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Compositional optimisation of silicon carbide for various fusion blanket designs

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Abstract

Recent changes to the European activation file cross-section library have prompted a re-evaluation of the neutron activation response of SiC. The new data suggest that both short-term and long-term activation levels are slightly improved compared with previous library releases. Using this new data, a compositional optimisation methodology based on real measurements is described, and a fully specified SiC composition calculated. The optimised composition is believed to be a compromise between a material that can be realistically fabricated while retaining much of the excellent radiological properties of pure SiC. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Within the Fusion community, there is a growing interest in composite material made from silicon carbide fibres in a silicon carbide matrix. Much of this interest stems from its inherently low remnant activation characteristics following exposure to fusion neutrons. Previous work [1] has indicated that SiC exhibits extremely attractive short-term residual activation properties in all measures examined. At longer time-scales, however, activation characteristics were found to be inferior to both vanadium alloys and reduced activation martensitic steels. Based on these observations, SiC would thus appear to be an ideal material if accident safety was a priority design issue, but not as well suited for recycling and waste management options as the other two low activation alternatives.

In recent years, two new developments have occurred prompting the need for a re-evaluation of SiC activation response. The first of these involves continuing improvements made in nuclear cross-section data derived from both theoretical and experimental sources. Cross-section data changes for target nuclides in the near vicinity of silicon and carbon are expected to lead to altered activation responses. The second development

centres on the use of a new tool in neutron activation analysis concerned with compositional specification. It is well established that tramp impurities are often the cause of degraded activation properties in otherwise excellent materials. The computer code OPTIMAT can be used to determine a chemical composition of any material that minimises the activation behaviour under a number of prescribed criteria. These new aspects of SiC activation will be examined and discussed in detail.

2. Activation of pure SiC

A number of variant plant model concepts using SiC as either a structural material or as a coolant channel flow liner were investigated as part of the SEAFP-99 study [2]. Neutronic and activation analyses were performed for all these variants, but we present here a single subset of this data for simplicity. Neutronics analyses were made for the entire PM-4 power plant model concept [3] using two neutron transport codes. The PM-4 concept employs a silicon carbide first wall and blanket together with an austenitic stainless steel for in-vessel shield and all ex-vessel structural components. For in-vessel components the Monte Carlo code MCNP-IV was used to produce energy spectra in the 175 Vitamin J group structure covering the range 10^{-5} eV–20 MeV. Ex-vessel analyses were made using the discrete-ordinates code, ANISN. For the purpose of the

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present study, we confine further analyses to the first wall position of the power plant, since this produces the hardest and most intense neutron spectrum.

Activation calculations were made using the EASY-99 system [4]. This comprises the FISPACT-99 neutron activation code [5] and the most comprehensive nuclear data libraries yet formulated, as part of the European Activation File – EAF-99 [6]. These libraries contain improved data for over twelve and a half thousand reactions, based on updated experimental measurements and new theoretical assessments, of which several thousand have been changed from earlier versions. In addition they include new sources of data, cover reactions involving actinides and contain uncertainty information on cross-section values, which can be used in inventory codes, but are not included here. As such, many of the cross-sections pertinent to the SiC system have altered from earlier versions of the library. To indicate these changes, calculations are presented comparing activation analyses made using EAF-99 and its antecedent EAF-3.1 (released in mid-1993) which is more directly equivalent to the FENDL/A-2.0 used by some other workers.

Theoretically pure SiC contains 30.1 wt% carbon and 69.9 wt% silicon. Activation of these two elements yields a number of daughter nuclides with decay half-lives ranging from milliseconds to millions of years, different combinations of which are important for the various time-dependent activation response properties. There is clearly a strong influence of cross-section library on activation response as can be seen for the important radiological measure of contact γ dose rate (other activation properties exhibit comparable changes) shown in Fig. 1. Here, four radionuclides – all derived from the ^{28}Si parent – dominate the activation response at different post-irradiation time-scales. While there is no significant variation in the generation of ^{28}Al between the two cross-section libraries, the abundance of both

^{24}Na and ^{26}Al is reduced by ~ 5 –6 times due to lower cross sections in the new library. Conversely, production of ^{22}Na is some 20 times higher in EAF-99, due in part to the successive ‘amplification’ effect in the three-step pathway.

3. Compositional optimisation

Although pure SiC possesses excellent low activation properties, it is important to consider the impurity elements inevitably present. Tolerable concentrations for these impurities need to be set at levels that do not impinge dramatically upon the activation response of the pure material, while at the same time are still credible from the manufacturing feasibility viewpoint. This can be accomplished to some extent by using the OPTIMAT computer code, for which we describe the essential algorithm below. Beginning with pure SiC, the remaining stable elements can be assigned maximum concentration limits based on the following methodology.

Using the same neutronics and activation systems described in Section 2, 83 files containing activation properties for all terrestrially occurring elements are produced. Each file consists of a rectangular j by k matrix, $A_{j,k}^i$, calculated for the i th chemical element, where j denotes the following six different activation properties (specific activity, decay power, dose rate, ingestion hazard, inhalation hazard, and clearance index) and k is the number of discrete decay time intervals at which the activation property is calculated. In the present study k ranges from 1 s to 5×10^6 years (in 43 steps), to cover all post-shutdown times of interest.

Next, the material to be optimised is chemically specified. This involves assigning the concentrations of the known elements in the given material. For a pure sample this would include simply the elements C and Si, but for a sample with some more detailed chemical analysis data available all the specified/measured impurities could also be added. To aid understanding of the optimisation algorithm, we will consider the pure case only, i.e., a material containing 30.1% and 69.9% of carbon and silicon, respectively.

The initial specification in weight fraction is represented by an 83-element vector quantity f^i , with all vector elements set to 0, except for carbon – set to 0.301 and silicon – set to 0.699. Clearly, the sum of the weight fractions should yield unity,

$$\sum_i f^i = 1. \quad (1)$$

Next, each of the 83 file matrices $A_{j,k}^i$ is cross-multiplied by its corresponding weight fraction in the f^i vector to yield another set of j by k matrices for each element,

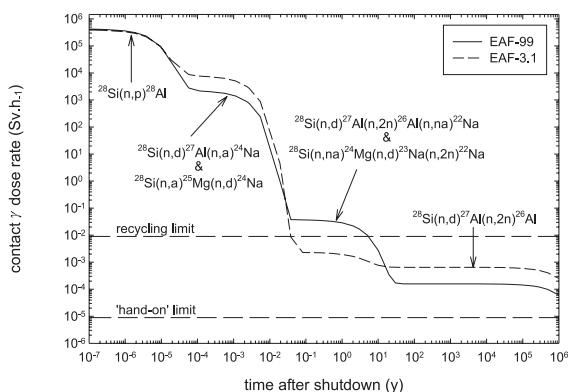


Fig. 1. Contact γ dose response of pure SiC in a first wall position.

$$P_{j,k}^i = f^i \times A_{j,k}^i. \quad (2)$$

Each new matrix quantity, $P_{j,k}^i$, provides the ‘partial’ contribution that the weight fraction of an initially specified element can contribute to the activation properties of the whole material. This implies that an impurity present at the 1 ppm level can only contribute 10^{-6} of the activation property compared with a material containing 100% of the same element.

For pure silicon carbide, the sum of the partial contributions from the carbon and the silicon components define the overall activation response of the mixture. In other words, the total time-varying activation response is given by another single j by k matrix $A_{j,k}^{\text{mat}}$ as follows:

$$A_{j,k}^{\text{mat}} = \sum_i P_{j,k}^i. \quad (3)$$

We next determine the limiting concentration levels for the remaining 81 elements not already specified in f^i . This begins with an operation whereby every matrix element in $A_{j,k}^{\text{mat}}$ is divided by its corresponding (same position) matrix element in each of the $A_{j,k}^i$ matrices. A concentration ratio matrix in j – k space may be defined as

$$R_{j,k}^i = \frac{A_{j,k}^{\text{mat}}}{A_{j,k}^i}. \quad (4)$$

For any given $R_{j,k}^i$, there exists some position in the j – k matrix where a minimum value occurs. Here, the minimum value in each $R_{j,k}^i$ is extracted and written into a new vector quantity M^i .

$$M^i = \text{Min}[R_{j,k}^i]. \quad (5)$$

We choose to extract the minimum value from $R_{j,k}^i$ in order to be as conservative as possible.

M^i is therefore a list of concentrations (in weight fraction) for all 83 elements similar to f^i except that there are now no vector elements with 0 values. As a point of detail, we do *not* allow the initially specified elements (C and Si) to be overwritten in the new M^i vector; but allow them to retain their initial values.

The importance of M^i can be fully appreciated when one examines the next logical extension of its use. Suppose that the $R_{j,k}^i$ matrix for an element i has a minimum concentration value of x wppm and that this value occurs at a single matrix position j, k . By definition, all other matrix positions will have a higher concentration value, and the single minimum value will be transferred to M^i . If we now add x weight fraction of element i to pure SiC, the additional activation contribution from i will be 100% at the corresponding j, k matrix position in the $A_{j,k}^{\text{mat}}$ matrix. In other words, for one value in j – k space the impurity i will double the activation response. Since a doubling of the activation response at this one

matrix element is probably too extreme, we factor in an additional term, such that

$$C^i = \alpha M^i. \quad (6)$$

Here, α is assigned a value of 0.1, meaning that the allowable concentration is set at 10% of that calculated from Eq. (5). This simple expedient is introduced to prevent the (highly unlikely) situation arising where several impurities could reinforce at the same j – k matrix element, each adding its full 100% contribution and thus seriously degrading the activation response of the entire material. Clearly the value for α can be altered as required and has no fundamental basis. The new vector quantity C^i thus gives a concentration limit for each chemical element in the host SiC material. As before, any chemical elements specified initially are not overwritten in C^i .

The next stage of the optimisation algorithm involves an iterative process where each of the remaining chemical elements is added to the host material in stages. This process begins by first examining the C^i vector in order to determine the identity of the i th single most restrictive chemical element (MRE), i.e., that element with the lowest calculated concentration. The concentration value of the MRE is next compared with the corresponding real chemical analysis measurements on SiC made by Scholz et al. [7]. It is nearly always the case that the MRE value for the i th element is many orders of magnitude lower than the real chemical analysis measurement, thus indicating that the theoretical calculation is unrealistically low. In this case, the measurement due to Scholz et al. is obviously a better estimation of a typical concentration for the i th element. We modify the starting composition, f^i , such that it now contains carbon, silicon and the i th element at the concentration indicated by Scholz et al. measurements. By adding in i at this ‘high’ concentration, the activation characteristics of the SiC material are significantly degraded. Consequently, the theoretical concentration levels calculated in Eqs. (1)–(6) for the remaining 80 elements are relaxed upwards, i.e., the concentrations for these elements are less restrictive than before.

We iterate again, determining the next (second) MRE, again comparing its value against real chemical analysis measurements. On each new elemental addition to f^i , the activation properties for the SiC material degrade further, and levels on remaining concentration relax upwards. We continue this process a number of times adding each new element to the starting composition at each cycle. At some point we find that the theoretical calculation for the MRE is comparable or higher than the real measurements and can therefore be used instead of Scholz et al. values. Convergence to a position where additions of new impurities no longer perturb the activation responses is reached after typically 10–20

iterations, at which point the procedure is terminated and we achieve a final C^i matrix. The resulting specification thus contains the full range of elements under consideration, and represents a compromise between a theoretically specified ‘lowest activation’ material with one containing realistic levels of impurities based on real measurements. The ‘optimised composition’ for the SiC materials is listed in Table 1.

4. Activation of optimised SiC

Using the optimised SiC composition presented in Table 1, the activation response at the first wall is re-calculated and shown in Fig. 2. Given that the optimised SiC material contains impurities at levels that can be justified by chemical analyses [7], and could therefore be realistically fabricated, it is noteworthy that there is still significant radiological deterioration for this material when it is compared with pure SiC. This reflects the superior quality of pure SiC rather than any deficiencies in the optimisation methodology. It should be clearly stated that the optimised SiC composition (or something that closely resembles it) represents the best that can be feasibly achieved industrially.

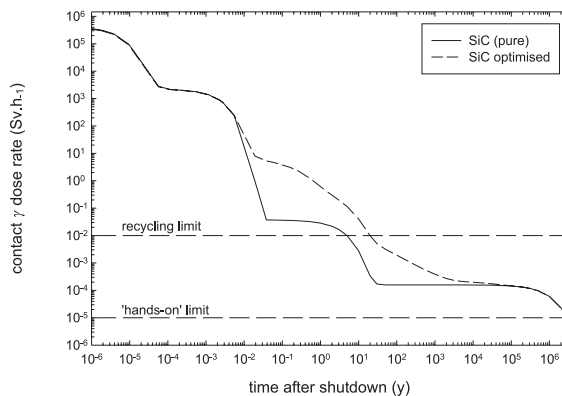


Fig. 2. Contact γ dose rate responses of pure and compositional optimised SiC in a first wall position.

5. Summary and conclusions

Re-evaluation of the activation response of pure SiC using the new EAF-99 cross-section library has highlighted differences in many radiological properties (in particular contact γ dose rate) when compared with earlier versions of the library. Additional analyses of

Table 1
Optimised chemical composition for SiC

Element	C^i (wt%)	Element	C^i (wt%)	Element	C^i (wt%)
H	1.00E-02	Cu	4.95E-05	Ce	7.79E-04
He	1.00E-02	Zn	5.54E-05	Pr	2.34E-03
Li	3.88E-05	Ga	3.10E-04	Nd	6.62E-04
Be	9.36E-03	Ge	8.74E-03	Sm	1.00E-07
B	3.85E-03	As	1.76E-04	Eu	4.00E-07
C	3.01E+01	Se	1.69E-03	Gd	1.30E-04
N	5.23E-04	Br	9.54E-05	Tb	1.00E-05
O	4.00E-00	Kr	3.78E-04	Dy	1.20E-04
F	1.06E-02	Rb	1.87E-04	Ho	5.00E-05
Ne	1.00E-02	Sr	2.55E-03	Er	3.35E-04
Na	1.06E-04	Y	5.00E-05	Tm	1.00E-04
Mg	3.25E-03	Zr	7.90E-04	Yb	2.94E-03
Al	1.06E-03	Nb	5.00E-05	Lu	1.48E-04
Si	6.58E+01	Mo	8.44E-04	Hf	8.48E-05
P	2.82E-02	Ru	3.73E-04	Ta	1.00E-06
S	7.18E-04	Rh	5.00E-05	W	5.29E-04
Cl	1.06E-03	Pd	2.22E-04	Re	3.40E-04
Ar	1.76E-04	Ag	4.14E-05	Os	4.05E-04
K	5.36E-05	Cd	1.13E-03	Ir	5.41E-05
Ca	1.34E-03	In	2.02E-04	Pt	6.12E-04
Sc	1.00E-07	Sn	1.93E-03	Au	2.16E-04
Ti	8.51E-04	Sb	5.30E-06	Hg	7.08E-03
V	2.31E-02	Te	8.75E-04	Tl	2.30E-04
Cr	5.87E-03	In	6.04E-04	Pb	1.42E-01
Mn	5.00E-05	Xe	6.33E-04	Bi	5.33E-05
Fe	3.80E-04	Cs	1.50E-06	Th	5.00E-05
Co	4.00E-07	Ba	7.43E-04	U	4.00E-06
Ni	5.00E-05	La	4.21E-04		

other plant model concepts, although not presented here, show similar trends.

Reductions in the magnitude of ^{24}Na generation as a result of using the new library indicate that the short-term activation response of pure SiC is even better than earlier calculations would suggest. This finding reinforces the notion that pure SiC is a good candidate material when accident safety considerations are paramount. Incorporation of impurities is unlikely to alter this observation notably, since their influence is not significant in the optimised composition until at least 1 day post shutdown.

Similar reductions in ^{26}Al generation are observed in using the new library. These changes will enhance the long-term activation properties of pure SiC, resulting in modest improvements regarding recycling and waste management options. The addition of impurities will degrade the radiological properties of the optimised material to some extent, but the time to cross the recycling limit for first wall material will still be feasible after 20 years.

At intermediate times, an increase in ^{22}Na production in pure SiC is noted from use of the EAF-99 library. However, this small enhancement will be completely masked by increased activation from impurity elements occurring over much of the same time interval.

Despite efforts to minimise impurity levels during the optimisation process, activity levels are still significantly elevated over that of the pure material in the time interval between ~ 1 day and 5×10^4 years. Nevertheless,

the composition suggested for the optimised SiC is believed to be the best that can be feasibly achieved from the manufacturing perspective.

Acknowledgements

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