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Long Lived Activation Products in Eurofer

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ABSTRACT

Two samples of eurofer steel were irradiated in TRIGA reactor at JSI. One sample was irradiated bare and one under Cd filter which absorbed almost all neutrons with energies below 0.5 eV. The samples were exposed to the (unperturbed) total neutron fluence of 2×10^{19} n/cm^2. After irradiation the samples were measured with gamma spectroscopy using HPGe detector. The highest contribution to the total activity is (n, gamma) reaction on ^{51}Cr and ^{182}Ta in both samples. The highest contribution (~ 80 %) to the total dose rate is due to neutron capture in ^{181}Ta.

1 INTRODUCTION

Eurofer is a candidate low activation steel as first wall and breeder blanket structural material for future fusion power plants. It is one of the key materials of the fusion technology as it is expected to be used in the quantities of several hundred tons in each fusion reactor. As the development of the eurofer is still in progress, there are still many issues to be solved. One of them is the activation of eurofer after irradiation in neutron field.

The 14 MeV neutron transmissions through the structural components of the fusion reactor depend on the material used for construction. While there is no accurate knowledge about the neutron spectrum in the future fusion reactors, materials used in the fusion reactors must be tested in various neutron spectrums. Some work has already been done relating eurofer activation in a 14 MeV neutron field [1]. In the present work we are interested in eurofer activation in prevailingly thermal neutron spectrum. The aim of our work is also to develop the method to determine the amount and composition of impurities and long-lived activation products in eurofer. The eurofer components of the future fusion reactor will be most probably manufactured in several different steelworks, meaning that each part will contain different impurities. Therefore it is expected that this method could become a standard for testing of structural components.

In order to determine the composition and activity of activation products several samples of eurofer were irradiated in the Jozef Stefan Institute (JSI) TRIGA research reactor [4]. The neutron spectrum in TRIGA reactor is thermal, while in future fusion devices the neutron spectrum will be hard due to much higher initial neutron energy (14 MeV) and lack of moderator. In order to assess the influence of neuron spectrum on activation, some samples were irradiated under cadmium filter (~1 mm thick), which absorbed all neutrons with energies below 0.5 eV. The irradiation took two months; the samples were exposed to the

unperturbed total neutron fluence of ~2×10^{19} n/cm^2. After irradiation, gamma spectrometry of the irradiated samples was performed in order to determine the composition of the activation products.

Two cylindrical samples (1 mm in diameter and 3 mm high), with a mass of ~ 0.2 g, were irradiated in the central channel of the TRIGA Mark II reactor core, where the highest flux of 2×10^{13} n·cm^{-2}·s^{-1} is achieved at 250 kW. One sample was irradiated under cadmium filter (denoted as 'eurofer under Cd') and the other sample was irradiated bare (denoted as 'eurofer'). In order to prevent the contamination of the eurofer samples by cadmium, they were wrapped in the aluminium foil.

Sample irradiation started on 30th of May 2007 at 8:45 and ended on 24th of July 2007 at 8:30. The position of the samples did not change during the time of irradiation. The effective time of irradiation at the power of 250 kW was 247 h and 54 min. The samples were left in the reactor core for approximately nine days after the irradiation. On the 2nd of August at 10:40 the samples were taken out of the reactor core and the measurements of the dose rate were performed at approximately constant intervals. The dose rate at a distance of 1 m was measured with two dose rate meters, BERTHOLD UMO LB 123 and AUTOMESS 6150AD.

2 RESULTS

2.1 Measurements

The results of the dose rate measurements are presented in Figure 1. The following conclusions important for application of eurofer in the fusion power plant can be made on the basis of the dose rate measurements soon after the irradiation:

1. The dose rate is relatively high, 10 Sv/h per ton of eurofer at distance 1m and after nine day cooling.
2. The dose-rate is decreasing relatively slowly (20% per month).

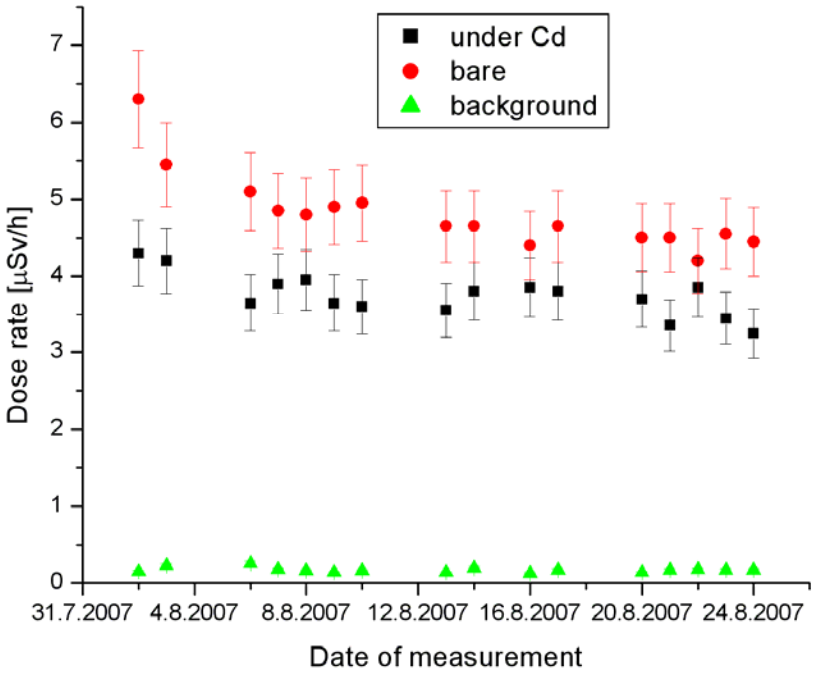

Figure 1: Dose rate measurements of eurofer samples at a distance of 1m.

Exposed even to relatively low fluence (less than 1% of expected fluence in a typical fusion power plant) the components made of eurofer are expected to become far too radioactive for approach and manipulation by workers, and will not cool in reasonable time.

One month after irradiation the samples were taken out of the aluminium foils to prepare the clean samples for gamma spectroscopy analysis. When we unwrapped the aluminium foil we observed small but visible quantity of black dust inside the foil. Possible explanation is corrosion of eurofer or chemical reaction between eurofer and aluminium.

The activity of samples was measured with gamma spectroscopy using HPGe coaxial detector produced by Canberra (relative efficiency of 40%). Due to relatively high activity the samples were positioned 1.5 m away from the detector. The efficiency for sample geometry was calculated with ISOCS/LABSOCS software [5]. The detector was characterized by the producer. The specific activity measurements are presented in Table 1. Note that the measured quantity was activity. Specific activity was obtained by dividing the measured activity by the sample mass, which is 0.20 g.

Table 1: Gamma spectroscopy measurements results (date: 30.8.2007). The highest contributions to the total specific activity are highlighted.

Isotope	**$T_{1/2}$**	**EUROFER Specific Activity (Bq/g)**	**% of total**	**EUROFER under Cd Specific Activity (Bq/g)**	**% of total**
Na-22	950 d	(4.44±0.34)E+05	0.20	(3.79±0.27)E+05	0.52
Cr-51	27.7 d	(1.40±0.19)E+08	61.87	(8.05±0.11)E+06	11.10
Mn-54	312.7 d	(3.92±0.29)E+06	1.73	(4.01±0.29)E+06	5.52
Fe-59	44.6 d	(7.80±0.05)E+06	3.45	(9.25±0.07)E+05	1.28
Co-60	5.27 y	(2.25±0.15)E+05	0.10	(5.35±0.06)E+04	0.07
Cd-109	464 d	(6.90±0.14)E+06	3.05	(4.61±0.92)E+06	6.36
Ta-182	114.4 d	(6.70±0.02)E+07	29.61	(5.45±0.01)E+07	75.15
total		(2.26±0.19)E+08		(7.25±0.02)E+07	

Dose rates at 1m distance were measured with GM counter AUTOMESS 6150 AD6. Contributions of different radionuclides based on their activities were calculated by MicroShield program (v6.02) [6].

Table 2: Dose rate measurements (date: 30.8.2007). Note that the dose was measured at a distance of 1 m from the sample, with a mass of 0.20 g.

Sample	**Dose rate at 1 m (µSv/h)**	**Ta-182 contributor %**
EUROFER	3.8±0.1	77
EUROFER under Cd	3.2±0.1	91

Two important conclusions can be drawn:

1. Main contribution to the dose-rate is due to Ta-182
2. Main contribution to the total activity is activation of Cr-51 by thermal neutrons and activation of Ta-182 by epithermal and fast neutrons.

Low contribution of thermal neutrons to the activation of Ta-182, the most important contributor to the total dose, indicates that our experiments are relevant also for the actual fusion reactor where the spectrum will be only harder. In spite of well thermalized spectrum in TRIGA reactor it is reasonable to use the reactor in activation experiments as the main

effects arise in the part of the spectrum that is believed to be similar in fission and fusion reactors.

Tantalum as the main contributor to the activity came as a surprise as it is not among main constituents of eurofer (less than 1/1000 by weight). It is not problematic with respect to long-term storing and disposal of eurofer as radioactive waste, as it will decay in a few years (114 days half-life). However, immediate dose-rates could be reduced almost for a decade if it was eliminated from the alloy.

3 CALCULATIONS

For a better understanding of the measured results calculations of the eurofer activation have been performed using the FISPACT code, part of the European Activation System (EASY) [2]. FISPACT is an inventory code for neutron-induced activation calculations for materials in fusion devices. It uses external libraries of nuclear data [3] for all relevant nuclides to calculate the number of atoms of each species at a specified time during the irradiation or after a decay time following shutdown. The various species are formed either by a direct reaction on a starting material, by a series of reactions some of which can be on radioactive targets, or by a decay or series of decays.

The essential input data for any activation calculation are the material composition of the irradiated sample and the neutron spectrum and fluence. The exact material composition of our sample was not available thus reported standard values were used [7], [8]. The elemental composition used for the calculations is presented in Table 3.

Table 3: The composition of eurofer used for activation calculations [7]

Element	Fe	Cr	C	Mn	P	S	V	B	N	O	W
Wt. %	88.963	9	0.11	0.4	0.005	0.005	0.2	0.001	0.03	0.01	1.1
Element	Ta	Ti	Nb	Mo	Ni	Cu	Al	Si	Co	Sn	As
Wt. %	0.075	0.01	0.001	0.005	0.005	0.005	0.01	0.05	0.005	0.005	0.005

The neutron spectrum in the central channel of the TRIGA MARK II reactor of the J. Stefan Institute in Ljubljana was determined by MCNP calculations and the flux normalized accordingly [9], [10]. The flux was calculated for the position in the central channel, where the samples were irradiated for two cases – with and without a Cd cover around the samples. The total neutron flux at the point of irradiation was found to be 2×10^{13} n/(cm^{2}s) for the case without the Cd cover and 1.1×10^{13} n/(cm^{2}s) for the case with the Cd cover. Both neutron spectra are presented in Figure 2.

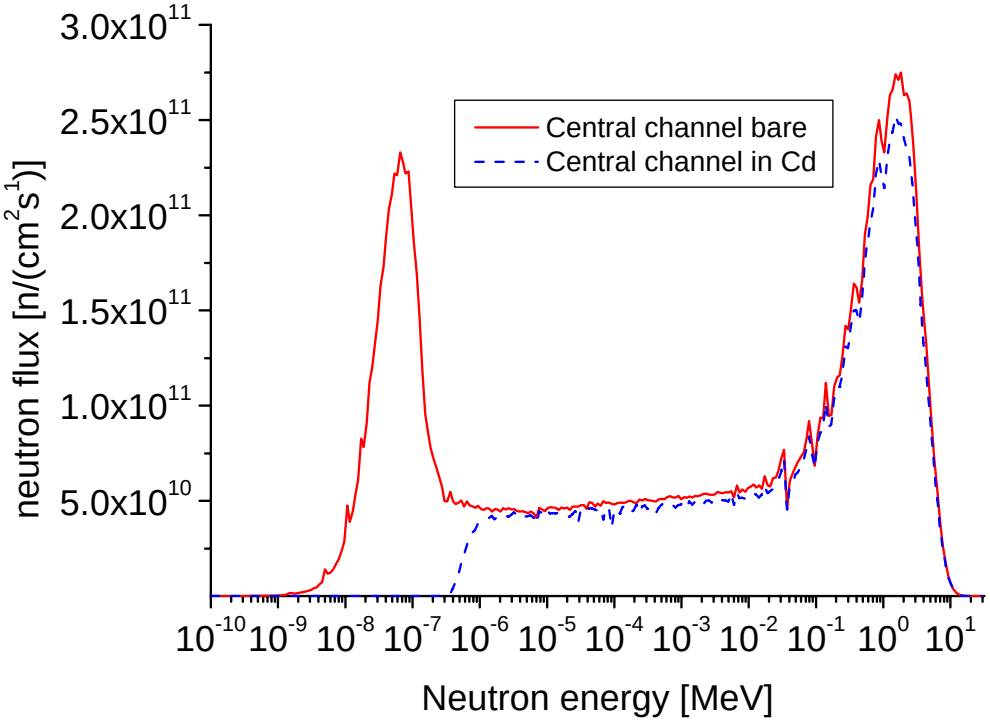

Figure 2: Neutron flux distribution in the central channel of the JSI TRIGA reactor calculated with MCNP.

The activation is in general dependent on the detailed irradiation history, i.e. on the exact time dependent flux levels. In our case the irradiation took place in several steps with the reactor power changing from 0 to the maximum power of 250 kW within a period of 55 days. The effective irradiation time at full power was 247h 54min. Although the results may depend on the flux levels in most cases only the fluence is important. It has been found out in our case that by changing the flux by a factor of 10 the activation per incident neutron for any of the relevant nuclides does not change for more than three percent. For the calculations the flux was thus raised three times to the maximal level and reduced to zero in the meantime in a way, that the overall irradiation time was 55 days and the effective one 247.9h .

The activation calculations were performed for a thin sample – FISPACT default – i.e. shading effects and neutron transport in the samples were neglected. Since the size of the eurofer sample, placed into the central channel was 1mm, this simplification is justified. A sample of 1g mass was used for the calculations and all results are scaled to this value.

The inventory and the doses were calculated at the times of 9 days and 30 days after the end of irradiation. The values for the activity and dose rate, each time for the 4 dominant nuclides, are presented in Table 4.

Table 4: Calculated specific activity and dose rates for the four most important nuclides for irradiation of a sample of 1g of eurofer. The cooling times were 9 and 30 days. Note that the totals are presented as well.

Bare samples (without cadmium cover)						
9 days after irradiation						
NUCLIDE	SPECIFIC ACTIVITY (Bq/g)	PERCENT ACTIVITY		NUCLIDE	DOSE RATE (Sv/hr)	PERCENT DOSE RATE
Total	4.5E+08			Total	1.2E-05	
Cr 51	3.3E+08	73.9		Ta182	7.3E-06	60.1
Ta182	4.6E+07	10.2		Cr 51	1.9E-06	15.4
Fe 55	3.2E+07	7.2		Fe 59	1.8E-06	14.8
Fe 59	1.2E+07	2.8		Mn 54	4.3E-07	3.6
30 days after irradiation						
NUCLIDE	SPECIFIC ACTIVITY (Bq/g)	PERCENT ACTIVITY		NUCLIDE	DOSE RATE (Sv/hr)	PERCENT DOSE RATE
Total	2.9E+08			Total	9.5E-06	
Cr 51	2.0E+08	67.4		Ta182	6.5E-06	68.2
Ta182	4.0E+07	13.9		Cr 51	1.3E-06	13.8
Fe 55	3.2E+07	10.9		Fe 59	1.1E-06	11.8
Fe 59	9.0E+06	3.1		Mn 54	4.1E-07	4.4

under cadmium cover						
9 days after irradiation						
NUCLIDE	SPECIFIC ACTIVITY (Bq/g)	PERCENT ACTIVITY		NUCLIDE	DOSE RATE (Sv/hr)	PERCENT DOSE RATE
Total	6.7E+07			Total	6.6E-06	
Ta182	3.5E+07	52.2		Ta182	5.6E-06	85.6
Cr 51	1.6E+07	23.1		Mn 54	3.9E-07	5.9
W 185	5.7E+06	8.5		Fe 59	2.3E-07	3.6
Mn 54	3.5E+06	5.3		W 187	2.1E-07	3.1
30 days after irradiation						
NUCLIDE	SPECIFIC ACTIVITY (Bq/g)	PERCENT ACTIVITY		NUCLIDE	DOSE RATE (Sv/hr)	PERCENT DOSE RATE
Total	5.2E+07			Total	5.6E-06	
Ta182	3.1E+07	60.1		Ta182	5.0E-06	89.0
Cr 51	9.2E+06	17.9		Mn 54	3.7E-07	6.7
W 185	4.7E+06	9.1		Fe 59	1.7E-07	3.0
Mn 54	3.4E+06	6.6		Cr 51	5.2E-08	0.9

As can be seen from the table the most important nuclides are ^{182}Ta and ^{51}Cr. They are produced in the reactions ^{181}Ta(n,γ)^{182}Ta and ^{50}Cr(n,γ)^{51}Cr . By comparison with the measured data relatively good agreement in the case of Cr is observed, whereas the calculated activity for the trace element Ta is in both cases lower than the measured value (note that the calculated values are presented per g of eurofer). The reason could be that in the measured sample the Ta content was higher than assumed from [7] or that the Ta cross sections are wrong.

In Figure 5 the calculated dose rates at a distance of 1 m from the sample for various cooling times are presented. As expected the doses are smaller for the Cd covered sample. However, the difference is relatively small indicating low importance of thermal neutron reactions.

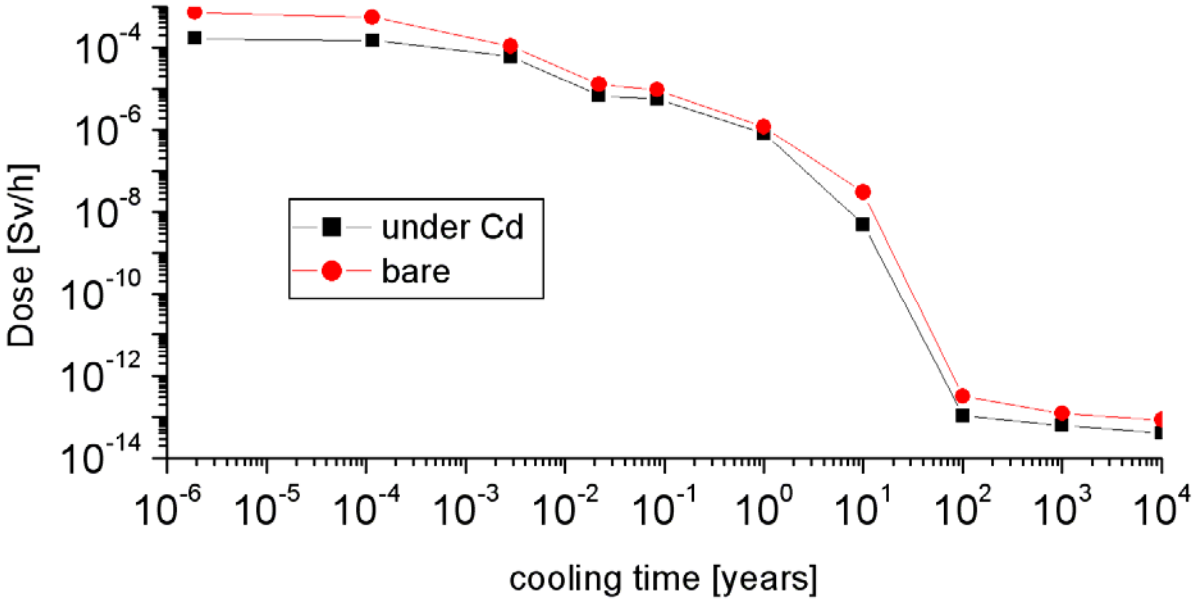

Figure 5: Dose rates at a distance of 1 m from the sample for cooling times from 1 min to 10000 years calculated with FISPACT.

4 CONCLUSIONS

Main isotopes contributing to the long-term activation have relatively short life-times (100 days to several years). This composition of eurofer is suitable from the radioactive waste disposal point of view. However one should be aware that dose rates inside and around the eurofer structures will be significant even several months of cooling. The highest contribution to the total activity (80%) in this time interval comes from ^{182}Ta. It may be speculated that further optimization of eurofer with respect to the activation could be achieved if the elemental composition were re-examined, in particular considering fast neutron reactions.

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