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Feasibility Study on Production of High-Purity Rhenium-185 by Nuclear Transmutation of Natural Tantalum

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Abstract: Rhenium-186 (Re-186) has attracted attention as a medical isotope. The feasibility of producing Re-185, the raw material for Re-186, using a fast reactor was evaluated using a continuous energy Monte Carlo code. The irradiation of natural tantalum (Ta) in the fast reactor can produce Re-185 with an isotopic purity of 99%. A two-step irradiation process with different moderators was found to improve the production rate of Re-185. Specifically, this can be achieved by using zirconium hydride ($ZrH_{1.7}$) as a moderator in the first transmutation process from natural Ta to tungsten (W), and then zirconium deuteride ($ZrD_{1.7}$) as a moderator in the second transmutation process from W to Re-185. Due to the two-step irradiation, the production rate of Re-185 from Ta can be increased up to a maximum of 470 times compared with irradiation without a moderator, and 2.3 g of Re-185 can be obtained from 1571 g of Ta in 1 year of irradiation. The proposed isotope production method is a new method that is different from the conventional electromagnetic enrichment process.

Keywords: rhenium-185; rhenium-186; tantalum; tungsten; transmutation; fast reactor; JOYO; Monte Carlo code; MVP version3/MVP Burn; medical isotope

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1. Introduction

Rhenium (Re) has excellent resistance to high temperatures; therefore, it is used in a variety of applications, as evidenced by the fact that it is used to make turbine blades for jet aircraft engines. On the other hand, Re is one of the least abundant elements in the Earth's crust, and it is not evenly distributed around the globe [1]. This means that it has a very high supply risk [2].

Re has attracted attention as a radioisotope for medical applications. Re-186 emits β-rays of maximum 1.08 (MeV) and γ-rays of 137 (keV) at 8.6% of decay with a short half-life of 89.3 h. This property allows for simultaneous treatment with β-rays and diagnosis with γ-rays [3,4]. Re-186 can be produced using nuclear reactors or particle accelerators [5]. The (n, γ) reaction of Re-185 in the nuclear reactor is 112 (barn) compared to ~0.6 (barn) for the (d, 2n) reaction of W-186 at ~15 (MeV) in the particle accelerator [6], which means that the production process in the nuclear reactor can produce large amounts of Re-186. On the other hand, the nuclear reactor process produces Re-186 with carriers, resulting in low specific activity [7]. This character is the main disadvantage compared to the particle accelerator process, which can produce carrier-free Re-186. However, this disadvantage may be overcome with techniques to isolate Re-186. Lapi et al. patented a method to isolate Re-186 by vaporizing a compound containing Re-185 and Re-186, ionizing it to form negatively charged molecules, and then using a positively charged collector to separate and recover the molecules by mass difference [8].

The nuclear reactor process requires the preparation of high-purity Re-185 (>94%), but Re-185 has a natural abundance of only 37.4%, as shown in Table 1, and it must be enriched. Electromagnetic isotope separation has long been used for isotope enrichment [9,10], and

isotope supply systems have been commercially established. However, due to the limited number of isotope suppliers [11], existing methods for the production of rhenium isotopes may not be able to meet the growing demand for radiotherapeutics in the future [12].

Table 1. Isotopic composition of natural Re.

Nuclide	Isotope Ratio	Half-Life
Re-185	37.4%	Stable
Re-187	62.6%	4.12×10^{10}y

In previous studies, the method of producing Re from tungsten (W) was discussed [13–15]. Re can be produced via the transmutation of W with a neutron capture reaction and subsequent β decays using a fast reactor. W is more abundant than Re in the Earth's crust, and its price is a few tenths of that of Re.

As shown in Figure 1, when natural W is used as the material for transmutation, Re-185 is produced mainly from W-184 with a natural abundance of 30.64%, and Re-187 is produced mainly from W-186 with a natural abundance of 28.43%.

$$^{184}\mathrm{W}(\mathrm{n},\,\gamma)^{185}\mathrm{W} \rightarrow {}^{185}\mathrm{Re} + \beta^{-}(75\mathrm{d})$$
$$^{186}\mathrm{W}(\mathrm{n},\,\gamma)^{187}\mathrm{W} \rightarrow {}^{187}\mathrm{Re} + \beta^{-}(24\mathrm{h})$$

Yokoyama et al. studied a method for producing Re with a higher ratio of Re-185 than the natural one by controlling the transmutation of W isotopes by moderating the energy of neutrons produced in a fast reactor with a moderator [16]. However, Re-185 with an isotopic purity of 65% is not pure enough as a raw material for the production of Re-186. We propose the use of tantalum (Ta) as a new raw material. Figure 2 shows the transmutation path of natural Ta to Re. Ta is similar to W in that it is a more abundant element in the earth than Re. The isotopic purity of Ta-181 in natural Ta is 99.9%. Each isotope of W is produced by the decay of higher-order Ta isotopes via successive neutron capture reactions of Ta-181. Since the half-life of the higher-order Ta isotope is short enough, the produced W will mainly contain W-182 or lower-order isotopes; thus, the amount of W-186 will be extremely small. As a result, the amount of Re-187 transmuted from W-186 in the produced Re is expected to be very small.

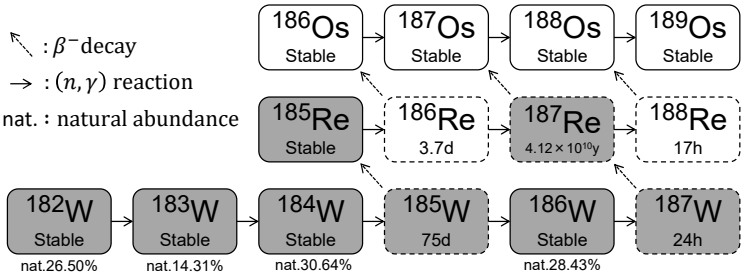

Figure 1. Transmutation path from natural W to Re. Through the neutron capture reaction with subsequent β decays, Re-185 is produced mainly from W-184. In contrast, Re-187 is produced from W-186.

Our objective is to investigate the feasibility of a method for producing high-purity Re-185 via the transmutation of natural Ta with a new isotope production process using a fast reactor. In a fast reactor, there are excess neutrons, and it is possible to generate electricity and produce plutonium, etc., at the same time. If this excess of neutrons can be used to produce Re-185, a new isotope production method can be realized. In addition, isotopes can be produced simultaneously with power generation without occupying a dedicated machine as in the conventional isotope production method, which may lead to a reduction in production costs. Furthermore, we propose a scheme from the transmutation of natural

Ta to the medical use of Re-186. Produced high-purity Re-185 is chemically separated from other elements, and Re-186 produced by the (n, γ) reaction in the nuclear reactor was used as a radiopharmaceutical. Since some solvent extraction or anion exchange methods, etc., are widely studied for their separation and purification, we will not discuss them in detail here.

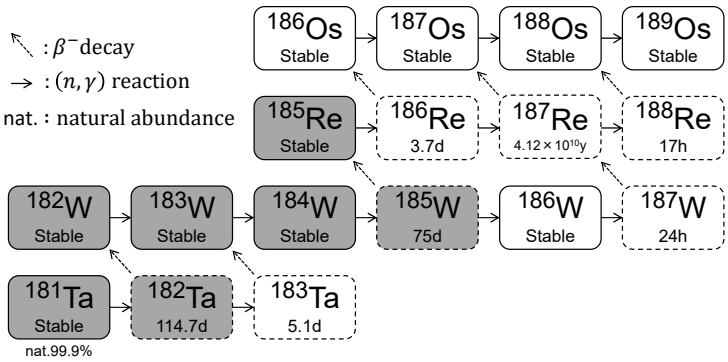

Figure 2. Transmutation path from natural Ta to Re. W is intermediately produced by the neutron capture reaction of Ta-181.

2. Analysis Methods

The feasibility of producing Re-185 via the transmutation of natural Ta was investigated using a 100 MWt fast reactor modeled on the experimental fast reactor JOYO with MK-III core. The energy spectrum of neutrons in the fast reactors is harder than that in light water reactors, and it may be possible to adjust the energy by moderator arrangement to achieve highly efficient transmutation, which has been used to study the transmutation of technetium-99 (Tc-99), a long-lived fission product [17].

The main reactor parameters are listed in Table 2. The system was sodium-cooled and uses plutonium-uranium mixed oxide (MOX) fuel. The irradiation target pin was placed in the center of the core, as shown in Figure 3. The cylindrical moderator can be placed around the sample. The candidate moderators were zirconium hydride ($ZrH_{1.7}$) and zirconium deuteride ($ZrD_{1.7}$); these were selected based on previous studies [13–16].

The production characteristics were evaluated using a three-dimensional continuous energy Monte Carlo code, MVP version 3 [18], with its burn-up calculation routine MVP-BURN [19]. To accurately simulate the production characteristics from Ta to Re, a transmutation chain model from Ta to osmium (Os), shown in Figure 4, was added to the burn-up chain model "u4cm6fp50bp16F", which is a standard set in MVP-BURN. The evaluated nuclear data library JENDL-5 [20] was used for the cross-section data. The number of histories and batches were set appropriately to keep the typical statistical error for k-effective below about 0.05% with a 1σ error.

Table 2. Main core parameters of the fast reactor in this study.

Specification		Data
Reactor Thermal Power	(MWt)	100
Number of driver fuel subassembly (Inner Driver Fuel and Outer Driver Fuel)		77
Equivalent core diameter	(cm)	80
Core height	(cm)	50
^{235}U enrichment	(wt%)	18
Pu content: Pu/(Pu+U)	(wt%)	16/21 *
Fissile Plutonium content: (^{239}Pu + ^{241}Pu)/(Pu+U)	(wt%)	12/15 *
Reflector/shielding		Steel Special Use Stainless (SUS)/B_4C

* Inner Core/Outer Core.

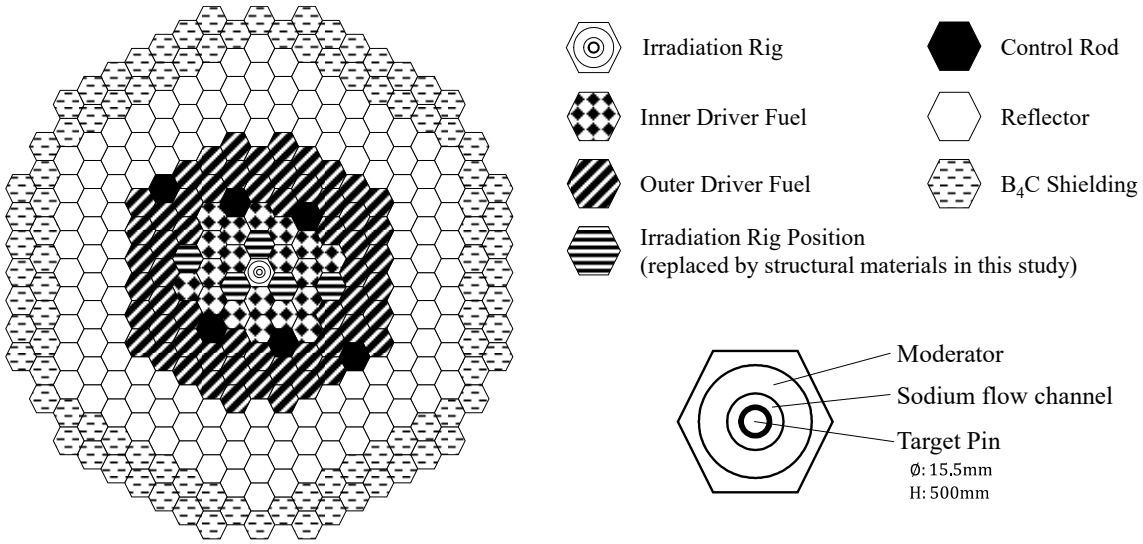

Figure 3. Irradiation target location. The irradiation target pin is placed in the center of the core. The power is set to 100 MWt.

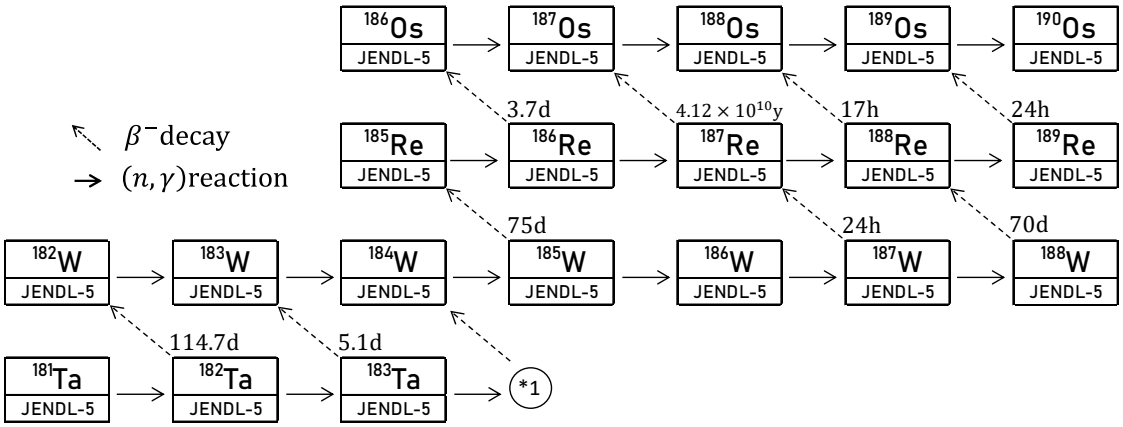

Figure 4. Transmutation chain model and cross-section library from Ta to Os.

3. Results and Discussion

3.1. Production of Re-185 via the Transmutation of Natural Ta Compared to Natural W

Table 3 shows the results of Re production via the transmutation of natural Ta and natural W after 1 year of irradiation without moderator and 30 days of cooling. The Re produced from natural Ta has Re-185 with an isotopic purity of 99.9%. On the other hand, the production of Re-185 from natural Ta is not large because it must pass through the production of W. Furthermore, Re-186 and Re-187 were also produced by the (n, γ) reaction of Re-185, albeit in very small amounts.

3.2. Improving the Production Rate of Re-185 from Natural Ta Using a Moderator

3.2.1. Comparison of Re-185 Production Using $ZrH_{1.7}$ or $ZrD_{1.7}$ as a Moderator

Table 4 shows the results of Re production from natural Ta when $ZrH_{1.7}$ or $ZrD_{1.7}$ is used as moderator, and Table 5 shows the energy-averaged one-group effective cross-section $\overline{\sigma}$ of the (n, γ) reaction for the main nuclides, which is defined as

$$\overline{\sigma} = \frac{\int \sigma(\mathrm{E})\phi(\mathrm{E})d\mathrm{E}}{\phi(\mathrm{E})d\mathrm{E}}$$

where $\sigma(E)$ is energy dependent microscopic cross section, and $\phi(E)$ is the energy-dependent neutron flux [21]. The neutron energy spectrum in the irradiation target pin is shown in Figure 5.

Table 3. Re production from natural Ta and natural W after 1 year of irradiation without a moderator and 30 days of cooling.

Nuclide	Irradiation to Natural Ta		Irradiation to Natural W	
	Initial (g)	after 1 Year of Irradiation and 30 Days of Cooling (g)	Initial (g)	after 1 Year of Irradiation and 30 Days of Cooling (g)
Ta-181	1571	1344	—	—
W-182	—	221.3	476.4	452.6
W-183	—	6.2	258.7	252.1
W-184	—	0.32	556.9	561.6
W-186	—	2.1×10^{-7}	522.3	505.1
Re-185	—	4.9×10^{-3} (99.96%)	—	22.0 (59.9%)
Re-186	—	8.3×10^{-8} (0.00%)	—	3.7×10^{-4} (0.00%)
Re-187	—	2.0×10^{-6} (0.04%)	—	14.8 (40.1%)
Os (total)	—	3.7×10^{-4}		6.6

Table 4. Re production when either $ZrH_{1.7}$ or $ZrD_{1.7}$ are used as a moderator after 1 year of irradiation and 30 days of cooling.

Nuclide	Initial (g)	After 1 Year of Irradiation and 30 Days of Cooling (g)		
		with $ZrH_{1.7}$ Moderator	with $ZrD_{1.7}$ Moderator	(without Moderator) *
Ta-181	1571	715.1	1170	1344
W-182	—	552.2 (64.9%)	381.5 (94.6%)	221.3 (97.1%)
W-183	—	202.7 (23.5%)	18.5 (4.5%)	6.2 (2.7%)
W-184	—	101.8 (11.6%)	3.5 (0.8%)	0.32 (0.1%)
W-186	—	1.0×10^{-3} (0.0%)	1.7×10^{-5} (0.0%)	2.1×10^{-7} (0.0%)
W (Total)	—	856.7 (100.0%)	403.5 (100.0%)	227.9 (100.0%)
Re-185	—	1.2 (80.4%)	0.11 (99.6%)	4.9×10^{-3} (99.96%)
Re-186	—	4.9×10^{-4} (0.03%)	5.5×10^{-6} (0.00%)	8.3×10^{-8} (0.00%)
Re-187	—	0.29 (19.5%)	5.0×10^{-4} (0.4%)	2.0×10^{-6} (0.04%)
Os (Total)	—	4.75	3.1×10^{-2}	3.7×10^{-4}

* From Table 3.

W (total) and Re-185 production increased with the use of the moderator. In particular, Re-185 production, in the case where the $ZrH_{1.7}$ moderator was used, increased by more than 200 times compared to the case without the use of the moderator. This is because the effective cross-sections of the major nuclides from Ta to Re tend to be larger in the low-neutron energy [22], and the softening of neutron energy by the moderator increases the (n, γ) reaction rate. On the other hand, in the case where the $ZrH_{1.7}$ moderator was

used, the production ratio of Re-185 was 80.4% of the total Re, which was smaller than in the other cases. This is mainly due to the larger effective cross-sections of W-185 and Re-185, which increased Re-187 production through the production paths shown below.

$$^{185}\mathrm{W}(\mathrm{n},\,\gamma)^{186}\mathrm{W}(\mathrm{n},\,\gamma)\,^{187}\mathrm{W}(\beta^-)\,^{187}\mathrm{Re}$$
$$^{185}\mathrm{Re}(\mathrm{n},\,\gamma)^{186}\mathrm{Re}(\mathrm{n},\,\gamma)\,^{187}\mathrm{Re}$$

Here, in addition to the above paths, there are also other paths in which Re-186 is produced by its own β decay to Os-186.

The $ZrD_{1.7}$ moderator case produced less Re-185 than the $ZrH_{1.7}$ moderator case but produced Re-185 with an isotopic purity of 99.6%. The effective cross-section of W-184, which directly affects the production of Re-185, was slightly smaller than that of the $ZrH_{1.7}$ moderator case, but there was no significant difference, while the effective cross-section of Re-185 was about 1/10 smaller than that of the $ZrH_{1.7}$ moderator case. This is advantageous in preventing the produced Re-185 from being transmuted to another nuclide. These results suggest that the $ZrH_{1.7}$ moderator is more efficient for the production of W from natural Ta and that the $ZrD_{1.7}$ moderator is more efficient for the production of Re-185 from W.

Table 5. One-group effective cross-section of main nuclides from Ta to Re with either $ZrH_{1.7}$, $ZrD_{1.7}$, or no moderator.

Nuclide	The One-Group Effective Cross-Section (Barn)		
	with $ZrH_{1.7}$ Moderator	with $ZrD_{1.7}$ Moderator	without Moderator
Ta-181	3.09	0.99	4.79
Ta-182	1285.17	3.34	0.73
Ta-183	19.71	1.89	0.37
W-182	1.68	0.28	0.16
W-183	2.31	1.27	0.38
W-184	0.40	0.36	0.18
W-185	4.80	1.32	0.36
W-186	11.21	0.89	0.13
W-187	8.04	0.87	0.17
W-188	0.66	0.06	0.03
Re-185	32.89	3.37	0.92
Re-186	33.13	4.00	1.04
Re-187	10.16	2.48	0.82

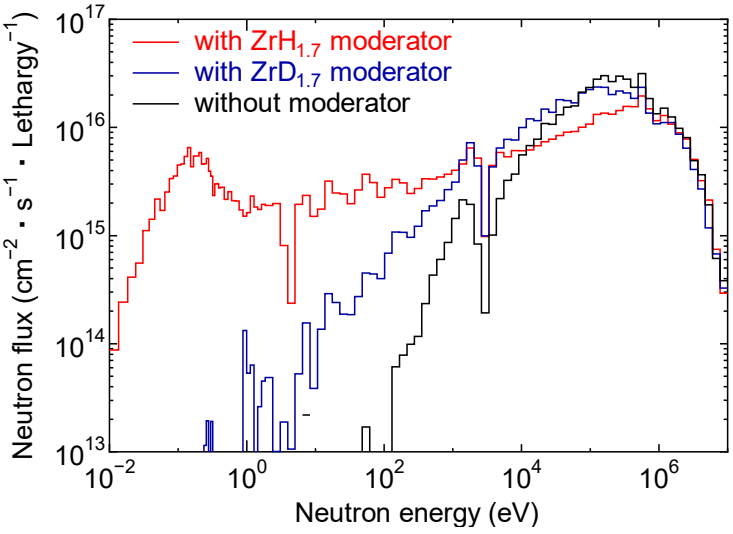

Figure 5. (Color online) Neutron energy spectrum in the irradiation target pin with $ZrH_{1.7}$, $ZrD_{1.7}$, or no moderator.

3.2.2. Improvement in the Re-185 Production Rate via Two-Step Irradiation with $ZrH_{1.7}$ and $ZrD_{1.7}$ Moderators

To further improve the production rate, a two-step production scheme is proposed, in which W is efficiently produced from natural Ta with the $ZrH_{1.7}$ moderator and Re-185 is efficiently produced from tungsten with the $ZrD_{1.7}$ moderator. The ratio of the irradiation period with the $ZrH_{1.7}$ moderator (first-step) and the subsequent irradiation period with the $ZrD_{1.7}$ moderator (second-step) up to 1 year was studied and the results are shown in Figure 6. Each irradiation period was followed by 15 days of cooling. The best balance for production of irradiation period between the first-step and the second-step was achieved when $(ZrH_{1.7} : ZrD_{1.7}) = (8 : 4)$. The Re-185 produced at this ratio was 2.3 g (about 0.15%/year from natural Ta) and that with an isotopic purity of 95.4%. This is about twice the production rate with the $ZrH_{1.7}$ moderator, and about 470 times the production rate without moderator over the entire irradiation period. In addition, this results also suggest that the isotopic purity of Re-185 can be controlled by adjusting the balance of irradiation period.

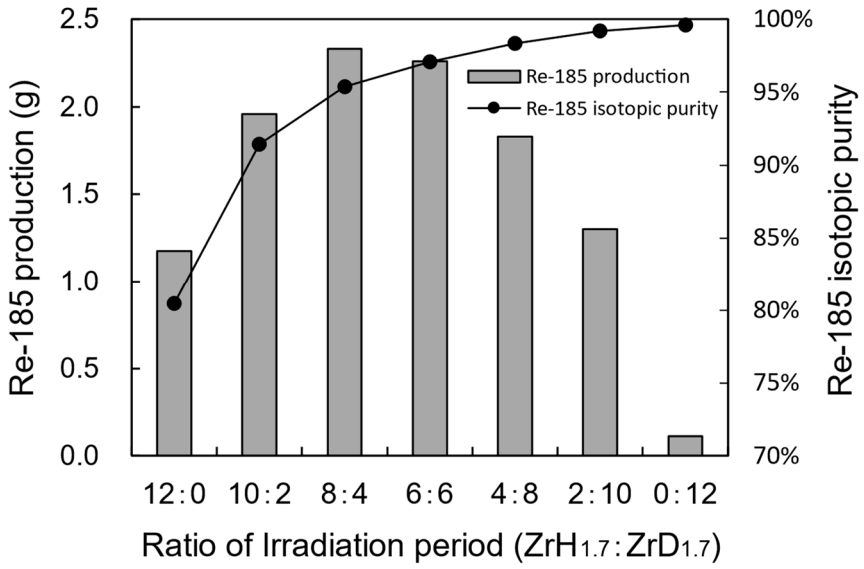

Figure 6. Re-185 production and isotopic purity with the ratio of irradiation period with $ZrH_{1.7}$ and $ZrD_{1.7}$ moderator up to 1 year irradiation.

3.3. Estimated Doses of Medical Re-186 Produced from Re185

The dose of Re-186 produced for medical use was estimated, assuming that the Re-185 produced in this study is irradiated in a thermal neutron reactor.

The High Flux Reactor in Petten, which is light-water-cooled and moderated (operated at 45 MWt), was employed, as it is commonly used to produce medical isotopes. The obtained specific radioactivity of Re-186 was 68 GBq/mg after 240 h of irradiation ($2.2–3 \times 10^{14}$ n/cm^2/s), with 97.4% Re-185 in that reactor [5]. Therefore, although the ratio of Re-185 is slightly different, the 2.3 g of Re-185 obtained in this study corresponds to more than 150,000 GBq of Re-186. In addition, the maximum tolerated dose of Re-186 HEDP (1-hydroxy-ethylidene-1,1 diphosphonic acid) complex, which is widely used for the palliative treatment of bone metastases caused by breast cancer and prostate cancer, is estimated to be 2.4 GBq [4,23]. Assuming that it takes approximately 1 week from the end of irradiation to the final product and accounting for the decay of radioactivity, the results of this study are equivalent to more than 17,000 doses.

3.4. Scheme for the Production of Medical Re-186 Starting from the Transmutation of Natural Ta

We propose a scheme of transmutation of natural Ta to Re-186 for medical use, as shown in Figure 7. In this study, a 100 MWt fast reactor was used to verify that Re-185

can be produced from natural Ta, but obviously the production Re-185 would be much higher if a large fast reactor were used (as in previous studies). In the chemical separation process, Re-185 could be separated from Ta, W, and Os; for example, Re, W, and Os would be co-separated from Ta by a conventional extraction with Tri-butyl Phosphate in low-concentration nitric acid media. Further, the mutual separation of Re and W would be possible using the separation process of the W-188/Re-188 generator [24]. Further investigations into Os behavior in this separation process are required. The purified Re-185 can be transmuted to Re-186 for medical use through using a nuclear reactor or accelerator-driven neutron sources with (n, γ) reaction. The remaining Ta and W should be recycled for the next irradiation, as it will increase the production rate of Re-185 due to its higher-order W content already produced in the previous irradiation.

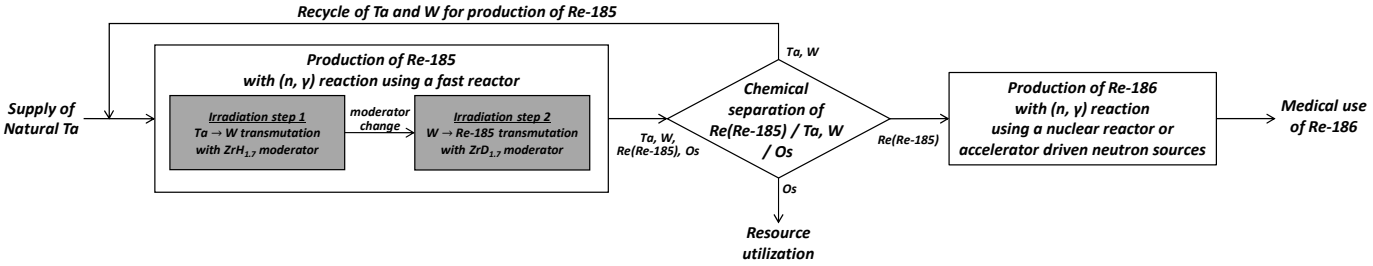

Figure 7. Production scheme of medical Re-186 starting from the transmutation of natural Ta. Using a fast reactor, irradiation is divided into two steps to improve the production rate of Re-185. After chemical separation, the remaining Ta and W should be utilized again for the production of Re-185, which will further improve the production rate.

4. Conclusions

The feasibility of producing rhenium-185 (Re-185) via nuclear transmutation was evaluated using a continuous energy Monte Carlo code MVP version 3/MVP-BURN. It was found that Re-185 with an isotopic purity of 99% can be produced through the transmutation of natural tantalum (Ta) using a 100 MWt fast reactor. In addition, it was found that the use of moderators is effective in improving the production rate of Re-185. This can be achieved with a zirconium hydride ($ZrH_{1.7}$) moderator in the main transmutation process from natural Ta to tungsten (W) and with a zirconium deuteride ($ZrD_{1.7}$) moderator in the main transmutation process from W to Re-185. Through two-step irradiation, first-step irradiation with a $ZrH_{1.7}$ moderator, and second-step irradiation with a $ZrD_{1.7}$ moderator, the production rate of Re-185 from natural Ta can be increased by up to 470 times compared with irradiation without a moderator, and 2.3 g of Re-185 can be obtained from 1571 g of natural Ta in 1 year of irradiation. This can be estimated as the raw material for more than 17,000 doses of Re-186 radiopharmaceutical treatment.

A cost evaluation for the practical application of Re-185 production was not performed in this study. We plan to investigate this in a commercial fast reactor and compare it to the conventional isotope production process.

Author Contributions: Conceptualization, Y.T., T.Y. and M.O.; software, T.Y.; investigation, Y.T.; resources, T.Y.; writing—original draft, Y.T.; writing—review & editing, T.Y. and M.O.; supervision, M.O.; project administration, M.O. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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