

ACTIVATION OF SILICON CARBIDE IN FUSION ENERGY SYSTEMS - H. L. Heinisch, Pacific Northwest Laboratory*

OBJECTIVE

The objective of this work is to review the information on activation of silicon carbide with respect to use as a low activation material in fusion energy systems.

SUMMARY

Because of production of ^{26}Al from Si, SiC irradiated in a fusion energy system first wall exceeds the limits for shallow land burial, based on 10 CFR 61, Class C, for irradiation doses typical of a first wall component service lifetime in DEMO, 12.5 MWy/m². However, if first wall activities can be averaged over entire components that include portions within the fusion machine where fluxes of high energy neutrons are smaller than at the first wall, production of ^{26}Al may stay under the shallow land burial limit for practical component service lifetimes. Realistic information on energy system design, waste disposal criteria, and decommissioning procedures is necessary to determine with certainty the role SiC can play as a low activation fusion energy system material. Sequential charged particle reactions have no significant effect on the residual radioactivity of SiC irradiated in a fusion energy system first wall.

PROGRESS AND STATUS

SiC as a Low Activation Material. Silicon carbide has enjoyed a reputation as a potential low activation structural material for fusion energy systems for about the past 15 years¹. Although there was uncertainty about the usefulness of monolithic SiC associated primarily with its being a brittle ceramic, lately, silicon carbide fiber-reinforced silicon carbide composites (SiC/SiC) with improved fracture toughness offer the promise of acceptable mechanical performance. In 1983, the report of a DOE panel on low activation materials for fusion applications² considered SiC to be a "very low activation" material (indeed, the panel's reference "low activation" Tokamak design consisted almost entirely of Al alloy and SiC). However, since at least 1988, the long term environmental limitations of neutron activated SiC, due to the production of the radioisotope ^{26}Al , have been well documented³⁻⁷.

Environmental Limits on ^{26}Al . It is likely that ^{26}Al (from irradiation of either Al or Si) was not recognized as a limited radionuclide earlier because it was not included in the original list of the dozen or so radionuclides in the 10 CFR 61 limits for Class C shallow land burial. In 1985, Maninger³ calculated an estimated Class C limit of 0.1 Ci/m³ for ^{26}Al using modified versions of the NRC computer codes. In 1988, Fetter, Cheng and Mann⁴, using the same methodology as the NRC, but somewhat different criteria, determined a limit of 0.09 Ci/m³. Thus, although 10 CFR 61 does not explicitly place limits on ^{26}Al , regulations similar to 10 CFR 61, applying to activated materials in fusion energy systems, will likely have such limits. It is also possible that regulations applying to shallow burial of fusion energy system wastes will be more conservative than 10 CFR 61, which was not originally developed for waste streams containing large amounts of activated structural materials.

Production of ^{26}Al . In neutron irradiated Al, ^{26}Al is made directly in the reaction $^{27}\text{Al}(n,2n)^{26}\text{Al}$, which has a significant cross section only for neutrons above about 13.5 MeV. In neutron irradiated Si, ^{26}Al is

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produced by either the two-step process $^{28}\text{Si}(n,d)^{27}\text{Al}(n,2n)^{26}\text{Al}$ or the alternate two-step process $^{28}\text{Si}(n,np)^{27}\text{Al}(n,2n)^{26}\text{Al}$. Because two reactions with neutrons are required, the production of ^{26}Al increases during irradiation approximately as the square of the fluence. Both reactions require high energy neutrons (> 10 MeV). Thus, the amounts of ^{26}Al produced from Si, relative to production of other radionuclides, will in general depend on both the neutron energy spectrum and the neutron fluence.

The production of ^{26}Al from Si (or its importance) may have gone unnoticed earlier on for several reasons. Earlier activation calculation codes did not often contain provisions for multiple reactions, so one would have to anticipate this two-reaction process and calculate it specifically. Furthermore, analyses with activation calculations that compared only the activities (not doses, biological hazard potential, etc.) would have shown that the major contributor to the activity of SiC is ^{14}C (which is not a gamma emitter and is allowed in relatively high concentration under 10 CFR 61). Also, ^{26}Al is not included in 10 CFR 61. Today multireaction processes are routinely addressed in the major activation codes, and the production of significant amounts of ^{26}Al from Si is widely known.

All activation calculations contain uncertainties due to incomplete knowledge of nuclear data and uncertainties in neutron spectra. The neutron cross sections for ^{27}Al production from ^{28}Si that are in present nuclear data bases are calculated from theoretical models. Some experimental measurements for the $^{28}\text{Si}(n,d)^{27}\text{Al}$ cross section exist, but they have not been evaluated nor incorporated into present data bases. Evaluation of Si cross sections over the entire energy spectrum of interest are in progress⁸. It is expected that the (n,d) data will be reviewed, and, if acceptable, used to scale the cross sections for the high threshold interactions. We speculate that any corrections to these cross sections will be within an order of magnitude of present calculated values, and conclusions drawn from present calculations might not be significantly changed. However, until the evaluations are done, we will not know for sure. It is important to include the best available information because of the potential technological importance of SiC.

Sequential Charged Particle Reactions. When a proton or alpha particle is a product of the interaction of an energetic neutron with a nucleus, the charged particle has usually been assumed to end up as a gas atom in the material. However, some of these charged particles are emitted with enough energy to enter into a nuclear reaction with another atom (with greater than negligible probability). These reactions are known as sequential charged particle reactions (SCPR), and they should not be confused with the process involving two separate neutron reactions that leads to ^{26}Al from Si, as discussed in the previous section. Because (n,p) and (n,a) reactions generally have high thresholds, SCPR are expected to be significant only for high energy neutrons. Until recent calculations were performed, it was not known whether they might make important contributions to activation or transmutation in fusion energy systems (or in other facilities having neutron energies > 10 - 15 MeV). Cierjacks^{9,10} developed the methodology and nuclear data libraries for including SCPR in activation calculations for fusion energy system materials. In 1994, Ehrlich, Cierjacks et al.¹¹ reported on the results of the investigation of all stable elements ($Z=1$ - 83) for the effects of SCPR. Table 1 of that paper lists all elements for which activity, dose rate or decay heat are increased by SCPR in a DEMO first wall spectrum at 12.5 MWy/m². Neither Si nor C have significant enough SCPR contributions to be included in the table. The only effect of SCPR for potential fusion materials is an increase in total activity of V after 1000 years, due to production of ^{53}Mn (which has no gamma, is not in 10 CFR 61, and is not limited for Class C disposal according to Fetter et al.⁴). Thus, even for vanadium alloys, the effects of SCPR are probably much less than effects of impurities.

The Cierjacks calculations were done for a DEMO first wall spectrum. The magnitude of the SCPR contribution at neutron energies higher than 14 MeV is not known. The chance that significant new SCPR interactions occur at energies in the 14-20 MeV range is probably small, but should be examined for the sake of completeness.

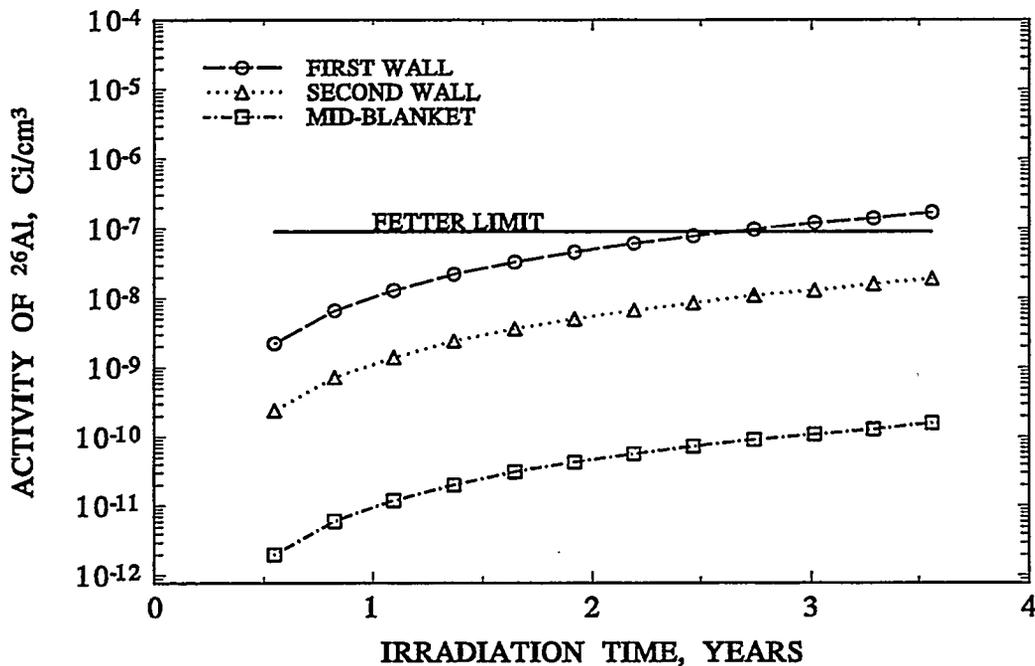


Figure 1. The buildup of ^{26}Al in SiC irradiated in STARFIRE for 3.5 years (12.5 MWy/m^2) in the first wall, second wall and mid-blanket positions. The limit for ^{26}Al activity calculated by Fetter et al.⁴ using 10 CFR 61 methodology is indicated.

Implications for Use of SiC in Fusion Energy Systems. With respect to waste disposal by shallow land burial, based on the present (Fetter) limits for ^{26}Al , SiC is at best marginal for use in the first wall region, at least based on recent activation calculations using DEMO and STARFIRE fusion machine designs. Levels of ^{26}Al obtained in exposures on the order of DEMO component lifetimes just exceed the best present guess for disposal limits. Figure 1 shows the results of calculations of the quantity of ^{26}Al produced in SiC irradiated in STARFIRE as a function of irradiation time. The calculations were done using the REAC*2 code over 12 time steps, for a total radiation time of 3.5 years, corresponding to 12.5 MWy/m^2 (the anticipated service life of DEMO first wall components). The results are compared to the Fetter et al.⁴ limit for ^{26}Al that was calculated using 10 CFR 61 methodology. In the STARFIRE first wall spectrum, the Fetter limit is exceeded after only 2.5 years of irradiation (9 MWy/m^2). The lower activities achieved in the STARFIRE second wall (6 cm behind the first wall) and mid-blanket (0.5 m behind the first wall) regions are primarily because of reduced total neutron fluxes there rather than the spectral differences. According to these results, at the second wall the Fetter limit will not be reached for about 25 years, while in the mid-blanket region the concentration of ^{26}Al will be two orders of magnitude under the Fetter limit for any fusion system lifetime. The activation of SiC in these STARFIRE locations should be seen as only qualitatively similar to a machine constructed of SiC (the STARFIRE first wall design uses PCA stainless steel backed by a Be neutron multiplier, with a LiAlO_2 breeder in the PCA supported blanket).

Figure 2 shows the residual radioactivity of SiC as a function of time after irradiation for the present STARFIRE calculations using REAC*2. The activity of ^3H is omitted from the plot, and the separate contributions of Si and C are shown. The contribution from C is exclusively from ^{14}C , while the major contribution from Si after 50 years is from ^{26}Al .

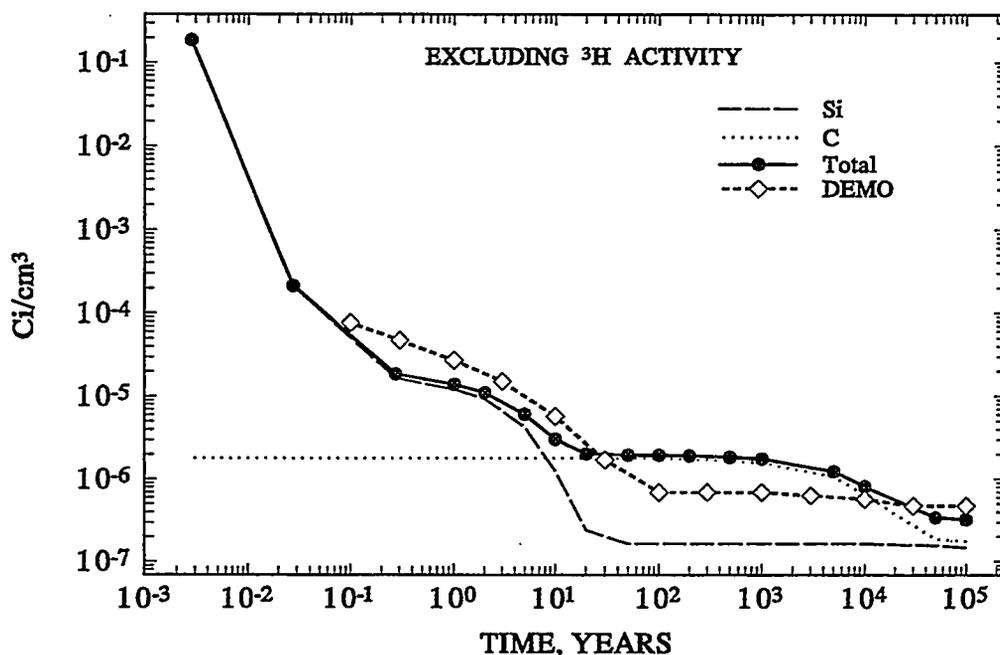


Figure 2. The residual radioactivity in Ci/cm^3 of SiC after irradiation in STARFIRE to a dose of $12.5 \text{ MWy}/\text{m}^2$ as a function of time after irradiation. The large dashed and dotted lines are the separate contributions of Si and C, respectively. The solid line with data points is the total for SiC. The curve with open diamonds is the result of a calculation by Butterworth⁵ for SiC in DEMO to the same dose, $12.5 \text{ MWy}/\text{m}^2$.

The STARFIRE results are compared in Fig. 2 with calculations for DEMO (to $12.5 \text{ MWy}/\text{m}^2$) by Butterworth⁵ (who apparently omitted activities of both ^3H and ^{14}C from his plot). The DEMO calculation shows residual activity of ^{26}Al four times greater than that from the present STARFIRE calculation. This difference could be due to differences in neutron spectra, differences in nuclear data bases or even computational methods.

By present computational methods of evaluation, SiC activation is essentially at the Class C limit in these "generic" fusion energy system spectra. For a more precise evaluation of whether or not SiC can meet Class C burial limits as a fusion material, it is important to have the most realistic energy system design and neutron spectrum information available. If it is safe and practical to average activities over large components for disposal purposes, then perhaps the higher concentrations of ^{26}Al in the first wall areas will not prohibit shallow land burial of SiC components containing the first wall. Fusion energy system waste

disposal criteria, waste stream characteristics and decommissioning procedures will be extremely important factors in determining if SiC can be considered a low activation fusion material.

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