

# Production of Rhenium by Transmuting Tungsten Metal in Fast Reactors with Moderator

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**Abstract:** The feasibility of rhenium (Re) production by irradiating tungsten (W) metal in a medium size fast reactor was evaluated by using a Monte Carlo code. The fast reactor can produce about 50 kilograms of Re per every 3 years, which corresponds 10% of Japanese domestic production. The specific activity of Re can be reduced below the exemption level or even the natural Re level if W and osmium is separated after the irradiation. The use of ZrD<sub>1.7</sub> moderator reduces the specific activity by half compared to that of ZrH<sub>1.7</sub> case, and even the no moderator case is permissible to produce the production of Re which has lower specific reactivity than that of natural Re.

**Key words:** Rhenium, tungsten, fast reactor, transmutation, MVP, ORIGEN2, specific activity.

## 1. Introduction

In recent years, the demand for rhenium (Re) is growing as an additive to super alloys for jet engines. However, rhenium is one of the rarest elements in earth. The abundance of Re is much smaller:  $10^{-9}$  times of Si and  $10^{-4}$  times of tungsten (W). The price of Re is about 500 times that of W at present. Therefore, active technology for creation of Re that we advocate has a great value.

Après ORIENT research program, newly initiated in 2011, includes transmutation of stable elements to create rare metals/RE (rare earth) elements by  $(n, \gamma)$  reaction with subsequent beta decays in the reactors [1-3]. Re can be produced by nuclear transmutation where W captures neutrons and disintegrates to Re [4].

The objective of this study is to clarify the feasibility of production of Re from W metal in fast reactors by using neutron moderator.

The neutron energy spectrum of FRs (fast reactors) shows faster or harder than that of LWRs (light water reactors) due to the lack of neutron moderators. The average number of emitted neutrons of FRs per absorption, where the main fissile nuclide is plutonium-239, is about 2.5, while that of LWRs is 2.1. Furthermore, the neutron loss of parasitic absorption of structure and coolant per fission neutron is 0.1 in FRs, while that of LWRs is 0.5. Thus more excess neutrons are expected to utilize for transmutation if the system is designed appropriately.

In our study, we focused two points for setting the transmutation system in FRs, amount of produced Re for practical use and specific activity of Re for radiation protection. The exemption level of specific activity, that is a radioactivity level under that market use is approved, is defined by IAEA for each radioactive nuclide as Bq/gram-element [5].

## 2. Candidates for Neutron Moderator in FRs

Fig. 1 illustrates the transmutation of W to Re with other related reactions in reactors. The isotope

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abundance of W indicates that, two main paths for Re production are:

- (1) W-184 (abundance: 31%) to W-185 by neutron capture to Re-185 by beta disintegration;
- (2) W-186 (abundance: 28%) to W-187 by neutron capture to Re-187 by beta disintegration.

As shown in Fig. 2, lower energy neutrons react with more W isotopes. Fig. 3 shows the scattering and absorption cross section of the typical neutron moderator nuclides, proton and deuteron. Proton has a larger scattering cross section than deuteron by 1 order while deuteron has much smaller absorption cross section. We have selected, thus, materials containing proton or deuteron as candidates for effective moderator, and zirconium hydride or zirconium

deuteride are employed because zirconium can retain much hydrogen (1.7 times Zr or more at 600 °C) and have some experiences for the reactor materials in FRs.

### 3. Analysis Method

The code system used in the analysis is shown in Fig. 4. A continuous energy 3-dimensional Monte Carlo code, MVP [6] with its burn-up calculation routine MVP-BURN [7] and JENDL-4.0 library [8] where the neutron energy spectra of each zone of reactors are exactly simulated as well as detailed configurations is used for evaluating the transmutation rate of W, the effective neutron cross sections and neutron flux for the use of ORIGEN2.

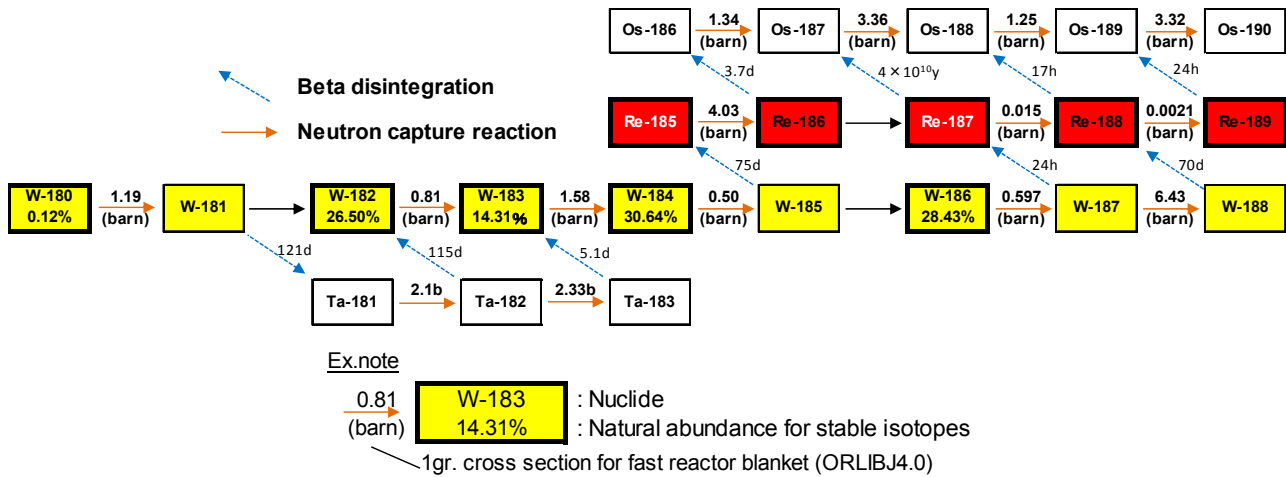


Fig. 1 Nuclear transmutation scheme of tungsten (W) to rhenium (Re).

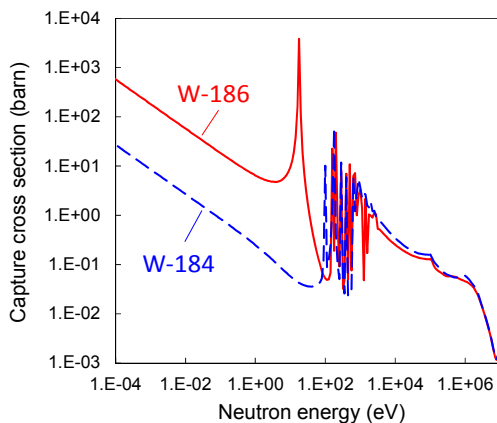


Fig. 2 Capture cross section of W isotopes.

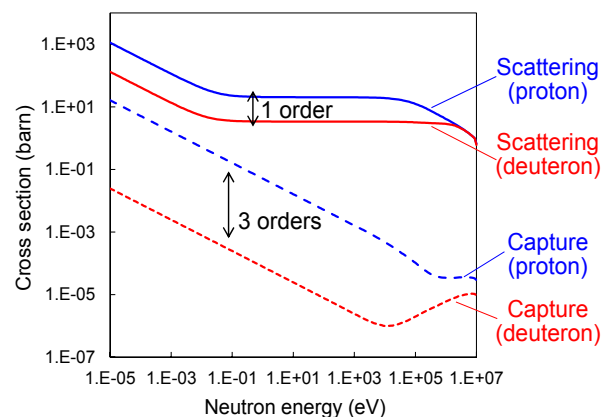


Fig. 3 Cross section of hydrogen and deuteron.

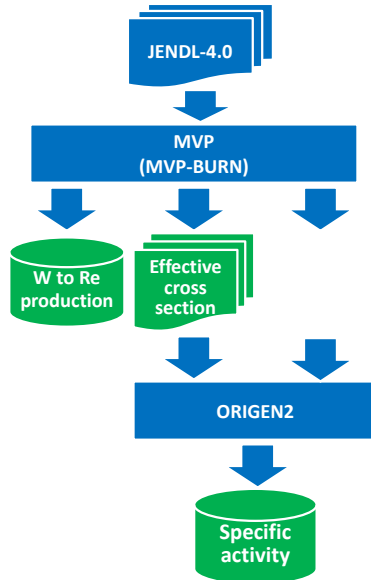


Fig. 4 Capture cross section of W isotopes.

The one-point model burn-up code ORIGEN2 with one group cross section system [9] is used for evaluating the specific activity of each isotope by applying the effective cross section and flux evaluated by the MVP code.

#### 4. Core and Assembly for Transmutation

The fast reactor core layout and the assembly for the transmutation are shown in Fig. 5. The thermal

power of the core is 710 MWt, the coolant is liquid sodium and the active core size is 1.8 m in diameter and 0.9 m in height.

The number of the target assemblies is 54 located at the peripheral of the core where the impact of the target on core performance can be minimized.

The pin layout of the target assembly is shown at the right hand in Fig. 5. Outer 43 pins contain tungsten (W) pellets, which total height is 0.9 m.

Inner 18 pins contain zirconium hydride ( $\text{ZrH}_{1.7}$ ) or zirconium deuteride ( $\text{ZrD}_{1.7}$ ) pellets, which face the active core.

We have set the no-moderator case for the comparison, where all of 61 pins contain W pellets only.

#### 5. Results and Discussion

##### 5.1 Cross Section and Neutron Flux at W Region

As shown in Table 1, the one group effective capture cross section and neutron flux at W region are evaluated by MVP for the use of ORIGEN2, where specific reactivity is calculated. The effective cross sections of moderator applied cases increase by a few times that of no moderator case (Case 5), while the fluxes reduce by about 0.6-0.9 time only.

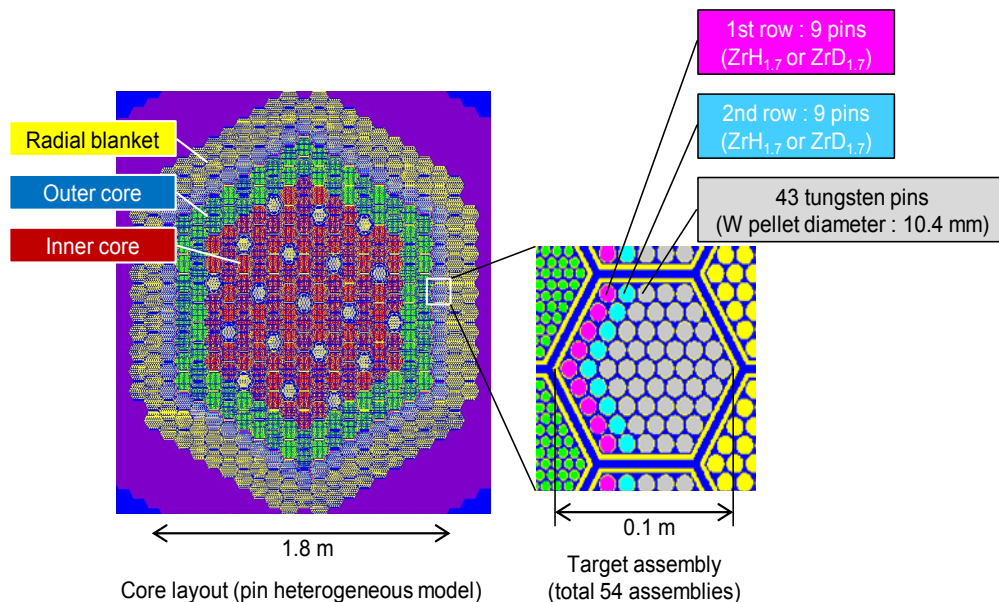
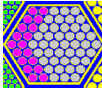
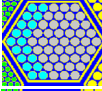
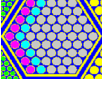
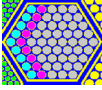
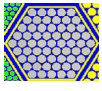
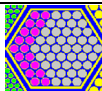
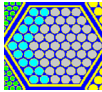
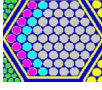
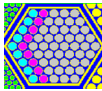
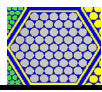


Fig. 5 Core layout and target assembly model for MVP calculation.

**Table 1 One group effective capture cross section and neutron flux at W region by MVP code.**

Case	Number of pins	Pin map	W-182 (barn)	W-183 (barn)	W-184 (barn)	W-186 (barn)	Re-185 (barn)	Re-187 (barn)	Neutron flux (n/cm <sup>2</sup> /s)
Case 1	ZrH <sub>1.7</sub> 18 pins W 43 pins		0.574	1.203	0.237	0.580	9.348	4.823	7.57E+14
Case 2	ZrD <sub>1.7</sub> 18 pins W 43 pins		0.268	0.792	0.230	0.176	2.355	2.361	1.09E+15
Case 3	ZrH <sub>1.7</sub> 9 pins ZrD <sub>1.7</sub> 9 pins W 43 pins		0.418	1.053	0.235	0.365	6.255	4.138	8.79E+14
Case 4	ZrD <sub>1.7</sub> 9 pins ZrH <sub>1.7</sub> 9 pins W 43 pins		0.442	1.115	0.24	0.399	6.728	4.39	8.65E+14
Case 5	W 61 pins (no moderator)		0.187	0.459	0.169	0.124	1.093	1.008	1.21E+15
Original ORLIBJ40 (infinite dilute model)			0.808	1.583	0.500	0.597	4.027	0.015	-

**Table 2 Production weight of Re after 3 years irradiation.**

Case	Number of pins	Pin map	W load (kg)	Re weight (kg)	Re/W (%)	Re-185 (kg)	Re-187 (kg)	Re-187 /Re-185 -
Case 1	ZrH <sub>1.7</sub> 18 pins W 43 pins		3,520	51.2	1.45	14.1	37.1	2.63
Case 2	ZrD <sub>1.7</sub> 18 pins W 43 pins		3,520	40.3	1.14	23.6	12.7	0.54
Case 3	ZrH <sub>1.7</sub> 9 pins ZrD <sub>1.7</sub> 9 pins W 43 pins		3,520	44.4	1.26	17.6	27.8	1.58
Case 4	ZrD <sub>1.7</sub> 9 pins ZrH <sub>1.7</sub> 9 pins W 43 pins		3,520	45.6	1.30	17.3	27.2	1.57
Case 5	W 61 pins (no moderator)		5,010	49.1	0.98	29.1	19.9	0.68

### 5.2 Production of Re after 3 Years Irradiation

The production weight of Re after 3 years irradiation in the core has been evaluated by MVP-BURN code. Table 2 lists the load weight of W, production weight of Re and the ratio with isotropic fractions.

The produced Re weight at 3 years irradiation will be more than 50 kg in prototype reactor, which corresponds to about 10% of Japanese total production of Re at present.

The Re production rate of moderator applied cases is 1.5 times that of no-moderator case at maximum.

The weight of Re-187 is larger than that of Re-185 in Case 1 (ZrH moderator), while Re-185 is larger than Re-187 in Case 2 (ZrD moderator).

This relation is investigated by analyzing neutron spectra as shown in Fig. 6. The softer neutron spectrum by ZrH<sub>1.7</sub> in Case 1 produces more Re-187 due to the higher W-186 cross section in lower energy. The harder spectrum by ZrD<sub>1.7</sub> in Case 2 produces less

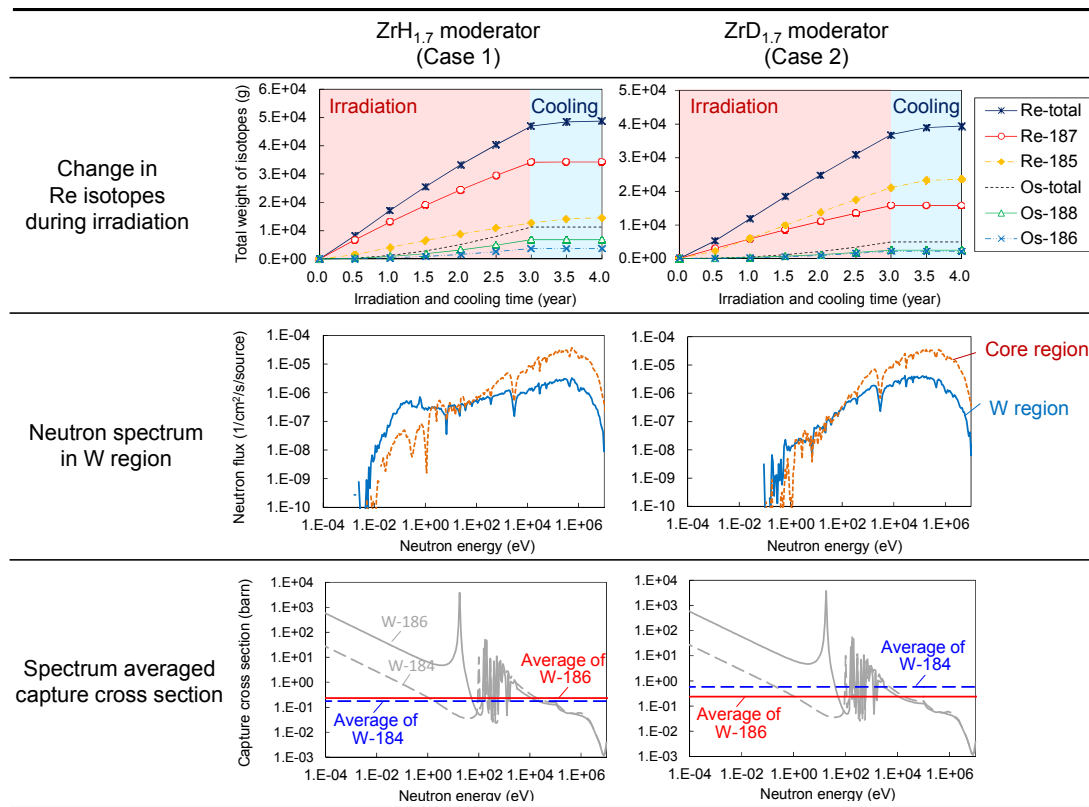


Fig. 6 Comparison of  $\text{ZrH}_{1.7}$  (Case 1) and  $\text{ZrD}_{1.7}$  (Case 2) moderator.

Table 3 Effective cross section of W isotopes.

Case (moderator)	W-184	W-186
Case 1 (ZrH)	0.237 b	0.580 b
Case 2 (ZrD)	0.230 b	0.176 b

Re-187. The effective cross section of W isotopes in the target region is shown in Table 3.

### 5.3 Specific Activity of Re

The specific activity of Re during irradiation and cooling has been evaluated by ORIGEN2 with the cross section and flux shown in Table 1.

Fig. 7 shows the specific activities of the isotopes of W, Re, and Os (osmium) in Case 1 where the weight of Re production is maximum among the study cases. The exemption levels of Re isotopes determined by IAEA [5] are listed in Table 4.

In Fig. 7, the main activity of Re is that of Re-188, the daughter of W-188, during the cooling time, if W and Re are mixed when the specific activity is 6 orders higher or more than the exemption level. Thus the

exemption level of Re specific activity is achieved in a half year cooling if W and Re is separated after the irradiation as shown in Fig. 8.

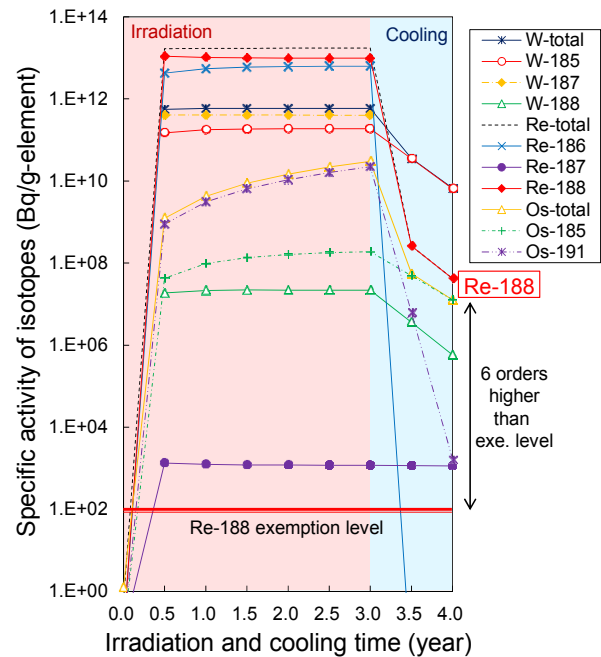


Fig. 7 Specific activity when Re is not separated from W.

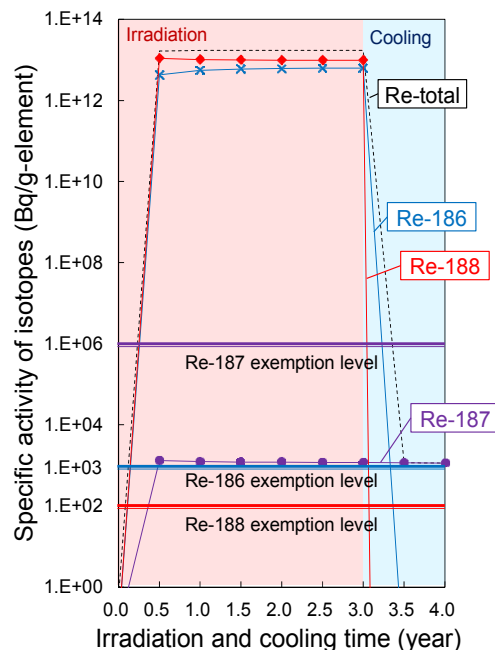
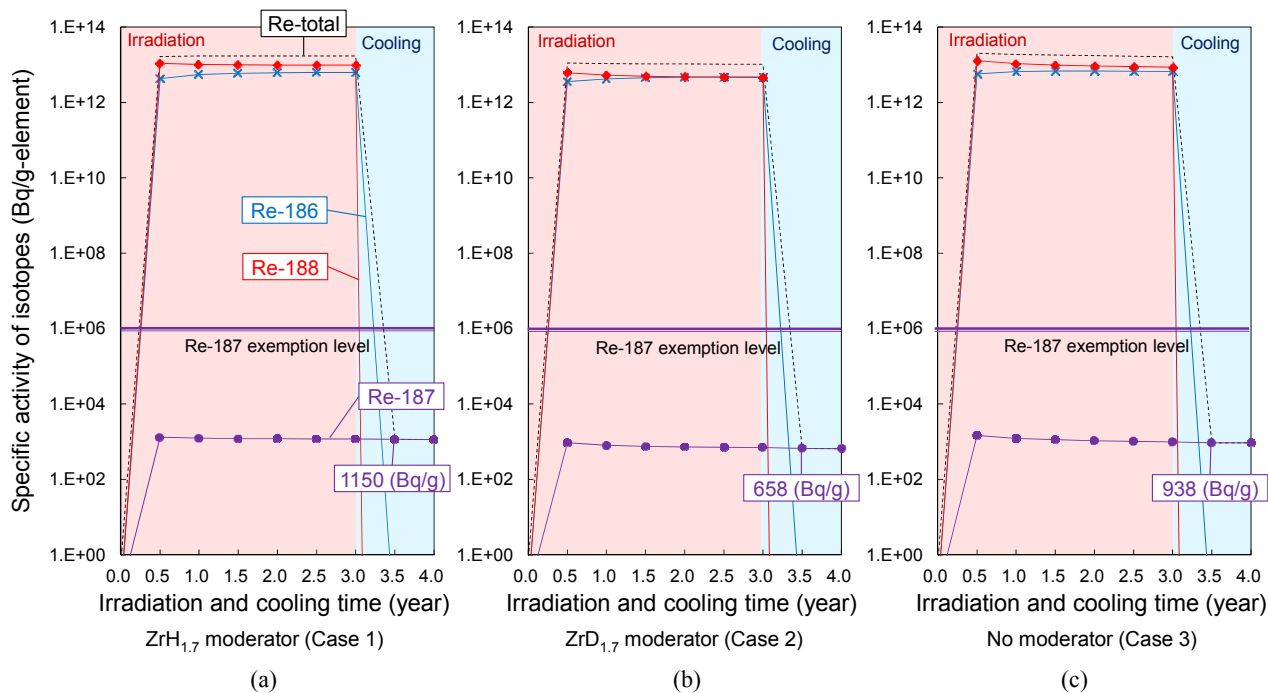
**Table 4** Exemption level of Re isotopes.

Isotope ( $T_{1/2}$ )	Bq/g
Re-186 (3.8 d)	$1 \times 10^3$
Re-187 ( $5 \times 10^{10}$ y)	$1 \times 10^6$
Re-188 (17 h)	$1 \times 10^2$

#### 5.4 Specific Activity for Various Moderators

Fig. 9 illustrates the specific reactivity of Re for various types of moderators when Re is separated from W and Os after the irradiation. The use of  $\text{ZrD}_{1.7}$  reduces the specific activity by half compared to that of  $\text{ZrH}_{1.7}$  or no moderator due to the decrease of the fraction of Re-187 ( $T_{1/2} = 4.33 \times 10^{10}$  years).

The specific activity of produced Re is lower than that of natural Re (2,610 Bq/g, Re-187 62.6%) for all cases in Fig. 9, which suggests the no moderator case is permissible for the production of Re in the fast reactor shown above.

**Fig. 8** Specific activity when Re is separated from W and Os after irradiation.**Fig. 9** Specific activity of Re vs. types of moderator.

## 6. Conclusions

The feasibility of rhenium (Re) production by irradiating tungsten (W) metal in a medium size fast reactor was evaluated by using the Monte Carlo calculation code MVP and ORIGEN2. The prototype fast reactor can produce about 50 kilograms of Re per every 3 years, which corresponds to about 10% of Japanese domestic production. The specific activity of Re can be reduced below the exemption level or even the natural Re level if W and Os is separated after the irradiation. The use of  $ZrD_{1.7}$  moderator reduces the specific activity by half compared to that of  $ZrH_{1.7}$  case, and even the no moderator case is permissible to produce the production of Re which has lower specific reactivity than that of natural Re.

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