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Material composition and nuclear data libraries' influence on nickel–chromium alloys activation evaluation: a comparison with decay heat experiments

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Abstract

The paper presents the activation analyses on Inconel-600 nickel–chromium alloy. Three activation data libraries, namely the EAF-4.1, the EAF-97 and the FENDL/A-2, and the FENDL/D-2 decay data library, have been used to perform the calculation with the European activation code ANITA-4/M. The neutron flux distribution into the material samples was provided by JAERI as results of 3D Monte-Carlo MCNP transport code experiment simulation. A comparison with integral decay heat measurement performed at the Fusion Neutronics Source (FNS), JAERI, Tokai, Japan, is used to validate the computational approach. The calculation results are given and discussed. The impact of the material composition, including impurities, on the decay heat of samples irradiated in fusion-like neutron spectra is assessed and discussed. The discrepancies calculations–experiments are within the experimental errors, that is between 6% and 10%, except for the short cooling times (less than 40 min after the end of irradiation). To improve calculation consistency with the experimental results, the knowledge of the material impurities content is mandatory.

1. Introduction

The neutron-induced activation must be taken into account in the choice of materials for fusion machines, both from a safety point of view and the treatment and disposal of radioactive wastes. The material elemental composition is an important factor since responses are element and isotope related. Nickel–chromium alloys are considered as candidate structural material for future tokamak machines. So, it is relevant to correctly estimate their activation due to irradiation in fusion neutron spectra for fusion machine radioactive inventories characterization. In addition, due to the extreme conditions under which these plants are operated, thermo-mechanical material properties can be strongly impacted by irradiation in fusion-like neutron spectra, e.g., due to the gas production and the dpa.

To validate the codes, data and calculation procedures used for those estimates, various experiments were performed in several countries. Recently, an international benchmark exercise was launched [1] by the ITER Joint Central Team (JCT) to compare experimental integral decay heat measurements of fusion-relevant structural materials irradiated in 14 MeV neutron spectra with calculations. Integral decay heat validation experiments were organized and performed using the Fusion Neutronics Source (FNS) [2] at the Japan Atomic Energy Research Institute (JAERI), Tokai, Japan. Decay heat calculations were performed by different teams, using various activation codes [3].

This paper presents the decay heat calculation results obtained by a joint ENEA and Bologna University team for the Inconel-600 nickel–chromium alloy, and discusses their comparison with the experimental ones. The impact of material composition, including content of impurities, on the decay heat of samples irradiated in the 14 MeV neutron spectra is assessed and discussed.

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2. Calculation approach methodology

Activation characteristics were calculated using the European activation code [4] ANITA-4/M. Neutron energy spectra (in the 175 Vitamin-J energy-groups structure) as results of a 3D Monte-Carlo MCNP transport code [5] experiment simulation and source neutron intensity were provided by the FNS-JAERI team. Three neutron activation data libraries, namely the European Activation Files [6,7] EAF-4.1 and EAF-97 and the Fusion Evaluated Nuclear Data Library [8] (FENDL/A-2), and the decay data library for fusion applications [9] (FENDL/D-2), have been used to perform the calculation.

3. Experiment description and results

The FNS was used [10] to irradiate samples of various materials in a 14 MeV neutron flux. Irradiations were performed in two series, with durations of either 5 min or 7 h. After irradiation, each sample was transferred to a Whole Energy Absorption Spectrometer (WEAS) for measurement of decay energy at a series of times (named in the following as cooling times), up to about 1 h, for the 5 min irradiation, and 100 days, for the 7 h irradiation. The experiments included measurements of decay heat in 32 different materials. The overall experimental uncertainty totals between 6% and 10%. A detailed description of the experiments is given in [10].

First, ANITA-4/M calculation-comparisons with experiment [11,12] were concentrated on those materials relevant to ITER safety studies, namely stainless steel 316, copper and tungsten. The present analysis is devoted to the Inconel-600 nickel–chromium alloy.

The wt% material composition of Inconel-600 samples irradiated at the FNS is Cr 15.97, Fe 7.82, Mn 0.39, and Ni 75.82. In the following, it will be named as I_600-reference. The measured isotope abundances (at.%/100) are as follows:

- Chromium: Cr^{50} (0.04345), Cr^{52} (0.83789), Cr^{53} (0.09501), Cr^{54} (0.02365);
- Iron: Fe^{54} (0.058), Fe^{56} (0.9172), Fe^{57} (0.022), Fe^{58} (0.0028);
- Manganese: Mn^{55} (1.0);
- Nickel: Ni^{58} (0.68077), Ni^{60} (0.26223), Ni^{61} (0.0114), Ni^{62} (0.03634), Ni^{64} (0.00926).

A slightly different Inconel-600 material composition was initially provided by the FNS-JAERI team: Cr 15.5, Fe 8, Mn 1, and Ni 75.5 wt%. In the following, it will be named as I_600-alternate. The isotope abundances are the same as the previous ones.

Since no measured content of material impurities (or tramp elements) was provided by FNS-JAERI, a hypothetical typical set of their values (based on [13]) has been considered to estimate the impact on the Inconel-600 activation characteristics. The weight impurity content that is considered is the following: Si 0.35%, Mo 100 ppm, C 60 ppm, Ti 0.2%, Al 0.16%, Nb 300 ppm, Sn 100 ppm, Co 500 ppm, Cu 600 ppm, Mg 300 ppm, Pb 10 ppm and N 50 ppm.

4. Calculation–experiment comparison

To estimate the discrepancy between the calculated (C) and the experimental (E) decay heat values, the percentage ratio (C–E)/E% has been evaluated. For the Inconel-600, they are plotted in Figs. 1 and 2, for the 5 min and the 7 h irradiation time, respectively. The plots show the calculation–experiment comparison both for the reference (R) and the alternate (A) composition, and considering different activation data libraries for the calculations.

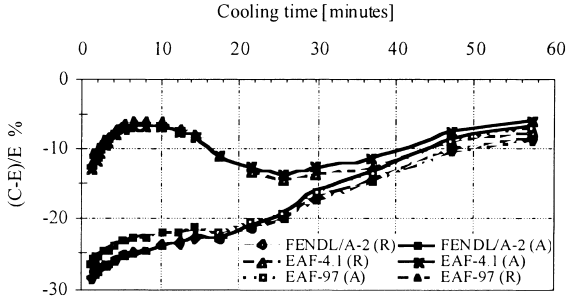

Fig. 1. Total decay heat vs cooling time of Inconel-600 samples irradiated for 5 min. Comparison of calculation with experiment (C–E)/E %, for reference (R) and alternate (A) compositions. Impact of the neutron activation data library.

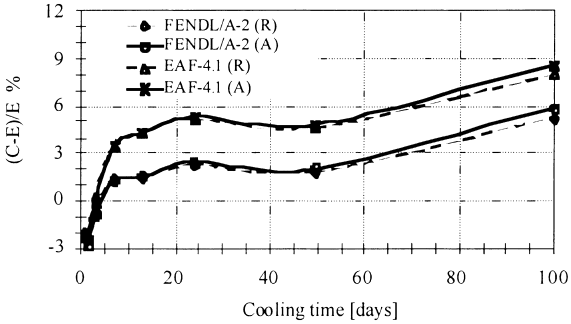

Fig. 2. Total decay heat vs cooling time of Inconel-600 samples irradiated for 7 h. Comparison of calculation with experiment (C–E)/E %, for reference (R) and alternate (A) compositions. Impact of the neutron activation data library.

5. Results of analysis

5.1. Samples irradiated for 5 min

The discrepancy between ANITA-4/M decay heat calculations and experimental results is around 12% when using the EAF-4.1 activation library, whilst it is around 25–30% for EAF-97 and FENDL/A-2, 1 min after the end of the irradiation (see Fig. 1). From about 40 min cooling time, the three activation libraries give quite comparable results with a discrepancy of about 10% with respect to the experimental values. The analysis of the calculation results points out the relevance of the V^{52} isotope contribution to the Inconel-600 decay heat in the first few minutes after the 5 min irradiation: about 85% on the total decay heat for 1 min cooling time and about 78% for 10 min cooling, when EAF-4.1 is used. Since significant differences in nuclear cross-section data for the V^{52} production reaction ^{52}Cr (n, p) ^{52}V exist among the three libraries, that reflect on corresponding differences in the decay heat calculation results for the first few minutes of cooling time. Indeed, for that reaction, the FENDL/A-2 uses data from the ADL-3 evaluation data file [14], whilst the EAF-4.1 refers to the EFF-2.4 evaluation data file of the EFF project library [15]. For that reaction, the EAF-97 refers to IRK data source [16]. The V^{52} is also produced by other reactions, i.e., ^{53}Cr (n, np) ^{52}V, ^{53}Cr (n, d) ^{52}V, ^{54}Cr (n, t) ^{52}V, ^{55}Mn (n, α) ^{52}V. For those reaction cross-sections EAF-4.1, FENDL/A-2 and EAF-97 use data from the EFF-2.4 data file (only for the ^{55}Mn (n, α) ^{52}V reaction the EAF-97 refers to Russia Dosimetry File FEI [17]). For 1 h cooling time, the V^{52} isotope contribution to the Inconel-600 decay heat is less than 1%. So, the differences in cross-sections for reactions producing that isotope became negligible. At 1 hour cooling, the relevant isotope, with respect to the Inconel-600 decay heat, is the Mn^{56} (72%). The three activation data libraries considered refer to the same evaluated data file for all the reactions contributing to the Mn^{56} production. The difference between the I_600-reference and I_600-alternate sample composition produces no relevant discrepancy in decay heat results, as shown in Fig. 1.

5.2. Samples irradiated for 7 h

The discrepancy between the ANITA-4/M decay heat calculations and experimental results is less than 3% both using the EAF-4.1 and the FENDL/A-2 activation libraries in the first few days after the 7 h irradiation (see Fig. 2). The discrepancy rises to about 5% and 8%, respectively, for FENDL/A-2 and EAF-4.1, for 100 days cooling. The relevant isotopes contributing to the Inconel-600 decay heat are: Ni^{57}, Co^{58}, and Co^{58m} for the cooling times of interest (i.e., between 1 and 100 days). The relevant reaction producing the Ni^{57} isotope is ^{58}Ni (n, 2n) ^{57}Ni. Both the EAF-4.1 and the FENDL/A-2 for that reaction refer to IRDF-90.2 data source. On the contrary, for the ^{58}Ni (n, p) ^{58}Co, i.e. is the relevant reaction for the Co^{58} production, the FENDL/A-2 uses data from the ADL-3, whilst the EAF-4.1 uses data from the IRDF-90.2. Also for the 7 h irradiation, the sample composition difference gives no relevant discrepancy in decay heat results.

5.3. Impurities impact on the Inconel-600 activation characteristics

To assess the impact of material impurities, ANITA-4/M evaluation of the activation characteristics of samples (having the I_600-reference composition) has been performed. The comparison of the experimental decay heat values with the calculated ones (with and without considering impurities and both for the FENDL/A-2 and the EAF-4.1 activation libraries) are shown in Figs. 3 and 4, respectively, for the 5 min and the 7 h irradiation.

For the shorter irradiation time, the results analysis clearly shows that including impurities, the calculations

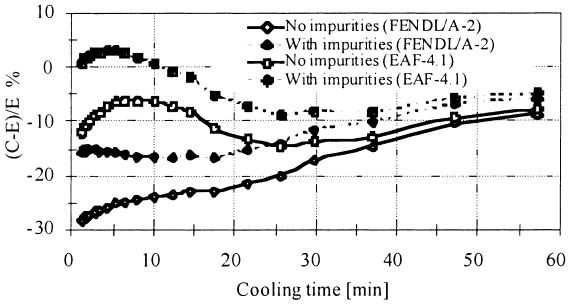

Fig. 3. Total decay heat vs cooling time of Inconel-600 samples irradiated for 5 min. Comparison of calculation with experiment (C–E)/E %. Impact of the material impurities.

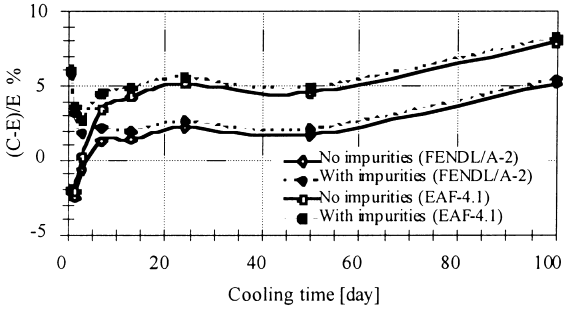

Fig. 4. Total decay heat vs cooling time of Inconel-600 samples irradiated for 7 h. Comparison of calculation with experiment (C–E)/E %. Impact of the material impurities.

provided decay heat closer to the experiments (see Fig. 3). This is especially true for the first minutes of cooling time and it is mainly due to the relevance of the Al28 (about 10% on the total Inconel decay heat). The isotope is produced by impurity reactions like ^{27}Al (n, γ) ^{28}Al and ^{28}Si (n, p) ^{28}Al. The Al^{28} contribution suddenly decrease below 0.1% after 20 min cooling.

For the longer irradiation time, the results analysis indicates no significant differences between calculation with and without including impurities for cooling times higher than about 10 days (see Fig. 4). On the contrary, relevant discrepancies appear in the first few days cooling time. This is due to the Na^{24} (about 7% on the total Inconel decay heat for 1 day cooling down to about 0.1% for 7 days cooling). The Na^{24} is produced mainly by the ^{27}Al (n, α) ^{24}Na and the ^{24}Mg (n, p) ^{24}Na reactions.

6. Conclusions

Activation analyses on Inconel-600 nickel–chromium alloy have been performed with the European activation code ANITA-4/M using three activation data libraries, the EAF-4.1, the EAF-97 and the FENDL/A-2, and the FENDL/D-2 decay data library. The decay heat calculation results have been compared with experimental ones obtained at FNS-JAERI on samples irradiated for 5 min or 7 h in a 14 MeV neutron flux.

The discrepancy between the ANITA-4/M decay heat calculations and the 5 min experimental results is around 12% when using the EAF-4.1 activation library, whilst it is around 25–30% for EAF-97 and FENDL/A-2, 1 min after the end of the irradiation. From about 40 min cooling time, the three activation libraries give quite comparable results. For the three libraries, the discrepancy with respect to the experimental value decrease to around 8% for 1 h cooling time.

The discrepancy between the ANITA-4/M decay heat calculations and the 7 h experimental results is less than 3% both using the EAF-4.1 and the FENDL/A-2 activation libraries in the first few days cooling and it arise to about 5% and 8%, respectively, for FENDL/A-2 and EAF-4.1, for 100 days cooling.

For the shorter irradiation experiment, the result analysis clearly shows that including impurities the calculations provided decay heat closer to the experiments both for the FENDL/A-2 and the EAF-4.1 libraries. For the longer irradiation experiment, the result analysis indicates no significant differences between calculation with and without including impurities for cooling times higher than about 10 days. On the contrary, relevant discrepancies appear in the first few days cooling time. To improve calculation consistency with experimental results, the knowledge of the material impurities content is mandatory.

Finally, the discrepancies between calculations and experiments are within the experimental errors, except for the short cooling time periods, i.e., up to 40 min from the end of the irradiation. For that time interval, the EAF-4.1 provided better results than the FENDL/A-2 and the EAF-97. This can be explained mainly for the differences in estimating the V^{52} production. Indeed, the EAF-4.1 takes the reaction cross-sections for V^{52} production from the EFF-2.4 data file, whilst FENDL/A-2 and EAF-97 refers to ADL-3 and IRK data files, respectively.

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