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Measurement and analysis of radioactivity induced in CuCrZr by D–T neutrons

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Abstract

CuCrZr is used in high heat flux components of ITER and other fusion reactor designs. A CuCrZr alloy of the European Fusion Technology Programme was irradiated with D–T neutrons, and the γ-activities of all nuclides which are relevant up to the recycling limit of the material were measured. The results were analysed with the European Activation System (versions EASY-2001 and EASY-2003). The calculated total activation property of the material was validated within 10%. The long-term radioactivity was estimated to be well below the hands-on limit with an uncertainty of 19%.

1. Introduction

CuCrZr alloys are candidate materials for the heat sink in the first wall of the blanket and in the divertor of fusion devices [1]. The radioactivity in a fusion reactor is mainly produced by fast neutrons of the 14 MeV fusion peak, where the number of open reaction channels is a maximum and by thermal neutrons because for some reactions the cross-section is large in this energy region. In the present work the γ-ray activities induced by the 14 MeV neutrons in this near-plasma material were investigated.

Generally, the radioactivity of the material is calculated with inventory codes and nuclear data libraries such as the European Activation System (EASY) [2]. The reliability of the calculated results depends on the quality of both the nuclear data (cross-section data of the relevant reactions and decay data of the produced nuclides) and the analysis of the elemental composition. The CuCrZr alloy investigated is a material used in the European Fusion Technology Programme. The constituents of the alloy were quantified before the irradiation by two different procedures. In a first step the contents of the major alloying elements Cr and Zr were determined by Optical Emission Spectrometry with Inductively Coupled Plasma Source (ICP-OES) as accurately as possible so as to check the relevant EASY cross-sections for neutron reactions on these nuclides. For this purpose the alloy was dissolved in acid and analyzed with calibration solutions matching the sample solutions in acid and copper contents. The results are presented with their uncertainties in the upper part of Table 1. In a second step the CuCrZr alloy was examined also by ICP-OES for another 42 metallic impurities, but with the use of only one unmatched multielement calibration solution. Therefore the results of the analyses (Table 1) include a relative uncertainty of ±20%. The copper content has been calculated by difference. Its uncertainty given in Table 1 takes into account the uncertainties of

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Table 1
Elemental composition of the CuCrZr samples

Element	Weight%
Cu	**98.6 ± 0.1**
Cr	**0.754 ± 0.006**
Zr	**0.163 ± 0.003**
Li	<0.0005
B	<0.0005
Na	**0.0004**
Mg	<0.0005
Al	**0.0059**
Si	<0.01
P	<0.003
K	<0.001
Ca	**0.0012**
Ti	**0.0002**
V	<0.0001
Mn	**0.0001**
Fe	**0.0088**
Co	<0.00002
Ni	<0.0001
Zn	<0.0001
As	<0.001
Se	<0.0005
Rb	<0.0005
Sr	<0.00001
Nb	<0.0002
Mo	<0.0001
Ru	<0.0001
Rh	<0.0005
Pd	<0.001
Ag	<0.005
Cd	<0.00002
Sn	**0.0053**
Sb	<0.0005
Te	<0.0005
Cs	<0.4
Ba	<0.00001
Hf	<0.0001
Ta	<0.0001
W	**0.0062**
Re	<0.0005
Os	<0.0005
Ir	<0.001
Pt	<0.0001
Au	<0.045
Pb	<0.007
Bi	<0.0015

the Cr and Zr content as well as 20% of the upper limit of the metallic impurities.

For planning the irradiation experiment, a calculation was made with EASY. The material was assumed to be irradiated at reactor conditions, i.e. with a flux density of the 14 MeV neutrons corresponding to a power density of 1.0 MW/m^2, for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation are shown in Fig. 1

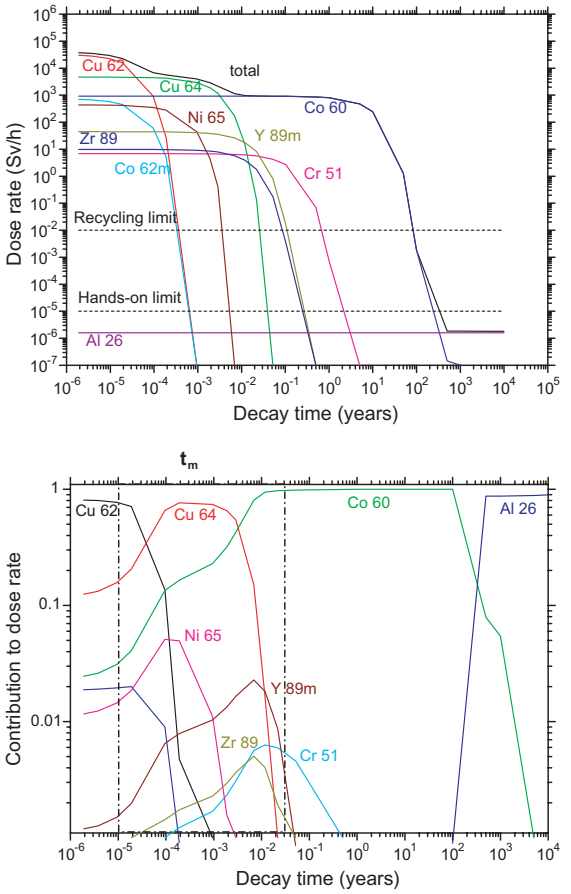

Fig. 1. Calculated contact dose rate (upper part) and contribution of the different radionuclides to the total dose rate (lower part) after irradiation of CuCrZr with fusion peak neutrons of 1.0 MW/m^2 power density for one year as a function of the decay time.

for a range up to 10^4 years. The safety-related short-term radioactivity is expected to be determined by the ^{62}Cu and ^{64}Cu isotopes. The recycling limit and the hands-on-limit of the material depend on the ^{60}Co activity. The long-term radioactivity which is important for the waste management mainly depends on the ^{26}Al activity. It is produced by the ^{27}Al(n,2n)^{26}Al reaction. Its dose rate is well below the hands-on-limit, if the Al impurity is as small as in Table 1.

Fig. 1 shows, that measurements of the γ-activities in the range of decay time labelled by t_m permit testing the calculated radioactivity induced by 14 MeV neutrons up to the recycling limit. Together with all available recent experimental data, the results will contribute to the validation process of the EASY data [3]. Furthermore, the irradiation experiment may be used for analyzing the nuclide content of some of the impurities.

2. Experiment

The irradiation of the CuCrZr alloy was performed at a D–T neutron generator of TUD. The spectral distribution of the neutron flux at the sample position (Fig. 2) was calculated with the Monte Carlo code MCNP [4] taking into account the kinematics of the D–T reaction for thick target conditions and neutron interactions with target backing, coolant and structure. The mean neutron energy of the fusion peak is $\langle E_n \rangle = 14.76$ MeV, the full width of the peak at half maximum is $\Delta E_n = \pm 0.40$ MeV. During an irradiation time of 203.0 min a total neutron fluence of 2.1×10^{12} cm^{-2} was applied. This was measured by simultaneous activation of niobium foils and evaluating the activity induced by the ^{93}Nb(n,2n)^{92m}Nb reaction. The cross-section of this reaction was considered to be constant in the range of 14–15 MeV with a value of 464 mb and an uncertainty of 4.2%. A possible background component of thermal neutrons was checked by ^{197}Au(n,γ) activation using thin foils. The ratio of thermal to 14 MeV neutrons was found to be less than 10^{-5}. The CuCrZr sample had a thickness of 1.02 mm, a front area of 98.47 mm^2 and a mass of 0.8334 g. The attenuation of the neutron and γ-ray fluxes in the sample as well as the geometry (source-sample and sample-detector) were determined by 3D calculations with MCNP.

Gamma-ray spectra were taken with an HPGe-spectrometer at a distance of 50 mm between irradiated sample and crystal at several decay times in the range labelled with t_m in Fig. 1. The γ-activities identified by energy and half-life were used to determine the nuclide activities using γ-yield data from JEF2.2 [5].

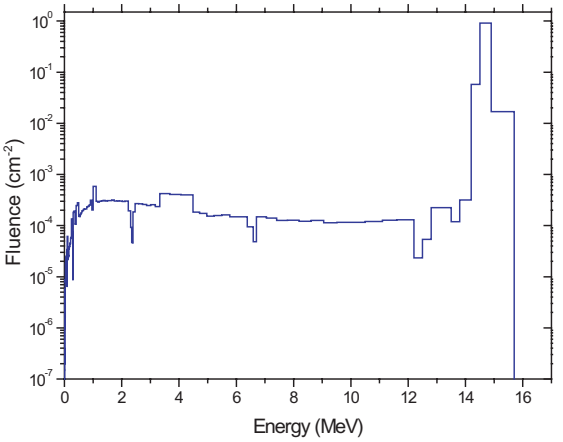

Fig. 2. Spectral distribution of the neutron flux at the sample position normalized to one source neutron and plotted as group fluence in Vitamin-J structure.

3. Results

The measured activities were analysed with EASY-2001 and the recent version EASY-2003 of the European Activation System [2]. In the calculations the sample was assumed to consist of CuCrZr as presented in Table 1. The impurities were taken into account at their upper limit, with the exception of the Cs content (see below).

The results of the analysis for individual activities are presented in Table 2. The uncertainties of the calculated activities ($\Delta C/C$) include both cross-section and half-life errors, as estimated by EASY. The uncertainties of the experimental values (E/E) take into account possible errors of the γ-activity measurements (statistical uncertainty of the γ-counting, the uncertainty of the efficiency calibration of the spectrometer including the geometry factor), of the sample mass and elemental composition, of the γ-yield data and of the neutron flux monitoring.

The radionuclides of Table 2 are each produced by one dominant reaction. Ratios of calculated-to-experimental activity (C/E) that deviate from unity by more than 50% and that are outside the experimental error bars were obtained with EASY-2001 for ^{61}Co and ^{62m}Co. The validation process of the EASY cross-section data [3] results in better agreement with the measured activity of ^{61}Co for EASY-2003, whereas the discrepancy for ^{62m}Co remains unchanged. For the other activities, C and E agree within the total uncertainties. Smaller $\Delta E/E$ values than the $\Delta C/C$ in some cases indicate possibilities for further improvements in the EASY data base.

The ^{89}Zr activity was determined by measuring the 909.2 keV γ-line intensity. This line is emitted by the isomeric transition ^{89m}Y (IT) ^{89}Y which follows the (β^-)-decay of ^{89}Zr and has a half-life of $t_{1/2} = 16$ s. The intensity of this γ-line was measured at decay times larger than 1 h, when the decay rates of ^{89}Zr and of ^{89m}Y are in equilibrium, and contributions from any direct production of ^{89m}Y by neutron reactions could be neglected. However, in dose rate calculations from EASY the 909.2 keV γ-rays are attributed to ^{89m}Y, and ^{89m}Y appears in Fig. 1 with a significant contribution.

The long-term radioactivity is dominated by the ^{26}Al activity produced mainly by the ^{27}Al(n,2n) reaction. The content of the Al impurity as given in Table 1 was checked by measuring the ^{24}Na activity, produced by the ^{27}Al(n,α) reaction, with 1368.63 keV γ-ray line. The Al-content was found to be less than 0.006 wt% with an uncertainty of 17.5%. The cross-section of the ^{27}Al(n,2n) reaction, given in EASY-2001 with an uncertainty of 46%, was recently measured by Accelerator Mass Spectroscopy with an uncertainty of only 5% [6]. The cross-section value presented in Ref. [6] is smaller than the EAF-2001 value at 14 MeV by a factor of 0.86, resulting in the value for the dose rate of ^{26}Al being smaller by this factor than that given in Fig. 1, with a

Table 2
Results obtained for the activity of radionuclides

Radio-nuclide	Half-life	E_γ (keV)	Reaction	Contribution (%)	C/E EASY-2001	C/E EASY-2003	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{51}Cr	27.7 d	320.2	^{52}Cr(n,2n)	100	1.16	1.16	5.0	10.8
^{60}Co	5.27 yr	1332.5 1173.2	^{63}Cu(n,α)	100	1.14	1.12	48.6	9.7
^{61}Co	1.65 h	320.2	^{65}Cu(n,n′α)	100	1.73	1.09	20.0	9.1
^{62m}Co	13.9 min	1163 2004	^{65}Cu(n,α)	100	1.53	1.53	62.0	11.4
^{65}Ni	2.5 h	1481.8 1116.1 366.5	^{65}Cu(n,p)	100	1.19	1.19	10.0	7.4
^{62}Cu	9.74 min	1163 875.7	^{63}Cu(n,2n)	100	1.09	1.09	5.0	11.3
^{64}Cu	12.71 h	1345.9	^{65}Cu(n,2n) ^{63}Cu(n,γ)	99.4 0.6	1.15	1.15	5.0	34.8
^{89}Zr	3.27 d	909.2	^{90}Zr(n,2n)	100	1.12	1.12	51.0	10.3

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.

total uncertainty of 19%, taking into account the uncertainty of the measurement, the impurity content, the cross-section and the gamma yield data. This confirms the long-term dose rate level is well below the hands-on limit.

The impurity content of Cs given in Table 1 as <0.4 wt% was checked by the intensity of the 667.08 keV γ-line from ^{132}Cs produced by the ^{133}Cs(n,2n) reaction. An upper limit of the Cs content of 0.014 wt% with an experimental uncertainty of 20% was found. With this value, no significant contribution of activities produced from Cs to the total radioactivity of the material is expected.

Since the contribution of the radionuclides of Table 2 to the total activity and the total dose rate of the irradiated material is very different, the sum of the measured activities (forming more than 98% of the total activity) at one decay time is compared in Table 3 with the corresponding calculated value. The deviation of 10% from unity is well within the uncertainty range. Furthermore, the C/E obtained for the individual activities were used to determine an experimental sum of the dose rate for checking the calculated value. Also the calculated and experimental dose rates agree within 10% to unity. For other decay times up to the recycling limit the activity and the dose rate becomes more and more dominated by ^{64}Cu and ^{60}Co, with the deviation from unity rising up to about 15%.

Table 3
Comparison of the experimental and calculated (EASY-2001) results for the sums of the radioactivity produced by the nuclides of Table 2 at a decay time of 6 min

	Activity (Bq)	Dose rate (Sv/h)
Calculation	$(3.79 \pm 0.16) \times 10^5$	6.44×10^{-7}
Experiment	$(3.44 \pm 0.61) \times 10^5$	5.84×10^{-7}
C/E	1.10 ± 0.2	1.10
Percent of the total calculated	98.2	98.7

4. Conclusions

The activation performance of a CuCrZr alloy of the European Fusion Technology Programme was investigated in a fusion peak neutron field. Calculations with the European Activation System predict the recycling limit of the material after irradiation under fusion power plant conditions (power density of 1 MW/m^2, irradiation of 1 year) to be reached after about 100 years.

All activities that are important up to the recycling limit were experimentally determined and compared with the calculated values. From the ratio of calculated-to-experimental total activity it may be concluded that the expected activation performance is validated within 15%.

The calculated long-term radioactivity dominated by ^{26}Al can be confirmed within an uncertainty of 19% with recent accurate measurements of the ^{27}Al(n,2n) cross-section and with the determination of the Al content by the ^{27}Al(n,α)^{24}Na activation. The dose rate produced by

fusion peak neutrons in the technological CuCrZr alloy under reactor conditions is well below the hands-on limit after 500 years.

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