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PERFORMANCE MODELING OF NEUTRON ABSORBERS *

F. J. Homan

Metals and Ceramics Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

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ABSTRACT

The components of a computer code to predict the performance of fast reactor control rods are presented and discussed. Predictions of integrated performance of a typical FTR-type control rod in row 2 of EBR-II are included to demonstrate the predicted influence of the ^{10}B enrichment and solid reaction product swelling. Models for helium gas bubble swelling and release are presented, which suggest minimum swelling from this source at about 1000°C . It is noted that control rods are predicted to fail very shortly after the fabricated gap between the absorber pellets and the cladding has closed, due to high contact pressure which develops with continued pellet swelling.

PERFORMANCE MODELING OF NEUTRON ABSORBERS

INTRODUCTION

Neutron absorber materials such as boron carbide (B_4C) are widely used as control elements in both thermal and fast reactors. Because of the obvious requirement that the performance of these materials during irradiation be well understood to insure safe and reliable reactor operation, extensive research programs are underway at the Hanford Engineering Development Laboratory and the Oak Ridge National Laboratory to characterize the fabrication techniques and irradiation performance of these materials. Part of the Oak Ridge program includes a modeling task whereby the means to predict mathematically the behavior of an absorber rod in-reactor are being developed. To accomplish this task a computer code called ABSORB has been written. It is anticipated that this code will have multiple applications including the planning and design of irradiation experiments on absorber materials, interpretation of the data from these tests, and as a means of tying together the different experimental areas of the absorber materials development programs.

Many aspects of fuel performance modeling have been utilized directly in the absorber performance code, such as swelling and mechanical performance of the cladding. Other models, such as swelling and gas release from the absorber material have required parallel development. This paper will describe the models used in

the code which are unique to absorber materials and will present some of the results obtained from the code. Emphasis is given to modeling the performance of small boron carbide rods for Fast Test Reactor (FTR) application, but consideration is also given to larger diameter rods for comparison. Since integrated testing of neutron absorber rods in fast flux environments has begun only recently, there is not an adequate body of data yet available for extensive testing of the model against experimental results. This task remains for the future.

At present the ABSORB code is set up to handle only boron carbide, but it could easily be converted to consider other absorber materials when sufficient materials data become available.

THERMAL PERFORMANCE

The methods used to calculate temperature distributions in neutron absorber rods under irradiation are identical to those used in analyzing fuel rods,^{1,2} assuming steady-state conduction with an internal heat source. A recently developed model³ to describe the heat transfer across a pellet-cladding gap eliminates the need for the code user to supply this parameter. For boron carbide control rods, the internal heat source is the $^{10}\text{B}(n,\alpha)\text{Li}$ reaction in the B_4C pellets (which generates 2.78 MeV per reaction⁴) and gamma heating in both pellets and stainless steel cladding. The heat generation rate within the boron carbide pellet will depend linearly upon the neutron flux, concentration (or enrichment) of

^{10}B atoms in the pellet, and the capture cross section of the ^{10}B atoms. The steady-state operating temperatures will depend upon the amount of heat generated and the ease with which it conducted out of the rod.

The thermal conductivity data for boron carbide found in the literature⁵ for unirradiated material indicate a function which decreases with temperature to about 1000°C, then increases with temperature. These data can be conveniently fit to a polynomial expansion in temperature to represent the thermal conductivity function for 100% dense material, with a Maxwell-Eucken correction for porosity, as shown in Fig. 1. Also shown in Fig. 1 are thermal conductivity data for boron carbide after irradiation at 500°C (Ref. 6). Comparison of these data with the unirradiated data reveals a substantial reduction in thermal conductivity with neutron exposure for boron carbide.

HEDL investigators⁷ have measured the thermal conductivity of the 92% dense boron carbide material being used to fabricate FTR control rods as a function of temperature and irradiation, and have fit their data to an equation of the form:

$$K(T,C) = \left[\frac{1}{A_1 + A_2 T} \right] \left[\frac{1}{1 + A_3 C} \right], \quad (1)$$

where

$K(T,C)$ = thermal conductivity,

T = temperature,

C = depletion,

A_1, A_2, A_3 = empirical constants .

Thermal Conductivity of B₄C

Fig. 1.

Equation (1), which is plotted in Fig. 2, is based on thermal conductivity measurements at temperatures up to 1400°F (760°C) and therefore does not reflect the minimum present in Touloukian's compilation at about 1000°C. When Eq. (1) is corrected from 92% density to 100% density using the Maxwell-Eucken correction shown in Fig. 1, the agreement with the polynomial expansion equation shown in Fig. 1 is excellent. Therefore, the thermal conductivity function used in the ABSORB code is the polynomial expansion equation shown in Fig. 1 to represent the temperature dependence of 100% dense boron carbide, a Maxwell-Eucken porosity correction, and the second term on the right-hand side of the equals sign in Eq. (1) to correct for the influence of depletion.

As indicated earlier, the steady-state temperature distribution in a cylindrical absorber rod is dependent upon the rate at which heat is generated and conducted out of the rod. The $^{10}\text{B}(n,\alpha)\text{Li}$ reaction in boron carbide is given in Eq. (2):

$$\dot{R} = [^{10}\text{B}] \sigma \phi, \quad (2)$$

where

$\dot{R}$ = the reaction rate (reactions/cm³-sec),

$[^{10}\text{B}]$ = the concentration of the ^{10}B isotope (atoms/cm³),

σ = the absorption cross section for the ^{10}B isotope (cm²),

ϕ = the neutron flux (neutrons/cm²-sec) .

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Thermal Conductivity of B₄C (92% TD)

Fig. 2.

Using values of σ and ϕ corresponding to row 2 of the EBR-II core at the reactor midplane ($1.15 \times 10^{-24} \text{ cm}^2$ and $3 \times 10^{15} \text{ neutrons/cm}^2\text{-sec}$, respectively)^{8,9} and the 2.78 MeV/reaction quoted earlier, the steady-state temperature distributions shown in Fig. 3 have been calculated, using different assumptions concerning thermal conductivity,¹⁰ ^{10}B enrichment, and pellet size. The 0.98 cm OD pellets represent a proposed FTR design;¹⁰ the 4 cm OD pellets represent the vented control rods in the French Rapsodie-Fortissimo Reactor;¹¹ and the 1.91 cm OD pellets (0.75 in.) represent an intermediate size included here for comparison. The conclusions which are drawn from Fig. 3 are that for large diameter, high ^{10}B enriched pellets, the operating temperatures can reach levels sufficient to cause restructuring, grain growth, and extreme plasticity in other ceramic systems. But for the small diameter FTR absorber pellet, which is of immediate interest, the operating temperatures are sufficiently low that these phenomena are not of concern. Since little is known about the restructuring, grain growth, and plasticity of boron carbide or other absorber materials, these aspects of performance modeling have been ignored for the present.

MECHANICAL PERFORMANCE

As indicated in the introduction, models describing the mechanical behavior of cladding are readily available in numerous fuel performance codes, and will therefore not be discussed here. The response of the absorber pellet to the temperature gradients described

Calculated B_4C Pellet Temperature for Different Pellet Sizes,
 Enrichments and Thermal Conductivity Assumptions

Fig. 3.

in Fig. 3 deserves some comment however. Using standard finite difference techniques,¹² the elastic stresses associated with the temperature gradients shown in Fig. 3 were calculated. For these calculations it was necessary to know the elastic modulus, Poisson's ratio, and the linear coefficient of thermal expansion as functions of temperature for boron carbide. Values for the elastic modulus and Poisson's ratio could only be found at room temperature (65.3×10^6 psi and 0.21, respectively).^{13,14} These values were used in the calculation which is equivalent to assuming that these properties are not dependent upon temperature. Thermal expansion¹⁵ and rupture modulus data¹³ from the literature (shown in Fig. 4 as functions of temperature) were also used in the calculation. Calculated stress gradients corresponding to the temperature gradients shown in Fig. 3 are plotted in Fig. 5. Notice that the thermal stresses developed in both of the larger pellets exceed the rupture modulus, while those for the smaller FTR pellet do not. Thus, it was concluded that pellet cracking was probably not of immediate concern for performance modeling of the FTR absorber rods. However, a cracking model which is described in detail in Ref. 16, was built into the ABSORB code for possible future use.

Temperature Dependent Properties of B_4C

Fig. 4.

Calculated Elastic Stresses due to Temperature Gradients in B₄C Pellets of Different Sizes and Enrichments Operating in Row 2 of EBR-II

Fig. 5.

HELIUM RELEASE

The $^{10}\text{B}(n,\alpha)\text{Li}$ reaction produces one atom of helium and one lithium atom per reaction. Both of these atoms are larger than the boron atom which they replace in the lattice,¹⁷ causing a potential swelling problem. The helium will form into gas bubbles, and some of this gas will be released from the boron carbide lattice which will increase the internal gas pressure. The need for analytical treatment of both swelling and gas release in a performance model is obvious.

There are abundant data in the literature describing helium release from boron carbide irradiated at low temperatures in a thermal flux.^{18,19} A recent analysis²⁰ of data obtained on both powders and pellets irradiated in a thermal flux at temperatures between 300 and 600°C to depletions from 7 to 26×10^{20} captures/cm³ has shown gas release to be dependent upon temperature, porosity, and form (powder or pellet). No dependence upon depletion could be detected. An equation describing the thermal flux helium release data is given below:

$$\% \text{ helium release} = A \exp(-Q/RT), \quad (3)$$

where

$$Q = 3600 \text{ cal/mole},$$

$$T = ^\circ\text{K},$$

$$A = \exp(C - 1.85 D),$$

$$C = 6.69 \text{ for pellets},$$

$$= 7.12 \text{ for powder},$$

$$D = \text{fractional density}.$$

In thermal flux irradiations most of the captures occur close to the pellet or powder surface because of the self-shielding characteristics of boron carbide. Because the path for conduction of heat out of the pin is much shorter in this situation than under fast flux conditions with the same reaction rate, operating temperatures are low. Also, the gas release characteristics in a fast flux may be entirely different, even at the same temperature, than those in a thermal flux. While few helium release data are available for boron carbide in fast reactor tests, HEDL investigators⁷ have developed an equation of the form given below to describe helium release from boron carbide irradiated in EBR-II.

$$R = \frac{\exp[A_1 (T - T_1)]}{1 + \exp[A_1 (T - T_1)]} + \frac{[A_2 - A_3 T] \exp[A_4 (C - C_1 + A_5 T)]}{1 + \exp[A_4 (C - C_1 + A_5 T)]}, \quad (4)$$

where

R = fraction helium released,

T = temperature,

C = depletion,

$A_1, A_2, A_3, A_4, A_5, T_1, C_1$ = empirical constants .

Figure 6 is a plot of this equation for depletions of 10×10^{20} and 20×10^{20} depletions/cm³. For comparison, Eq. (3) is also shown for 92% dense pellets. Agreement between Eq. (3) and the HEDL equation seems quite reasonable at low temperatures, and the conclusion that there is no detectable influence of depletion on gas release in the thermal flux data examined appears to be born out by the HEDL equation, at least at low temperatures.

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Helium Release from Irradiated B_4C (92% TD)**Fig. 6.**

Because of the greater range of temperatures used in developing the HEDL gas release equation and its applicability to fast flux conditions, this equation is used in the ABSORB code to describe helium release.

SWELLING

It is necessary to consider swelling of boron carbide during irradiation in terms of a combination of solid reaction products and helium gas bubbles retained in the lattice. This is because the solid component of swelling will be much less temperature sensitive than the component due to retained gas. While this may be the logical way to model swelling of boron carbide during irradiation, the means to separate the two components when analyzing experimental data are not readily available. Most of the swelling data reported to date represent thermal irradiation tests conducted at low temperatures. Figure 7 shows such data²¹ for natural, enriched, and depleted B_4C irradiated in a variety of thermal flux facilities at temperatures between 93 and 277°C. While there appears to be considerable scatter in these data, it should be noted that the materials ranged in density from 90 to 100% of theoretical, with much of the data from the more porous materials falling at the bottom of the scatter band in Fig. 7. This indicates the possibility of accommodation of swelling in fabricated porosity. The curves drawn on Fig. 7 to represent boundaries of the data are

Solid Reaction Product Swelling in B_4C

Fig. 7.

straight lines. Close examination of the data indicates an apparent discontinuity in the swelling vs depletion relationship at about 100×10^{20} depletions/cm³, with the data at higher depletions being represented by curves of higher slopes than those shown.

To use data such as that shown in Fig. 7 to predict performance at higher operation temperatures, the Greenwood-Speight model²² as modified by Goldberg et al.²³ was used. This model assumes homogeneous nucleation of gas bubbles within the solid lattice and a random walk mechanism of bubble motion. These assumptions are probably much more applicable to the boron carbide case where the radial temperature gradient is lower than it is for oxide fuel modeling, where the model is also used.^{1,23} This gas swelling model is quite simple and therefore very attractive for performance modeling. However, numerous materials parameters are required for its use. The values used for these parameters and their sources are tabulated in Table 1. Several of the required values could not be found for boron carbide; so the values suggested by Goldberg et al.²³ were used.

The swelling behavior predicted by the modified Greenwood-Speight-model for 100% dense boron carbide irradiated at constant temperature and hydrostatic pressure, and assuming all the helium generated is retained in the lattice, is shown in Fig. 8. Notice that the swelling rates predicted by this model increase very rapidly with temperature beginning in the range 800 to 1000°C, and that increases in hydrostatic pressure are predicted to have a relatively minor influence on swelling.

Table 1. Materials Parameters Required in Greenwood-Speight
Gas Swelling Model for Boron Carbide

Parameter	Value	Reference
Atomic volume boron	$0.523 \times 10^{-24} \text{ cm}^3$	17
Debye frequency	$3.22 \times 10^{12} \text{ sec}^{-1}$	23
Entropy factor	1.0	23
Number of new neighboring sites contacted by a bubble in diffusing through a distance equal to its diameter	4	23
Atomic spacing boron	$1.8 \times 10^{-8} \text{ cm}$	24
Activation energy for surface diffusion	$3.81 \times 10^{12} \text{ erg/g-mole}$	23
Van der Waal's constants for helium		
a	$2.360 \times 10^{10} \text{ dyne cm}^4/\text{g mole}$	
b	19.54 cm^3	
Helium atoms for reaction	1.0	
Surface tension of B_4C	500 dyne/cm	23
Diffusion coefficient for helium in B_4C	$D = 1.4 \times 10^{-9} \exp\left(\frac{-29000}{RT}\right)$ $T = ^\circ\text{K}$	25

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Calculated Swelling in Boron Carbide due to Helium Bubbles
(assuming no gas release)

Fig. 8.

If the HEDL gas release function given in Eq. (4) and plotted in Fig. 6 is applied to the gas swelling model, the results are as shown in Fig. 9. From Fig. 6 it can be seen that the HEDL equation predicts gas release approaching 100% at just about 1000°C. It is doubtful that 100% of the gas will be released at any temperature. So for the purposes of the model, it is assumed that a small percentage of gas remains in the boron carbide at temperatures above 1000°C. The predicted swelling is very sensitive to the amount of gas assumed to be retained; however, for purposes of modeling FTR absorber rods, where temperatures will be well below 1000°C, this aspect of the analysis is of little interest.

Comparing Fig. 7 with Fig. 9 in the range 93 to 277°C reveals that most of the swelling associated with the data plotted in Fig. 7 comes from retained helium gas (assuming, of course, that the treatment of gas swelling is valid). From this comparison we have concluded the component of swelling due to solid reaction products to be 0 to 0.1% $\Delta V/V$ per 10^{20} depletions/cm³. For 100% dense boron carbide, this is equivalent to 0 to 1.1% $\Delta V/V$ per 1% boron atoms reacted. Because of the discontinuity noted earlier in Fig. 7, this swelling rate is not expected to apply beyond about 100×10^{20} depletions/cm³.

Calculated Swelling in Boron Carbide due to Helium Bubbles
 (after gas release)

Fig. 9.

From the results shown in Fig. 9 it would appear advantageous to operate boron carbide at temperatures near 1000°C where swelling due to helium gas appears to be at a minimum. Such operation would, of course, result in more helium gas being released than lower temperature operation; but this gas could be accommodated in larger gas plenums or through the design of a vented pin.

PREDICTIONS OF INTEGRATED PERFORMANCE

The performance predictions discussed here are for an absorber rod containing boron carbide pellets clad in cold-worked type 316 stainless steel. The fabrication parameters, which are listed in Table 2, represent the FTR control rod design.¹⁰ The FTR design calls for natural enrichment of the boron (19.8 at. % ^{10}B); however, a case assuming 90% enrichment is considered here as well, for comparison. Because of the higher concentration of ^{10}B atoms in 90% enriched material, the reaction rate (and thus the heat generation rate) is higher than for the naturally enriched B_4C . It is assumed that the absorber rod being studied is irradiated in row 2 of EBR-II, where the neutron flux and capture cross sections of the ^{10}B isotope are known.^{8,9}

Figure 10 shows the time changes in both depletion and heat rate for both cases. The reaction rate (and the heat rate) decreases with time and depletion as shown in Eq. (2). Even as the heat rate is decreasing with time, the pellet center temperatures

Table 2. Fabrication Data for FTR Boron Carbide Absorber Rod

Cladding

Type: Cold worked type 316 stainless steel

OD: 0.475 in.

Thickness: 0.040 in.

Clad-pellet gap (diametral): 0.009 in.

B₄C Pellets

OD: 0.386 in.

Density: 92% theoretical

Theoretical density: 2.52 g/cm³¹⁰B enrichment: 19.8% ¹⁰B

Pellet stack: 36 in.

Gas Plenum

Length: 18 in.

Fill gas: Helium at 1 atm.

(plotted at the top portion of Fig. 10) are increasing. This is expected from the nature of the thermal conductivity relationships shown in Fig. 2.

Figure 11 shows the time-dependent increases in helium gas pressures for the two enrichment cases, and the resulting cladding stresses and strains. Because helium is being generated and released much more rapidly in the 90% enrichment case, the cladding is placed under greater stress. The higher operating temperatures in the 90% enrichment case cause higher gas release as shown in Fig. 6, which adds to this effect.

Perhaps the most useful performance prediction presented here is that gap closure is predicted for three out of the four cases shown in Fig. 12. The swelling vs time relationship is plotted for both natural and 90% ^{10}B enrichment, assuming values of 0 and 0.1% $\Delta V/V$ per 10^{20} reactions/cm³ for the swelling contribution of solid reaction products. Recall that these values represent the range for solid swelling determined earlier by subtracting the gas bubble swelling shown in Fig. 9 from the total swelling shown in Fig. 7. The gap closure characteristics associated with these swelling assumptions are shown at the top of Fig. 12. The code predicts pin failure very soon after the gap closure, due to the high contact pressures which develop between the pellet and the cladding. Unlike oxide fuels, the boron carbide pellets do not have a hot plastic core which can be deformed to prevent the buildup of such high contact pressures.

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Heat Rates and Operating Temperatures
for FTR Control Rods in Row 2 of EBR II
Fig. 10.

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Helium Gas Pressure and Subsequent Cladding Stresses
for FTR Control Rods in Row 2 of EBR-II

Fig. 11.

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Swelling and Gap Closure Characteristics
for FTR Control Rods in Row 2 of EBR II

Fig. 12.

SUMMARY

A computer code called ABSORB has been developed to predict the performance of neutron absorber materials for control rod application in fast reactors. The individual models used in this code have been developed from experimental data generated in the absorber materials development programs at HEDL and ORNL. Several of these models have been presented and discussed.

Integrated performance predictions have been made using the ABSORB code for a typical MTR control rod, irradiated in row 2 of EBR-II. Several performance parameters have been presented and discussed. One important performance observation is related to the sensitivity of predicted performance to the swelling characteristics of the boron carbide pellets. The code predicts control rod failure very shortly after the fabricated gap between the pellet and the cold-worked type 316 stainless steel cladding has closed. This is due to the high contact pressures which develop between the pellet and the cladding due to continued swelling of the boron carbide pellets. Unlike oxide fuels, there is no hot plastic center within the boron carbide pellet to deform in response to growing contact pressures at the pellet cladding interface. Thus, it would appear that pellet swelling is the most immediate problem related to control rod performance. It was pointed out that the helium gas component of pellet swelling may be minimized by operation at temperatures approaching 1000°C where gas release fractions are high, and the swelling rates from residual gas are still low compared to the rates at higher temperatures.

It should be noted that the models which have been developed for incorporation into the ABSØRB code are based on very limited data. Swelling rates and helium release characteristics at higher temperatures need to be investigated. In addition, performance predictions from ABSØRB need to be checked against experimentally measured performance to establish the veracity of the code.

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