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Activation experiment with tungsten in fusion peak neutron field

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Abstract

Tungsten is the preferred material for the divertor plates of fusion devices and a constituent of reduced activation structural materials. Samples of pure tungsten were irradiated with D-T fusion neutrons. The radioactivity following irradiation was determined several times during decay by γ-spectroscopy. The results were analysed with the European Activation System. Ratios of calculated-to-experimental values for individual activities are discussed in connection with the expected activation performance of the material on fusion power plant conditions.

1. Introduction

The radioactivity induced by neutrons in the materials of a fusion device represents a central topic of safety-related investigations [1]. Radionuclides with a broad range of half-lives have to be included in the corresponding analyses. The short-term radioactivity (half-life ranging from the order of magnitude of minutes to weeks) is mainly of interest with respect to heat production and shut-down dose rates, whereas long-term radioactivity (half-life of the order of 10–100 years or more) determines the waste management [2]. Calculations of the radioactivity are based on inventory codes and nuclear data libraries such as the European Activation System (EASY) [3] that need to be validated experimentally to assure they give reliable results when applied in design calculations. This is necessary for the variety of nuclides constituting the materials to be used in a fusion device.

Tungsten is the preferred material for the divertor and constituent in other components of fusion devices. The validation of EASY-2001 [4] with the experimental data available at that time revealed for W the necessity for further improvements.

The spectrum of the neutron flux in a fusion device consists of two parts, a D-T fusion peak at 14 MeV and a continuum ranging down to thermal energies. The radioactivity is mainly produced at 14 MeV neutron energy where the number of open reaction channels is a maximum, and at thermal energy where cross-sections are large. In the present work, the radioactivity induced by 14 MeV neutrons in W was investigated experimentally and analysed with EASY-2001 and the following version EASY-2003.

2. Experiment

In a calculation with EASY, pure W was assumed to be irradiated with 14 MeV neutrons of a flux density corresponding to the power density of 1.0 MW/m^2, for a period of one year. The result obtained for the contact dose rate as a function of decay time after irradiation is presented in Fig. 1.

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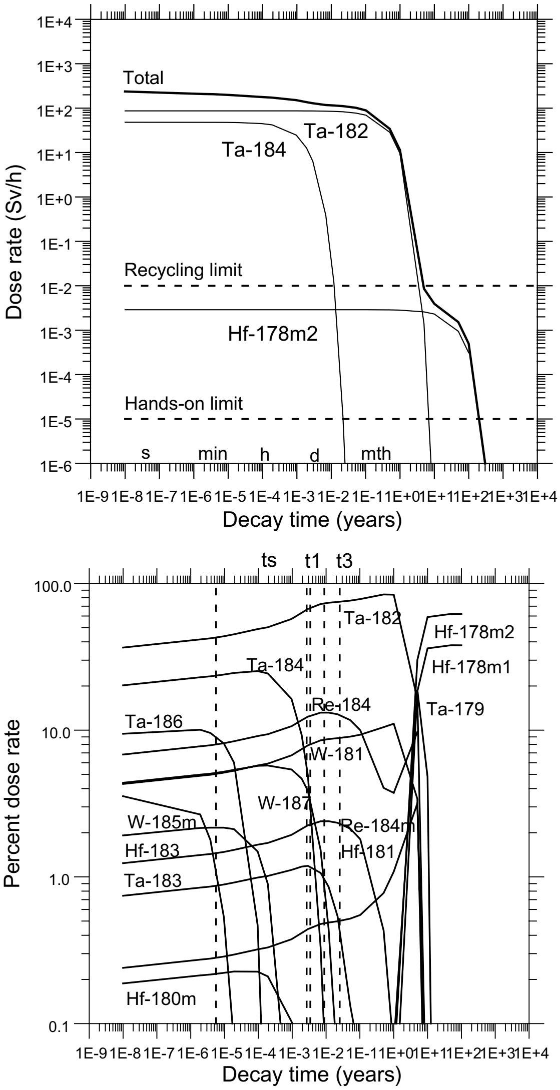

Fig. 1. Contact dose rate (upper part) and contribution of the different radionuclides to the total dose rate (lower part) after irradiation of W with 14-MeV neutrons of 1.0 MW/m^2 power density for one year, as a function of the decay time.

After about 5 years the dose rate is expected to be below the recycling limit and after about 200 years below the hands-on limit. To investigate the activity of all nuclides that dominate before the recycling limit is reached, two different irradiations were carried out. A short irradiation followed by activity measurements at decay times in the range labelled by '*t*s' in Fig. 1 (lower part) was directed to the short-term radioactivity; and after a longer irradiation with activity measurements at times '$t_1, \ldots, t_3$', the production of the other nuclides was measured.

The irradiations were performed at the high-intensity D-T neutron generator SNEG-13 [5] at Sergiev Posad. The parameters of three irradiations are given in Table 1. The 14 MeV neutron peak had at the sample position a mean energy of $\langle E_n \rangle = 14.93$ MeV and a spread of $\Delta E_n = \pm 0.27$ MeV during the irradiation 1 and 2. The incidence energy was shifted in the third experiment to (14.37 ± 0.12) MeV to check the influence of threshold reactions on the γ-radioactivity. The applied neutron fluences were determined via activation measurement of niobium foils by the reaction ^{93}Nb(n,2n)^{92m}Nb. The cross-section of this reaction was considered constant in the energy range 14–15 MeV and equal to 464 mb with an uncertainty of 4.2%. A possible background component of thermal neutrons was checked by ^{197}Au(n,γ) and by ^{115}In(n,γ) activation using thin foils. No statistically significant count rate was found for the decay of ^{198}Au. The very large integral capture cross-section of ^{115}In resulted in count rates, which showed that the flux of low-energy neutrons at the sample position is 10^{-5} of the total. The W samples had dimensions of 10×10 mm and a thickness of the order of 1 mm. γ-ray spectra were taken from the samples several times in the range '*t*s' after irradiation 1 and at t_2, t_3 and t_4 in the case of irradiations 2 and 3. The detection efficiency of the Ge(Li)-spectrometer used had an uncertainty of 2.5–3.0% in the energy range of measurements (60 keV–3 MeV). γ-activities identified by energy and half-life were used to determine nuclide activities with γ-yield data from EASY.

3. Results

The measured activities were analysed with the versions EASY-2001 and EASY-2003 of the European Activation System. Results are presented in Table 2. The uncertainty of the calculated activity includes both cross-section and half-life errors as estimated by EASY-2001. The uncertainty of the experimental values given

Table 1
Irradiation of samples with D-T neutrons

Number	Sample mass (g)	Neutron energy (MeV)	Irradiation time	Neutron fluence (cm^{-2})
1	1.816	14.93 ± 0.27	10.0 min	6.20×10^{12}
2	1.816	14.93 ± 0.27	13.93 h	2.32×10^{14}
3	1.725	14.37 ± 0.12	13.93 h	4.00×10^{13}

in Table 2 represent the root-mean-square of the uncertainties of the neutron fluence monitoring, the γ-yield data, the efficiency calibration of the γ-spectrometer and the γ-counting statistics. As the experimental uncertainties are lower than the uncertainty estimations by EASY, the experimental results can be used for improving the EASY data base.

For the ^{187}W activity, ratios of calculated-to-experimental activity (C/E) were obtained that are significantly below 1.0. This may mainly be attributed to a contamination of the neutron field by D-D self-target neutrons [6], the fluence of which on the sample was not exactly measured and the $\Delta E/E$ for this activity does not include this effect. Hence, these C/E ratios were not used for validating EASY. D-D neutrons did not affect the other activities identified. They are produced by reactions with threshold well above the D-D neutron energy.

The other activities investigated, typically show C/E of about 1.1–1.7, if they are analysed with EASY-2001. These overestimations are generally in agreement with the results obtained in previous benchmark experiments [4]. The reactions by which the activities were produced are of the type (n, charged particle). Their cross-sections at 14 MeV are of the order of 1 mb due to the high Coulomb barrier of W (atomic number 74), i.e. 14 MeV neutrons are just above the reaction threshold. The representation of these parts in the library data has to be improved. This is indicated also by the energy dependence of the C/E values. Significant progress is already achieved with the version EASY-2003.

As the contributions of the several nuclides to the radioactivity of the material are very different, sums of the dose rate produced by the nuclides investigated in the present work are compiled in Table 3 for irradiation conditions of a fusion power plant and three different decay times. The sum of their dose rates comprises about 80–85% of the total dose rate calculated with EASY-2001. Dividing the dose rates calculated for the

Table 2
Results of irradiations No. 1 and 2 (upper part) and of No. 3 (lower part)

Radio-nuclide	Half-life	E_γ (keV)	Reaction Contr. (%)	C/E EASY-2001	C/E EASY-2003	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{181}Hf	42.4 d	482	^{184}W(n,α) 100	1.66	1.29	12	7.5
^{183}Hf	64 min	459 784	^{186}W(n,α) 100	1.33	0.89	18	15
^{182} Ta	114.7 d	1121 1189	^{182}W(n,p) 86.5 ^{183}W(n,d) 6.5	1.60	1.26	52	6.8
^{183}Ta	5.09 d	354	^{183}W(n,p) 64.0 ^{184}W(n,d) 20.7 ^{186}W(n,α)β$^-$ 15.3	1.36	1.29	23	7.8
^{184}Ta	8.7 h	253 318 414 921	^{184}W(n,p) 98.0 ^{186}W(n,t) 2.0	1.34	1.17	10	6.5
^{186}Ta	10.5 min	738	^{186}W(n,p) 100	1.10	1.10	14	16
^{185m}W	1.67 min	132 174	^{186}W(n,2n) 100	0.88	0.88	61	7.2
^{187}W	23.7 h	480 686 773	^{186}W(n,γ) 100	0.61	0.61	50	5.9
^{181}Hf	42.4 d	482	^{184}W(n,α) 100.	1.42	1.54	12	18
^{182}Ta	114.7 d	1121 1189 1221	^{182}W(n,p) 86.5 ^{183}W(n,d) 6.5	1.48	1.14	52	21
^{183}Ta	5.09 d	246 354	^{183}W(n,p) 71.1 ^{184}W(n,d) 15.8 ^{186}W(n,α)β$^-$ 13.1	1.08	1.09	18	9.5
^{187}W	23.7 h	480 618 686 773	^{186}W(n,γ) 100	0.22	0.22	50	6.1

Radionuclides identified, their half-life and the γ-radiation used to determine the activity, the neutron reactions producing these radionuclides, the ratios of calculated-to-experimental activity (C/E), obtained with EASY-2001 and EASY-2003, and the uncertainties of both the calculated (EASY-2001) and the experimental activities.

Table 3
Dose rates produced by the radionuclides investigated (Table 2) after irradiation of W with 14 MeV neutrons of 1.0 MW/m^2 power density for one year at different decay times

Decay time	10 min	1 day	1 year
Calculation EASY-2001 (Sv/h)	$(1.54 \pm 0.51) \times 10^2$	$(1.01 \pm 0.38) \times 10^2$	9.59 ± 4.93
Fraction of total (%)	85.3	81.1	84.8
Experiment (Sv/h)	$(1.15 \pm 0.09) \times 10^2$	$(6.92 \pm 0.47) \times 10^1$	6.00 ± 0.41
C/E EASY-2001	1.34 ± 0.45	1.47 ± 0.56	1.60 ± 0.83
C/E EASY-2003	1.19 ± 0.35	1.17 ± 0.36	1.26 ± 0.38

individual nuclides by the C/E values of Table 2 for the activities, dose rates are obtained that represent an experimental value. The sum of these data is compared with the results of both EASY-2001 and EASY-2003 calculations.

The C/Es obtained with EASY-2003 are closer to unity than those for EASY-2001, and the uncertainty interval is obviously reduced.

4. Concluding remarks

The activation performance of pure W irradiated with fusion peak neutrons was investigated. The activities of radionuclides that are dominant (producing 80–85% of the total γ-ray dose rate) up to the recycling limit of the material calculated for fusion reactor conditions were measured and compared with the corresponding results of calculations with the European Activation System versions EASY-2001 and EASY-2003. An overestimation of the radioactivity induced by the neutrons of up to 60% by EASY-2001 is reduced to about 25% by EASY-2003, indicating the level of validation for this range of decay time.

The hands-on-limit is determined by radionuclides mainly produced by the reactions $^{182}\mathrm{W}(n,n'\alpha)^{178}\mathrm{Hf}^*$ and $^{180}\mathrm{W}(\mathrm{n,2n})\ \beta^+ \rightarrow {}^{179}\mathrm{Ta}$ that should be further investigated.

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References

[1] G. Marbach, I. Cook, Fus. Eng. Des. 46 (1999) 243.
[2] E.T. Cheng et al., Fus. Technol. 34 (1998) 721.
[3] R.A. Forrest, J.-Ch. Sublet, Reports Culham Science Centre, UKAEA FUS 450 and UKAEA FUS 451, April 2001; UKAEA FUS 484 and UKAEA FUS 485, December 2002.
[4] R.A. Forrest et al., Fus. Eng. Des. 63&64 (2002) 295.
[5] V.D. Kovalchuk et al., Report Russian Research Centre Kurchatov Institute, IAE- 5589/8, Moscow, 1992.
[6] K. Seidel et al., Report Technische Universität Dresden, TUD-IKTP/02-01, April 2002.