NEW HEAD PROCESS FOR NON-HEU ^{99}Mo PRODUCTION BASED ON THE OXIDATION OF IRRADIATED UO$_2$-PELLETS FORMING SOLUBLE U$_3$O$_8$

by

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All fission-based ^{99}Mo producers worldwide are required to convert their ^{99}Mo production processes from using highly enriched uranium to low-enriched uranium. At a recent IAEA meeting in Vienna, problems related to bottlenecks and target modification and optimization of low-enriched uranium-based ^{99}Mo production processes were discussed. Ceramic UO$_2$-pellets (as used in fuel) were excluded from the discussion with the argument that this material cannot be dissolved under practically applicable conditions. In this paper, we suggest transforming the non-soluble ceramic UO$_2$ fuel-pellets into the U$_3$O$_8$ form by simple oxidation and the use of the soluble U-oxide modification as the starting material for the ^{99}Mo production processes. Due to the absence of Al, larger target quantities could be processed and the waste volume could still be kept small. The approach is known and proven in nuclear technology. In principle, this new head process can be connected to any of the presently used ^{99}Mo production procedures.

Key words: low-enriched uranium, higly enriched uranium, UO$_2$, U$_3$O$_8$, ^{99}Mo, voloxidation

INTRODUCTION

Worldwide progress in nuclear medicine is historically based on the use of ^{99}Mo/^{99m}Tc generators, where the ^{99}Mo is produced from fission of ^{235}U. The overwhelming fraction of ^{99}Mo is, hereby, still produced using highly enriched U (enrichment in ^{235}U up to 93 %). Recently, after it became clear that low-enriched uranium (LEU) could be used for industrial scale production of ^{99}Mo [1], as well as for reasons of non-proliferation of nuclear materials, it became widely accepted that the production technology using highly enriched uranium (HEU) shall be converted to LEU-targets. A comprehensive overview of all activities related to the non-HEU production of ^{99}Mo is given in [2].

Due to the short irradiation time, the burn-up in targets for ^{99}Mo production is only about 1 to few %, which is significantly less (by a factor of 10 to 20), as compared to the burn-up in reactor fuel. Nonetheless, targets used in ^{99}Mo production are manufactured according to fuel specifications, with the consequence that these targets are still qualified as high burn-up. This concerns all types of dispersion targets such as U-Al-alloy, silicide and nitride (see, for example, [3-6]), whereas most of ^{99}Mo produced worldwide is still generated from U-Al-alloy dispersion targets cladded with Al and the use of highly enriched uranium (HEU, enrichment in ^{235}U up to 93 %). The main characteristics of those HEU-targets are: uranium-density in the meat: 1.0-1.2 g/cm^3, thickness of the Al-cladding: ~0.3 mm [2].

The conversion of HEU to LEU for industrial scale ^{99}Mo production is not trivial, since the irradiated LEU-targets contain about 2.5 times less ^{99}Mo as compared to HEU-targets, assuming the same chemical target composition considering an increased meat density of 2.6 g U/cm^3. This means that one would need to process by a factor of 2.5 more target plates to produce the same ^{99}Mo activity as done before with HEU. Consequently, considerable additional investment is required for installing additional processing units. Also, additional irradiation capacity and corresponding R&D needs would be needed to assure routine supply of ^{99}Mo to the nuclear medicine environ-

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ment without any interruptions. All of the said issues were the motivation for several IAEA meetings in Vienna (see [3]).

Intense R&D is going on aimed at increasing the ^{235}U-content (as LEU) in the target plates by enhancing meat thickness or U-density in Al-clad dispersion targets, as practiced in the development of high-density atomized U-Al$_x$-dispersion targets [7], or by using U$_3$Si$_2$-targets according to Sameh [8]. In all of these cases, the Al-content determines the technical limitations of the production technology. Al-containing targets are usually digested in alkaline solvents. Due to the relatively poor solubility of Na-aluminates (2.1 mol/dm^3), the process volumes and, consequently, waste volumes become large.

There are several versions of Al-free ^{99}Mo production processes based on fission. The use of metallic U-pellets as targets has been demonstrated [9]. It has been shown that the modified CINTICHEM process based on metallic U-foil targets (density ~19 g/cm^3) works [10, 11]. Other Al-free targets are U-oxides: UO$_2$ and U$_3$O$_8$. UO$_2$-pellets have been used in Australia over the years (1982-2006) as target material (enrichment in ^{235}U was 1.7 %, and from the mid 1990s, 2.2 %) [12].The pellets were sintered only at relatively low temperatures, providing sufficient mechanical stability and keeping the solubility properties in HNO$_3$. A special annular UO$_2$-powder target has been developed by B & W for Canada, but it has never been used [2]. Kotschkov *et al*. (Obninsk, Russia) are using a similar target approach. They employ HEU-U$_3$O$_8$ „diluted" with ZnO as target material because of better solubility in HNO$_3$ compared to UO$_2$ [13].

So far, ceramic-like UO$_2$ fuel pellets that are sintered at high temperature (~1600 °C) have not been considered as target material in ^{99}Mo production. The said ceramic material is practically not attacked by anything, neither conc. alkaline nor conc. HNO$_3$ [3]. In the present paper, we suggest using a simple oxidation step for the transformation of ceramic UO$_2$-fuel pellets into soluble U$_3$O$_8$-powder. In fuel reprocessing technology, this process is known as "voloxidation" [14, 15].

OXIDATION OF CERAMIC-LIKE INSOLUBLE UO$_2$ TARGET PELLETS TO SOLUBLE U$_3$O$_8$ POWDER

Ceramic UO$_2$-pellets, 5 % enriched in ^{235}U, are the most widely distributed fuel type in nuclear energetics, thus the corresponding target technology is relatively easy to qualify. Reprocessing of spent light water fuel in form of UO$_2$ starts with a simple oxidation step known as voloxidation (derived from volume oxidation). There are two reasons for applying this process: first, separating the tritium and certain volatile fission products (^{129}I, ^{85}Kr for instance) from the spent fuel and, second, obtaining a fine U$_3$O$_8$-powder that can be relatively easily dissolved in nitric acid for further processing. The oxidation reaction is as follows eq. (1)

$$3\,UO_2 \quad O_2 \quad U_3O_8 \qquad (1)$$

The reaction enthalpy is ΔH° = –98.75 kJ/250 g UO$_2$ meaning that the reaction is strongly exothermic and quantitative. For 250 g UO$_2$-material, we theoretically need 34.7 dm^3 (STP) air resulting in 259.9 g U$_3$O$_8$. The voloxidation process usually takes place at 450-600 °C, [14-16]). According to Cadieux and Stone [14], under the given conditions (490 °C, air as reaction gas) ^{85}Kr is practically quantitatively released. This finding is somehow in conflict with data reported by Jubin [15], which states that only 1 % of the ^{129}I and 5 % of the ^{85}Kr are released. At higher temperatures, these volatile fission products are released more efficiently, which was the motivation for developing an „Advanced voloxidation" process which works at temperatures around and above 1200 °C [17-19]. At a temperature this high, the release of volatile fission products is quantitative [19].

As said before, via the voloxidation of UO$_2$, one obtains a fine black powder of U$_3$O$_8$ whose crystalloid structure has been studied by Park *et al*. [20]. All above stated facts concern UO$_2$-fuel after long exploitation in power reactors and a cooling down period of >5 years. From shortly irradiated UO$_2$-fuel used as targets in ^{99}Mo production we expect a similar behavior concerning the formation of soluble U$_3$O$_8$ and similar behavior in release properties of fission gases.

TEST EXPERIMENTS

Test experiments have been performed in the former CINR, Department of Nuclear Fuel Research. The said experiments demonstrated the feasibility of using the standard VOLOXIDATION approach for transforming ceramic-like UO$_2$-fuel pellets into U$_3$O$_8$ powder which could easily be dissolved in nitric acid. The obtained solution would than be suitable for ^{99}Mo separation. Figure 1 illustrates the obtained results: on the left, the original pressed UO$_2$-pellet which was an-

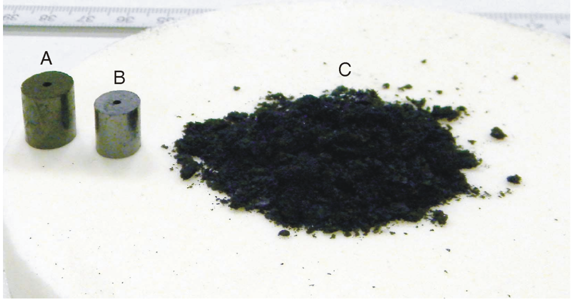

Figure 1. UO$_2$-pellets before and after a heat treatment similar to the standard voloxidation process; (A): standard fuel pellet (dummy) from nat-UO$_2$, annealed at 600 °C; (B): fuel pellet (A) made from nat-UO$_2$ after annealing at 1600 °C; (C): U$_3$O$_8$-powder obtained after heating of the fuel pellet (B) at 500 °C in oxygen-containing atmosphere (in this case, ordinary air)

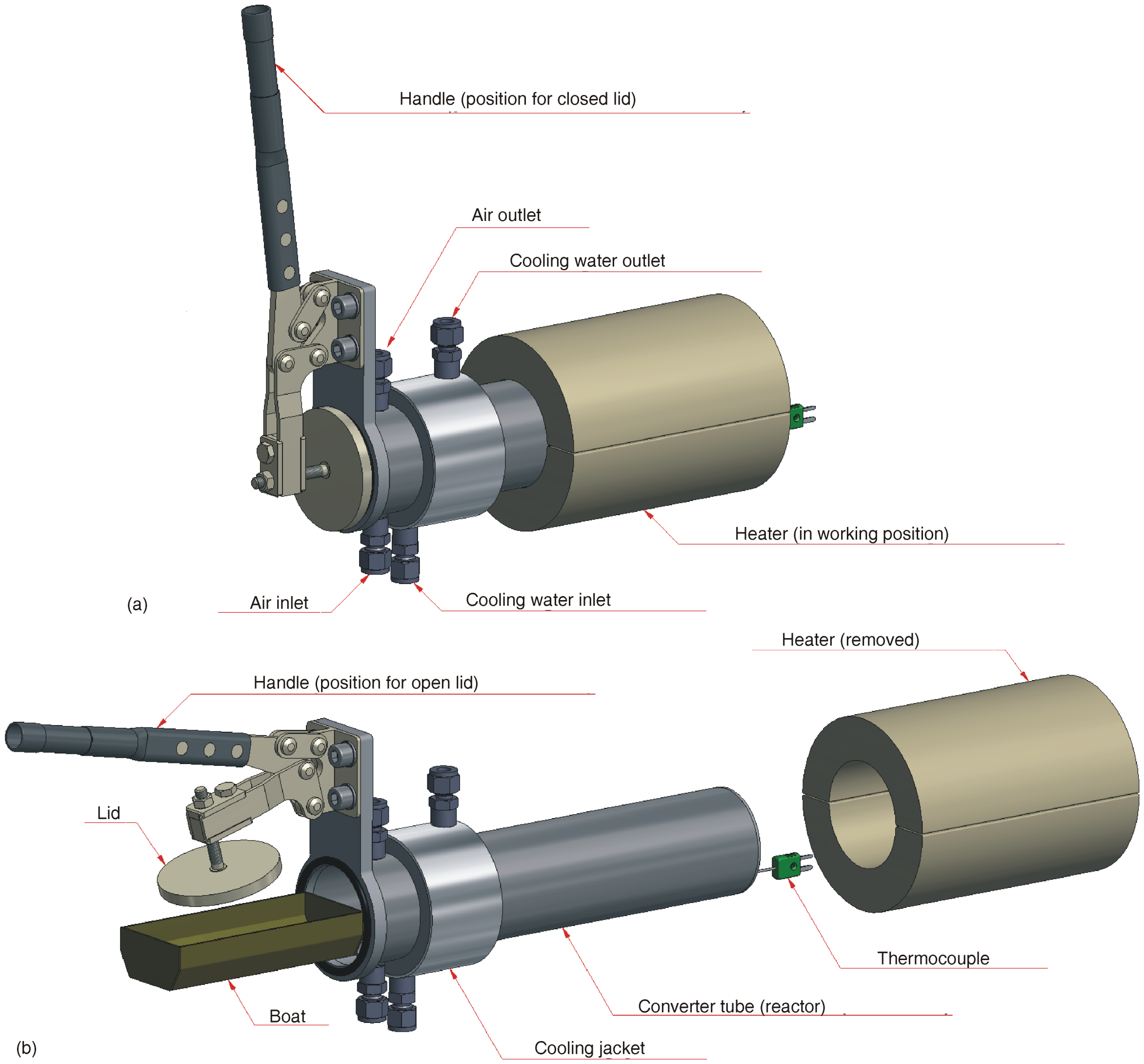

Figure 2. Special furnace for conversion of UO_2-target pellets into U_3O_8 consisting of a special removable furnace, flange for opening with handle, gas inlet, gas outlet, and thermocouple; (a) converter in working position, (b) loading/unloading of the boat with UO_2/U_3O_8 with the heater removed

nealed at 600 °C. These pellets were thereafter annealed for one hour at 1600 °C in inert gas (Ar); the result is shown in fig. 1 (middle). In this annealing process the volume of the pellet becomes reduced by about 30 %. Right side of fig. 1: the black U_3O_8 powder obtained after the voloxidation process. One of the ceramic-like UO_2-pellets was placed on a porcelain plate and just heated for 1 hour at 500 °C in an ordinary furnace at normal atmospheric pressure. The increased volume of the U_3O_8 is clearly seen. The conversion reaction was quantitative, with no residue of UO_2-species detected.

Under practical conditions (max. temperature 600 °C) molybdenum remains quantitatively with the U_3O_8, as will be outlined further down.

For this purpose, Spencer [16] suggested a special rotary cylindrical Voloxidizer which is operated horizontally in the process and vertically during loading and unloading procedures. In fig. 2, we illustrate a special furnace that may be used for the conversion of around 250 g UO_2-target pellets to U_3O_8. The furnace is operated horizontally only while loading and unloading. The unit consists of a cylindrical reactor (made from INCONEL) and a special cylindrical furnace, consisting of two half-segments that can be opened. Alternatively, a tube furnace could be used that can be moved horizontally over the reaction tube. The system is equipped with a gas inlet (with valve), gas outlet (with valve) connected to the off-gas treatment line and a thermocouple for temperature monitoring. For the online determination of oxygen content, a solid-phase oxygen monitoring sensor is installed in the off-gas line further away from the furnace.

The target material will be introduced into a boat that is moved into the middle of the reactor with a special magnetic tool (operated by the manipulator); the same goes for the removal of the boat loaded with the reaction product.

A small blower would help cool the reactor tube down to near room temperature. Only thereafter, the reactor is opened and the boat with the obtained U_3O_8 removed.

The oxidation procedure (potential operation algorithm)

- The irradiated pellets (with or without cladding) are transferred into the converter.
- The converter is tightened and the gas treatment line connected.
- The furnace is switched on to reach a temperature of 500 + 50 °C.
- Ordinary air could be used as reaction gas. An air stream of about 1.5 dm^3/min is adjusted and the oxygen content in the reactor gas volume is monitored. The off-gas passes through a corresponding gas treatment line and is finally collected in an adequate gas storage tank.
- The temperature is kept and controlled at ~600 °C.
- The partial pressure of oxygen in the converter is monitored. When the oxygen content is back to nearly the initial value, the system remains heated for a few minutes more to complete the reaction. The reaction time is expected to be around 1.0-1.5 hour for 250 g pellets.
- Then, the heating is switched off and the heating jacket removed from the converter.
- The converter is cooled down to about room temperature within ~20 min.
- After the temperature is down, the converter can be opened and the U_3O_8 transferred into a dedicated dissolver vessel.
- The converter is, in principle, ready for the next cycle.

The cladding material, usually Al or Zircalloy or Zr-Nb [15], does not disturb neither the voloxidation process nor the dissolving process thereafter.

The dissolving procedure

The obtained U_3O_8-powder could be easily dissolved in HNO_3 according to the following reaction eq. (2)

$$U_3O_8 \quad 8\,HNO_3 \quad 3\,UO_2(NO_3)_2 \quad 2\,NO_2 \quad 4\,H_2O \tag{2}$$

This process is routinely used in Obninsk (Russia), starting from irradiated U_3O_8 – powder [13]. Since one needs to consider the presence of radio-iodine and radio-Xe (mainly from the ^{133}I decay), the off-gas from the dissolving process needs to undergo a gas treatment. The system may consist of following components: reflux condenser, two successive alkaline traps for scrubbing the NO_x gases followed by a gas collection tank.

The amount of HNO_3 used for the dissolving process may be determined from the reaction equation in such a way that the excess of free acid after complete dissolution is of the order of ~0.5 M. In case off any excess, HNO_3 can be destroyed by adding formaldehyde which transforms NO_3^- under acidic conditions to N_2 according to the following eq. (3)

$$4\,HNO_3 \quad 5\,H_2CO \quad N_2 \quad 5\,CO_2 \quad 7\,H_2O \tag{3}$$

The obtained uranium fission product solution may now be used for any of the existing ^{99}Mo-separation processes. Generally, due to the Al-free process, the total waste volume becomes drastically reduced compared to the original ROMOL-99® [21] or the Sameh KSA process [22], both employing UAl_x-Al-clad dispersion targets.

Example 1 – adaptation to the ROMOL® (wet process)

The obtained uranium fission product solution is directly used for ^{99}Mo separation using the ROMOL®-process technology following these steps.

- Feeding the weak acid U-solution through an Al_2O_3-column, (alternatively TiO_2-column). Under these conditions, the $[^{99}MoO_4]^{2-}$ is retained at the column, while the uranium and nearly all of the fission products are washed out (standard practice, see for example [9, 12]).
- Washing the column with 0.5-1 M HNO_3 and, thereafter, with water and 0.01 M NH_4OH solution.
- The ^{99}Mo is then stripped off with 1-2 M NH_3 solution.
- This solution is fed directly through a DOWEX-1 column, as in the original ROMOL® process [21].
- Evaporation and sublimation.

Example 2 – adaption to the KSA (wet process)

In this case, the ^{99}Mo is separated after precipitating the U as $Na_2U_2O_7$ from the basic solution.

- The primarily obtained acidic target solution is neutralized and made alkaline with NaOH (or KOH) and U precipitates as $Na_2U_2O_7$. The obtained $Na_2U_2O_7$, which simultaneously collects the bulk of unwanted fission products, is separated by filtration.
- The obtained basic solution is fed through a DOWEX-1 column and further separation and purification of the ^{99}Mo can then be done according to the original KSA-process developed by Sameh [22].

Example 3 – dry process

As said previous, the Mo remains with the U_3O_8 when temperatures of max. 600 ºC are applied. Eichler

Table 1. Volatilization of MoO_3 from UO_2 matrix at elevated temperature [26]

T [ºC]	710	840	880	950	1040	1140
% of volatile MoO_3	1.4	2.0	5.2	7.2	8.5	22.5

et al. [23] studied the volatilization of Mo from UO_2 in an airstream and found the relationship presented in tab. 1.

It is clear from these findings that for separating Mo from the irradiated fuel pellets by "dry" distillation one would need to apply temperatures well above 1200 °C. In order to volatilize Mo-species at lower temperatures, one needs to add another component into the reaction gas. Recently, Brown [24] described a new "Method and apparatus for selective gaseous extraction of ^{99}Mo and other fission product radioisotopes" from irradiated U_3O_8-targets at the ^{99}Mo-Topical Meeting held in Boston [25]. In this patent, General Atomics (GA) suggest using a porous U_3O_8-target positioned inside a reactor and the release of gaseous species of Mo through a long, heated gas transfer line to a ^{99}Mo condenser column. It is assumed to operate the process either on-line or off-line. Indeed, Mo forms a relatively volatile compound MoO_2Cl_2, which sublimes at 184 °C, practically without melting. Thus, it seems feasible to transport ^{99}Mo in the gas phase through slightly heated transfer tubes. Nevertheless, there are strong concerns related to the long-term stability of the on-line target inside the irradiation position.

In this paper we suggest a third option, that of combining the voloxidation process with the GA approach for the gaseous extraction of ^{99}Mo during the exothermic process of converting the irradiated UO_2-targets to U_3O_8-powder. The process could look like the following.

- Start the voloxidation process as described above.
- Use a gas mixture consisting of the gas composition proposed by GA and add 20 vol. % O_2 for voloxidation. In this case, ^{99}Mo will form the volatile MoO_2Cl_2 which is trapped in the first columns (at room temperature) of the gas treatment line.
- Proceed with gas processing according to the GA approach without alterations.

DISCUSSIONS

Ceramic-like UO_2-pellets are suggested as target material for a new ^{99}Mo production processes [26]. Since this material is the most commonly used fuel for nuclear power reactors, one can expect its easy qualification as target material for ^{99}Mo production. The cladding material does not disturb the voloxidation process, meaning that the process can be carried out with or without it.

The productivity of the process is, to a great extent, based on the available irradiation conditions. Depending on the local conditions, one is able to adjust the enrichment of the ceramic fuel. It is known that in power reactors the thermal power of such fuel elements is up to 250 W/cm. There seems to be no technical hurdle for processing up to 1 kg of UO_2-target pellets for ^{99}Mo production; uranium concentration would still be at an acceptably low level for the alumina column process. Our own experiences have shown that one can separate ^{99}Mo from uranium solutions with a U-concentration of up to 2 M by use of an alumina column process. Depending on irradiation conditions, this approach seems to be equally suitable for industrial scale ^{99}Mo production, as well as for a very low production scale involving small research reactors. With a typical enrichment of 5 % in ^{235}U, one would have ~44 g of ^{235}U, which is ~1.7 times more than in a typical target batch used in a conventional fission ^{99}Mo production process. This means (assuming the same irradiation geometry in the reactor) that the average productivity per batch can be increased by a factor of ~1.7, as compared to the currently used HEU-based production processes, although very low-enriched target material is used in the proposed process.

Three approaches for separating ^{99}Mo from the obtained uranium fission product solution have been suggested above. The first two propositions are classical wet separation techniques. In the first example, ^{99}Mo is separated directly from the acidic uranium containing the target solution (as, for instance, in [9] and [12]). In the second version, uranium is first separated by precipitation as $Na_2U_2O_7$ and ^{99}Mo obtained thereafter from the alkaline U-free solution (as seen in [21, 22]). The potential third approach would be a dry technology, not yet applied in praxis.

The advantage of the first approach is that the process is straightforward and does not require an additional filtration process. The total volume of liquid waste is smaller compared to version two (alkaline media). The drawback is that we obtain a waste solution that contains both components: uranium, as well as the bulk of the fission products. Historically, it has been demonstrated that this simple alumina column process in presence of U, in combination with sublimation, can provide high purity ^{99}Mo preparations that fulfill the current demanding quality parameters perfectly well.

The second suggested process makes use of the co-precipitation of most of the metallic fission products with the $Na_2U_2O_7$ precipitate from an alkaline solution. After filtration, one obtains a solution that, besides ^{99}Mo, contains alkaline nuclides, some Ru and traces of contaminants. The separation of ^{99}Mo from this solution is easier and safer. Another advantage is that the slightly larger liquid waste volume is of significantly lower radiation danger, that critical fission products are co-precipitated with the U and that, as such, they are already configured in the solid waste (the precipitate). However, the volume of the $Na_2U_2O_7$

precipitate is significantly larger as compared to common large-scale ^{99}Mo production processes, due to larger U-content in the target material (the ceramic-like UO_2 pellets are only enriched up to 5 % in ^{235}U). It needs to be shown that the filtration process of the enlarged U-quantities does not create practical problems.

The proposed dry process makes use of the advantages of the voloxidation process for converting the irradiated, insoluble UO_2 target pellets, into soluble U_3O_8-powder. During this process, in addition to the gaseous fission products, ^{99}Mo can be evaporated by adding Cl_2-gas to the reaction gas for the voloxidation process, as shown by GA [25].

The described approach for making use of the voloxidation process by transforming the ceramic-like UO_2-fuel pellets into soluble U_3O_8 has been formulated as a patent application [26].

CONCLUSIONS

We have shown that ceramic-like UO_2-pellets (used as nuclear fuel) can be easily transformed by a simple oxidation process into the U_3O_8 form, a well soluble U-oxide modification. It suggests the creation of a new, Al-free, fission-based ^{99}Mo production process by using standard ceramic-like UO_2-fuel pellets, 5 % enriched in ^{235}U as target material. Due to the absence of Al, larger target quantities can be processed and the waste volume still kept small. The approach of converting the non-soluble ceramic-like UO_2 fuel pellets into a soluble U_3O_8 configuration has been proven in nuclear technology and known as voloxidation. This conversion process could be adapted to any of the presently used ^{99}Mo production procedures.

The proposed possibility of forming a gaseous ^{99}Mo-compound that is released during the voloxidation process when the oxygen-containing process gas is mixed with chlorine deserves, to our opinion, to be seriously considered.

AUTHORS' CONTRIBUTIONS

R. Muenze and G. J. Beyer have a lifetime experience in fission-produced ^{99}Mo separation and purification acquired at the Research Center Rossendorf; they developed the well-known ROMOL® process. The idea to combine the voloxidation process with existing ^{99}Mo separation and purification processes (particularly the ROMOL® process) was launched by G. J. Beyer who was the author of the draft for this paper. B. Eichler and T. Reetz are to be meritted for modifications to the voloxidation process suitable for ^{99}Mo production. The credit for the development of the high temperature furnace for the voloxidation process goes to J. J. Čomor. All authors participated in the preparation of the final version of the manuscript.

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НОВИ ПРИМАРНИ ПРОЦЕС ЗА ПРОИЗВОДЊУ ^{99}Mo НА ТЕМЕЉУ ОКСИДАЦИЈЕ ОЗРАЧЕНИХ ГРАНУЛА UO_2 ПРЕВОЂЕЊЕМ У РАСТВОРЉИВ ОКСИД U_3O_8, БЕЗ УПОТРЕБЕ ВИСОКО ОБОГАЋЕНОГ УРАНИЈУМА

Од произвођача широм света се очекује да промене своје процесе производње ^{99}Mo тако да уместо високо обогаћеног уранијума користе ниско обогаћени уранијум. Недавно је одржан састанак у МААЕ у Бечу на коме су разматрана ограничења и проблеми везани за модификацију мета и оптимизацију производње ^{99}Mo засноване на ниско обогаћеном уранијуму. Керамичке грануле UO_2 какве се користе у горивима нису узете у обзир уз аргумент да тај материјал не може бити растворен под практично примењивим условима. У овом раду предлажемо превођење нерастворних керамичких UO_2 гранула једноставном оксидацијом у U_3O_8 облик и употребу овог растворљивог уранијумоксида као почетног материјала за производњу ^{99}Mo. Због одсуства алуминијума, веће количине мета могу бити обрађене, при чему количина радиоактивног отпада остаје мала. Овај поступак је познат и доказан у нуклеарној технологији. Нови примарни процес може се надовезати на било коју процедуру производње ^{99}Mo која је тренутно у употреби.

Кључне речи: ниско обогаћени уранијум, високо обогаћени уранијум, UO_2, U_3O_8, ^{99}Mo, волоксидација