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Section 13. Safety, transmutation, activation and waste management issues

Measurement and analysis of radioactivity induced in steels and a vanadium alloy by 14-MeV neutrons

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Abstract

Samples of the structural material of the International Thermonuclear Experimental Reactor (SS316), of the low-activation steels MANET and F82H, and of the vanadium alloy V4Ti4Cr were irradiated with D–T fusion neutrons. The radioactivities induced were determined after irradiation several times during decay by γ-spectroscopy. The results were analysed with the European Activation System (EASY-97). In order to validate EASY-97, the total activities of the samples are compared, and ratios of calculated-to-experimental values for the individual activities are derived and discussed.

1. Introduction

The attractiveness of fusion power with respect to safety and environmental aspects is connected with the possibility of using fusion reactor materials with low-activation characteristics, i.e. with short half-lives and low total activity of the radionuclides produced by the fusion neutrons in first wall, blanket, vacuum vessel and other components of the device. The radioactivity induced in materials by neutrons is calculated with code and data library packages such as the European Activation System (EASY) [1], where the elemental composition of the material and the spectral neutron flux are used as input, and activity (as well as nuclear heat and dose rate) as a function of time during and after irradiation are the output. EASY, which has been adopted as a reference system for the design of the International Thermonuclear Experimental Reactor (ITER), consists of the inventory code FISPACT and the European Activation File (EAF) which contains large sets of reaction cross-section data, decay data and other related files.

The aim of the present paper is the experimental validation of EASY calculations for steels irradiated with D–T fusion neutrons. Samples of the stainless steel 316 (AISI 316LN(IG)), which is the structural material of ITER, and of the low-activation steels MANET and F82H are used for benchmarking. As vanadium alloys are potential structural materials, because of their low-activation behaviour, V4Ti4Cr samples are also included.

The work is complementary to irradiations of the same materials at FZ Karlsruhe with a spectrum of fast neutrons [2] and to activation data benchmarks with pure element samples at FNS in the framework of a USDOE–JAERI collaborative programme [3].

2. Experiment

The compositions of the materials used are given in Table 1. Samples of materials with masses of 0.6–1.3 g and with an area of 10 mm × 10 mm were activated at the high-intensity neutron generator SNEG-13 [4] at Sergiev Posad. The geometrical relations are outlined in

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Table 1
Elemental composition of the samples in wt%

	MANET	SS316	F82H	V4Ti4Cr
B	0.0089	0.0025		
C	0.11	0.031	0.089	0.00187
N	0.03	0.06	0.0043	0.00234
O			0.0125	0.00975
Al	0.006		0.0022	0.00833
Si	0.27	0.39		0.0449
P	0.005	0.023		
S	0.004	0.002		
Ti			0.0008	3.92
V	0.20	0.09	0.159	92.15306
Cr	10.3	17.7	7.70	3.77
Mn	0.94	1.77	0.159	
Fe	86.7629	65.3485	89.65625	0.0245
Co	0.006	0.09	0.0026	
Ni	0.62	12.1	0.020	
Cu	0.007	0.09	0.0054	0.00625
As	0.010			
Zr	0.009			
Nb	0.15		0.00035	
Mo	0.56	2.29	0.0046	0.0590
Sn	0.001			
Sb	0.0002			
Ta		0.01	0.014	
W			2.17	
Pb		0.003		

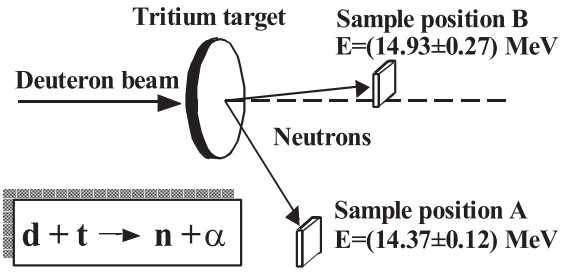

Fig. 1. Irradiation geometry.

Fig. 1. Each material was irradiated at two positions, at an angle of 4° with respect to the deuteron beam direction, where the mean neutron energy is 14.93 MeV with a spread of 0.54 MeV fwhm, and at 73° with (14.37 ± 0.12)-MeV neutrons. The 14-MeV neutron flux was monitored by ^{93}Nb(n,2n) activation with thin Nb foils attached in front and at the back of the sample. The fluences applied were of the order of 10^{14} neutrons/cm^2. The background component of thermal and intermediate-energy neutrons was measured by ^{115}In(n,γ)-activation at the sample positions and found to be 10^{-5} of the total flux.

Gamma spectra were taken from the samples with Ge(Li)-spectrometers several times during decay from 4 min up to about 500 d after irradiation. The activities of the nuclides ^{24}Na, ^{48}Sc, ^{51}Cr, ^{54}Mn, ^{56}Mn, ^{59}Fe, ^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co, ^{57}Ni, ^{89}Zr, ^{95}Nb, ^{96}Nb, ^{99}Mo, ^{182}Ta, ^{183}Ta and ^{187}W, identified by gamma energies and half-life, were determined for the steels. They produce after 8 h of decay more than 99% of the nuclear heat and dose rate. After one year of decay they are the origin of more than 90% of the heat and more than 80% of the total dose rate. In the vanadium alloy the activities of ^{24}Na, ^{27}Mg, ^{28}Al, ^{46}Sc, ^{47}Sc, ^{48}Sc, ^{45}Ti, ^{51}Ti, ^{52}V, ^{51}Cr, ^{54}Mn, ^{57}Co, ^{58}Co, ^{92m}Nb and ^{99}Mo were measured. These nuclides produce after 3 min of decay more than 99% of the total heat and gamma dose rate, and after one year of decay, when the activity of the V4Ti4Cr sample is further decreased by almost six orders of magnitude, they are the origin of about 92% of the heat and of more than 99% of the gamma dose rate.

Calculations with EASY, as released in 1997, were carried out for each of the investigated samples taking into account the history of irradiation and using the chemical composition of the material as determined by X-ray fluorescence analysis (Table 1). A 175-group cross-section library (VITAMIN-J structure) produced with a flat micro-flux weighting spectrum was used to avoid falsifying influences of a weighting spectrum with a fusion peak at 14.1 MeV for reactions with thresholds in the 14-MeV region [5], and it was collapsed with the neutron flux spectrum at the sample positions.

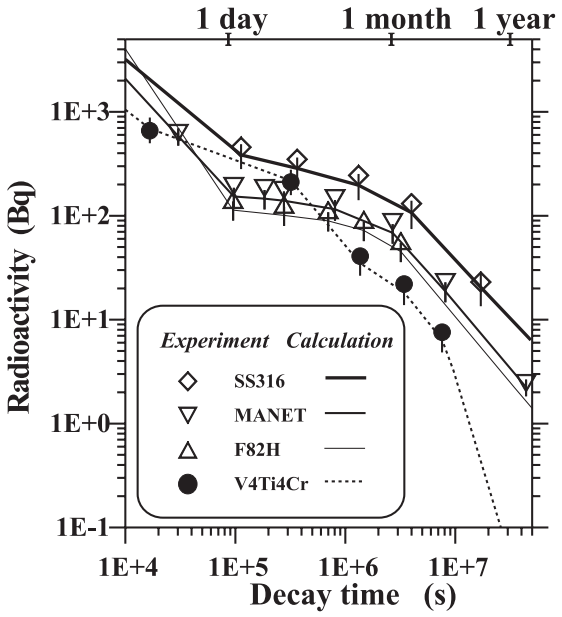

Fig. 2. Total gamma activity of steels and of the vanadium alloy V4Ti4Cr, both at sample position B, normalised to a sample mass of 1 g and to a neutron fluence of 1×10^{12} neutrons/cm^2. The error bars represent the uncertainties of the calculated activities. The experimental uncertainties are smaller than the symbol size.

3. Results

The total gamma activities calculated for the four materials in sample position B are compared in Fig. 2 with the sum of the experimentally determined activities, after normalisation to a sample mass of 1.0 g and a neutron fluence of 1.0×10^{12} cm^{-2}, for those decay times at which γ-spectra were measured. The activities of the steels show similar values for decay times up to about one day. For larger times the expected low-activation behaviour of MANET and of F82H compared to the ITER SS316 is confirmed by the experimental data. The gamma activity of the vanadium alloy, having a shorter effective half-life, is more than one order of magnitude lower than the activity of the steels after one year of decay.

The uncertainties in the calculated activities, which include both cross-section and half-life uncertainties, are shown in Fig. 2 by error bars. A typical value is 30%. All of the experimental points are within or very close to these bars, and a general validation of EASY-97 for activation of steels by 14-MeV neutrons can be concluded. More detailed results for vanadium alloys are presented in [7].

However, there are compensations of over- and underestimated activities in the total sample activity. The

Table 2
Radionuclides in steel, their half-life, the relative contribution of various nuclear reactions and the ratio of C/E activity obtained including experimental uncertainties for sample position A (average neutron energy $\langle E\rangle = 14.37$ MeV) and sample position B (average neutron energy $\langle E\rangle = 14.93$ MeV)

Radionuclide	Half-life	Reaction	$\langle E\rangle = 14.37$ MeV		$\langle E\rangle = 14.93$ MeV	
			Contr. (%)	C/E	Contr. (%)	C/E
^{24}Na	14.96 h	^{27}Al(n,α)	69.1	0.75 ± 0.13	69.1	0.87 ± 0.16
		^{27}Al(n,α)m	30.9		30.9	
^{48}Sc	43.67 h	^{51}V(n,α)	98.5	0.88 ± 0.09	98.6	0.91 ± 0.10
		^{48}Ti(n,p)	1.5		1.4	
^{51}Cr in F82H	27.7 d	^{52}Cr(n,2n)	83.9	0.69 ± 0.04	86.7	0.78 ± 0.04
		^{54}Fe(n,α)	16.1		13.3	
^{54}Mn in MANET	312.2 d	^{54}Fe(n,p)	69.1	0.96 ± 0.05	63.7	0.92 ± 0.04
		^{55}Mn(n,2n)	30.8		35.8	
^{56}Mn	2.58 h	^{56}Fe(n,p)	99.5	1.04 ± 0.05	99.0	1.00 ± 0.05
		^{57}Fe(n,d)	0.5		1.0	
^{59}Fe in SS316	44.5 d	^{62}Ni(n,α)	62.2	0.64 ± 0.07	64.9	0.63 ± 0.08
		^{59}Co(n,p)	36.4		33.7	
		^{58}Fe(n,γ)	1.3		1.4	
^{56}Co in F82H	77.26 d	^{56}Fe(p,n)	100	1.72 ± 0.10	99.8	2.11 ± 0.17
^{57}Co	271.8 d	^{58}Ni(n,d)	99.7	0.80 ± 0.04	99.6	0.89 ± 0.07
^{58}Co	70.86 d	^{58}Ni(n,p)	77.0	0.97 ± 0.07	75.8	0.90 ± 0.05
		^{58}Ni(n,p)m	20.6		21.0	
		^{59}Co(n,2n)	1.9		2.5	
		^{59}Co(n,2n)m	–		0.6	
^{60}Co	5.272 a	^{60}Ni(n,p)	52.6	0.90 ± 0.07	52.1	0.86 ± 0.07
		^{60}Ni(n,p)m	45.5		43.0	
		^{61}Ni(n,d)	1.3		2.3	
^{57}Ni	36.0 h	^{58}Ni(n,2n)	100	0.96 ± 0.07	100	0.96 ± 0.07
^{89}Zr	78.4 h	^{90}Zr(n,2n)	53.4	0.62 ± 0.06	51.4	0.83 ± 0.07
		^{90}Zr(n,2n)m	10.0		13.2	
		^{92}Mo(n,α)	26.0		25.6	
		^{92}Mo(n,α)m	10.6		9.8	
^{95}Nb	34.97 d	^{95}Mo(n,p)	79.3	0.97 ± 0.09	71.9	1.15 ± 0.10
		^{96}Mo(n,d)	18.0		25.0	
^{96}Nb	23.4 h	^{96}Mo(n,p)	97.2	0.82 ± 0.08	95.4	1.26 ± 0.12
		^{97}Mo(n,d)	2.8		4.5	
^{99}Mo	66.0 h	^{100}Mo(n,2n)	99.6	0.93 ± 0.07	100	0.93 ± 0.07
^{182}Ta	114.4 d	^{182}W(n,p)	78.9	0.76 ± 0.31	79.8	0.76 ± 0.25
		^{183}W(n,d)	2.8		2.7	
^{183}Ta	5.0 d	^{183}W(n,p)	69.9	1.02 ± 0.52	62.1	1.26 ± 0.55
		^{184}W(n,d)	15.0		20.2	
		^{186}W(n,α)β^-	15.1		18.0	
^{187}W	23.72 h	^{186}W(n,γ)	100	0.64 ± 0.06	100	0.93 ± 0.08

ratios of calculated-to-experimental values (C/E) for the activities analysed in the steels are shown in Table 2 for both sample positions. They represent the average of the C/E obtained at several times for the three samples. The uncertainty of the C/E values includes the total experimental uncertainties only, but not the calculated ones. The former are estimated on the basis of the statistical and systematic errors of γ-spectroscopy, of neutron flux monitoring via ^{95}Nb(n,2n), of the uncertainties of the γ-yield data and of the elemental content of the sample. Usually they are smaller than the uncertainties of the calculated activities, and may be used for improving the calculation tools.

The largest overestimation is obtained for ^{56}Co, which is produced in a two-step process: protons from various (n,p)- and (n,pn)-reactions have sufficient energy for (p,n)-reactions on ^{56}Fe. It was calculated with the model for the treatment of sequential charged particle reactions developed at Karlsruhe [6].

The largest underestimation is observed for ^{59}Fe. About two thirds of ^{59}Fe is produced by the ^{62}Ni(n,α)-reaction, one third by the ^{59}Co(n,p)-reaction. The evaluated cross-section of the ^{62}Ni(n,α)-reaction was enhanced by 40% in the version of the EAF as released in 1999 (EAF-99, [8]) in comparison to EAF-97. When applying this correction, the C/E value will increase. The reaction cross-section of the ^{59}Co(n,p)-reaction as given by EAF-97 describes experimental cross-sections in the neutron energy region between 13 and 15 MeV rather well, but with the tendency of a small underestimation.

As shown in Fig. 2, there is a weak tendency of underestimation of the total gamma activity for decay times greater than one day. This underestimation is mainly due to ^{51}Cr.

The comparison of C/E obtained for the two neutron energies reveals some threshold effects of reaction cross-sections, especially for (n,2n)-, (n,np)- and (n,d)-reactions.

4. Concluding remarks

The EASY, version EASY-97, has been validated with an uncertainty range of typically $\pm 30\%$ for the calculation of radioactivity profiles induced by 14-MeV neutrons in steels for decay time ranges between minutes and about one year. The materials used are candidates for fusion reactor structures. For further improvements, the cross-sections of reactions producing activities with larger deviations in C/E values should be checked in detail.

The radioactivity of the vanadium alloy V4Ti4Cr is significantly lower than the radioactivity of the investigated steels after cooling times greater than one month.

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