MEASUREMENTS OF FUSION NEUTRON INDUCED ACTIVATION AND DECAY HEAT IN RHENIUM. COMPARISON WITH CODE PREDICTIONS

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Tungsten is a common material used when heavy thermal flux load must be supported. The International Thermonuclear Experimental Reactor (ITER) will make use of large quantities of Tungsten in the high thermal flux zones like the plasma divertor. The possibility to use W on the first wall, facing the plasma is considered as well. The activation properties of the materials used for next generation of fusion facilities are carefully studied in order to reduce the radiation inventory and hazard. Specific "ad hoc" computer codes and databases are under development in the fusion community to predict the radiation issue in ITER. The activation databases for the most important ITER structural materials have been already validated. Presently studies of the activation properties of the most important material impurities are under way. Rhenium is one of the most common impurity elements in Tungsten. In this paper the activation characteristics of Rhenium have been studied, irradiating some samples with the 14 MeV neutrons produced with the Frascati Neutron Generator (FNG) and measuring the induced gamma and beta activation. The experimental results have been compared with the FISPACT code prediction, using the latest version of the European Activation File, EAF-2007. The results of the comparison and the uncertainty analysis are presented in the paper.

I. INTRODUCTION

Neutron activation of materials produces an energy release during the subsequent radioactive decay. In a fusion power plant this energy release is of the order of MWs. Accurate prediction of this decay heat is fundamental for the design of a D-T fusion power plant, especially for the safety analysis. Measurement of this decay power is not trivial since the existing 14 MeV neutron generators are able to induce only very low levels of decay power, in the order of few μW. A very efficient detector system able to measure both beta and gamma heats simultaneously and separately has been developed at ENEA Frascati and has been already used to validate the predictions of a computer code developed for this purpose.[1,2,3]

II. DESCRIPTION OF THE DECAY HEAT MEASURING SYSTEM

A good knowledge of the decay heat of radionuclides is necessary to design large devices for energy production, like a fusion power plant, the future proposed Energy Amplifier or a fission reactor. Experimental studies to measure decay heat following fission of actinide samples have been performed previously using direct detection of temperature rises of irradiated samples[4] or by measuring the energy spectra of beta and gamma-rays independently and deducing the decay heat from measured spectra.[5-9] Experimental data of total decay heat for materials irradiated in a simulated fusion device environment have been published by a Japanese team.[10] At ENEA Frascati, Italy, we have developed a very efficient spectrometer to measured the beta and gamma decay heats produced by neutron activation.

The spectrometer consists of a large cylindrical (22.8 × 22.8 cm) well-type CsI(Tl) scintillator and a small (1.9 cm diameter, 2.6 cm high) plastic BC400scintillator inserted in the well (Fig. 1). The CsI(Tl) crystal has larger density and atomic number compared to more common NaI(Tl) scintillators. The dimensions chosen permit to detect with high efficiency (≈100%) almost all photon radiation while electrons are adsorbed and eventually converted into x-rays in the entrance window. The BC400

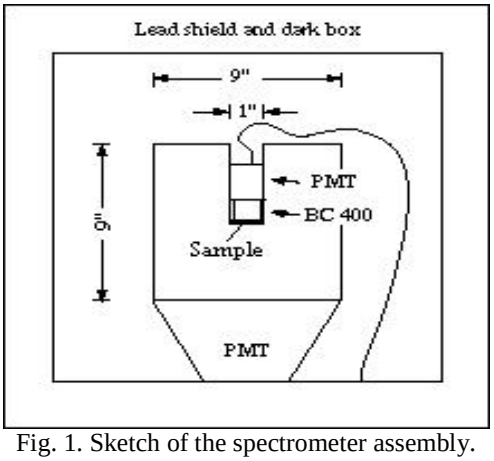

Fig. 1. Sketch of the spectrometer assembly.

scintillator is window-less; it has low density and atomic number. Its dimensions where chosen to detect virtually only β radiation and conversion electrons with about 50% efficiency (2π geometry). Time response of this detector is very fast, a 0.5 μs shaping constant was used to reduce any coincidence effect.

Both detectors have been energy calibrated using standard radiation sources, while accurate detection efficiency is calculated by modelling the spectrometer and the material sample under study using the MCNP-4C radiation transport code.[11] Thin material samples (disc-shaped, ≤ 25 μm thick, 1.8 cm diameter) are used to reduce radiation self-adsorption. The very high sensitivity to photons and electrons of the two scintillators permits the determination of the absolute activity (from β decay) and the total photon and electron heats.[12]

Both scintillators are thermally stabilised using a suitable unit to avoid a temperature dependent light response.[13] From the pulse-height spectra (PHS) recorded by multichannel analysers, both beta and gamma decay heats (in units of μW/g) are deduced according to the following formula:

$$\text{Decay heat} = 1.6\times10^{-19}\times f_i\times S_i(C_i\times E_i)/(T_c\times m).$$

Each count (C_i) of the PHS in the channel-i is multiplied by the corresponding energy (E_i) and summed to obtain total deposited energy. The total energy is divided by the counting time T_c and the sample mass m; f_i is the correction factor for the efficiency calculated with MCNP-4C code and corresponds to the fraction of the energy lost.

Typical backgrounds in the spectrometer are about 0.3 Bq for activity detection and 1.4×10^{-5}, 2.5×10^{-8} μW for photon and electron heat measurements, respectively. More detailed information about the measuring accuracy and the uncertainty analysis of this detector system can be found in Ref 14.

III. IRRADIATION AND MEASUREMENTS HISTORY

Two high purity Rhenium samples 25 μm thick were irradiated at a fixed position in front of the FNG neutron target. The neutron yield was monitored by the associated particle method[15] while the neutron distribution was measured with several activation foils and unfolded using the SAND-II computer code[16] and the IRDF-2002 dosimetry library.[17] An "ad hoc" neutron reflector has been used behind the samples under irradiation to produce an ITER "First Wall" like neutron spectrum. The monitor foils plus the two Rhenium samples were irradiated with the 14.8 MeV neutrons produced through the d(T,α)n fusion reaction by the 260 keV FNG deuteron beam for about 16000 s. After the irradiation the monitor foils and one Rhenium sample were measured using an absolute calibrated, 40% relative efficiency, HPGe detector while the other Rhenium sample was measured with the decay heat measuring system described above.

The neutron spectrum at the samples position has been first calculated with the MCNP-4C code using a special developed source subroutine and target geometry which accurately describes the FNG neutron emission.[18] The calculated spectrum was normalized using the total neutron yield measured by the associated α particle and then used as input trial spectrum of the SAND-II unfolding code. The list of the reactions used for the unfolding procedure, the Experimental to Calculated ratio after the unfolding and the overall experimental errors are gathered in Table I.

TABLE I. Spectrum Unfolding Results

Reaction	E/C Sand	Exp. Error
Al27(n,α)Na24	0.990	1.58%
In115(n,n')In115m	0.994	2.52%
In115(n,γ)In116m	1.000	1.50%
Au197(n,2n)Au196	1.016	1.53%
Au197(n,γ)Au198	1.000	1.60%

The reactions used cover the whole energy neutron spectrum, from the 14.8 MeV source neutron energy down to thermal energy. The unfolded adjusted spectrum was grouped in 175 neutron groups (Vitamin-J type) ready to be used as input spectrum in the FISPACT code[1] The spectrum is shown in Fig. 2.

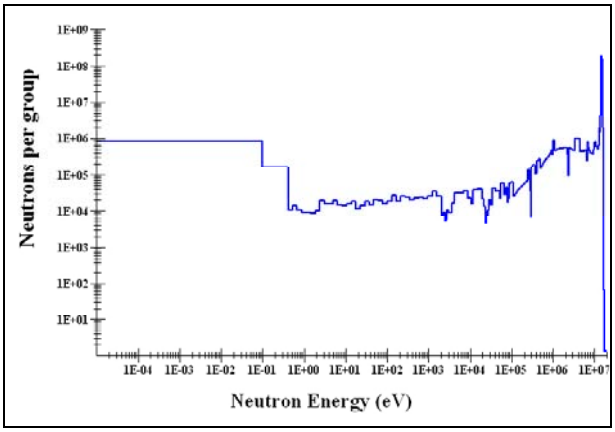

Fig. 2. Neutron spectrum at the sample irradiation position.

This spectrum was first of all used as input spectrum in the EASY-2007 package to recalculate the activity of the monitor foils used during the unfolding. The purpose of this job was to verify the adequacy of the spectrum in

reproducing the foil activities. The result (summarized in Table II) is satisfactory, well below the uncertainties in the EASY-2007 library for these reactions[1].

TABLE II. E/C Calculated by EASY-2007

Reaction	E/C EASY	Uncertainty
Al27(n,α)Na24	0.990	45.6%
In115(n,n')In115m	1.040	7.0%
In115(n,γ)In116m	1.041	54.9%
Au197(n,2n)Au196	1.026	9.4%
Au197(n,γ)Au198	1.059	25.5%

The measure of the Rhenium samples range from few minutes after the irradiation up to about one month of decay time.

IV. RESULTS

The results of C/E comparison between the measured beta heat and EASY-2007 predictions vs. the decay time are given in Fig. 3. In Fig. 4 the contribution of the different radionuclides to the total beta heat, as predicted by the code, is also given.

The gamma heat results are show in Figs 5, 6. For both measured heats the trend is an underestimation of the calculation for decay times up to about one day.

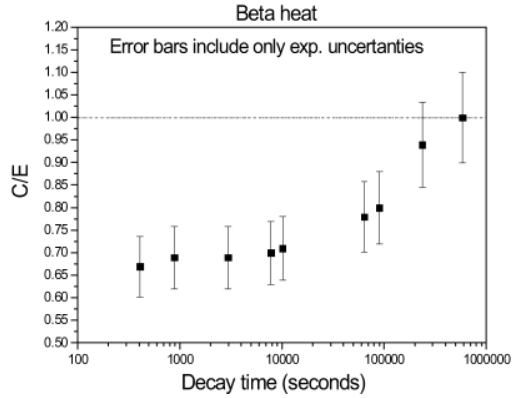

Fig. 3. C/E comparison for beta heat.

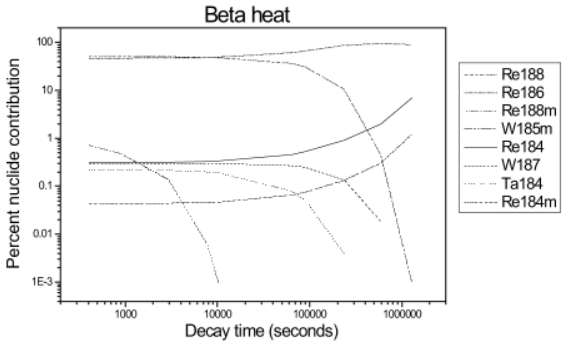

Fig. 4. Percent contribution of the radionuclides.

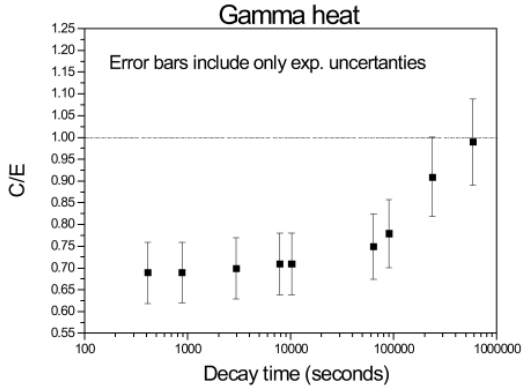

Fig. 5. C/E comparison for gamma heat.

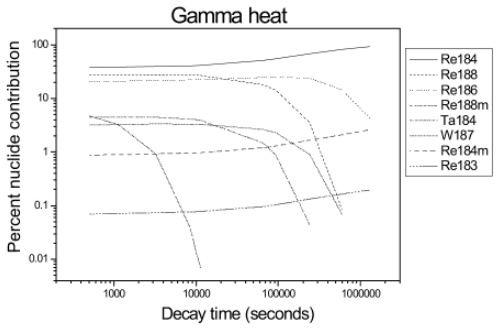

Fig. 6. Percent contribution of the radionuclides.

From the pathways analyses performed with EASY-2007 the main radionuclide responsible of the C/E discrepancy is Re188 for both beta and gamma heat. This radionuclide was produced in our experiment through the reactions:

$$\mathrm{Re187(n,\gamma) \rightarrow Re188}$$
$$\mathrm{Re187(n,\gamma) \rightarrow Re188m(IT)\,Re188}$$

Our results indicate that the cross-section and/or the decay data used in EAF-2007 databases, which are part of EASY-2007 package, are underestimated.

The gamma spectroscopy performed on the second Rhenium sample exposed simultaneously have been used to clarify the discrepancy found. The HPGe gamma spectroscopy results are summarised in Table III while the C/E comparison is given in Table IV. The uncertainties quoted in Table IV are those from the calculation and the experiment in quadrature summed. In this table also the Quality Score of each identified reaction are given The Quality Score (QS) is a value from 0 to 6 indicating the degree to which the EAF-2007 data are backed up by experiments available in literature (Ref. 2); 0=no experimental data, 1=weak disagreement with differential data, 2=weak agreement with differential data, 3=strong disagreement with differential data, 4=strong agreement

with differential data, 5=conflicting differential and integral data, 6=agreement between differential and integral data (validated).The decay heat measurements have indicate the Re188 radionuclide as responsible of the disagreement between our experimental data and EASY-2007 prediction. The reactions producing Re188 show an underestimation of the C/E ratio also for gamma spectroscopy. This fact implies that are the cross-section data used in EASY-2007 the reason of the discrepancy in our integral experiment. This fact is in agreement also looking at the quality score 4 and 5 already assigned in EASY-2007 by the authors at the reactions producing Re188.

For the other radionuclides identified the results of the C/E comparison are satisfactory, within the total uncertainties, apart for the reaction:

$$Re185(n,3n) \rightarrow Re183$$

This reaction however has QS of zero which means no differential data exits in literature and thus the cross-section used in EASY-2007 was derived only by code evaluation.

TABLE III. Gamma Spectroscopy Results

Reaction	Path	T/2	Activ. Calc.	Err %	Activ. Meas.	Err %
Re187 (n,g) **Re188m**	100%	19 m	1193	63	1254	11
Re187 (n,a) **Ta184**	100%	8.7 h	68	20	73	10
Re187 (n,g) **Re188**	97%	17 h	4557	22	7178	1.5
Re187 (n,g) Re188m (IT)**Re188**	3%					
Re187 (n,p) **W187**	100%	24 h	180	9	196	12
Re185 (n,g) **Re186**	11.7%	90.6 h	15568	14	16129	1.5
Re187 (n,2n) **Re186**	88.3%					
Re185 (n,2n) **Re184**	100%	38 d	808	15	1100	1.5
Re185 (n,3n) **Re183**	100%	70 d	11.7	20	18	23
Re185 (n,2n) **Re184m**	100%	165 d	55	20	50	8

TABLE IV. C/E from Gamma Spectroscopy

Reaction	C/E	Err.	QS
Re187(n,g)Re188m	0.95	63%	5
Re187(n,a)Ta184	0.93	22%	4
Re187(n,g)Re188	0.63	22%	4
Re187(n,g)Re188m(IT)Re188			5
Re187(n,p)W187	0.92	15%	4
Re185(n,g)Re186	0.97	14%	4
Re187(n,2n)Re186			6
Re185(n,2n)Re184	0.73	15 %	5
Re185(n,3n)Re183	0.64	31 %	0
Re185(n,2n)Re184m	1.10	22%	6

V. CONCLUSIONS

The data presented in this work will be used, together with other future integral data eventually available, for the continuous improvement of the EASY code system databases. The experiment on the Rhenium samples has indicate that only some minor revision of Rhenium data in EAF-2007 are probably necessary. Our results will also contribute to reduce the uncertainties used in the databases.

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