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IRRADIATION TESTING OF THE RERTR FUEL MINIPLATES WITH BURNABLE ABSORBERS IN THE ADVANCED TEST REACTOR

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ABSTRACT

Based on the results of the reactor physics assessment, conversion of the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL) can be potentially accomplished in two ways, by either using U-10Mo monolithic or U-7Mo dispersion type plates in the ATR fuel element. Both designs, however, would require incorporation of the burnable absorber in several plates of the fuel element to compensate for the excess reactivity and to flatten the radial power profile. Several different types of burnable absorbers were considered initially, but only borated compounds, such as B₄C, ZrB₂ and Al-B alloys, were selected for testing primarily due to the length of the ATR fuel cycle and fuel manufacturing constraints. To assess and compare irradiation performance of the U-Mo fuels with different burnable absorbers we have designed and manufactured 28 RERTR miniplates (20 fueled and 8 non-fueled) containing fore-mentioned borated compounds. These miniplates will be tested in the ATR as part of the RERTR-13 experiment, which is described in this paper. Detailed plate design, compositions and irradiations conditions are discussed.

1. Introduction

The Advanced Test Reactor (ATR) at the Idaho National Laboratory is a 250 MW_{th} water moderated and cooled reactor, whose primary mission is irradiation testing of fuels and materials. The reactor has been successfully operating since 1967 with Highly Enriched Uranium (HEU) fuel, however to reduce the risk of nuclear materials proliferation it is imperative to convert ATR to Low Enriched Uranium (LEU) fuel, without any compromise in its safety and performance.

Initial reactor physics calculations performed by G. Chang [1] demonstrated that ATR can be converted from HEU to LEU using either monolithic U-10Mo fuel or dispersion U-7Mo fuel. In both cases, however, it would be required to incorporate Burnable Absorber in the inner/outer plates of the fuel element to hold off initial reactivity and flatten the power profile. The new LEU ATR Fuel Element has to be designed and tested to meet the following criteria:

- contain sufficient amount of burnable absorber in the inner/outer plates to suppress excess reactivity and flatten radial power profile
- withstand peak burnup in fissions/(cc fuel meat) that will be established for LEU fuel
- provide sufficient power to maintain enough reactivity through fuel life
- operate safely without losing integrity at nominal and peak operating temperatures
- provide enough safety margin to failure during off-normal (transient) events

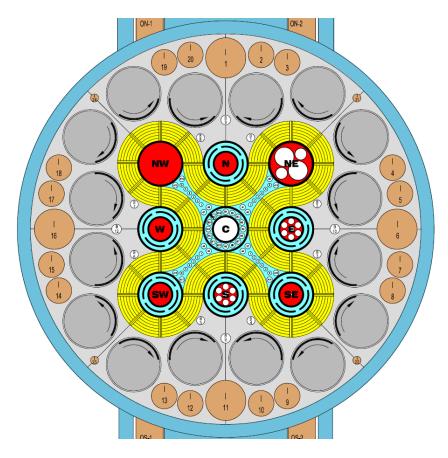


Figure 1. Cross-sectional View of the ATR Core

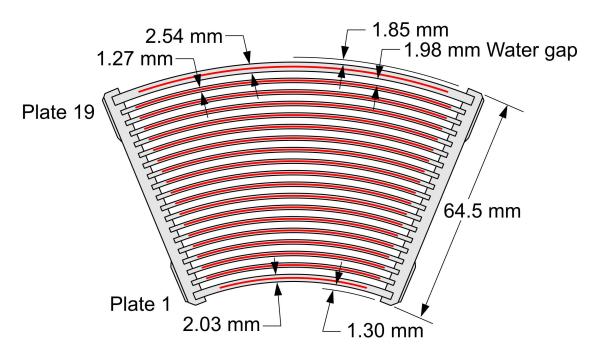


Figure 2. Cross-sectional View of the ATR Fuel Element [2]

Table 1. Composition of the Borated Plates in Mark VII ATR HEU Fuel Element

HEU Plate	Fuel Meat Thickness (mil)	Fuel Meat Volume (cm ³)	U density (HEU) (g/cm³)	U-235 Density (g/cm³)	U-235 Mass (g)	B-10 Mass (g)	B-10 Density (g/cm³)	B-10/U- 235 Ratio
Plate 1	20	23.69	1.103	1.026	24.3	0.063	0.0027	0.0026
Plate 2	20	29.54	1.059	0.985	29.1	0.078	0.0026	0.0027
Plate 3	20	31.12	1.337	1.243	38.7	0.044	0.0014	0.0011
Plate 4	20	32.70	1.328	1.235	40.4	0.045	0.0014	0.0011
				•••	•••			
Plate 16	20	51.69	1.331	1.238	64.0	0.071	0.0014	0.0011
Plate 17	20	53.27	1.330	1.237	65.9	0.073	0.0014	0.0011
Plate 18	20	54.22	1.067	0.992	53.8	0.143	0.0026	0.0027
Plate 19	20	52.64	1.074	0.999	52.6	0.143	0.0027	0.0027

2. Selection of Burnable Absorber for ATR LEU Fuel Element

Many parameters play significant role in selection of BA. These include, but are not limited to, initial negative reactivity worth, thermal-physical properties, available space for loading, ease of fabrication, cost of BA, compatibility with fuel/cladding/coolant materials and associated hazards.

However the most important criterion that essentially drives the selection process is the rate of the burnout of BA:

BR = N
$$\sigma(E) \Phi(E)$$
 (1)

where

BR is Burnout Rate of BA, atoms/(sec cm³)

N is atom density of BA, atoms/cm³

 $\sigma(E)$ is absorption cross-section of BA, cm²

 $\Phi(E)$ is neutron flux, neutrons/(sec cm²)

E – is the neutron energy spectrum

As seen from equation (1) the burnout rate of BA in a given neutron flux depends on its atom density and neutron absorption cross-section in a specified neutron energy spectrum. The elements with high absorption cross-section in a given reactor energy spectrum are typically used as BA to minimize the loading of the absorber material, especially when space is limited. The rate of burnout can also be adjusted to some extent by varying configuration of the burnable absorber in the fuel; for instance, due to self-shielding, thick lumps of BA can deplete slower than thin layers of BA.

While it is critical to have the BA at the beginning of fuel life to suppress initial excess reactivity and power peaking, the presence of the BA towards the end of fuel life, when fuel reactivity naturally goes down due to fissile material depletion and fission products buildup, is not desirable as it would only shorten the life of fuel. Therefore the length of the fuel life in the given reactor essentially defines the range of the acceptable depletion rates for the

BA and lays out the basis for its selection. The difficulty in choosing BA comes from the fact that in most cases an element is selected with its natural abundance of the isotopes. While some of the naturally occurring isotopes of the element can have a suitable burnout rate, other isotopes or their daughters may deplete rather slowly and thus would remain at the end of fuel life, contributing to the negative reactivity penalty of the fuel. To compensate for such effect, it would be typically required to either increase fuel enrichment or fuel loading in the core, neither of which seems attractive. One possible solution is to enrich the BA element in the appropriate isotope of interest; however the enrichment process is usually associated with the increased cost.

Some examples of BA used in current fleet of the Light Water Reactors (LWR) and Research Reactors are shown in Table 2. Compared to the typical LWR, where the fuel life is 1 to 2 years on average, the life of the ATR fuel is rather short, 150-200 days. This means that only fast burning elements that deplete in 50 to 100 days would be suitable for ATR applications. Based on reactor physics assessment conducted by G. Chang [3], only boron and cadmium were found to have satisfactory burnout rates. However the use of cadmium in fuel fabrication, while not completely impossible, is considered to be quite problematic due to its relatively low melting temperature (350°C) and, largely, its carcinogenic properties. As a consequence, the BA options for ATR were pretty much limited to boron containing compounds.

Table 2. Examples of Burnable Absorbers in Current Fleet

	PWR	Research Reactors	ATR
Fuel Life	1-2 years on	Varies	150-200 days
	average		
BA in use	B ₄ C, ZrB ₂ , Gd ₂ O ₃ , Er ₂ O ₃	B ₄ C (HFIR, BR-2), B (Petten HFR, Grenoble RHF), Sm ₂ O ₃ (BR-2, ATI-TRIGA) Er (TRIGA) Hf (FRM-2)	B ₄ C
Other BA considered	Sm, Dy, Hf, Eu	Cd (BR-2, Petten HFR)	Cd, CdO, ZrB ₂ , Al-B alloys

3. Properties of Boron and Borated Compounds

Natural Boron has only two naturally occurring isotopes: B-10 at 19.9 at.% abundance and B-11 at 80.1 at.% abundance. It is in fact the B-10 isotope responsible for the good absorbing properties of Boron. B-10 has relatively high neutron absorption cross-section for the (n, α) reaction, that varies as $1/\sqrt{}$ (E). As the result of this reaction lithium and helium atoms are produced:

accompanied by the release of Q=2.792 MeV(Li⁷ ground) or Q=2.310 MeV(Li⁷ *) energy [4].

Production of helium gas in the fuel (in addition to fission gas) during irradiation can be of concern as it might affect fuel performance. Helium is insoluble in aluminum and would tend to precipitate into bubbles. The bubbles may agglomerate/grow in size and eventually lead to either significant swelling or blistering. In fact, it was established earlier that the addition of the borated compounds to the fuel has lowered their blister anneal temperature [5].

Boron can be added to the ATR metallic plates in the form of an alloy, for instance, as a thin Al-B foil. It can also be directly mixed with U-10Mo or U-7Mo to create a ternary U-Mo-B system. Alternatively, boron can be alloyed with the Al 6061 cladding. However, past experience with U-Al-B alloy shows that it might be difficult to achieve good control over boron loading in such alloys [6]. Moreover, the addition of boron can potentially affect mechanical and corrosion properties of the cladding material and would require cladding qualification besides fuel qualification.

Another attractive option is to incorporate boron into the ATR plates in the form of the dispersion of borides in metallic matrix. Among borated compounds boron carbide (B_4C) is perhaps the most widely used burnable absorber, though some other borides, such as ZrB_2 , are also being utilized [7, 8]. Most of the borides are refractory, inert and have high melting temperature. They also are non-reactive with Aluminum matrix. Based on tests conducted by Holden et al. [9], most diborides exhibit better helium retention properties during irradiation as compared to tetraborides and hexaborides. This fact is explained by the more compact crystal structure of diborides, where helium is more effectively trapped. For comparison, some of the properties of borated materials are listed in Table 3. Materials with the high boron content and density are preferred as burnable absorbers, since the addition of significant amounts of ceramics to the metallic fuel plate can alter its mechanical properties and make plate fabrication process more challenging.

Table 3. Properties of Borides [10]

Compound	Wt.% B	Density	B density	Melting T
		(g/cm³)	(g B/cm ³)	(°C)
B ₄ C	78.3	2.52	1.97	2350
ZrB ₂	19.2	6.17	1.18	3050
TiB ₂	31.1	4.38	1.36	3225
AIB ₂	44.5	3.19	1.42	Decomposes above 920°C

4. Quantities of Boron for ATR LEU Fuel Element

The required amounts of B-10 in the inner/outer plates of the monolithic and dispersion type LEU ATR fuel elements were estimated by Chang [3]. He also performed a separate study to minimize the number of borated plates in the ATR LEU fuel element down to four. Results of these calculations are listed in Tables 4 - 7. The different uranium loading in the monolithic plates is achieved by varying the thickness of the U-10 Mo foil, while in 25 mil

thick dispersion plates by varying the amounts of the U-7Mo in the fuel meat. It should be pointed out that both monolithic and 25 mil thick dispersion-type LEU ATR fuel elements will contain more U-235 per plate than in current HEU fuel elements. In addition, LEU elements will also require higher B-10 loadings per plate compared to HEU case.

Table 4. Compositions of the Eight Borated Plates in ATR LEU Monolithic Fuel Element

LEU	Fuel Meat	Fuel	U	U-235	U-235	B-10	B-10	B-10/
Plate	Thickness	Meat	density	Density	Mass	Mass	Density	U-235
	(mil)	Volume	(LEU)	(g/cm ³)	(g)	(g)	(g/cm ³)	Ratio
		(cm ³)	(g/cm ³)					
Plate 1	8	9.19	15.3	3.02	27.73	0.063	0.007	0.002
Plate 2	10	15.30	15.3	3.02	46.17	0.178	0.012	0.004
Plate 3	12	18.20	15.3	3.02	54.92	0.044	0.002	0.0008
Plate 4	12	19.11	15.3	3.02	57.68	0.05	0.0003	8.7E-05
Plate 16	12	30.20	15.3	3.02	91.16	0.001	3.31E-05	1.10E-05
Plate 17	10	27.58	15.3	3.02	83.23	0.033	0.0012	0.0004
Plate 18	8	21.06	15.3	3.02	63.56	0.133	0.0063	0.002
Plate 19	7	17.49	15.3	3.02	52.77	0.343	0.0196	0.0065

Table 5. Compositions of the Four Borated Plates in ATR LEU Monolithic Fuel Element

LEU Plate	Fuel Meat Thickness (mil)	Fuel Meat Volume (cm ³)	U density (LEU) (g/cm ³)	U-235 Density (g/cm³)	U-235 Mass (g)	B-10 Mass (g)	B-10 Density (g/cm ³)	B-10/ U-235 Ratio
Plate 1	8	9.47	15.3	3.02	28.57	0.058	0.006	0.002
Plate 2	8	11.82	15.3	3.02	35.68	0.149	0.013	0.004
		•••			•••			
Plate 18	8	21.7	15.3	3.02	65.49	0.111	0.005	0.002
Plate 19	8	21.49	15.3	3.02	64.85	0.317	0.015	0.005

Table 6. Compositions of the Six Borated Plates in ATR LEU Dispersion Fuel Element

LEU	Fuel Meat	Fuel	U	U-235	U-235	B-10	B-10	B-10/
Plate	Thickness	Meat	density	Density	Mass	Mass	Density	U-235
	(mil)	Volume	(LEU)	(g/cm ³)	(g)	(g)	(g/cm³)	Ratio
		(cm ³)	(g/cm ³)					
Plate 1	25	29.49	5.5	1.10	32.44	0.070	0.0024	0.002
Plate 2	25	36.84	6	1.20	44.20	0.178	0.0048	0.004
Plate 3	25	38.81	8	1.60	62.09	0.044	0.0011	0.0007
				•••	•••			
Plate 17	25	66.376	6.5	1.30	86.29	0.033	0.0005	0.0004
Plate 18	25	67.571	6.5	1.30	87.84	0.133	0.002	0.0015
Plate 19	25	66.942	5.5	1.10	73.64	0.380	0.0057	0.005

Table 7. Compositions of the Four Borated Plates in ATR LEU Dispersion Fuel Element [3]

LEU Plate	Fuel Meat Thickness (mil)	Fuel Meat Volume (cm ³)	U density (LEU) (g/cm ³)	U-235 Density (g/cm³)	U-235 Mass (g)	B-10 Mass (g)	B-10 Density (g/cm³)	B-10/ U-235 Ratio
Plate 1	25	29.49	5.5	1.08	31.96	0.058	0.002	0.002
Plate 2	25	36.84	6	1.18	43.54	0.149	0.004	0.003
Plate 17	25	67.57	6	1.18	79.87	0.111	0.002	0.001
Plate 18	25	66.94	5	0.99	65.94	0.317	0.005	0.005

5. Potential Locations of BA in the ATR LEU Fuel Element

Depending on the location, BAs are divided into two groups: Differential Burnable Absorbers (DBA) and Integral Burnable Absorbers (IBA). DBA are located outside of the fuel plate, for example, either in the side plates of the fuel element or separate plates/rods within reactor. For ATR case, we have considered an option of placing burnable absorber in the side plates of the fuel element, but found this location to be non-optimal for flattening power profile and also challenging for fabrication. As the name suggests, IBAs are the integral part of the fuel plate itself and are placed in the fuel region, cladding region or fuel/cladding interface. We have evaluated each particular region within the fuel plate for ATR LEU monolithic and dispersion types of fuel.

Fuel region

This option is most suitable for the dispersion type of LEU ATR fuel, where burnable absorber such as B_4C (or other borides) can be directly added to the fuel meat region, composed of a mixture of U-7Mo in Al-Si matrix. In fact, this is exactly how current borated HEU ATR plates are made today. The difference between the HEU and LEU plates will be in the type of fuel alloy (U-7Mo in LEU instead of U-Alx in HEU), slightly different matrix composition (higher Si in LEU) and in higher loadings of borated material per plate in LEU plates. The higher B_4C or ZrB_2 loadings can be problematic in fabrication especially at high U-Mo fuel loadings, since ceramic material tends to reduce ductility of the metallic plates.

Fuel-cladding interface

In monolithic plates the fuel-cladding interface is a sensitive area [11], protected from interaction with Zr diffusion barrier. Therefore an addition of another layer of BA to this interface would not be desirable. In dispersion plates this can be done, however dispersion fuel meat is significantly thicker than monolithic, which leaves less space in the cladding to accommodate Burnable Absorber.

Cladding region

The Al 6061 cladding on both dispersion and monolithic plates is protected from water corrosion by thin (0.00006" – 0.0003") prefilmed boehmite layer, for this reason it would not be desirable to add another layer of borated material on the surface of the cladding.

In monolithic type of fuel, due to the relatively thin fuel zone (8-13 mils), it is possible to incorporate BA as a thin layer within the cladding. This layer can be either made from B_4C or ZrB_2 dispersion in aluminum or Al-B alloy foil.

6. Irradiation Testing of Burnable Absorbers in ATR (RERTR-13 Experiment)

It is extremely important to select such burnable absorber for the ATR LEU fuel element that does not degrade fuel performance in the reactor and maintain sufficient safety margin. To assess performance of different borated burnable absorbers and assist in the BA selection process for both monolithic and dispersion types of fuels, we have initiated an RERTR-13 experiment in ATR. The test consists of 28 borated miniplates, 8 of which are non-fueled material test plates, 8 dispersion plates and 12 monolithic plates. The following burnable absorber materials were chosen for testing: B_4C dispersion in aluminum, ZrB_2 dispersion in aluminum, Al-1.5B (wt.%) and Al-4.5B (wt.%) alloys. One of the main contributing factors behind selection of these particular compounds was the availability of materials and the ability to fabricate desired plates in the relatively short period of time. The B-10/U-235 ratios in the miniplates were prototypic of the monolithic (0.0065 and 0.002) and dispersion (0.004 and 0.002) LEU ATR plates.

The prime phenomena of interest in these plates are helium behavior, fuel swelling, blister anneal failure temperature and interaction layer behavior.

6.1 Irradiation Conditions

The miniplates were loaded in 4 capsules, with 2 stacks of 4 miniplates per each capsule. The capsules will be inserted in ATR position B-10 for two irradiation cycles, with the miniplates positioned in the "edge-on" configuration towards the reactor core center. The test was designed not to exceed surface heat flux limit of 350 W/cm², however it should be realized that boron suppresses heat flux within fuel up to 30% at the beginning of life. The RERTR-13 fuels and materials will be tested in the range of fission rates and coolant temperatures and will be exposed to the different levels of burnup. This is accomplished by varying U-235 enrichments and axial positions of the plates within the test assembly. It is anticipated that B-10 will be depleted 70-99% at the end of irradiation. U-235 will be depleted 30-40% in LEU plates and 65-100% (LEU equivalent) in HEU plates.

6.2 Design of the RERTR-13 Miniplates

Borated Monolithic Miniplates

Borated monolithic miniplates consist of 0.010" (0.254 mm) thick U-10Mo foil coated with $\sim 0.001"$ (0.0254 mm) layer of Zr on both sides at the fuel-cladding interface (See Fig. 3). Uranium is 19.7 and 66% enriched in U-235. A 0.005" (0.127 mm) thick borated layer is incorporated in the cladding region, physically separate from the fuel region. In such arrangement helium will be generated in the different region than fission gas. Two of the monolithic plates contain Al-1.5B alloy (95.9 wt.% enriched in B-10), 6 plates contain 2 vol.% B₄C dispersion in Al (97.58 wt. % enriched in B-10) and 4 plates contain 4.5 vol.% ZrB₂

dispersion in AI (75.17 wt.% enriched in B-10). The AI-1.5B alloy procured from Ceradyne, Inc. and rolled to thickness at the INL. The B_4C and ZrB_2 particles (400 mesh, less than 37 micron) from Ceradyne, Inc. were incorporated in the AI dispersion matrix and rolled into a thin foil at the INL. The enriched B-10 compounds were used to minimize the loading of refractory borides in the aluminum matrix. It is worth mentioning that to achieve equivalent boron loading per plate more ZrB_2 material required than that of B_4C .

The borated layer was incorporated in the Al-6061 cladding using the standard roll technique. The whole fuel assembly was clad using the Hot Isostatic Press (HIP) process.

Borated Dispersion Miniplates

Borated dispersion miniplates consist of 0.025" (0.635 mm) thick fuel meat. The meat is composed of U-7Mo alloy (with the density of 6 g U/cc) dispersed in Al-4 wt.% Si matrix. Uranium is 25 and 69% U-235 enriched. Four of the dispersion plates contain 1.5 vol.% B_4C (19.9 at.% B-10) and the other four 2.5 vol.% ZrB_2 (19.9 at.% B-10). Both borides were directly added to the dispersion matrix. Dispersion plates were clad in Al-6061 by rolling. All of the dispersion plates were manufactured at Argonne National Laboratory (ANL).

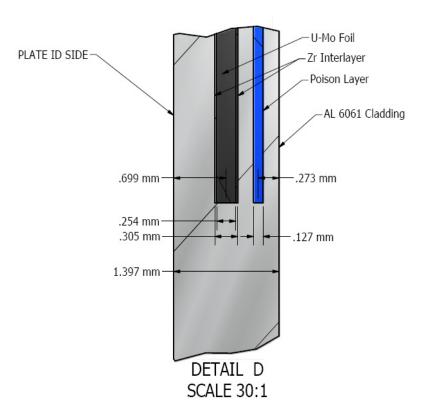


Figure 3. Borated Monolithic Miniplate

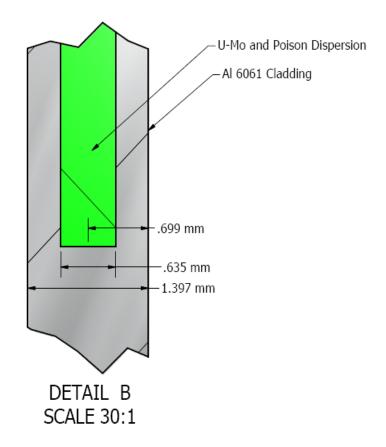


Figure 4. Borated Dispersion Miniplate

Borated Material Test Miniplates

The objective of testing borated material miniplates is to examine behavior of the Burnable Absorbers in the absence of fuel and potentially decouple effects of helium behavior from that of fission gas. Moreover, it is anticipated that material test plates will have significantly lower exposure rates after irradiation than plates containing fuel and thus can be submitted for the destructive post irradiation examination earlier.

All material test plates contain a 0.005" (0.127 mm) thick borated layer composed of one of the following: dispersion of 2 vol.% B₄C in Al (97.58 wt.% enriched B-10), dispersion of 4.5 vol.% ZrB₂ (75.17 wt.% enriched B-10) in Al, Al-1.5B (95.9 wt.% enriched B-10) and Al-4.5B (98.5 wt.% enriched B-10) alloy.

The Al-B alloys were procured from Ceradyne, Inc. To ensure acceptable workability and homogeneity of these alloys, the Al-4.5B composition was made at Ceradyne by the addition of boron halide to molten aluminum. As a result, boron is present in a dispersed phase of AlB $_2$ in Al-4.5B. The Al-1.5B was manufactured by adding a mixture of boron-halide and titanium-halide compounds to molten aluminum. Therefore boron is located in a dispersed TiB $_2$ phase in Al-1.5B alloy. These alloys were rolled to nominal thickness at the INL and clad in Al-6061 by HIP process.

The B_4C and ZrB_2 particles (400 mesh, less than 37 micron) from Ceradyne, Inc. were incorporated in the Al dispersion matrix and rolled into a thin foil at the INL.

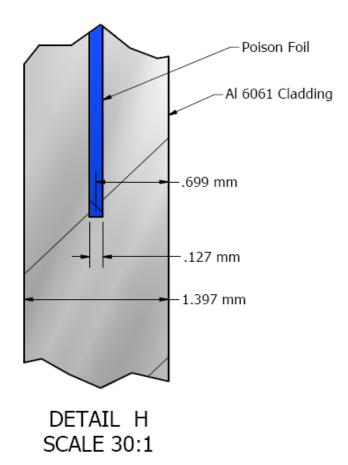


Figure 5. Borated Material Test Plate

6.3 Post-Irradiation Examination (PIE)

The selection of the Burnable Absorber will be made based on results of the post-irradiation examination of the RERTR-13 miniplates. Tentatively, post-irradiation examination will include visual examination, profilometry, oxide thickness and immersion density measurement, gamma scanning and determination of the blister failure temperature during annealing of the irradiated plates. In addition metallography, Scanning and Tunneling Electron Microscopy (SEM and TEM), and determination of burnup will be performed on irradiated fuel sample.

As mentioned earlier, production of helium from boron during irradiation can affect fuel performance. Therefore one of the objectives of PIE will be to evaluate helium behavior in all of the RERTR-13 miniplates. Of particular interest are helium retention in the lattice structure of different diborides and/or its precipitation in aluminum, the size of the bubbles formed, diffusion at operational temperatures and ultimately comparison of helium performance with that of fission gas. For this reason we have estimated helium generation

rates at the beginning of life and the total amounts of helium that will be produced at 100% B-10 depletion in the borated region of the plates. Results are presented in Tables 8 and 9.

Table 8. Estimated Helium Generation Rates in the RERTR-13 Miniplates

Type of the plate	Volume of borated region, (cm ³)	Helium generation rate at the beginning of life, atoms/ (cm³ borated region)/sec
Material Test Plate	0.2	0.54 - 1.99 x 10 ¹⁵
Monolithic Fuel Plate	0.2	4.02 - 6.69 x 10 ¹⁴
Dispersion Fuel Plate	0.999	3.97 - 7.71 x 10 ¹³

Table 9. Estimated Total Amounts of Helium Produced in the RERTR-13 Miniplates at 100% B-10 Depletion

Type of the miniplate	Borated region	Volume of the borated region, (cm ³)	Mass of B-10 in borated region, (g)	Atoms of helium in borated region
Dispersion plate	B ₄ C or ZrB ₂ in the fuel meat	0.999	0.0054	3.28 x 10 ²⁰
Monolithic fuel plate and Material test plate	B ₄ C or ZrB ₂ dispersion layer or Al-1.5B layer	0.2	0.0079	4.77 x 10 ²⁰
Material test plate	Al-4.5B layer	0.2	0.0235	1.42 x 10 ²¹

7. Summary

We have evaluated Burnable Absorbers for ATR LEU Fuel Element based on general selection criteria and ATR specific limitations. Due to ATR short fuel life and fuel fabrication constraints the choice of BA is limited to borated compounds. From the initial results of the ATR reactor physics calculations we have selected compositions of the borated fuel plates for testing and methods of incorporation of Burnable Absorbers into the Fuel Element. To access performance of different borated compounds we have designed and fabricated 28 RERTR-13 borated miniplates that will be tested in ATR for two irradiation cycles. To predict the levels of gas generated during irradiation we have estimated helium generation rates and total Helium production in the RERTR-13 miniplates. Selection of the Burnable Absorber for ATR-LEU will be based on the results of RERTR-13 PIE.

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