thermal cycling may act as penetration paths through which there can be an interdiffusion reaction of the uranium with a coating layer, such as Al or Ni, which serves as a protector against the reaction of the uranium with a fixed tube in an irradiation target.

[0005] Rare earth magnet foils are already manufactured through twin-roll casting and under vacuum according to JP-A-5-287 307.

SUMMARY OF THE INVENTION

[0006] The present invention aims to alleviate the problems as described above. Low or high enrichment uranium and uranium alloy foil are fabricated through twin-roll casting of a uranium melt without a hot-rolling process and heat-treatment process. An improvement in productivity and process economics due to process simplification and better quality from the absence of any residual stress on the foil are expected.

[0007] Accordingly, the said objects of the present invention can be achieved by providing a uranium foil solidified rapidly by twin-roll casting as defined in the method according to claim 1, and in apparatus as defined in claim 2 for fabricating the same.

[0008] More specifically, the present invention is achieved by providing an apparatus and a method for fabricating the uranium foil wherein low or high enrichment uranium and alloy element material are weighed and then charged into a heat-resistant crucible equipped with a tundish having a slot. The crucible is mounted at the inner part of a vacuum chamber in which a vacuum of 10^{-3} torr or higher can be maintained by a vacuum pump system. Then, the uranium and the alloying elements are melted by high frequency electric power. The alloy melt is fed through the slot to the space between side dams and two rotating rolls driven by an electric motor, and thin foil is formed by pressing the partly solidified melt with the cold rolls. The foil produced is further cooled by supplying inert cooling gas.

BRIEF DESCRIPTION OF THE DRAWING

[0009] The above objects, features and advantages of the present invention will be more clearly understood from the following detailed description taken in conjunction with the accompanying drawing, in which:

> Fig. 1 shows a schematic view of a configuration of an apparatus for fabricating the uranium foil according to the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0010] With reference to Fig. 1, there is an apparatus for fabricating the uranium foil by twin rolls according to the present invention.

[0011] The apparatus of the present invention comprises: a heat-resistant crucible (2), a tundish having a slot (1), an induction coil (3) connected to a high-frequency generation apparatus (not shown) to increase the temperature of the crucible (2), a vacuum pump system (5) for evacuating the chamber (4) to an appropriate degree of vacuum, or twin-roll casting device with a roller (6) installed in the chamber (4), side dams with a resistance heater (not shown) for preventing a rapid decrease of the uranium melt temperature after pouring from the crucible and for guiding the uranium melt into the roller (6), a gas-feeding valve (7) for supplying a cooling gas into the chamber (4), and a recovery container (8) for collecting the fabricated foil.

[0012] Uranium and alloy elements are charged at the lower part of the heat-resistant crucible (2) mounted at the upper part of the chamber (4). Also a stopper (9) is installed in the crucible. The induction coil (3) mounted around the heat-resistant crucible (2) superheats the charged material to about 200 °C higher than the melting temperature by the high frequency generation apparatus.

[0013] The vacuum pump system (5) allows an inner part of the chamber (4) to be evacuated to a suitable vacuum of 10^{-3} torr or higher and is connected to one side of the chamber (4).

[0014] The roller (6) is positioned in the same central plane as the slot (1) in the chamber (4). Also, the roller is operated by an electric motor (not shown) to manufacture the uranium foil from an alloy melt poured through the slot (1). The preheated side dams (not shown) prevent the rapid decrease of the uranium melt temperature after pouring from the crucible and guide the uranium melt into the roller (6).

[0015] The alloy melt fed through the slot (1) is fabricated into uranium foil by the rotating rolls driven by the electric motor, and the resulting foil is rapidly cooled under argon or helium cooling gas.

[0016] The gas-feeding valve (7), connected to the chamber (4), feeds inert argon or helium cooling gas into the chamber (4) to rapidly cool the uranium foil fabricated by the cooling roller (6).

[0017] The inert argon or helium cooling gas, flowing into the chamber (4) through the gas-feeding valve (7), is injected into the chamber (4) through gas nozzles mounted at the upper part of the chamber (4).

[0018] The recovery container (8) at the bottom part of the chamber (4) collects the thin foil (10) manufactured in the chamber (4).

[0019] A better understanding of the present invention may be obtained in light of the following examples which are set forth to illustrate, but are not to be construed to limit, the present invention.

EXAMPLE 1

[0020] To fabricate low or high enrichment uranium foil, uranium material is charged into a heat-resistant crucible (2) having a slot (1). The crucible (2) and an insulation material (not shown) are assembled in proper order in a fabrication apparatus.

[0021] Then, the steel chamber in the apparatus equipped with the crucible (2) is evacuated to a vacuum of 10^{-3} torr or higher by a vacuum pump system (5).

[0022] A high frequency generator (3) is operated to superheat the charge material of the crucible to a temperature of about 200 °C higher than the melting temperature of the uranium.

[0023] When the rotation speed of the roller (6) driven by the electric motor is stabilized at about 300 rpm, a stopper (9) placed in the crucible (2) is lifted upward to discharge an alloy melt.

[0024] The poured alloy melt passes through the slot (1) of 1-mm width and then is fed to the space between the preheated side dams (not shown) and the roller (6) rotating at about 300 rpm to form the thin foil (10).

[0025] Then, a gas-feeding valve (7) is operated to supply the chamber (4) with the inert cooling gas, whereby the foil (10) fabricated by the roller (6) is rapidly solidified (at a rate greater than 10^3 °C/sec).

[0026] Because a uranium foil having fine and randomly orientated grains is directly obtained by such a rapid solidification effect, it is not necessary to heat-treat the foil and quench from about 800 °C to form fine grains. The uranium foil is collected within a recovery container (8) installed at the bottom of the chamber (4).

[0027] The foil in the recovery container (8) is about 125 μm thick and the foil with an acceptable thickness is recovered at about 90% yield.

[0028] A piece of uranium foil prepared as previously described is assembled with other components to fabricate an irradiation target, in order to charge into a reactor for producing fission isotope ^{99}Mo, the only parent nuclide of ^{99m}Tc, which is an extremely useful tool for medical diagnosis.

EXAMPLE 2

[0029] This invention is applied to a uranium alloy of low or high enrichment uranium [U-(A)Q-(B)X-(C)Y (Q: Al, Fe, Ni, Si, Cr, Zr elements, X: Al, Fe, Ni, Si, Cr, Zr elements, Y: Al, Fe, Ni, Si, Cr, Zr elements, $Q \neq X \neq Y$, $(A) \leq 1$ wt%, $(B) \leq 1$ wt%, $(C) \leq 1$ wt%)] foil for the irradiation target.

[0030] For the fabrication of U-500ppm Fe-1200ppm Al-500ppm Ni alloy foil, uranium and alloy elements including Fe, Al and Ni are appropriately weighed according to the desired alloy composition and charged to the crucible. The steel chamber (4) is evacuated to 10^{-3} torr or higher using the vacuum pump system (5) as described in a fabrication procedure for uranium foil.

[0031] As such, when the crucible (2) and the insulation material (not shown) are assembled in the proper order in the apparatus, the high frequency generator (3) is operated to superheat the charge material of the crucible to a temperature about 200 °C higher than the melting temperature of the uranium alloy.

[0032] When the rotation speed of the roller (6) operated by the electric motor is stabilized at about 300 rpm, the stopper (9) placed in the crucible (2) is lifted upward to feed the alloy melt.

[0033] The discharged alloy melt passes through the slot (1) of 1.2 mm width and then is fed to the space between the preheated side dams (not shown) and the roller (6) rotating at the high speed of 300 rpm to form the thin foil (10).

[0034] Then, the gas-feeding valve (7) is operated to inject the inert cooling gas into the chamber (4), whereby the foil (10) fabricated by the roller (6) is quickly solidified (10^3 °C/sec or faster).

[0035] Because the uranium alloy foil having fine and randomly orientated grains is directly obtained by such a rapid solidification effect, it is not necessary to heat-treat the hot rolled foil and quench from about 800 °C to form fine grains. The uranium alloy foil is collected within a container (8) installed at the bottom of the chamber (4).

[0036] The foil in the recovery container (8) is about 150 μm thick, and foil with a suitable thickness is recovered at a 90% or higher yield.

[0037] A piece of uranium foil prepared as previously described is assembled with other components to fabricate an irradiation target, in order to charge into a reactor for producing fission isotope ^{99}Mo, the only parent nuclide of ^{99m}Tc, which is an extremely useful tool for medical diagnosis.

[0038] The fabrication process of uranium alloy foil by the present invention is greatly simplified compared to the conventional fabrication method, which includes a vacuum induction melting process, a repetitive hot-rolling process, a washing/drying process for removing impurities, such as surface oxides, and a heat-treatment process for obtaining fine isotropic grains.

[0039] Since the melt of uranium or uranium alloy is rapidly cooled to directly fabricate the uranium foil, the uranium or uranium foil, being difficult to roll due to its high toughness, can be easily fabricated.

[0040] A long time period is required to conduct the repetitive troublesome conventional hot-rolling process to adjust the thickness of a uranium ingot. In contrast, the alloy melt may be cast at once to fabricate large amounts of the foil in a few minutes by the present invention, thereby having a high productivity.

[0041] In addition, because the uranium is lacking in ductility, the uranium foil may be damaged and cracked owing to an induced stress during the hot-rolling process, which leads to a low yield and a reduced economic efficiency.

[0042] However, the foil fabrication process by rapid solidification of the present invention has a 90% or higher yield through which several kilograms of the foil can be directly fabricated in a few minutes. The foil fabrication process, using preheated side dams and a cooling roller, facilitates control of the width of the foil. Accordingly, the yield of uranium is very high and the economics are highly favorable because enriched uranium is very expensive.

[0043] Furthermore, foil fabricated by twin rolling has smaller stress than foil obtained through repetitive hot rolling of a uranium plate. Accordingly, deformation or cracking of the foil generated by thermal cycling during the irradiation process can be prevented. Defects in deformation areas or cracks can act as penetration paths for elements in the coating layer of the target. The interaction between coating layer and target will be enhanced by the defects or cracks. However, the foil fabricated by the present invention does not have such paths.

[0044] Commonly, uranium foil undergoes large anisotropic growth during irradiation in a reactor. However, the uranium foil of the present invention has homogeneous and fine grains with random orientation so as to prevent the uranium foil from excessively growing during irradiation.

Claims

1. A method for fabricating a uranium foil of thickness 100-200 μm having fine randomly oriented grains, **characterised by** rapidly solidifying through twin-roll casting a melt of uranium or uranium alloy under controlled vacuum and further cooling by inert cooling gas, directly using a cooling roller, wherein the uranium alloy is U-(A)Q-(B)X-(C)Y, in which Q is selected from the group consisting of Al, Fe, Ni, Si, Cr, and Zr elements, X is selected from the group consisting of Al, Fe, Ni, Si, Cr, and Zr elements, and Y is selected from the group consisting of Al, Fe, Ni, Si, Cr, and Zr elements, and $Q \neq X \neq Y$, $(A) \leq 1$ wt.%, $(B) \leq 1$ wt.%, $(C) \leq 1$ wt.%.

2. A twin-roll casting apparatus for fabricating a uranium foil having fine randomly oriented grains by solidifying rapidly a melt of uranium or uranium alloy directly using a cooling roller, comprising

   a chamber (4),

   a heat-resistant crucible (2) within said chamber and equipped with a tundish having a slot (1),

   an induction coil (3) connected to a high frequency generating apparatus to heat the crucible (2),

   a vacuum pump system (5) for obtaining an appropriate vacuum within the chamber (4),

   a twin-roll casting device with a cooling roller (6) and preheated side dams localised between said slot and said twin-roll casting device and both installed in the chamber (4),

   a gas-feeding valve (7) for feeding gas into the chamber (4) and

   a recovery container (8) for collecting the fabricated foil.

Patentansprüche

1. Verfahren zur Herstellung einer Uranfolie mit einer Dicke von 100 bis 200 μm und mit feinen willkürlich

ausgerichteten Körnern, **gekennzeichnet durch** schnelles Verfestigen **durch** Doppelwalzengießen einer Schmelze von Uran oder Uranlegierung unter gesteuertem Vakuum und weiteres Abkühlen **durch** inertes Kühlgas bei direktem Verwenden einer Kühlwalze, wobei die Uranlegierung U-(A)Q-(B) X-(C)Y ist, wobei Q aus der Gruppe bestehend aus Al-, Fe-, Ni-, Si-, Cr- und Zr-Elementen ausgewählt ist, X aus der Gruppe bestehend aus Al-, Fe-, Ni-, Si-, Cr- und Zr-Elementen ausgewählt ist und Y aus der Gruppe bestehend aus Al-, Fe-, Ni-, Si-, Cr- und Zr-Elementen ausgewählt ist und Q ≠ X ≠ Y, (A) ≤ 1 Gew.-%, (B) ≤ 1 Gew.-% und (C) ≤ 1 Gew.-%.

2. Doppelwalzengießgerät zur Herstellung einer Uranfolie mit feinen willkürlich ausgerichteten Körnern durch schnelles Verfestigen einer Schmelze von Uran oder Uranlegierung bei direktem Verwenden einer Kühlwalze, umfassend:

eine Kammer (4),

einen wärmebeständigen Tiegel (2) innerhalb der Kammer und ausgestattet mit einem Zwischenbehälter, welcher einen Schlitz (1) aufweist ,

eine Induktionsspule (3), welche mit einem Hochfrequenzerzeugungsgerät verbunden ist, um den Tiegel (2) zu erwärmen,

ein Vakuumpumpensystem (5) zum Erhalten eines angemessenen Vakuums innerhalb der Kammer (4),

eine Doppelwalzengießvorrichtung mit einer Kühlwalze (6) und vorgeheizten Seitenüberläufen, welche zwischen dem Schlitz und der Doppelwalzengießvorrichtung angeordnet und beide in der Kammer(4)eingebaut sind,

ein Gaszuführventil (7) zum Zuführen von Gas in die Kammer (4) und

einen Rückgewinnungsbehälter (8) zum Sammeln der hergestellten Folie.

Revendications

1. Procédé de fabrication d'un ruban en uranium d'une épaisseur comprise entre 100 et 200 μm à grains fins orientés au hasard, **caractérisé par** une solidification rapide à l'aide d'une coulée entre cylindres d'une coulée d'uranium ou d'alliage d'uranium sous un vide contrôlé et par un refroidissement à l'aide d'un gaz de refroidissement inerte, en utilisant directement un cylindre refroidisseur, et dans lequel l'alliage d'uranium est U-(A)Q-(B)X-(C)Y, où Q est choisi parmi le groupe comprenant les éléments Al, Fe, Ni, Si, Cr et Zr, X est choisi parmi le groupe comprenant les éléments Al, Fe, Ni, Si, Cr et Zr et Y est choisi parmi le groupe comprenant les éléments Al, Fe, Ni, Si, Cr et Zr, et Q ≠ X ≠ Y, (A) ≤ 1 % en poids, (B) ≤ 1 % en poids, (C) ≤ 1 % en poids.

2. Appareil de coulée entre cylindres permettant de fabriquer un ruban d'uranium à grains fins orientés au hasard par une solidification rapide d'une coulée d'uranium ou d'alliage d'uranium en utilisant directement un cylindre refroidisseur comprenant
une chambre (4) ,
un creuset réfractaire (2) placé dans ladite chambre et doté d'un panier de coulée ayant une fente (1),
une bobine d'induction (3) raccordée à un appareil générateur de haute fréquence pour chauffer le creuset (2),
un système de pompe sous vide (5) pour obtenir un vide approprié au sein de la chambre (4),
un dispositif de coulée entre cylindres ayant un cylindre refroidisseur (6) et des seuils latéraux préchauffés localisés entre ladite fente et ledit dispositif de coulée entre deux cylindres, les deux étant installés dans la chambre (4),
une vanne d'alimentation en gaz (7) pour amener le gaz dans le chambre (4) et
un récipient de récupération (8) pour collecter le ruban fabriqué.

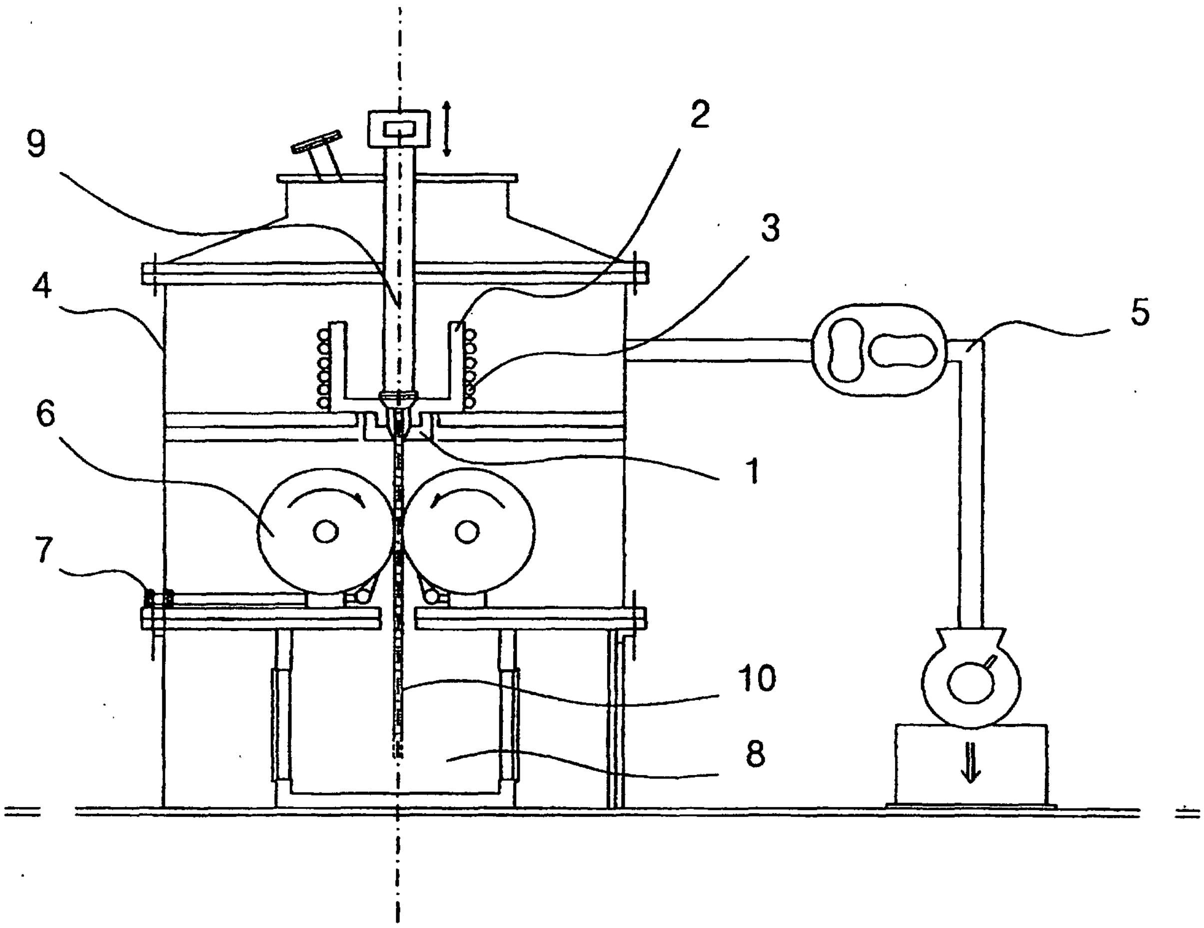

FIG. 1