BORON CONCENTRATION GRADIENT FOR IMPROVED THERMAL REACTOR
PERFORMANCE OF BORON-STAINLESS STEEL CONTROL RODS*

R. J. Beaver and A. E. Richt
Metals and Ceramics Division, Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

MASTER

SUMMARY

The performance of conventional 2 to 3 wt % ^{10}B -stainless steel alloys and powder dispersions is limited in thermal reactors because the $^{10}\text{B}(\text{n},\alpha)$ reactions are localized at and near the surface of the material. Significant swelling occurs after a relatively short exposure, and additional exposure creates drastic damage to the microstructure in the localized regions. By designing a symmetrical boron concentration gradient in which the ^{10}B is limited at the surface and increases in a prescribed manner to higher concentrations below the surface, the performance capability of boron-stainless steel neutron absorbers can be increased substantially.

We applied this generalized concept to the SM-1 Neutron Absorber Section Assembly, which is a rectangular parallelepiped consisting of four 0.090-in.-thick plates clad with 0.032-in.-thick austenitic stainless steel. We limited the ^{10}B concentration at the surface to 1 wt % and in a programmed manner increased the concentration to a maximum of 3 wt % ^{10}B 0.024 in. below the surface. For an intended maximum fluence of 4.3×10^{20} nvt (thermal), this technique distributes the $^{10}\text{B}(\text{n},\alpha)$ reactions throughout 50% of the material and limits the ^{10}B consumption in any volume increment to 41×10^{20} atoms/cm³. This increases the

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allowable average ^{10}B burnup to 20 at. %, without significantly reducing control rod worth, compared with the 5 at. % ^{10}B average burnup limitation for 3 wt % ^{10}B -Fe dispersions. This improvement was demonstrated by the postirradiation examination of a boron-gradient neutron absorber, which contained ^{10}B dispersed in type 200 austenitic stainless steel and operated for six years in the central region of the active lattice of the 10 MW SM-1 Reactor.

INTRODUCTION

Boron alloyed in stainless steel or dispersed in iron has limited application as neutron absorbers in thermal reactors. Conventional fabrication techniques for manufacturing these materials cause the boron to form intermetallic compounds with the iron, nickel, and chromium of the metal matrix. The result is a decrease in the ductility of these materials (1-3). Beard et al. (4) found that boron-stainless steel alloys containing in excess of 2.5 wt % B tend to crack and are virtually impossible to fabricate by conventional methods.

Swelling caused by the $^{10}\text{B}(n,\alpha)$ reaction limits the service life of boron-metal dispersions. Richt et al. (5) found that in 0.156-in.-thick stainless-steel-clad plates with a 0.090-in.-thick core containing 3 wt % B (92% enriched in ^{10}B), swelling initiates at an average ^{10}B burnup of only 5 at. % and that the neutron damage progresses to severe destruction of the surface region at an average ^{10}B burnup of 16 at. %. This drastic effect is illustrated in Fig. 1 for a composite plate that swelled 6%. Approximately 25% of the neutron absorber has been extensively damaged, and obviously the ^{10}B burnup in this region is several times greater than the average. Such damage can be eliminated by a

design that limits the burnup of ^{10}B atoms in each incremental layer through the plate to a constant acceptable value. These considerations result in the ^{10}B concentration gradient profile through the thickness illustrated in Fig. 2.

We tested this method by an experiment in one of the highest regions of thermal neutron flux in the 10 MW SM-1 Reactor. Our experimental absorber was a neutron absorber section assembly like the design specified for the Army's stationary medium-power reactors (6) -- namely, a rectangular parallelepiped whose sides are composite stainless-steel-clad plates in which the core and the cladding are nominally 0.090 and 0.032 in. thick, respectively. Testing this full-scale neutron absorber section assembly in the operating reactor offers the advantage of achieving a burnup profile from the tip end, where the burnup is highest, toward the opposite end, where the burnup is the lowest. Thus, at the termination of the test we can assess the ability of our absorber to withstand the thermal neutron damage as a function of burnup of ^{10}B atoms. The term of irradiation was aimed at achieving near the tip end an average burnup of 20 at. %.

We chose type 200 austenitic stainless steel powder as a substitute for the iron powder previously used as the dispersant for the boron because of its higher corrosion resistance and its superior absorption of epithermal neutrons compared to iron. We prepared the dispersions using powder metallurgy techniques for convenience in arranging a boron gradient concentration profile, and the fabrication procedures were essentially the same as established previously by Beaver et al. (7).

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DESCRIPTION OF THE CONCEPT

The primary objective of the boron concentration gradient method is to distribute the $^{10}\text{B}(n,\alpha)$ reactions throughout as much of the neutron absorber as is feasible, with the concentrations of ^{10}B increasing from the surface inward to assure an essentially constant burnup of ^{10}B atoms in any volume increment. The data reported by Richt et al. (5) indicated that we could allow complete burnup of a dispersion of 1 wt % ^{10}B in iron (47×10^{20} atoms $^{10}\text{B}/\text{cm}^3$) without encountering swelling of stainless-steel-clad plates or undesirable damage to the metallurgical bond between the core of the plate and the cladding. We also limited the maximum concentration within the material to 3 wt % ^{10}B because this concentration is within existing technology. Thus, the concentration at the surface was established at 1 wt % ^{10}B , and the maximum concentration below the surface was limited to 3 wt % ^{10}B .

The intermediate concentrations (Fig. 2) are established with a computer program based on the theoretical burnup behavior of ^{10}B . The boron concentration profile is symmetrical for maximum service where the thermal neutron current penetrating each side of the absorber is essentially the same. The cross-hatched portions of the illustration represent the regions where the $^{10}\text{B}(n,\alpha)$ reactions principally occur. The vertical distance between the initial concentration profile and the profile after irradiation represents the ^{10}B burnup in any volume increment, in this design 34×10^{20} atoms $^{10}\text{B}/\text{cm}^3$, with a tolerance of $\pm 5\%$. The total cross-hatched area represents the average ^{10}B burnup anticipated and is 20 at. %.

THEORETICAL CONSIDERATIONS AND MATHEMATICAL MODEL

The principal objective of the boron gradient is to establish an initial concentration of ^{10}B in each finite increment of plate thickness to compensate for the decrease in neutron current from the surface inward. Theoretical considerations resulted in a rather complicated formula for the interior concentrations, and simplifying assumptions were resorted to.

Consider a parallel current of neutrons impinging perpendicularly on the surface of a layer. If S_i is the number of ^{10}B atoms/cm² of surface area in layer i , then the burnup rate of the ^{10}B atoms per unit area of surface layer is

$$\frac{dS_i(t)}{dt} = - \int_{x_i}^{x_{i+1}} I(x,t) N(x,t) \sigma(x,t) dx, \quad (1)$$

where

x = distance through region i , cm

t = time, sec

$N(\bar{x},t)$ = ^{10}B atoms/cm³ at x and t in region i

$I(x,t)$ = neutron current, neutrons cm⁻² sec⁻¹

$\sigma(x,t)$ = microscopic cross section for the absorption of thermal neutrons by ^{10}B , cm² nucleus⁻¹

Now consider space-averaged values of $I(x)$ and $N(x)$, and assume σ is constant with both time and space. Then

$$\frac{dS_i(t)}{dt} = - I_i(t) N_i(t) \sigma \Delta x_i. \quad (2)$$

Since the rate of change of S_1 and N_1 are the same,

$$\frac{dN_1(t)}{dt} = -I_1(t) N_1(t) \sigma, \quad (3)$$

which has the solution

$$N_1(t) = N_1(0) \exp\left(-\int I_1(t) \sigma dt\right). \quad (4)$$

The average number of ^{10}B atoms destroyed per unit volume is a measure of swelling and is the limiting quantity to be specified. It is calculated from

$$C_1(t') = N_1(0) - N_1(t') = N_1(0) \left[1 - \exp\left(-\int_0^{t'} I_1(t) \sigma dt\right) \right], \quad (5)$$

where t' is the desired life of the rod.

The space-averaged neutron current in each region can be estimated as follows:

$$I_i(t) = I_{i-1}(t) \left[1 - N_{i-1}(t) \sigma \Delta x_{i-1} \right] \text{ for } i > 1. \quad (6)$$

This model neglects scattering, absorption in anything but ^{10}B , changes in angular distribution of neutrons with x , and penetration of neutrons from the other side of the plate. Furthermore, it implies that one must know what the "perpendicular" neutron current is in region 1. To solve Eqs. (5) and (6) for $N_1(0)$, start with region 1 and assume that I_1 is independent of time and will be specified. Since $C_1(t')$ and t' can also be specified, $N_1(0)$ can be directly calculated:

$$N_1(0) = C_1(t') / (1 - e^{-I_1 \sigma t'}) \quad (7)$$

Substituting Eq. (7) into (4) gives

$$N_1(t) = \frac{C_1(t')}{(1 - e^{-I_1 \sigma t'})} e^{-I_1 \sigma t} \quad (8)$$

Substituting Eq. (8) into (6) gives

$$I_2(t) = I_1 \left[1 - \left(\frac{C_1(t')}{1 - e^{-I_1 \sigma t'}} \right) e^{-I_1 \sigma t} \right] \quad (9)$$

$N_2(0)$ is obtained by substituting Eq. (9) into (5) and integrating.

This process of back-substitution is repeated to obtain all values of $N_i(0)$.

In the actual application, a numerical analysis was used in which t' was divided into very short time steps. In Eq. (5) the exponential term can be expanded and all but the first two terms discarded. Furthermore, a time-averaged value of I_1 can be considered. The result is

$$\Delta C_1 = N_1(0) \bar{I}_1 \Delta t \sigma, \quad (10)$$

where $N_1(0)$ is the concentration at the beginning of each new time interval and

$$\bar{I}_1 \Delta t = \bar{I}_1 \Delta t - \sum_2^1 \Delta C_{i-1} \Delta x_{i-1} \quad (11)$$

Substituting Eq. (11) into Eq. (10) gives

$$N_1(0) = \frac{\Delta C_1 / \bar{I}_1 \Delta t \sigma}{\left[1 - \frac{\sum_2^1 \Delta C_{i-1} \Delta x_{i-1}}{\bar{I}_1 \Delta t} \right]} \quad (12)$$

$N_1(0)$ can be specified as the maximum permissible ^{10}B atoms destroyed, and therefore ΔC_1 for each of the time steps ($I_1 \Delta t$) can be calculated. Having established C_1 values, using the same time steps ($I_1 \Delta t$), and assuming a value for $N_2(0)$, ΔC values can be calculated for region 2. If the first trial is not successful, repetitions of this step will ultimately result in proper selection of the $N_2(0)$ value. The succeeding regions are similarly treated, and all initial concentrations can be established.

Our first trial at developing $N_1(0)$ from the described considerations was a relationship from which an average boron concentration gradient was approximated, as follows:

$$C_1(t') = \sum_0^{t'} \Delta C_1 = \sum_0^{t'} N_1(0) \bar{I}_1 \sigma \Delta t \quad (13)$$

By considering average values of $N_1(0)$ and $\bar{I}_1$ over t' , Eq. (13) becomes

$$C_1(t') = \bar{N}_1(0) \bar{\bar{I}}_1 \sigma \sum_0^{t'} \Delta t, \quad (14)$$

where

$$\bar{\bar{I}}_1 t' = \bar{I}_1 t' - \sum_2^1 C_{i-1}(t') \Delta x_{i-1} \quad (15)$$

Substituting Eq. (15) into Eq. (14) yields

$$N_1(0) = \frac{C_1(t')/\bar{I}_1 t' \sigma}{\left[1 - \frac{\sum_{i=2}^1 C_{i-1}(t') \Delta x_{i-1}}{\bar{I}_1 t'} \right]} \quad (16)$$

As indicated in connection with the derivation of Eq. (6), these equations neglect scattering of the neutrons and thus overestimate the penetration of the neutron current. In our first application of the ^{10}B distribution technique, we roughly compensated for scattering by assuming that the perpendicular current was reduced by 1.3 times the number of absorptions. In addition to our basic requirement that $C_i(t')$ for each incremental region be equal and that each thickness increment be equal, we also assumed that $\bar{N}_1(0) = N_1(0)/2$. Thus, Eq. (16) becomes

$$\bar{N}_1(0) = \frac{1.3 \bar{N}_1(0)}{[1 - (i-1) \bar{N}_1(0) \Delta x \sigma]} \quad (17)$$

Having established this time-averaged distribution, the initial ^{10}B concentrations were calculated using successive short time steps, making corrections for changes in I_1 and N_1 at each time step. The total times used in the back-calculation to arrive at the initial concentrations were the times required to increase $\bar{N}_1(0)$ to $N_1(0)$. It was assumed that all values of $N_1(0)$ occurred after the same lapse of time. Our results indicate that this method for approximating the $N_1(0)$ values for Eq. (12) produces a first-trial boron concentration gradient in which the ^{10}B atom burnup in each increment is reasonably constant.

In our specific design we specified that the surface layer be 1 wt % ^{10}B (47×10^{20} atoms/cm³) and the maximum concentration in the underlying regions be 3 wt % ^{10}B (142×10^{20} atoms/cm³). As a conservative measure we limited the ^{10}B burnup in each incremental region to 34×10^{20} atoms ^{10}B /cm³ and therefore the maximum thermal neutron fluence was to be 4.3×10^{20} neutrons/cm². With these criteria, when the first-trial boron concentration gradient was subjected to the specified thermal neutron fluence (dividing t' into short time steps as described previously), each region had the same ^{10}B atom burnup within a tolerance of $\pm 5\%$, and we did not believe further trials were merited.

DESIGN AND FABRICATION OF NEUTRON ABSORBER SECTION ASSEMBLY

Specific design details of the experimental Neutron Absorber Section Assembly are listed in Table I. It is not economically practical to attempt to achieve the smooth boron gradient profile previously illustrated, and "staircasing" the profile is a reasonable and feasible approach. The "staircase" profile illustrated in Fig. 3 describes the actual layering of the specific ^{10}B concentrations and is symmetrical about the thickness midplane. The cross-hatched area describes the anticipated average burnup of 20 at. % upon which the gradient is based. The total ^{10}B content of 9.36 g per plate complies with the "worth" requirements for the SM-1 control rod.

Tests demonstrated complete chemical and fabrication compatibility between boron and type 200 stainless steel to concentrations of 4.5 wt % B fabricated by conventional powder metallurgy and hot roll

bonding. The exact quantities of elemental boron (with 92% enrichment in the ^{10}B isotope) and type 200 stainless steel powder were blended with no additive for 2 hr, pressed in a die at 33 tons/in.², sintered in dry hydrogen at 1100°C, and coined in the same die at 33 tons/in.². Billets for encapsulating these compacts and rolling into composite plates provided for evacuation of the interior space before rolling. Figure 4 shows the arrangement of the components before encapsulation. The "barrier" foil in this case was used simply to assure a snug fit of the core. The rolling practice specified previously for SM-1 neutron absorber plates was used; basically, the billet was reduced in thickness by a factor of 10.5. Preheating and reheating temperatures were 1100°C. The leading end of the plate was identified and was maintained as the lead end throughout the rolling operations. The objective of this schedule was to preserve as well as possible the boron gradient at this end, which is the end of the neutron absorber that receives the highest exposure. The rolling schedule gives these plates a minimum thickness at the end scheduled for the lowest ^{10}B burnup and a maximum at the end scheduled for the highest burnup.

The plates were welded together to form the configuration illustrated in Fig. 5.

IRRADIATION PERFORMANCE

The boron gradient neutron absorber served as an actual operating neutron absorber assembly in the central control rod location of the 10 MW SM-1 pressurized water reactor (450°F, 1200 psia) for six years, the entire lifetime of Core II. During this period the reactor produced 13.07 MWyr of thermal energy, and it is estimated that this neutron

absorber was exposed to 1.2 effective full-power years of reactor service.

The ^{10}B burnup profile along a typical plate with the highest neutron exposure is listed in Table II. Visual inspection of all four plates did not reveal any defects or unusual corrosion effects. Ultrasonic scanning of all four plates indicated complete integrity of the cladding-core bond, with the possible exception of the bond at the very tip of the core.

The increase in plate thickness (swelling) caused by the irradiation is illustrated in Fig. 6. Swelling becomes noticeable at an average ^{10}B burnup of 16%, and at 20 at. % burnup it is a definite 1%.

Figure 7 illustrates the hardness increase caused by the irradiation. It is related to the specific boron composition in the gradient as well as the average ^{10}B burnup. The embrittling effect reported previously for irradiated boron-stainless steel alloys is obvious but is not surprising in view of previously reported results for boron-bearing dispersions. Nor is the increased hardness of the irradiated cladding surprising.

The microstructure of the 1.0 wt % ^{10}B portion of the boron gradient after average burnups of 16 and 24 at. % ^{10}B is shown in Fig. 8. Inspection of the photomicrographs confirmed the integrity of the cladding-core bonding shown by the nondestructive ultrasonic inspection. The porosity present is generally less than 0.0004 in., which is typical of the unirradiated powder metallurgically prepared gradient.

DISCUSSION OF RESULTS

The primary objectives of the boron gradient method were to (1) increase the ^{10}B burnup capability of boron-stainless steel dispersions and (2) eliminate the degradation of the metallurgical bond between the cladding and the boron-bearing core. Table III summarizes the results of our experiments and compares these results with the performance of a conventional plate-type neutron absorber containing 3 wt % ^{10}B (92% enriched) dispersed in iron. These experimental results show, at least from the SM-1 Reactor application, that the boron gradient principle has increased the resistance to any noticeable swelling fourfold. A more remarkable improvement is the tenfold decrease in swelling of the boron gradient plate compared to the iron-base dispersion after an average burnup of ^{10}B atoms of 24%.

To assure the integrity of the metallurgical bond between the boron-bearing core and the cladding, one of the fundamental criteria was that the boron concentration adjacent to the cladding not exceed 1 wt % ^{10}B , because complete destruction of all ^{10}B atoms in such a concentration, 47×10^{20} atoms/cm³, is not sufficient to create adverse damaging effects. The metallographic results show that this bond is preserved at average burnups as high as 24 at. %, and in addition, the ultrasonic nondestructive inspections for bond integrity indicated that this bond was preserved at an average ^{10}B burnup interpolated to be 35 at. %.

The increases in hardness of the dispersions of boron in type 200 stainless steel caused by the irradiation were not unexpected, but

the rapid rate during the first 5 at. % burnup increment shows that during most of their reactor tenure, these boron-stainless steel dispersions approach zero in ductility. This phenomenon emphasizes the necessity for encasing these boron-stainless steel dispersions. The increase in the hardness of the type 304 stainless steel cladding was not unexpected, and the obvious decrease in ductility is not considered a damaging adverse effect within the neutron exposure encountered.

It was not the primary purpose of this initial experiment to determine the accuracy of our theoretical ^{10}B burnup predictions in each volume increment of this boron concentration gradient. However, we found no evidence to indicate that the maximum permissible incremental ^{10}B atom burnup was exceeded at the average ^{10}B burnup of 20 at. % for which this specific boron concentration gradient was designed, and in this first attempt our assumptions and simplifications of the theoretical equations did not create any significant adverse effects. We believe that additional detailed investigations could result in a more precise verification of the boron gradient theory and could lead to additional improvements in the reactor performance capability of boron-stainless steel dispersions.

A noteworthy aside of our investigations was the fabrication of a dispersion containing 4.5 wt % in type 200 stainless steel with no incompatibility or fabrication problems. Heretofore, stainless steel alloys with concentrations exceeding 2.5 wt % B have been virtually impossible to fabricate by conventional processing. Our success is directly attributable to the use of powder metallurgy. This development permits the designer of a boron gradient the latitude to consider a

maximum concentration in the gradient of 4.5 wt % B for applications where longer lifetimes are desired and control rod worth maintained.

CONCLUSIONS

The limitation in the thermal reactor performance capability created by excessive $^{10}\text{B}(n, \alpha)$ reactions in the region near the surface of neutron absorbers containing conventional dispersions of boron in stainless steel can be alleviated by a boron concentration gradient. In such design an acceptable concentration near the surface is 1.0 wt % ^{10}B , increasing to 3.0 wt % 0.024 in. below the surface, and programmed to restrict the destruction of ^{10}B atoms in any volume increment to 34×10^{20} atoms/cm³ at a limiting average ^{10}B burnup of 20 at. %. Such a program increased the life expectancy of a boron-stainless steel neutron absorber in the 10-MW pressurized water SM-1 reactor from approximately 0.3 to 1.2 full-power years.

In our specific experiment tested in the SM-1 reactor, swelling initiated at a ^{10}B burnup of 16 at. % but reached only 1.6% at an average ^{10}B burnup of 24 at. %. No disintegration of the microstructure was detected, nor was the integrity of the cladding-core bond impaired.

In the past, alloys containing concentrations in excess of 2.5 wt % ^{10}B in stainless steel have been essentially impossible to fabricate. We obtained concentrations to 4.5 wt % B in type 200 austenitic stainless steel using conventional powder metallurgical practices and hot roll bonding procedures with no chemical incompatibility or fabrication problems.

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FIGURE CAPTIONS

- Fig. 1. Typical Structure of High-Burnup Fe- ^{10}B Miniature Absorber Specimen. Etched. 100X
- Fig. 2. Boron Gradient Designed to Achieve an Average ^{10}B Burnup of 20 at. %.
- Fig. 3. Actual Boron Gradient Profile in Manufactured Composite Stainless Steel Plates for Experimental Neutron Absorber Section Assembly BG-1.
- Fig. 4. Components of the Billet for Manufacturing the Experimental Stainless Steel Plate-Type Boron-Gradient Neutron Absorber.
- Fig. 5. Locations of Dimensional Measurements of Welded BG-1 Neutron Absorber Section for SM-1 Test.
- Fig. 6. Comparison of Preirradiation and Postirradiation Thickness Measurements, Plate G-4.
- Fig. 7. Hardness of Various Layers in Boron Gradient Absorber Plates of Absorber Section Assembly BG-1 as a Function of Average Burnup of ^{10}B Atoms.
- Fig. 8. Transverse Sections of Irradiated Boron Gradient Composite Plate G-1, Demonstrating Integrity of Bonded Interface Between Core and Cladding. (a) Average ^{10}B burnup, 16 at. %; polished; 50X. (b) Average ^{10}B burnup, 16 at. %; etched; 500X. (c) Average ^{10}B burnup, 24 at. %; polished; 50X.

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Fig. 8. Transverse Sections of Irradiated Boron Gradient Composite Plate G-1, illustrating Integrity of Bonded Interface Between Core and Cladding. (a) Average ^{10}B burnup, 16 at. %; polished; 50X. (b) Average ^{10}B burnup, 16 at. %; etched; 500X. (c) Average ^{10}B burnup, 24 at. %; polished; 50X.

TABLE I. Pertinent Design Data of Experimental Boron Gradient Absorber Section, BG-1.

Gradient Profile

Layer Type	Distance of Layer from Surface (in.)	Thickness of Layer (in.)	^{10}B Weight in Layer (g)	^{10}B Concentration in Layer (wt %)
1	0 to 0.012	0.012	0.65	1.00
2	0.012 to 0.020	0.008	0.48	1.13
3	0.020 to 0.024	0.004	0.43	1.95
4	0.024 to 0.045 ^a	0.021	3.22	3.00
	Total	Total ^{10}B	9.56	
		Total boron	10.34	

Materials

Core of Plate:

Boron	Elemental, enriched to the 92% level in the ^{10}B isotope.
Stainless steel	Type 200 (0.03 C-20.70 Mn-16.37 Cr-0.49 Ni, wt %).

Frame and Cladding:

Stainless steel	Type 304L (low cobalt)
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Nominal Dimensions, in.

Composite Plate:

	<u>Overall</u>	<u>Core</u>
Length	2.13	20 13/16
Width	2.464	2.214
Thickness	0.156	0.090
Length from end of plate to core end:	Tip end, 1/4 Opposite end, 5	

Absorber Section Assembly:

	<u>External</u>	<u>Internal</u>
Length	26 1/16	26 1/16
Width	2.620 x 2.620	2.308 x 2.308

^aMidthickness. The loading is symmetrical about this depth.

TABLE II. Analytical ^{10}B Burnup Results from Plate G-4.

Sample Identity	Content (wt %)		Average ^{10}B Burnup (at. %)
	B	Li	
B-5.25	0.44	0.005	1.7
B-16.25	0.71	0.027	5.6
B-20.75	0.81	0.098	16.0
B-23.75	0.86	0.17	23.9

TABLE III. Comparison of Swelling in Plates Containing Boron Gradient with Swelling in Plates Containing Conventional Boron-Iron Dispersion

Average Burnup (at. %)	Thickness Change			
	Conventional Fe-3% B		Boron Gradient	
	(in.)	(%)	(in.)	(%)
4	none	zero	none	zero
5	0.001	0.60	none	zero
12	0.003	1.90	none	zero
16	0.009	5.70	negligible	negligible
24	0.026	16.70	0.0025	1.62