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Section 6. Low activation materials and other structural materials. 1. Ferritic steels

Improved activation tests of fusion structural materials with a deuteron–beryllium neutron source

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Abstract

In the framework of the European Activation File (EAF) task of the European Fusion Technology Programme, the EAF data relevant to the steel types F82H-mod and MANET-2 were benchmarked experimentally. Samples were activated in an intense neutron field produced in a thick beryllium target by a 19-MeV deuteron beam of the Karlsruhe Isochron-Zyklotron. The irradiated samples were investigated by γ spectrometry. The results were compared with calculations using the FISPACT code, the EAF-4.1 library and the source neutron spectrum as measured by activation foil dosimetry. Calculation-over-experiment ratios between 0.96 and 1.19 are found for the more important medium-halflife radionuclides produced in F82H-mod. Similar values are found for MANET-2.

1. Introduction

The FISPACT code [1] together with the European Activation File (EAF) [2] is the European reference software for calculating the neutron-induced activation of fusion reactor materials. Experimental verifications (benchmark tests) of such results can be performed with either a 14-MeV neutron source or a 'white' source, and either on pure constituent elements or on the actual structural materials (alloys or composites). Tests with a white source on actual structural materials are more or less analogous to the 'integral' experiments used in benchmarking nuclear data for neutron transport.

In the present work, a deuteron–beryllium neutron source employing a 19-MeV deuteron beam of the Karlsruhe isochronous cyclotron is used. It produces a neutron flux density up to about $3 \times 10^{11}/\text{cm}^2/\text{s}$ averaged over a 1 cm^2 sample (higher on smaller samples). Since the neutrons are released mainly by the 'direct', non-resonant processes of deuteron stripping and deuteron breakup, their energy spectrum is continuous and very different from a deuterium–tritium fusion spectrum (see below). This is considered an advantage because, together with similar experiments using deuteron–tritium sources, it should result in a broader data base for validating the evaluated activation cross sections. The d+Be neutron emission is strongly forward peaked, which is advantageous for irradiating small, flat samples. Since the d+Be neutron spectrum is relatively difficult to calculate, it requires to be measured in order to fully exploit the information from the irradiated samples

2. Activation experiment

Fig. 1 shows schematically the target with an irradiation sample in place. It is an 'internal' target, inserted into the cyclotron on a radial position corresponding to the selected deuteron energy. The typical sample size is 10 mm × 10 mm × 1 mm. The irradiation time was 85 h for the F82H-mod sample and 10 h for the MANET-2 sample. Fluence monitor foils such as Nb or Al, customary in d–T neutron irradiations, were not used, because their activity induced by the d–Be spectrum

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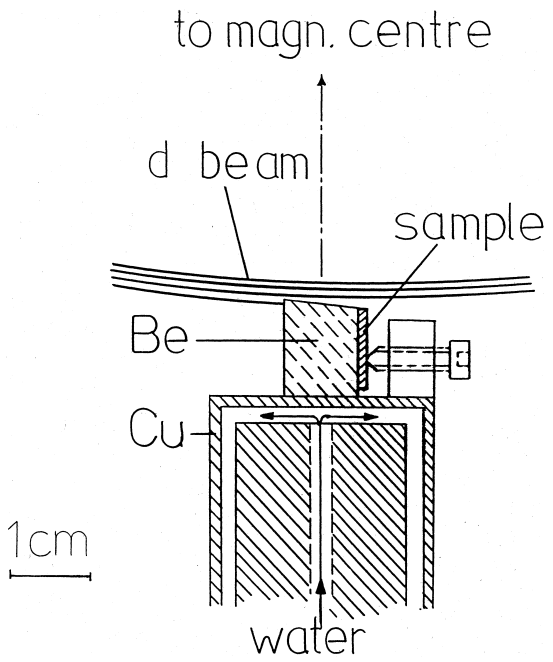

Fig. 1. Beam, target and irradiation sample.

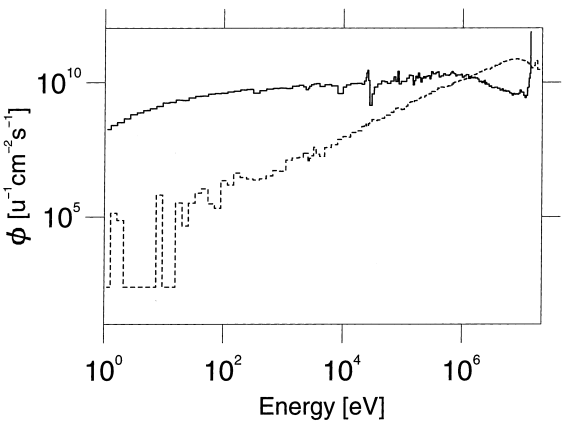

Fig. 2. DEMO (full line) and d + Be (dashed) neutron spectra (Neutrons per unit lethargy, cm^2 and s).

cannot readily be interpreted. For the F82H-mod irradiation, the absolute fluence per μC of beam charge was extracted from the measurement of the neutron spectrum (see below). The current in the steel sample activations was 10 μA, yielding 1.6×10^{11} neutrons/cm^2/s averaged over the sample. Corrections for some beam interruptions during the irradiations (relevant only for halflifes not much longer than the irradiation) were calculated and applied.

The γ activity of the samples was measured repeatedly after different cooling times using a 150 cm^3 high-purity germanium detector with a personal-computer based multichannel analyzer. An energy range of either 0–2 or 0–4 MeV was resolved into 4096 channels. The system was calibrated for both energy and efficiency using several calibrated γ ray sources. The samples or sources were placed 93 mm or more from the detector face, which corresponds to a solid angle of $\leqslant 0.26$ sr.

3. Neutron spectrum

The d + Be neutron spectrum for use in the FISPACT calculations was obtained experimentally by activation foil dosimetry. The reaction rates of 22 different activation reactions were measured by irradiation in the same arrangement as the steel samples, and subsequent γ spectrometry. The neutron spectrum was unfolded from these rates using the SAND-II code [3]. The a-priori guess spectrum required as input to the unfolding procedure was obtained by Monte-Carlo calculations using the MCNP-4A code [4] together with the McDeLi neutron source routine [5]. These calculations also served for scaling the result from the 5 mm × 5 mm dosimetry foils to the 10 mm × 10 mm steel samples, in view of the pronounced anisotropy of the neutron source (see above). The unfolded and scaled spectrum is shown in Fig. 2, together with a typical spectrum of the DEMO reactor for comparison.

4. Gamma spectra analysis

The spectra were analyzed automatically, using the same commercial software system (ORTEC 'GammaVision') which also serves for data acquisition and calibrations. The ESMASTER library of decay data (halflives, line energies, branching ratios etc.) used by this software is not identical with EAF decay data. However, at the present level of precision, this is a comparably minor source of systematic errors. Similarly, coincidence-loss corrections for nuclides emitting multiple γ rays should not be important with the detection geometry used (see above). Only γ lines above 100 keV were taken into account, so that γ self-absorption in the 1-mm thick samples is also small, and γ-induced X rays from the lead shield are not disturbing.

In the automatic analyses, some erroneous nuclide assignments were found, that could be clarified by observing the decay of spectral lines between subsequent measurements.

5. Calculations

The calculations were performed with the FISPACT-4.1/00 code and EAF version 4.1(0) in the well known

VITAMIN-J 175 energy group scheme. The steel compositions used are given in Tables 1 and 2. The F82H-mod composition was averaged from analyses made at Karlsruhe on five pieces from the same batch as the irradiated sample. The minute concentrations of Nb and Mo in F82H-mod were measured by a dedicated activation experiment [6]. The MANET-2 composition was taken from the manufacturer's certificate for the production batch [7].

6. Results and discussion

Table 3 gives the results for F82H-mod in the form of calculation/experiment ratios at two different cooling times.

Table 4 gives the results for MANET-2, also at two cooling times. The MANET-2 results can only be given as relative values, arbitrarily normalized to Mn-54. The absolute fluence for this irradiation, in which a somewhat different beryllium target was used, cannot be extracted from the neutron spectrum measurement. Also the spectral shape may differ slightly from the one measured with the standard target; this may explain the generally larger deviations from $C/E = 1$.

The tables contain all the radionuclides found experimentally with reasonable confidence, with the following exceptions:

1. Nuclides erroneously assigned to γ lines by the analysis software, as indicated by a time decay not fitting the known halflife,
2. nuclides above a mass number of $A = 150$,
3. Sc-48 and V-48, which decay to the same daughter nuclide so that their more important γ lines are identical,
4. Mn-52 and Co-56. For these, sequential charged particle reactions are probably important, which could not be taken into account with the FISPACT version used.

Each of the omitted nuclides is $\leqslant 1\%$ of the sample activity at the cooling times considered, with the possible exception of activation products around $A = 180$ from the 2% W content of F82H-mod. These are probably more important and are seen in the γ spectra. However, their clean analysis requires additional work because of low and partially duplicate γ line energies.

For any nuclide in Tables 3 and 4 that constitutes $\geqslant 0.1\%$ of the sample activity at the respective cooling time, C/E is between 0.96 and 1.19 for F82H-mod, and between 0.68 and 1.28 for MANET-2. We conclude from this that the EAF-4.1 library is reasonably well suited to predict the induced activity in the investigated steels for the range of cooling times considered here.

The most long lived nuclide detected in the present analyses is Co-60, which is known to dominate the activity of steels at cooling times of about 10–100 years. In Tables 3 and 4, the C/E for Co-60 are below unity and decrease with increasing cooling time. This indicates a possible systematic experimental error, caused by a Co-60 calibration source stored with insufficient shielding near the spectrometer, which may have introduced an unknown Co-60 background in the different measurements. The steel samples will be remeasured under cleaner conditions for this important nuclide.

Table 1
Composition of F82H-mod steel

Element	*w*/0
Fe	89.6
Cr	7.8
W	2.0
Mn	0.16
V	0.16
C	0.09
Ni	0.02
Ta	0.016
O	0.012
N	0.006
Cu	0.006
Co	0.002
Al	0.002
Mo	0.005
Ti	0.001
Nb	0.00025

Table 2
Composition of MANET-2 steel

Element	*w*/0
Fe	86.8
Cr	10.3
Mn	0.94
Ni	0.62
Mo	0.56
Si	0.27
V	0.20
Nb	0.15
C	0.11
N	0.03
As	0.01
Zr	0.009
B	0.0089
Cu	0.007
Al	0.006
Co	0.006
P	0.005
S	0.004
Sn	0.001
Sb	0.0002

Table 3
Calculation/experiment ratios *C/E* for F82H-mod

Nuclide	$T_{1/2}$	$T_c = 181$ h		$T_c = 1263$ h	
		f_a %	*C/E*	f_a %	*C/E*
Cr-51	28d	53.3	1.19	29.2	1.10
Mn-54	312d	18.1	1.05	27.7	1.02
Fe-59	44d	0.03	0.85	0.02	0.79
Co-58	71d	0.22	1.02	0.25	0.96
Co-60	5.3a	*	0.85	0.0008	0.77
Ni-57	36h	*	1.12	**	– [b]
Zr-89	78h	*	0.41	**	– [a]
Nb-92m	10d	0.01	1.91	0.001	– [a]
Mo-99	66h	0.01	2.45	**	– [a]

$T_{1/2}$: Halflife; T_c: Cooling time; f_a: Fraction of total sample activity at T_c.
* <0.02%.
** <0.0008%.
[a] Below exptl. threshold.
[b] Not given by FISPACT.

Table 4
Calculation/experiment ratios *C/E* for MANET-2, normalized to Mn-54

Nuclide	$T_{1/2}$	$T_c = 87$ h		$T_c = 676$ h	
		f_a %	$(C/E)_{norm}$	f_a %	$(C/E)_{norm}$
Cr-51	28d	53.5	1.02	47.4	1.28
Mn-54	312d	13.4	(1.00)	20.7	(1.00)
Fe-59	44d	*	0.68	0.03	0.70
Co-57	272d	0.4	0.72	0.06	0.87
Co-58	71d	5.2	0.98	6.7	0.82
Co-60	5.3a	*	0.52	0.02	0.13
Zr-89	78h	0.2	1.08	**	– [a]
Zr-95	64d	*	0.41	0.008	– [a]
Nb-92m	10d	2.7	1.22	0.8	1.25
Nb-95m	87h	0.05	1.02	**	– [a]
Mo-99	66h	3.9	0.68	0.01	– [a]

$T_{1/2}$: Halflife; T_c: Cooling time; f_a: Fraction of total sample activity at T_c.
* <0.3%. ** <0.008%. [a] Below exptl. threshold.

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