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Validation of induced radioactivity calculations for candidate fusion materials through measurements in a graphite-centered assembly

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Abstract

Induced radioactivity measurements were carried out jointly by the USA and Japan in a graphite-centered assembly, in the framework of ITER Task T-218 entitled 'Shielding Blanket Neutronics Experiments'. An intense, accelerator-based D-T rotating target neutron source at JAERI, with a nominal intensity of 5×10^{12} s^{-1}, was used. Two locations, providing different neutron energy spectra, were chosen for irradiating samples of a range of materials of interest to ITER. Three independent experimental campaigns were conducted so as to accommodate a large number of samples, on one hand, and as many short and long half-life products as possible, on the other. The total neutron fluence ranged from 4.7×10^{11} to 1.5×10^{14} n cm^{-2}. Altogether, samples of Mg, Al, SiO_2, Ti, V, Cr, Mn, Fe, Co, Ni, FH82 steel, SS316LN steel (ITER grade), Cu, Zn, KCl, Zr, Nb, Mo, Ag, In, Sn, Dy, Ta, Hf, Re, Au, Ir, and Pb were irradiated. The irradiated samples were cooled for variable periods ranging from 30 s to 250 days before their decay gamma-ray spectra were counted on high purity intrinsic germanium detectors. The half lives of the observed radioisotopes have ranged from 18.7 s (^{46m}Sc from Ti) to 5.3 years (^{60}Co from Ni). The neutron energy spectra for the two locations were calculated using Monte Carlo code MCNP with FENDL-1 and ENDF/B-VI data libraries. The analysis of isotopic activities, expressed in Bq cc^{-1}, of the irradiated materials has been carried out using REAC-3 radioactivity code, with FENDL-2A and FENDL-1A activation cross-section and decay data libraries. Typically, C/E lies in a band of 0.5–1.5 for the results being reported.

1. Introduction

In the framework of ITER Task T-218, entitled 'Shielding Blanket Neutronics Experiments', induced radioactivity measurements were conducted jointly by the USA and Japan by irradiating small samples of a number of materials in a graphite-centered assembly. Major aims of this effort have consisted of compiling experimental database for measured radioisotopic activities, on one hand, and, their use in the validation of calculations, on the other, along the lines of work reported in [1–5]. Three independent experimental campaigns were conducted so as to accommodate a large

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number of samples, on one hand, and as many short and long half-life products as possible, on the other. In all, specimens of Mg, Al, SiO_2, Ti, V, Cr, Mn, Fe, Co, Ni, FH82 steel, SS316LN steel (ITER grade), Cu, Zn, KC1, Zr, Nb, Mo, Ag, In, Sn, Dy, Ta, Hf, Re, Au, Ir, and Pb were irradiated. The irradiated samples were cooled for variable periods before their decay gamma-ray spectra were counted on high purity intrinsic germanium detectors and converted to decay rate in Bq/cc. The half lives of the observed radioisotopes have ranged from 18.7 s (^{46m}Sc from Ti) to 5.3 years (^{60}Co from Ni).

Analysis of the radioactivity measurements has been carried out using state-of-the-art transport code, transport cross-sections, activation cross-section and decay data libraries, and calculations compared to the experimental measurements of decay radioactivity. Typically, C/E lies in a band of 0.5–1.5.

2. Experiments

A schematic cross-sectional view of the experimental assembly is shown in Fig. 1. The central zone of the assembly consisted of graphite blocks with a central channel hole of 50 mm diameter and a length of 300 mm. Six graphite blocks, each measuring 48 mm diameter by 48 mm length, were placed one after another in this channel. The cross-section of the central zone of graphite measured 150×150 mm. This region was enveloped by a $\sim$17.5 cm thick graphite shell and a 5 cm thick polyethylene layer. The outer dimension of the stainless steel 304 container measured $600 \times 600 \times 600$ mm. The experimental assembly was kept directly in front of the rotating target D-T neutron source at JAERI, at a distance of 14 mm. This D-T neutron source is accelerator-based and has a nominal intensity of 5×10^{12} s^{-1}. The axis of the central channel was aligned to the d^+ beam direction in order to take advantage of the geometrical symmetry during subsequent analysis phase of the experiments. Two locations were chosen for irradiating samples of a range of materials of interest to ITER. These locations provided different neutron energy spectra. The first location was directly after the first graphite block, at a depth of 50.2 mm in the central channel, and was identified as location A. The second location was directly behind the third graphite block, at a depth of 151.2 mm in the central channel, and was characterized as location B.

Samples of Mg, Al, SiO_2, Ti, V, Cr, Mn, Fe, Co, Ni, FH82 steel, SS316LN steel (ITER grade), Cu, Zn, KC1, Zr, Nb, Mo, Ag, In, Sn, Dy, Ta, Hf, Re, Au, Ir, and Pb were irradiated at the two locations. Three independent experimental campaigns were conducted so as to accommodate a large number of samples and also, as many short and long half-life products as possible. Table 1 lists information regarding the salient characteristics of the experiments. The first experimental campaign consisted of a short irradiation of 15 min followed by a long irradiation of 8 h. The second campaign was similar to the first one, except that different sets of samples were irradiated at the two locations. The third campaign was aimed at very short half life products, and was made up of 16 very short irradiations, each with a duration of 2 min. An individual irradiation consisted of a sample material foil accompanied by a monitoring aluminum foil. A pneumatic tube transported the foil set to just behind the first graphite block, corresponding to location A. The irradiated samples were cooled for variable periods ranging from 30 s to 250 days before their decay gamma-ray spectra were counted on high purity intrinsic germanium detectors. The half lives of the observed radioisotopes have ranged from 18.7 s (^{46m}Sc from Ti) to 5.3 years (^{60}Co from Ni). Measured decay gamma spectra were converted to decay rates for each radioisotope and expressed in units of Bq cc^{-1} of the irradiated material to enable subsequent, direct comparisons to the calculated decay rates.

3. Analysis

Neutron energy spectrum calculations were carried out using Monte Carlo code MCNP [6], with two cross-section libraries, ENDF/B-VI and FENDL-1 [7–9]. Calculated neutron energy spectra for both locations, i.e. A and B, are shown in

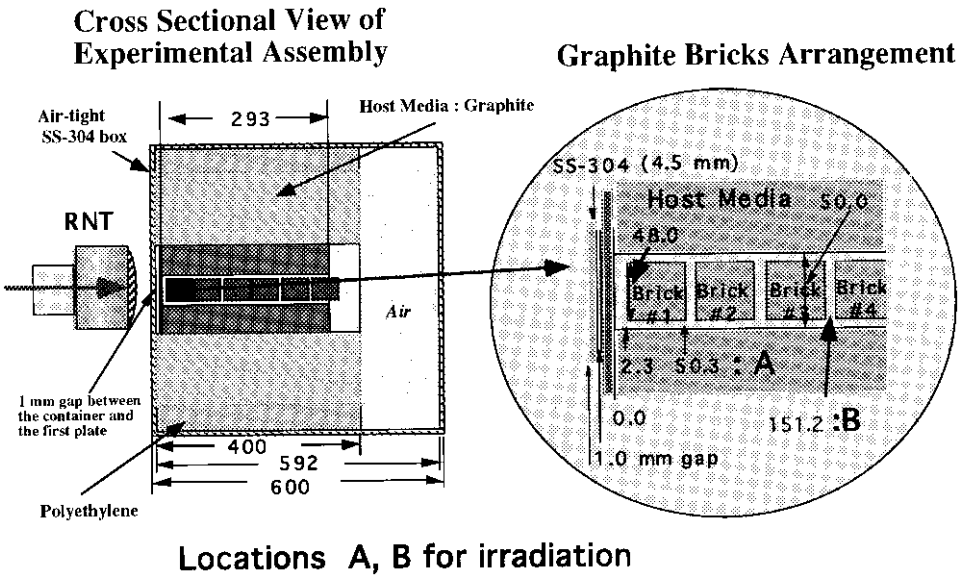

Fig. 1. Schematic representation of graphite-centered experimental assembly.

Table 1
Information on irradiation experiments in graphite-centered assembly

Assembly	Graphite-centered
Neutron source	Rotating target D-T neutron source at FNS, JAERI
Source neutron intensity	3×10^{12} s^{-1}
Irradiation locations	A: between 1st and 2nd graphite blocks, at 51 mm inside the central graphite region. B: between 3rd and 4th graphite blocks, at 152 mm inside the central graphite region
Irradiation periods	2 min, 15 min, 8 h
Total neutron fluence (calculated)	4.7×10^{11}–1.5×10^{14} n cm^{-2}
Cooling times of irradiated samples	30 s–250 days
Irradiated materials	Mg, Al, SiO_2, Ti, V, Cr, Mn, Fe, Co, Ni, FH82 steel, SS316LN (316 stainless steel), Cu, Zn, KC1, Zr, Nb, Mo, Ag, In, Sn, Dy, Ta, Hf, W, Re, Ir, Au, Pb
Half lives' range	18.7 s (^{46m}Sc from Ti)–5.3 years (^{60}Co from Ni)
Experimental error	Variable, largely controlled by counting statistics for a radioisotope of interest, from 1 to 20%

Fig. 2. There is a dominant D-T neutron peak around 14 MeV—the peak flux at A being almost an order of magnitude larger than that at B. However, at lower neutron energy, the spectra for the two locations get closer. Thus, B offers a significantly softer spectrum than A. In other words, isotopic activities produced by neutron capture reactions at the two locations will be much closer to one another than the activities generated by high energy neutrons. The calculated flux was then used, along with FENDL-1A [7–9] and FENDL-2A [7,8] activation cross-section and decay data libraries, as input to radioactivity code REAC-3 [10] to calculate isotopic activities in units of Bq cc^{-1}. A fine energy group structure of 175 neutron groups was chosen for the activation cross-section libraries [11]. Note that FENDL-2A is the most recent activation and decay data library released by IAEA [7,8,11]. The comparison of calculation to experimentally measured isotopic

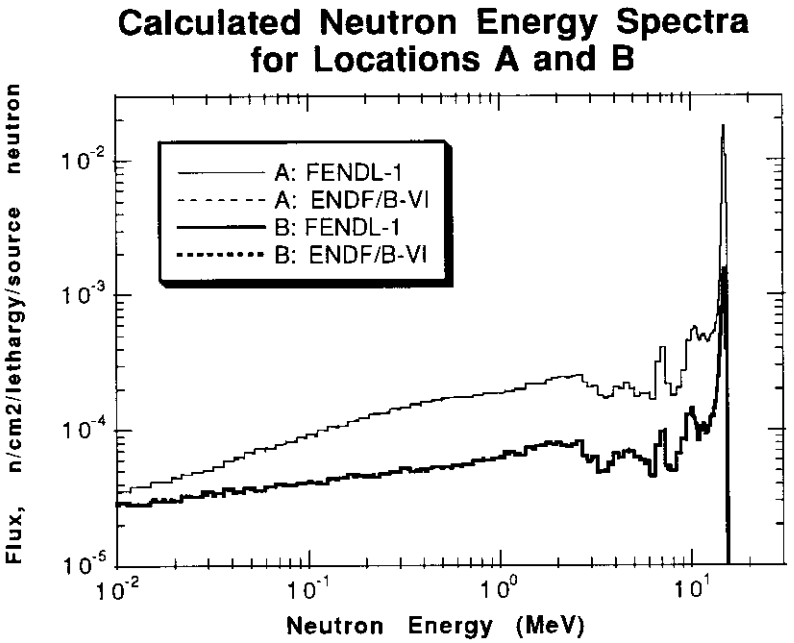

Fig. 2. Calculated neutron energy spectra (n/cm^2/lethargy) for irradiation locations A and B using Monte Carlo code MCNP with FENDL-1 and ENDF/B-VI libraries.

activities, i.e. C/E, was then obtained for all irradiated material samples. Experimental error was factored in while getting the C/E ratios—the experimental error being largely dominated by counting statistics for each individual radioisotope [1–5].

4. Results and discussion

Calculations of MCNP with FENDL-1 and ENDF/B-VI yielded same neutron spectra—within statistical error. This was confirmed by decay rate calculations with REAC-3. In addition, no observable difference was noticed between the calculated decay rates by FENDL-2A and FENDL-1A activation and decay data libraries for the materials analyzed. Hence, it was determined to focus entirely on the calculated results obtained using FENDL1 neutron spectra and FENDL-2A activation and data libraries while obtaining C/E ratios.

Fig. 3 is an overall plot of C/E ratios for isotopic products from some materials as a function of each material. The experimental error is less than 5% on the data points included in this plot. Data from remaining materials would be reported in future. The results for both of the locations, A and B, are included in the plot. Typically, C/E's lie in a band of 0.5–1.5. However, some data points for titanium (^{51}Ti), vanadium (^{51}Ti), and molybdenum (^{96}Nb) push C/E below 0.5, some data points for titanium (^{51}Ti), iron (^{53}Fe), cobalt (^{60m}Co), and nickel (^{61}Co) push C/E above 1.5.

Figs. 4–10 provide C/E data for radioisotopes of chromium, iron, nickel, molybdenum, FH82 steel, 316 stainless steel (SS316LN), and tungsten respectively. As for chromium in Fig. 4, ^{53}V, ^{52}V, ^{51}Ti, ^{49}Cr, and ^{51}Cr are included. C/E's greater than 1.1 are seen for ^{53}V, ^{52}V, and ^{51}Ti. The lowest C/E, 0.5–0.7, is seen for ^{49}Cr. Experimental data has considerable dispersion for ^{53}V and ^{51}Cr. As for iron (Fig. 5), C/E's appear to have tendency towards overprediction (>1), the products included being ^{58}Mn, ^{57}Mn, ^{53}Fe, ^{56}Mn, ^{51}Cr,

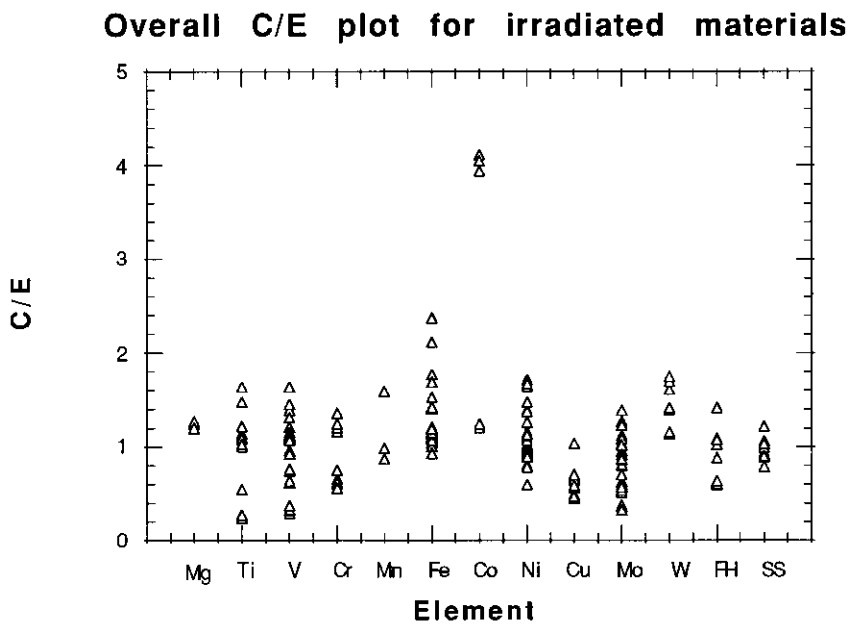

Fig. 3. Overall plot of ratio of calculation to experimental (C/E) measurement of activity of various radioisotopes produced by irradiation of different materials. REAC-3 code used with FENDL-2A activation cross-section and decay data libraries.

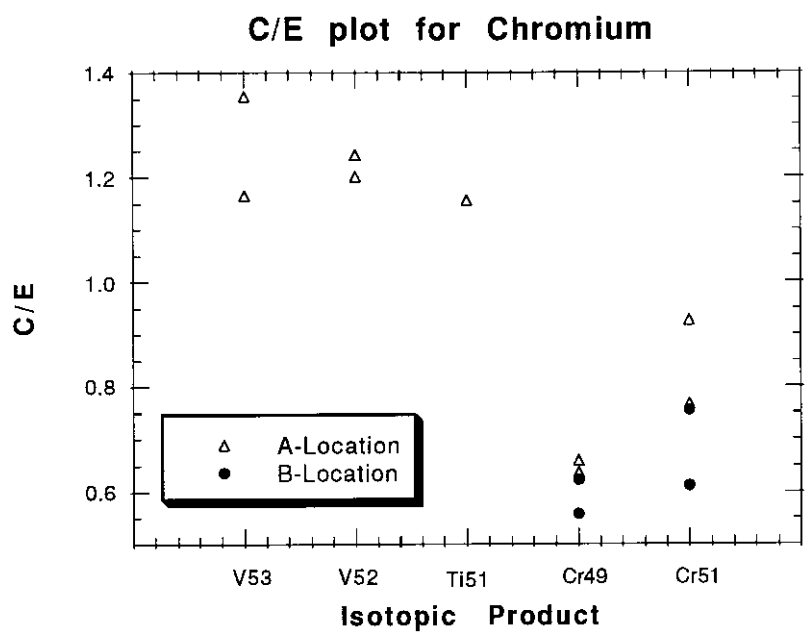

Fig. 4. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by the irradiation of chromium.

and ^{54}Mn. The largest C/E is observed for ^{53}Fe, the experimental data has quite large dispersion for this isotope. Nickel products span C/E's from 0.6 to 1.7, as seen in Fig. 6. Data for ^{62g}Co, ^{60m}Co, ^{62m}Co, ^{61}Co, ^{57}Ni, ^{58}Co, ^{57}Co, and ^{60}Co are covered. Largest C/E's are seen for ^{61}Co, whereas

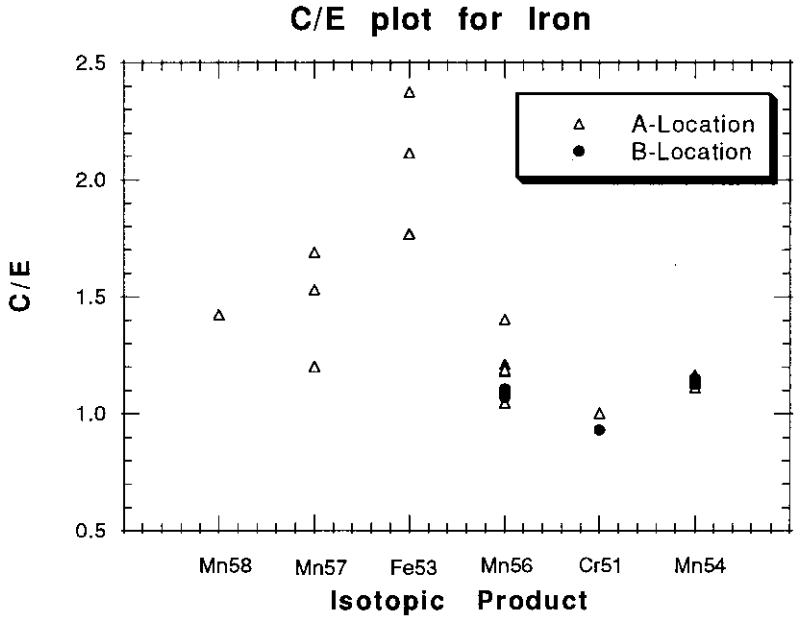

Fig. 5. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of iron.

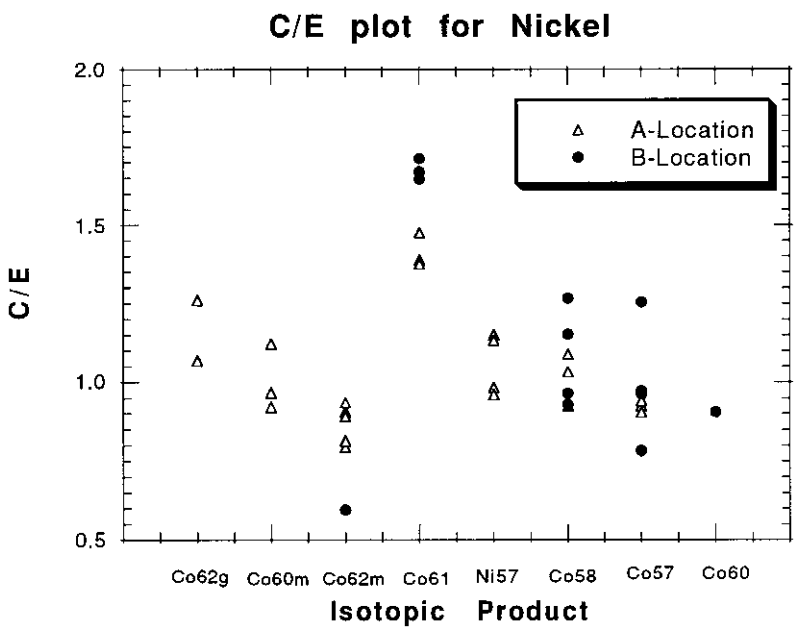

Fig. 6. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of nickel.

^{62m}Co has the lowest C/E corresponding to one data point. For molybdenum, C/E's are spread over a range of 0.35–1.4. The isotopes shown in Fig. 7 include ^{97m}Nb, ^{91m}Mo, ^{89m}Zr, ^{101}Mo, ^{98m}Nb, ^{97}Nb, ^{96}Nb, and ^{99}Mo. The lowest C/E's are seen for ^{96}Nb, whereas largest C/E's are found for ^{89m}Zr.

Fig. 8 is a plot of C/E's for FH82 steel. The

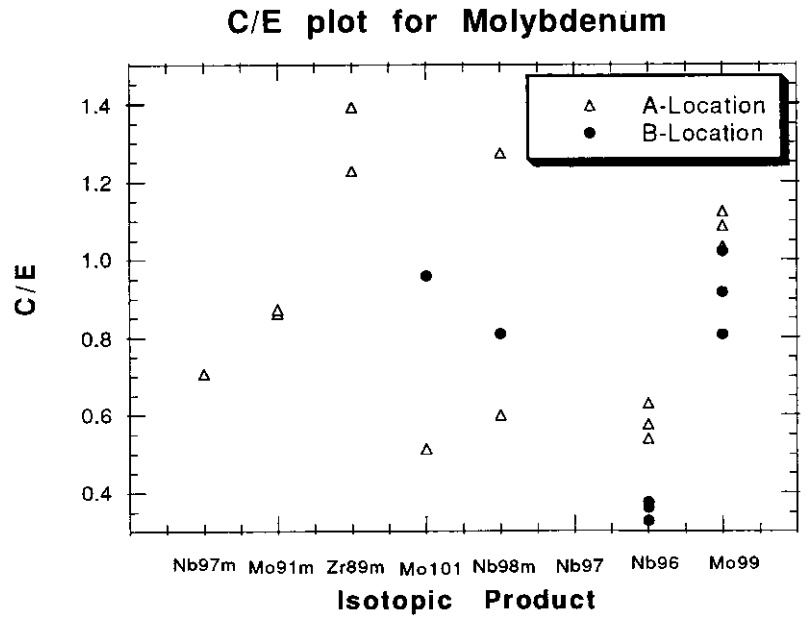

Fig. 7. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of molybdenum.

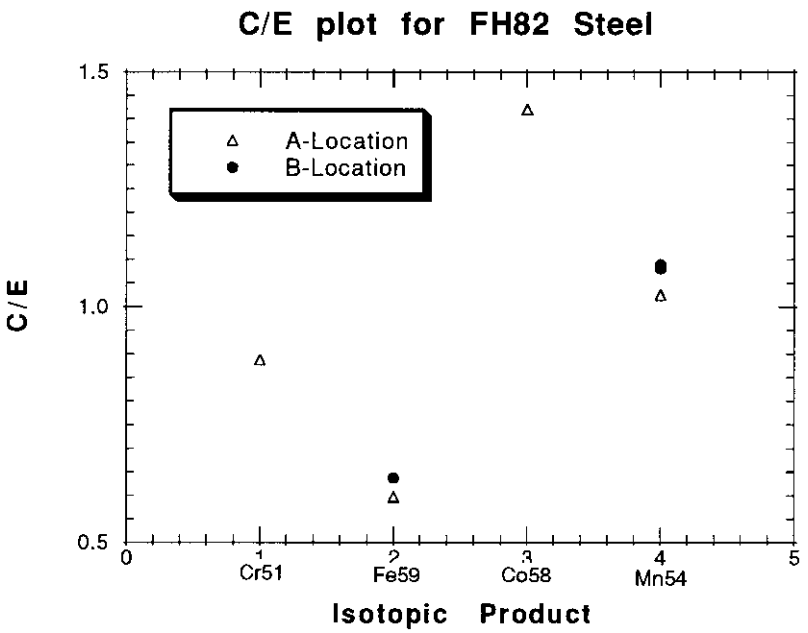

Fig. 8. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of FH82 steel.

products shown include ^{51}Cr, ^{59}Fe, ^{58}Co, and ^{54}Mn. C/E's for these products range from 0.9 (^{51}Cr) to 1.4 (^{58}Co). Fig. 9 is a C/E plot of 316 stainless steel (SS316LN). ^{92m}Nb, ^{51}Cr, ^{58}Co, ^{57}Co, ^{54}Mn, and ^{60}Co data are shown. C/E's range is from 0.8 to 1.2. Tungsten data are in Fig. 10.

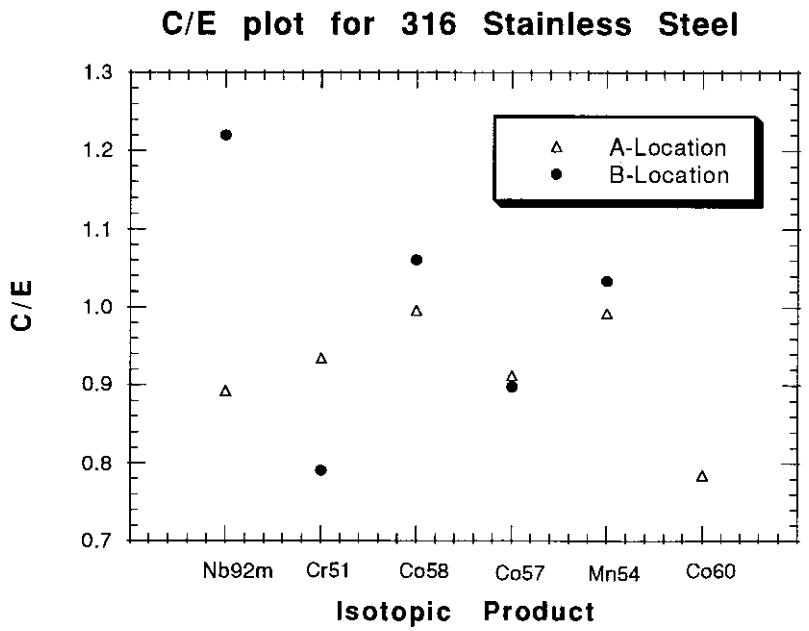

Fig. 9. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of 316 stainless steel (SS316LN).

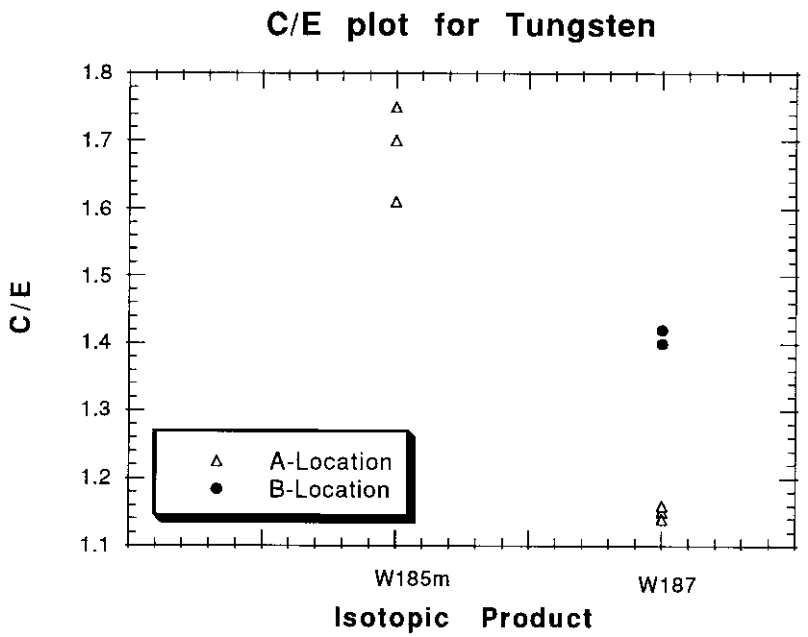

Fig. 10. Ratio of calculation to experimental (C/E) measurement of activity of the radioisotopes produced by irradiation of tungsten.

Only two isotopes are shown, i.e. ^{185m}W and ^{187}W. C/E's range from 1.15 to 1.75. C/E's for ^{185m}W are rather high.

5. Summary and conclusions

Induced radioactivity experiments were con-

ducted on a number of materials of interest to ITER in a graphite-centered assembly in the framework of ITER Task T-218. Samples of Mg, Al, SiO_2, Ti, V, Cr, Mn, Fe, Co, Ni, FH82 steel, SS316LN steel (ITER grade), Cu, Zn, KC1, Zr, Nb, Mo, Ag, In, Sn, Dy, Ta, Hf, Re, Au, Ir, and Pb were irradiated. The total neutron fluence ranged from 4.7×10^{11} to 1.5×10^{14} n cm^{-2}. The samples were cooled for varying periods, from half a minute to almost 9 months. Decay gamma-ray activity of each material was obtained in units of Bq cc^{-1} from the experiments, and was compared to the calculations using REAC-3 radioactivity code with two different activation libraries, i.e. FENDL-1A, and FENDL-2A. The decay activities calculated with the two libraries were found to be same. Typically, the ratio of calculation to the experimental measurement of the isotopic activity is between 0.5 to 1.5. Deviations from this range were found for a number of materials that include titanium, vanadium, iron, nickel, and molybdenum. It should be noted that these are important components of one or the other kind of steel suggested for use as a structural material. Further improvement in C/E values for the discrepant isotopic activities would need two-fold effort—additional experiments, on one hand, and activation data library improvement, on the other.

Acknowledgements

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