(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) **EP 3 671 762 A1**

(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 153(4) EPC

(43) Date of publication:
24.06.2020 Bulletin 2020/26

(21) Application number: **18836226.3**

(22) Date of filing: **23.04.2018**

(51) Int Cl.:
G21G 1/06 *(2006.01)*

(86) International application number:
PCT/RU2018/000258

(87) International publication number:
WO 2019/035736 (21.02.2019 Gazette 2019/08)

(84) Designated Contracting States:
AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR
Designated Extension States:
BA ME
Designated Validation States:
KH MA MD TN

(30) Priority: **14.08.2017 RU 2017129007**

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(54) METHOD OF PRODUCING THE RADIONUCLIDE NICKEL-63

(57) The invention relates to the field of production of radioactive isotopes and more specifically to the technology of production of the radioactive isotope Nickel-63 that is used to manufacture beta-voltaic current sources.

The method for producing Nickel-63 radionuclide involves obtaining an initial nickel target enriched in Nickel-62 to get the content as high as 98% or more, the bombardment of the target in the reactor and the enrichment of the exposed product into a light fraction. The initial nickel enriched in Nickel-62 is added to the light fraction until it reaches a content of 98% or more, and is used to make a secondary nickel target. The remaining heavy fraction is converted into a metal form and used for the manufacture of beta-radiation sources that are applied in beta-voltaic current sources.

After the bombardment, the solution of the nickel target is subjected to radiochemical cleaning to remove Cu-65 and gamma-active isotopes, in particular Fe-59 and Co-60.

The technical result consists in the most complete loading of the reactor cell with Nickel-62 isotope and an increase in the amount of Nickel-63 radionuclide produced.

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Description

[0001] The invention relates to the field of production of radioactive isotopes and more specifically to the technology of production of the radioactive isotope Nickel-63, mainly for use in beta-voltaic current sources.

[0002] A method of producing a nickel-63 radionuclide for beta-voltaic current sources is known, including the production of a nickel-62 enriched target from original nickel with a nickel-64 content of over 2%, bombardment of the target in the reactor and subsequent enrichment of the exposed product using nickel-63 until their content in the enriched product is 75% or more (see RF Patent No. 2 569 543, G 21 G1/00).

[0003] The implementation sequence of the known method is as follows.

[0004] Nickel of the natural isotopic composition in the form of nickel tetrafluorophosphine - Ni $(PF_3)_4$ is sent for isotopic enrichment. The enrichment process is implemented on Nickel-62, while the content of Nickel-64 may be more than 2%. Nickel-62 enriched tetrafluorophosphine nickel is converted to metal and sent to a reactor for bombardment. After 2 years of bombardment, Nickel-63 is accumulated in the target due to neutrons captured by atoms of Nickel-62. At the same time, the Nickel-64 content in the target being bombarded decreases to a greater extent due to its burning out in the reactor upon being exposed and the formation of a short-living isotopes Nickel-65 with a half-life of 2,57 hours, rather than increases due to the burning out of Nickel-63 formed during bombardment of Nickel-63 with neutrons. Exposed nickel metal is being dissolved and cleaned from decomposition products of Nickel-65, especially from Cu-65, by using chemical methods, for example, by sorption, and then transferred to nickel tetrafluorophosphine and sent for enrichment. The exposed product is being bombarded until the content of Nickel-63 in the enriched product reaches the level of 75% or more, which ensures the preservation of the characteristics of beta-voltaic current sources during their established service life.

[0005] Nickel-63 radionuclide can be produced on an industrial scale by bombarding a nickel target installed in a cell of an industrial reactor instead of a fuel assembly (FA). For example, nickel with a mass comparable to that of a fuel assembly of about 200 kg can be placed in a RBMK-1000 cell.

[0006] Cost analysis of commercial production of Nickel-63 radionuclide showed that the cost of bombardment of a target enriched in Nickel-62 in an industrial power reactor for 2 years (in RBMK-1000, for example), would be almost half the cost of producing Nickel-63 radionuclide to compensate for the reduction in the electrical energy generation in the occupied reactor cell. Therefore, the profitable production of the Nickel-63 radionuclide may be ensured when only the reactor cell is loaded with the Nickel-62 isotope as fully as possible.

[0007] The disadvantages of the known method include the fact that when loading a nickel target with a Nickel-64 content of more than 2% into the reactor for bombardment, the cell volume of the reactor is not used efficiently, as a result of which the amount of Nickel-63 radionuclide is reduced.

[0008] A method for producing Nickel-63 radionuclide is known, which includes producing a nickel target enriched with Nickel-62 with a Nickel-64 content not exceeding 2%, bombarding the target in the reactor and subsequent enrichment of the exposed Nickel-63 product, in which the Nickel-64 isotope is extracted from the exposed product (see RF Patent No.2 313 149, G 21 G1/06).

[0009] The implementation sequence of the known method is as follows.

[0010] Nickel of the natural isotopic composition in the form of nickel tetrafluorophosphine - Ni $(PF_3)_4$ is sent for isotopic enrichment. The enrichment process is implemented in such a way that the Nickel-64 content is not more than 2%. This restriction allows for enrichment of nickel up to 50% or more, but the main isotopic impurity is lighter than Nickel-62 isotopes. Nickel-62 enriched tetrafluorophosphine nickel is converted to metal and sent to a reactor for bombardment. In the known method, nickel of an average enrichment of 50-80% is recommended to be use for bombardment rather than highly enriched nickel. After 2 years of bombardment, 6.4% of Nickel-63 is accumulated, and the content of Nickel-64 increases to 1.5% due to burnout of Nickel-63. The exposed metallic nickel is transferred to nickel tetrafluorophosphine and sent for enrichment. Nickel-63 radionuclide is enriched to produce a heavy fraction, and at the same time Nickel-64 is extracted from the exposed material. The enriched heavy fraction of nickel tetraphosphine is converted into metallic form and is used in beta-voltaic current sources, for example. The remaining light fraction contains Nickel-60, 61, and 62, residues of Nickel-63, and no Nickel-64 virtually. This product may be re-directed to the reactor for bombardment.

[0011] The known method was chosen by the applicant as a prototype.

[0012] The disadvantages of this method include the fact that during primary loading of nickel of medium enrichment into the reactor for bombardment and secondary loading of the remaining light fraction containing more than 20% of Nickel - 60 and Nickel 61, the cell volume of the reactor is not used efficiently, resulting in a reduction in the amount of Nickel-63 radioisotope produced and, hence, its production becomes unprofitable.

[0013] Besides, the extraction of Nickel-64 from the exposed material leads to additional costs and losses of the Nickel-63 radionuclide, a part of which will be distilled off along with the extracted Nickel-64.

[0014] This invention is designed to ensure the possibility of large-scale profitable production of Nickel-63 radionuclide, especially for the production of beta-voltaic current sources.

[0015] The technical result consists in the most complete loading of the reactor cell with Nickel-62 isotope and an increase in the amount of Nickel-63 radionuclide produced.

[0016] To obtain this technical result, the known method of production of Nickel-63 radionuclide, which includes the production of a nickel target made of initial nickel and enriched in Nickel-62, the bombardment of the target in the reactor, the enrichment of the exposed product and the return of the light fraction back to the reactor for bombardment, uses initial nickel enriched in Nickel-62 until it reaches a content of 98% or more to make a primary nickel target. The exposed product is enriched to get a light Nickel-62 fraction and achieve a Nickel-62 content of 98% or more. The initial nickel enriched in Nickel-62 is added to the light nickel fraction until it reaches a content of 98% or more, and is used to make a secondary nickel target.

[0017] The remaining heavy fraction is converted into a metal form and used for the manufacture of beta-radiation sources, in particular for beta-voltaic current sources.

[0018] In a particular case of applying the method, the solution of the exposed nickel target is subjected to radiochemical cleaning to remove Cu-65 and gamma-active isotopes such as Fe-59, Co-60 and other radioactive impurities.

[0019] The use of the initial nickel target enriched in Nickel-62 to the content of 98% or more for the production of the primary nickel target makes it possible to load the reactor cell with the Nickel-62 isotope as fully as possible and to obtain, after bombardment, the maximum possible content of Nickel-63 radionuclide in the exposed target. Also, the use of the initial nickel enriched to such an extent allows to obtain the content of Nickel-63 radionuclide of more than 75% in the heavy fraction remained when enriching the exposed product into the light fraction subsequently.

[0020] The enrichment of the exposed product into the light fraction of Nickel-62 until it reaches a content of 98% or more allows the enrichment process to be implemented at a smaller number of separation production stages as compared to the enrichment into the heavy fraction due to the high Nickel-62 content in the exposed product and, thus, to reduce enrichment costs.

[0021] Also, when enriching the exposed product, 92-93% of the exposed product is removed into the light fraction and contains mostly Nickel-62, a partially unseparated Nickel-63 radionuclide, while Nickel-64 being practically absent.

[0022] The remaining heavy fraction is 7-8% of the exposed product and contains the radionuclide Nickel-63, Nickel-64 and partially not separated Nickel-62, and the content of the radionuclide Nickel-63 in it is more than 75%, which allows to use the heavy fraction for the manufacture of the beta radiation source without extracting Nickel-64.

[0023] Supplementing Nickel-62 contained in the light fraction with the initial nickel enriched in Nickel-62 to its content of 98% or more, and its further use to make a secondary nickel target allows to increase the amount of the Nickel-62 isotope and make a secondary target and, thus, to ensure the fullest loading of the reactor cell with the Nickel-62 isotope and, after bombardment, to obtain the maximum possible content of Nickel-63 radionuclide in the exposed target.

[0024] Conversion of the remaining heavy fraction into a metallic form and its use for the manufacture of beta-radiation sources, especially, for beta-voltaic current sources, eliminates the operation of extracting Nickel-64 from the heavy fraction, allows to reduce the loss of Nickel-63 radionuclide removed when Nickel-64 is extracted and thereby increase the amount of Nickel-63 radionuclide produced.

In the case of manufacturing a secondary target with the restoration of the initial volume of the Nickel-62 isotope, 7-8% of the initial nickel enriched in Nickel-62 isotope with its content over 98% shall be added to Nickel-62 remaining in the light fraction. In this case, the Nickel-64 content in the secondary exposed nickel target will be determined by only its content in the 7-8% of the enriched initial nickel that were added and the Nickel-63 burnout during the bombardment process.

After the exposed product is enriched, the content of Nickel-63 radionuclide in the remaining heavy fraction will be more than 75%.

[0025] In a case of manufacturing a secondary target with an excess of the initial volume of Nickel-62 isotope due to adding of more than 8% of the initial nickel with enriched Nickel-62 content of 98% or more to the Nickel-62 remaining in the light fraction, the Nickel-63 content in the remaining heavy fraction will be in the range of 75-85%, depending on the volume of added initial nickel enriched in Nickel-62.

[0026] Radiochemical cleaning of the nickel target to remove Cu-65 and gamma-active isotopes of Fe-59 and Co-60 after bombardment improves radiation safety for further operations with the exposed product and reduces the amount of impurities in the final product

[0027] The proposed method will be implemented in the following order.

[0028] The metallic nickel that was highly enriched in Nickel-62 isotope using the known method, the Nickel-62 content in which is 98% or more, is loaded in the reactor for bombardment. During the bombardment, Nickel-62 is accumulated in the target due to neutrons captured by atoms of Nickel-62. After a 2-year-long exposure, 6.4% of the Nickel-63 radionuclide is accumulated, and the Nickel-64 content increases slightly due to the Nickel-63 radionuclide burnout in the process of bombardment.

[0029] The exposed metallic nickel is dissolved and subjected to radiochemical cleaning to remove Cu-65 and gamma-active isotopes, in particular Fe-59, Co-60, and others. As soon as settled from the solution, the exposed nickel is converted to nickel tetrafluorophosphine and sent for enrichment.

[0030] The exposed product is enriched in Nickel-62 into the light fraction until its content reaches 98%, while 92-93% of the exposed product are removed into the light fraction, and 7-8% of the exposed product remain as a heavy fraction. The extracted light fraction contains Nickel-62, light nickel isotopes and a partially non-separated Nickel-63 radionuclide and almost no Nickel-64.

Then, the heavy fraction's nickel tetrafluorophosphine is converted to a metallic form and used as a beta-radiation source, in particular, in beta-voltaic current sources, because the content of Nickel-63 radionuclide in the heavy fraction is more than 75%, which allows using it for the manufacture of a beta source-radiation without extracting Nickel-64.

[0031] The light fraction's nickel tetrafluorophosphine is also converted into a metallic form, the initial nickel enriched in Nickel-62 isotope up to its content of 98% or more is added, and this is used to make secondary and subsequent nickel targets.

[0032] In the secondary and subsequent nickel targets, the content of Nickel-64 isotope will decrease to the level of its content in the added enriched nickel.

[0033] The company began the pilot project to produce Nickel-63 radionuclide using the proposed method. At present, a highly enriched target with a nickel isotope content indicated in line 2 of the table is exposed in the research reactor of the Institute of Reactor Materials JSC for bombardment. The table shows the actual and calculated data of the implementation of the proposed method for the case of the initial nickel that is highly enriched in Nickel-62 up to its content of 99.36%.

Table

	Product	Nickel isotopes,%					
		58	60	61	62	63	64
1	the isotopic composition of natural nickel,%	67.27	27.10	1.13	3.59	-	0.91
2	the isotopic composition of highly enriched nickel loaded for bombardment, %	0.01	0.44	0.00	99.36	-	0.18
3	the isotopic composition after being bombarded %	0.00	0.40	0.04	92.86	6.40	0.30
4	the isotopic composition of nickel in the light fraction enriched with nickel-62, %	0.00	0.40	0.04	99.36	0.19	0.01
5	the isotopic composition of nickel in the heavy fraction	0.00	0.42	0.04	8.76	86.50	4.28

[0034] Line 3 of the table shows the calculated data of JSC "Institute of Reactor Materials" on the isotopic composition of the nickel target after its bombardment. After a 2-year-long exposure, 6.4% of the Nickel-63 radionuclide is accumulated in the target, and the Nickel-64 content increases due to the Nickel-63 radionuclide burnout in the process of bombardment. The exposed product's enrichment in Nickel-62 into the light fraction to achieve the content of 99.36% will reduce the content of Nickel-62 in the remaining heavy fraction and lead to an increase in the content of the Nickel-63 radionuclide in it to 86.5%.

[0035] The isotopic compositions of the light fraction and the remaining heavy fraction are given in lines 4 and 5 of the table.

Claims

1. A method of producing Nickel-63 radionuclide including the production of an initial nickel target enriched in Nickel-62, the bombardment of the target in the reactor, the enrichment of the exposed product and the return of the light fraction back to the reactor for bombardment, **characterized in that** the initial nickel enriched in Nickel-62 to get the content as high as 98% or more is used to produce the primary nickel target, the exposed product is enriched into a Nickel-62 light fraction until the content of 98% or more is reached; nickel enriched in Nickel-62 is added to the light fraction's nickel until the content of 98% or more is reached, and then it is used to make a secondary nickel target, and the remaining heavy fraction is converted to a metallic form and used for the manufacture of beta-radiation sources for beta-voltaic current sources.

2. Method as per clause 1 **characterized in that** the solution of the exposed nickel target is subjected to radiochemical cleaning to remove Cu-65 and gamma-active isotopes, in particular Fe-59 and Cobalt-60.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/RU 2018/000258

A. CLASSIFICATION OF SUBJECT MATTER
G21G 1/06 (2006.01)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
G21G 1/06, 1/00, 4/00, B01D 59/00, 59/20

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
PatSearch (RUPTO internal), Esp@cenet, DWPI, PAJ, USPTO, CIPO, PubMed

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	RU 2313149 C1 (FEDERALNOE GOSUDARSTVENNOE UCHREZHDENIE ROSSYSKY NAUCHNY TSENTR "KURCHATOVSKY INSTITUT") 20.12.2007)	1-2
A	RU 2569543 C (FEDERALNOE GOSUDARSTVENNOE UNITARNOE PREDPRIYATIE "GORNO-KHIMICHESKY KOMBINAT" et al.) 27.11.2015	1-2
A	RU 2344084 C1 (FEDERALNOE GOSUDARSTVENNOE UNITARNOE PREDPRIYATIE "GOSUDARSTVENNY NAUCHNY TSENTR ROSSYSKOI FEDERATSY - NAUCHNO-ISSLEDOVATEL-SKY INSTITUT ATOMNYKH REAKTOROV") 20.01.2009	1-2
A	US 2013/0170593 A1 (THE SOUTH AFRICAN NUCLEAR ENERGY CORPORATION LTD.) 04.07.2013	1-2

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
13 July 2018 (13.07.2018)	13 September 2018 (13.09.2018)
Name and mailing address of the ISA/ RU	Authorized officer
Facsimile No.	Telephone No.

Form PCT/ISA/210 (second sheet) (April 2005)

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- WO 2569543 A **[0002]**
- WO 2313149 A **[0008]**