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Journal of Nuclear Materials 329–333 (2004) 1147–1150

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Impact of transmutation issues on interpretation of data obtained from fast reactor irradiation experiments

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Abstract

The subject of fission–fusion correlation is usually cast in terms of reactor-to-reactor differences, but recently the fusion community has become aware of the impact of differences within a given surrogate facility, especially in constant time experiments when different dose levels are attained in different positions of one reactor. For some materials, it is not safe to assume that in-reactor spectral variations are small and of no consequence. This point is illustrated using calculations for fusion-relevant materials that were irradiated in the Fast Flux Test Facility–Materials Open Test Assembly (FFTF–MOTA) over a wide range of in-core and out-of-core positions spanning more than two orders of magnitude in dpa rate. It is shown that although both the neutron spectrum and flux changes, the spectral effectiveness factor, dpa/10^{22} n/cm^2 ($E > 0.1$ MeV), remains remarkably constant over this range. The transmutation rate per dpa varies strongly with reactor position, however.

1. Introduction

The use of surrogate irradiation facilities, especially fission reactors, to simulate fusion environments requires that attention be paid to the trade-offs and penalties associated with the differences in neutron spectra and fluxes between the two environments. A variety of mixed spectrum reactors, such as the High Flux Isotope Reactor (HFIR), and fast reactors, such as FFTF and the Experimental Breeder Reactor (EBR-II) have been used to simulate fusion reactor environments. Although these facilities have very different neutron spectra, the concept of displacements per atom (dpa) has generally been very successful in correlating material effects between different reactors. However, some caution is needed since transmutation effects can be very different not only between facilities, but more importantly within a given reactor environment due to neutron spectral variations. Frequently, data are derived from various positions in a given reactor, each at a different dpa rate. If the radiation-induced property change of interest depends not only on the accumulated dpa level, but also on the dpa rate and/or transmutation-induced changes in composition, it is not valid to treat the data derived from such 'constant time' experiments as representing the response to a single variable. For instance, it has recently been shown that constant time FFTF–MOTA experiments on void swelling of Fe–Cr–Ni alloys which do not transmute strongly, are significantly impacted by differences in dpa rate [1,2]. In some cases the effect of dpa rate can overwhelm the impact of the dpa [3]. However, other fusion-relevant materials such as Cu, W, Re and V are known to transmute strongly, even in fast reactor spectra, requiring that the influence of spectral variations be examined more closely.

We have previously used the concept of the spectral effectiveness factor to characterize the neutron spectra. This factor is simply the number of dpa produced by the neutron fluence above 0.1 MeV in units of 10^{22} n/cm^2. However, in irradiations involving materials with

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doi:10.1016/j.jnucmat.2004.04.272

relatively high thermal or epithermal neutron cross-sections, transmutation may be so sensitive to small changes in the neutron spectrum that transmutation effects may be significant even though the spectral effectiveness factor shows little change. This point is easily illustrated by comparisons of spectral effectiveness factors with transmutation rates for various materials irradiated at different locations in FFTF–MOTA assemblies.

2. Results and discussion

2.1. Irradiations in FFTF

MOTA irradiation experiments were conducted in FFTF from 1983 through 1992. Calculations discussed in this paper were derived from reactor dosimetry measurements performed in the MOTA-2A experiment which was conducted from January 1990 to March 1991 for a total of 299.7 effective full power days (EFPD) at an average power level of 291 MW and for the MOTA-2B experiment from May 1991 to March 1992 for 203.3 EFPD. Irradiation histories for all of the FFTF experiments have been previously published [4]. Reactor dosimetry capsules were placed at various elevations in all of the MOTA experiments. Each capsule contained a number of monitors that were analyzed to determine reaction rates which were used to determine the neutron fluence spectrum at each point in the MOTA assemblies. The measurements and adjusted neutron spectra for the MOTA-2A and -2B experiments were published previously [5,6]. The adjusted neutron fluence spectra were used to determine the neutron fluences, dpa, and transmutation values shown in this paper. Neutron spectra are shown in Fig. 1 for above-core, midplane, and below-core positions in the MOTA-2A experiment. The neutron spectra are softer at the out-of-core positions and the increased neutron flux in the epithermal region can produce enhanced transmutation rates, as discussed later. The neutron spectra in the below-core (−62.9 cm) and above-core (+66.6 cm) locations have more lower-energy neutrons, compared to that of in-core locations, especially in below-core locations. This difference produces essentially the same dpa per fast neutron but very different transmutation rates in each location.

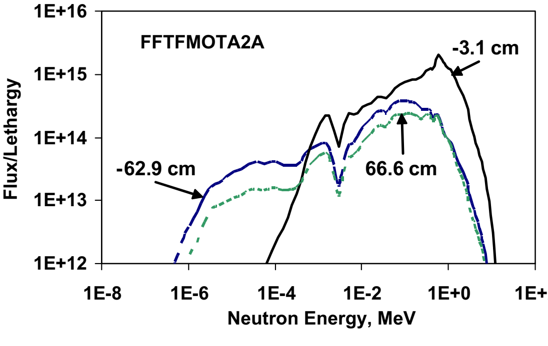

Fig. 1. Neutron flux spectra are shown at the below-core (−62.9 cm), level 3 (−3.1 cm), and above-core (66.6 cm) positions in the FFTF MOTA-2A and 2B assemblies. Note the increased epithermal flux at out-of-core positions, especially below core.

2.2. Transmutation and spectral effectiveness ratios

The dpa in 304 stainless steel as well as spectral effectiveness factors, defined by the ratio of the dpa produced by the neutron fluence above $E > 0.1$ MeV are shown for V, Cu, W and 304 stainless steel in Fig. 2. Note that although the dpa rates are changing nearly two orders of magnitude, as shown in Fig. 3, the spectral effectiveness factor is rather constant. The scatter in the ratio is most likely due to uncertainties and variations in the local neutron fluence spectra. The dpa values for various elements depend on the threshold energies and generally decrease with the mass of the recoil atom.

Transmutation rates were calculated by integrating neutron activation cross sections from ENDF/B-VI over the adjusted neutron flux spectra, such as those shown in Fig. 1, as determined by neutron dosimetry measurements [5,6]. The transmutation rates show significant differences at different elevations in the MOTA assemblies, as shown in Figs. 4 and 5. This effect is most pronounced for elements that have significant epithermal neutron cross-sections for transmutation, as shown for the transmutation of Re to Os and the burnup of boron in Fig. 4 (top). Similar dramatic differences are shown for the transmutation of V to Cr and W to Re in Fig. 4 (bottom). The transmutation effects are shown as a function of dpa in

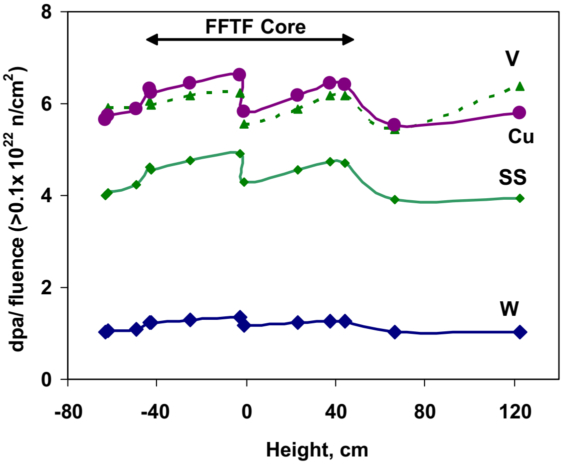

Fig. 2. The spectral effectiveness factors for 304 stainless steel, V, Cu, and W in terms of dpa per 10^{22} n/cm^2 ($E > 0.1$ MeV) as a function of height in FFTF MOTA-2A. Note that the ratio is nearly constant in spite of significant dpa, flux and spectral changes.

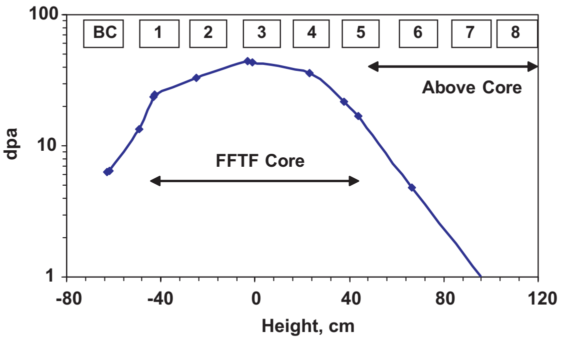

Fig. 3. Dpa rate variation as a function of axial position. The MOTA-2A capsule positions where irradiations are conducted in, below and above core are shown at the top of the figure.

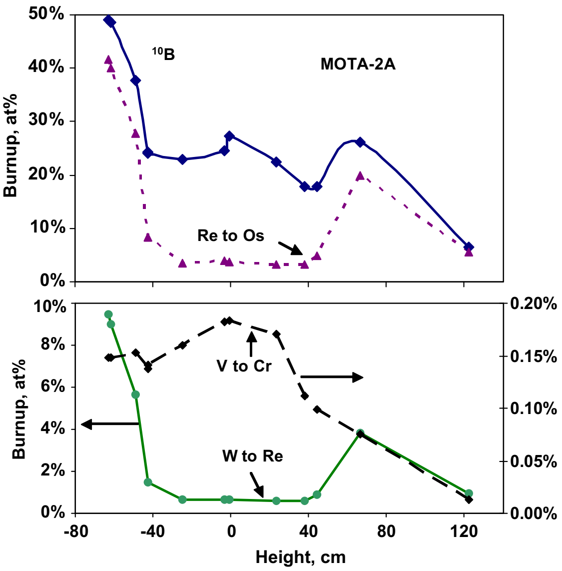

Fig. 4. Burnup of ^{10}B and transmutation of V to Cr as a function of height in the FFTF MOTA 2A experiment. Also shown are the transmutation of Re to Os and W to Re.

Fig. 5 which shows that samples having the same dpa exposure can have very different levels of transmutation. Fig. 6 shows that the He to dpa ratio for the alloy Fe–15Cr–16Ni similarly shows a significant difference between the below-core and above-core positions due to the extra helium produced by the well-known Ni-58, Ni-59 reaction series [7], arising from the increase of epithermal neutrons in the below-core position.

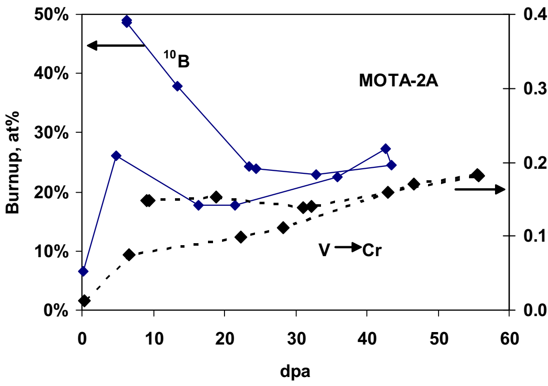

Fig. 5. Burnup of ^{10}B and transmutation of V to Cr as a function of dpa in the FFTF MOTA 2A experiment. For boron the dpa level used is that of the stainless steel in which the boron was dissolved.

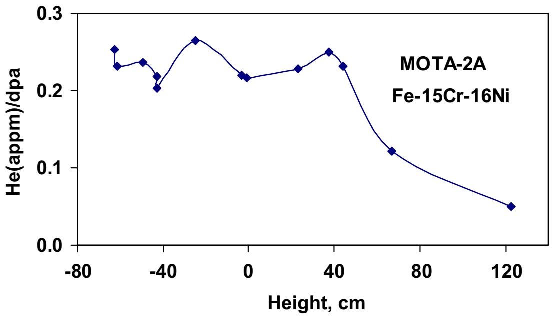

Fig. 6. He/dpa ratio for Fe–15Cr–16Ni as a function of height in the FFTF MOTA-2A experiment. The increased helium generation rate at below-core positions, compared with that above core, arises from the increased influence of the ^{59}Ni reaction.

2.3. Impact of transmutation on the interpretation of materials experiments

The impact of transmutation on materials experiments must be carefully evaluated since transmutation can lead to significant changes in materials that may well be more important with respect to a material property than the displacive effects of irradiation. Such effects have been shown in previous publications for Cu [8,9], V [10,11], Re [12], W [13], Mo and others [14,15]. In some cases such as in V and Re, the transmutation leads to significant changes in lattice parameter, obscuring accurate measurement of void swelling. In another case, the production of Cr in V leads to an acceleration of swelling relative to that of pure V.

In other cases the effect of two spatially dependent variables such as dpa rate and transmutation rate can compete in opposite directions so as to obscure the action of either one. A good example is provided in another paper in these proceedings concerning swelling of simple Fe–Cr alloys [16]. Whereas a strong effect on swelling of dpa rate differences between in-core regions of EBR-II and FFTF was previously observed on swelling of Fe–Cr binary alloys, no effect of dpa rate was observed when the experiment was conducted only in FFTF [17,18]. In this case the progressive spectral softening near the edge of the FFTF core leads to lower helium generation rates, unlike the behavior shown in Fig. 6 for nickel-bearing alloys. Therefore, the tendency for lower dpa rates to shorten the transient regime of swelling is countered by the concurrent lower helium generation rates, producing an apparent independence of swelling on dpa rate.

3. Conclusions

Consequently, caution is required in the interpretation of experiments conducted at different dpa rates in the same reactor since transmutation effects may invalidate the assumption that the dpa and dpa rate are the only significant variables operating in such experiments.

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