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Radioactivity of the vanadium-alloy induced by D–T neutron irradiation

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Abstract

In order to evaluate the induced activity of the V-alloy, DT irradiation experiments were performed for the V-alloy prepared by NIFS, US DOE and SWIP. The radioactivities were evaluated as a function of the cooling time. The dominant radioactive nuclei are the ^{51}Cr for 4–200 d, ^{46}Sc for 200–1200 d, the ^{57}Co and ^{54}Mn for a cooling time of over 1200 d after 151 h irradiation. Radioactivities of ^{57}Co and ^{54}Mn in the specimen by NIFS are 2–8 times smaller than those by other sources. From the induced activities, impurity concentrations can be clarified. It can be confirmed that the impurity concentration of Al in the ingot by NIFS is lower than the targeted number.

1. Introduction

The V-alloy is a candidate structure material for the liquid lithium breeding blanket, due to its impressive mechanical properties at high temperature, chemical compatibility with liquid lithium, and low activation property [1]. The induced activity of the V-alloy, however, would be enhanced by impurities. It is important to evaluate the induced activity taking into account the impurities included in the fabrication process. In the study of the previous design, Dolan et al. [2] and Cheng et al. [3] evaluated the induced activities of the V-alloy, and proposed the design targets for the elements that should produce long-lived radioactive nuclei. The examples of the design target are 0.036, 91, 21 and 0.17 wppm for Ag, Al, Mo and Nb, respectively, which produce long-lived radioactive nuclei of ^{108m}Ag (half-life, $T_{1/2}$: 418 y), ^{26}Al ($T_{1/2}$: 7.4×10^5 y), ^{94}Nb ($T_{1/2}$: 2.0×10^4 y) and ^{94}Nb, respectively. Ikeda et al. also roughly estimated the impurity concentrations of the V-alloy (#832864, #832865, #832394) prepared by the Teledyne Wahchang Albany Co. using D–T neutron irradiation experiments [4]. Mo and Nb impurity concentrations were reported, unlike the Ag and Al. Those were higher than the design targets by 1–2 orders of magnitude.

The fabrication study has been conduced by NIFS [5,6] to reduce the impurity concentrations producing long-lived radioactive nuclei. Thus, the D–T neutron irradiation experiments were performed for the V-alloy (V–4Cr–4Ti, Cr: 4.19 wt%, Ti: 3.61 wt%) prepared in the reference fabrication process proposed by NIFS in the present study. The induced activities were thereafter evaluated. The induced activities were also evaluated for the V-alloys by other sources (US DOE, Cr: 3.25 wt%, Ti: 4.05 wt% (#832665) [7], and SWIP, Cr: 3.61 wt%, Ti: 4.11 wt% [8]) for comparison purposes. Based on measurements of the induced activities, impurity concentrations were also elaborately evaluated.

2. Experimental method

Three test specimens, extracted from the ingots, using a variety of methods described above, were prepared. The test specimen has an area of a 10×10 mm^2 square and a thickness of 0.1 mm, the weight being about

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0.04–0.06 g. As a neutron fluence monitor, niobium foils with an area of 10×10 mm^2 and a thickness of 0.1 mm were attached to the specimens.

D–T neutron irradiations were performed using the 0° beam line of fusion neutronics source (FNS) [9] at JAERI. The DT neutron yield at the target was monitored by the ^{232}Th fission chamber located at the ceiling. Fifteen continuous irradiations, each about 10 h long, were performed, the integrated irradiation period being 151 h in total. Waiting time between each irradiation period was about 14–86 h. Total and average neutron yields were 1.2×10^{18} neutrons and 2.1×10^{12} neutrons per second, respectively.

In order to measure the short-lived radioactivities, 7 h, 25 and 10 min irradiations were also conducted. The specimens were attached to the Ti–T target assembly, with a distance from the target to the specimens limited to the range of 10–30 mm.

After these irradiations, induced radioactivities were measured during the cooling down process of several min to 1.5 years in length, using the gamma ray spectroscopy with the high purity Ge detector.

3. Results

The measured γ-ray spectra, subtracted from the background data, are shown in Figs. 1 and 2 for the test specimen by NIFS (NI) at the time of specimen cooling of about 4 and 320 d, respectively, after 151 h irradiation. Activities of the niobium foil attached to NI, those by US DOE (UD) and SWIP (SW) were 1.5×10^5, 7.4×10^4 and 9.1×10^4 Bq/g, respectively, just after irradiation. The measured activities of the niobium foil, and the neutron spectrum obtained by the Monte Carlo calculation indicate, that the neutron flux with an energy range of 13.9–15.0 MeV in NI is 4.4×10^9 cm^{-2} s^{-1}, the energy-integrated neutron flux being 4.8×10^9 cm^{-2} s^{-1}. The main observed radioactive nuclei, and the induced radioactivities are listed in Table 1 with the $T_{1/2}$, and a cooling time after irradiation. The induced radioactivities in UD and SW are normalized by multiplying the measured values in UD and SW by the ratio of the reaction rate of the niobium foil attached to the NI to those to the UD and SW.

Based on the measured data, the radioactivities in the NI, UD and SW were evaluated by each radioactive nucleus as a function of the cooling time after 151 h irradiation. The evaluated result for NI is shown in Fig. 3. The dominant radioactive nuclei are the ^{48}Sc for a period of 0–4 d, the ^{51}Cr for 4–200 d, the ^{46}Sc for 200–1200 d, and the ^{57}Co and ^{54}Mn for a cooling time longer than 1200 d for all specimens. The activities of the ^{54}Mn in the UD and SW are 3 and 8 times more intense, respectively, than that in the NI, and the activities of the ^{57}Co in UD and SW are 2.2 and 1.6 times more intense,

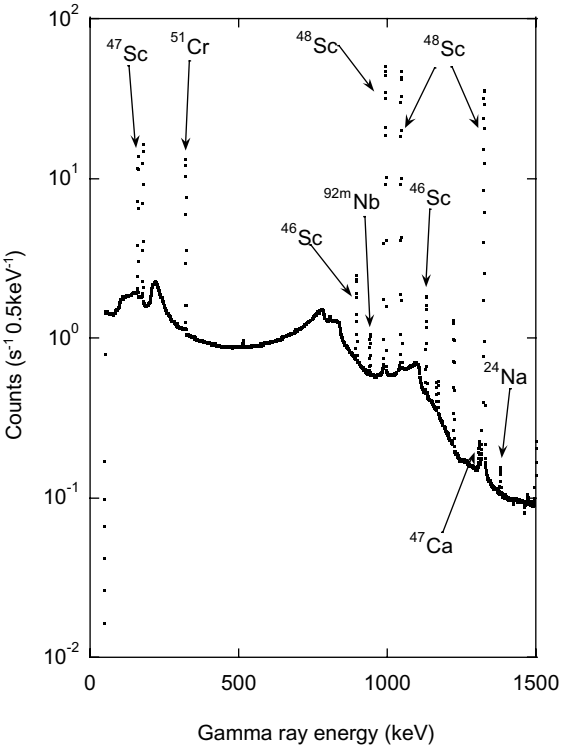

Fig. 1. Measured γ-ray spectrum for the test specimen by NIFS (NI) at the time of specimen cooling of about 4 days.

respectively, than that in the Nl. It can be expected that there will be a significantly less residual radioactivity in the NI compared with the radioactivities of the UD and SW in a cooling time of over 1200 d; thus it is certain that the NI shows a lower activation property.

4. Discussions

We calculated the radioactivity for 2 years irradiation, which corresponds to the actual operation condition assumed in the DEMO blanket [10], with the following formula:

$$A_n(t) = A_{n,m}(1 - e^{-\lambda_n t})/(1 - e^{-\lambda_n t_e}),$$

where the $A_n(t)$, $A_{n,m}$, λ_n, t and t_e represent the induced radioactivity of the nucleus n in the irradiation period t, the measured value of the nucleus n in the present experiment, the decay constant of the nucleus n, the irradiation period in the calculation, and that in the experiment itself. The values of t and t_e are 2 years and 151 h, respectively.

The final result is shown in Fig. 4. The dominant radioactive nuclei are the ^{51}Cr for a period of 0–150 d, the ^{46}Sc for 150–1000 d, and the ^{57}Co and ^{54}Mn for a

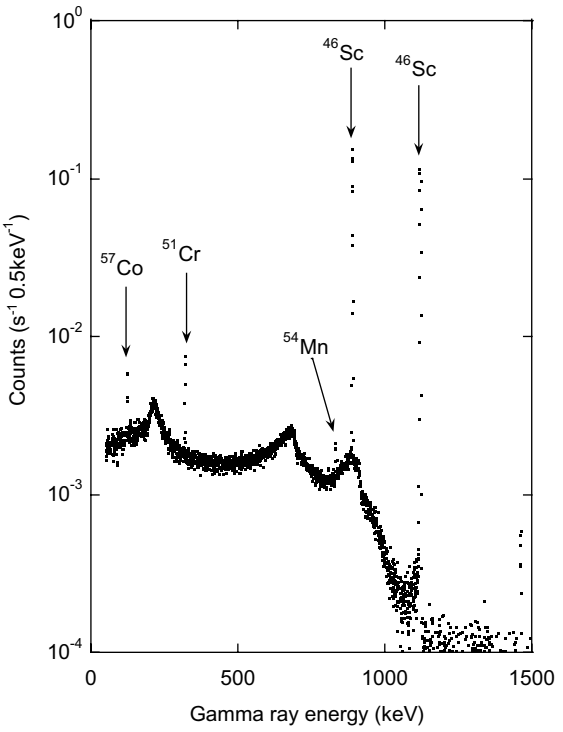

Fig. 2. Measured γ-ray spectrum for the test specimen by NIFS (NI) at the time of specimen cooling of about 320 days.

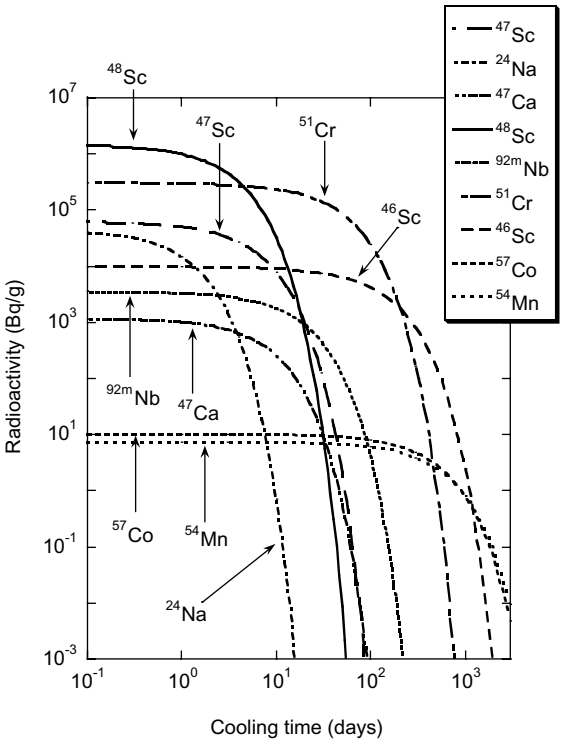

Fig. 3. Radioactive nuclei for the test specimen by NIFS (NI), as a function of the cooling time after 151 h irradiation. Based on the measured data, the radioactivities are evaluated by each radioactive nucleus.

cooling time of over 1000 d. The impact of the radioactive nuclei with a long half-life on the dominant radioactive nuclei has a higher significance in the actual operation condition of the nuclear fusion reactor.

It is expected, that the ^{46}Sc, ^{47}Sc, ^{48}Sc and ^{51}Cr are generated by the primary nuclear reaction via the ^{46}Ti (n, p), ^{47}Ti (n, p), ^{51}V (n, α) and the ^{52}Cr (n, 2n) of the major constituents in the V-alloy. On the other hand, it is predicted, that the ^{54}Mn is generated by the nuclear reaction via the ^{55}Mn (n, 2n) and the ^{54}Fe (n, p) in the impurity, and ^{57}Co is generated by that via ^{58}Ni (n, np) and ^{58}Ni (n, d) in the impurity. It is expected, that the impurity concentrations of Mn, Fe and Ni in the UD and SW are larger than those in the NI. The induced activities, generated by the major constituents, are negligibly small compared with those by the impurities for a

Table 1
Main observed radioactive nuclei and induced radioactivities

	NI		UD		SW		$T_{1/2}$
	Cooling (days)	Activity (Bq/g)	Cooling (days)	Activity (Bq/g)	Cooling (days)	Activity (Bq/g)	
^{24}Na	4	5.6E+2[a]			4.9	3.0E+2	15 h
^{47}Ca	4	6.4E+2	17.6	8.9E+1	4.9	5.4E+2	4.5 d
^{46}Sc	4	9.4E+3	10.8	8.3E+3	4.9	9.5E+3	83.8 d
^{47}Sc	4	2.7E+4	10.8	6.4E+3	4.9	2.3E+4	3.4 d
^{48}Sc	4	3.2E+5	10.8	2.3E+4	4.9	2.3E+5	43.7 h
^{51}Cr	4	2.7E+5	10.8	1.9E+5	4.9	2.5E+5	27.7 d
^{54}Mn	319.5	3.7E+0	449.8	6.8E+0	449.8	2.2E+1	312.5 d
^{57}Co	319.5	4.6E+0	449.8	6.4E+0	449.8	5.2E+0	271 d
^{92m}Nb	4	2.7E+3	10.8	1.5E+3	4.9	1.9E+3	10.15 d

[a] Read as 5.6×10^{2}.

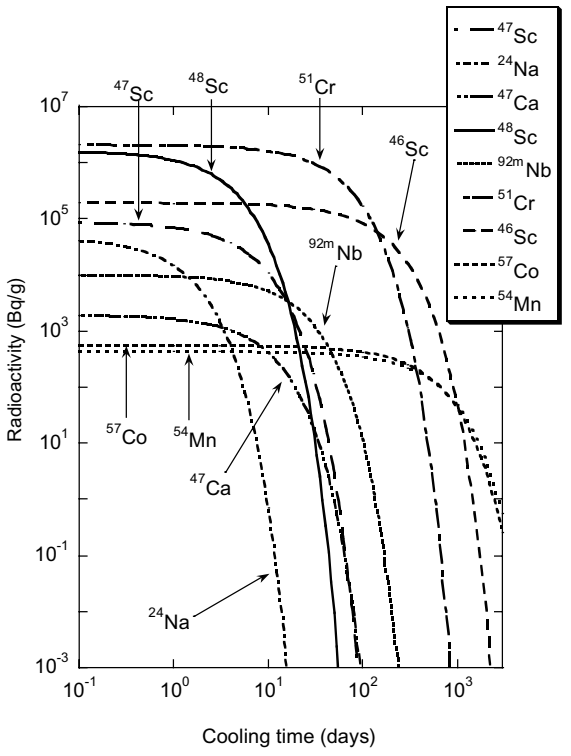

Fig. 4. Estimated radioactive nuclei for the test specimen by NIFS (NI), as a function of the cooling time after 2 years irradiation. Based on the measured data, the radioactivities are evaluated by each radioactive nucleus.

cooling time of over 1200 d. Thus, it is concluded that the V-alloy is very useful as a structural material from the viewpoint of the low activation property, though the radioactivities induced by the impurities are required to be suitably small.

The ^{47}Sc can be generated by the β-decay of the ^{47}Ca, generated by the nuclear reaction via the ^{51}V (n, α). The induced activities of the ^{47}Sc are more intense than those of the ^{47}Ca by more than one order of magnitude, for a cooling time of 0–30 d (as indicated in Fig. 3). Therefore, it is concluded that most of the ^{47}Sc are generated by the nuclear reaction via the ^{47}Ti (n, p). This occurs due to the activation cross-section of the ^{47}Ti (n, p) being higher than that of ^{51}V (n, α) by more than one order of magnitude. The half-life of ^{47}Ca is longer than that of the ^{47}Sc; thus the ratio of the ^{47}Sc generated by the β-decay of ^{47}Ca is increased as the cooling time is extended.

^{51}Cr and ^{54}Mn can be generated by sequentially charged particle reactions, which are defined as reactions induced by secondary charged particles, via the ^{51}V (p, n) and the ^{51}V (α, n), respectively. In the previous studies [11,12], the effective activation cross-sections were evaluated to be about 2×10^{1}–6×10^{2} barn for the reaction of the ^{51}V (p, n) against the incident DT neutron. The activation cross-section of the ^{51}V (p, n) ^{51}Cr, due to the sequentially charged particle reaction, is much lower than the rate of the ^{52}Cr (n, 2n) ^{51}Cr, since the primary neutron reaction exceeds the former by more than 2–3 orders of magnitude. It can be concluded that radioactivities by the sequentially charged particle reactions, are negligibly small, compared with the primary neutron reaction. Similarly it is expected to have negligible impact of the production of ^{54}Mn due to ^{51}V (α, n) reaction.

Weak γ-lines, due to the ^{24}Na and the ^{92m}Nb were detected in the measured γ-ray spectrum (as presented in Fig. 1). These nuclei are generated by the nuclear reaction of the impurities. In addition, very weak γ-lines due to the ^{28}Al, ^{56}Mn, ^{58}Co and the ^{89}Zr could also be detected. It is expected that the ^{24}Na is generated by the nuclear reaction via ^{24}Mg (n, p), ^{25}Mg (n, np) and ^{27}Al (n, α), ^{92m}Nb via ^{93}Nb (n, 2n) and ^{92}Mo (n, p), ^{58}Co via ^{59}Co (n, 2n), ^{28}Al via ^{28}Si (n, p), ^{56}Mn via ^{56}Fe (n, p) and ^{59}Co (n, α), ^{89}Zr via ^{90}Zr (n, 2n) and ^{92}Mo (n, α).

From measurements of the radioactivities generated by the impurities, impurity concentrations of Mg, Al, Si, Mn, Fe, Ni, Co, Zr, Nb and Mo could be evaluated (as seen in Table 2). As for the generation of the ^{92m}Nb, we cannot distinguish any contributions, due to the nuclear reaction via ^{92}Mo (n, p) and that via ^{93}Nb (n, 2n). The estimated temporary maximum values are shown in Table 2 for concentrations of the Mo and Nb. Impurity concentrations of Al, Mg, and Zr also show the temporary maximum values, while most of the ^{24}Na are expected to be generated by ^{27}Al (n, α) based on the chemical analysis. As for the generation of the ^{56}Mn, the contribution cannot be distinguished, due to the nuclear reaction via the ^{56}Fe (n, p) and ^{59}Co (n, α). The generation of the ^{59}Co (n, α) ^{56}Mn from the impurity concentration of Co is estimated, based on the generation of ^{59}Co (n, 2n) ^{58}Co. Thereafter, the impurity concentration of Fe is evaluated by subtracting the contribution of the generation from the ^{59}Co (n, α) ^{56}Mn. The impurity concentration of Mn was evaluated by the same procedure. Impurity concentrations evaluated by the chemical analysis are also shown in Table 2, with values only roughly consistent with those in the present study.

It can be confirmed, that the impurity concentration of Al in the ingot fabricated by NIFS, is less than the design target of 91 wppm proposed by Dolan et al. [2], though those by US DOE and SWIP can have a concentration, exceeding the target by 2–4 times. The impurity concentrations of Mo and Nb in all specimens can be higher than the design targets of 21 and 0.17 wppm, respectively, except for Mo in SWIP, though they are still significantly lower than the V-alloy (#832864, #832865, #832394) produced by the Teledyne Wahchang Albany Co. evaluated by Ikeda et al. Further reduction of these impurities is necessary.

Table 2
Impurity concentrations analyzed by induced radioactivities, evaluated in the present study

	NI		UD		SW	
	PS[a]	CA[b]	PS	CA	PS	CA
Mg	<53	–	<1.3E+2[c]	–	<2.9E+2	–
Al	<70	59	<1.7E+2	1.9E+2	<3.8E+2	3.4E+2
Si	2.7E+2	2.7E+2	9.1E+2	7.3E+2	1.3E+2	1.5E+2
Mn	<2.0	<1	<5.6	<1	<16	17
Fe	<9.9E+1	4.9E+1	<2.8E+2	2.0E+2	<8.0E+2	6.5E+2
Ni	6.4	7	14	9	10	9
Co	1.0	0.7	0.2	0.2	0.4	0.2
Zr	<5.8	2.5	<36	40	<14	<10
Nb	<7.0	0.8	<70	54	<1.6	0.2
Mo	<5.4E+1	2.4E+1	<5.4E+2	3.0E+2	<11	4

Unit: wppm.
[a] Present study.
[b] Chemical analysis.
[c] Read as 1.3×10^2.

5. Conclusion

The DT neutron irradiation experiments were performed for the V-alloy, prepared by NIFS, US DOE and SWIP. Radioactivities of these specimens in the experiment were reviewed. The dominant radioactive nuclei are the ^{57}Co and the ^{54}Mn for a cooling time of over 1200 d. Radioactivities of ^{57}Co and ^{54}Mn, in the specimen prepared by NIFS, are 2–8 times smaller than those by US DOE and SWIP. From the induced activities, impurity concentrations could be clarified. It is possible to confirm, that the impurity concentration of Al in the ingot, fabricated by NIFS, is lower than the design target.

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