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PRELIMINARY EVALUATION OF BETA-SPODUMENE AS A FUSION REACTOR STRUCTURAL MATERIAL

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Beta-spodumene was investigated as a candidate material for use in fusion reactor environments. Properties which support the use of beta-spodumene include good thermal shock resistance, a very low coefficient of thermal expansion, a low-Z composition which would result in minimum impact on the plasma, and flexibility in fabrication processes. Specimens were irradiated in the Advanced Test Reactor (ATR) to a fluence of 5.3×10^{22} n/m², $E > 0.1$ MeV, and 4.9×10^{23} n/m² thermal fluence in order to obtain a preliminary evaluation of the impact of irradiation on the material. Preliminary data indicate that the mechanical properties of beta-spodumene are little affected by irradiation. Gas production and release have also been investigated.

1. INTRODUCTION

Many of the proposed applications of ceramic materials for fusion systems are dictated by the need for electrical insulators. Magnetic coils, toroidal current breaks, neutral beam injectors, RF heating systems, and direct converter systems in magnetic confinement reactors, require highly electrically resistant materials which can maintain their electrical integrity under relatively high temperatures, high neutron fluences, and some mechanical stress. Low-Z ceramic materials have also been proposed for first wall (or liner) applications which theoretically would reduce plasma impurity radiation and neutron-induced radioactivity, as well as allow higher operating temperatures [1]. However, there are serious questions as to the long-term reliability of ceramic materials, owing to degradation of properties induced by the environment. Certainly, one of the more potentially detrimental aspects of this environment to ceramics is the thermal cyclic nature of pulsed fusion systems. Subsequently, thermal shock fracture and/or thermal fatigue leading to fracture could occur in ceramic components resulting in a premature shutdown of the entire system.

The propensity of a material for thermal shock and fatigue damage is directly proportional to its coefficient of thermal expansion. Beta-spodumene, $\text{Li}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2$, having the unique property of an extremely low thermal expansion, $0.7 \times 10^{-6}/\text{K}$, would be little affected by thermal gradients [2]. Furthermore, the fabrication of beta-spodumene ceramic ware by casting and refiring, a

technique used by several manufacturers, allows fabrication of relatively intricate shapes.

However, the lithium content in beta-spodumene raises the question of radiation stability vis-a-vis the $\text{Li}(n,^3\text{H})^4\text{He}$ transmutation reaction, as well as structural damage effects. While there are other materials considerations which will eventually need to be addressed, this paper presents a preliminary evaluation of the effects of irradiation upon the mechanical strength and gas production in beta-spodumene.

2. BETA-SPODUMENE

Beta-spodumene ($\text{LiAlSi}_2\text{O}_6$), as well as beta-eucryptite (LiAlSiO_3), can be considered as "stuffed beta-quartz" structures with spiral chains of relatively rigid Si-O and Al-O tetrahedra arranged around fourfold screw axes.

The framework is basically 5-membered rings of alumina-silica tetrahedra which form channels along (010) and (100) with the lithium ions stuffed in the 3A diameter zeolite-like channels. The lithium ion occupies the 8b positions of the space group $P4_32$ [3-5].

The anisotropical thermal expansion of beta-spodumene, by which the a-axis contracts while the c-axis expands upon heating, is a result of changes in bond angles between neighboring silica and alumina tetrahedra. An analogy to the mechanical helical spring has been proposed for the thermal behavior of both beta-eucryptite and -spodumene. The relatively rigid helical spring reacts under torsional stress by contracting. The torsional stress in beta-spodumene is introduced as the spring material, the Si-O and Al-O framework, attempts to expand upon heating. By addition of SiO_2 , which enters into solid solution with

K20

beta-spodumene, a balance of a- and c-axis behavior can be obtained resulting in a near zero unit cell expansion [5-6].

3. EXPERIMENTAL PROCEDURE

3.1 Specimen Preparation

Specimens were prepared by melting, at 1923 K, blended oxide powders with the composition given in Table 1. The composition was selected to closely approximate commercially available products such as Cervit or Corning 9608 [2].

TABLE 1. COMPOSITION OF GLASS-CERAMIC

	Wt. %
SiO ₂	70
Al ₂ O ₃	18
MgO	3
Li ₂ O	3
TiO ₂	5
As ₂ O ₅	1

The melt was poured onto a flat surface and quenched into glassy plates 5.0 mm thick. Specimens were cut from the plates via a diamond saw into coupons 127 x 127 mm. The coupons were devitrified in two stages, first at 1073 K for 16 hrs, then at 1273 K for 24 hrs, and subsequently cooled to room temperature.

3.2 Phase Identification

The crystalline phases were identified by X-ray diffraction scans. Beta-spodumene was identified as the primary crystalline phase present with a minor amount of beta-eucryptite.

3.3 Specimen Irradiation

Prior to irradiation, thermal analysis calculations were made to determine the temperature profile over the specimens and the type of thermal bond to be used during the irradiation. Individual specimens were encapsulated in 15.87 mm diameter aluminum tubes with a helium gas at one atmosphere pressure as the thermal bond. With a gamma heating of 1.55 W/g, a peak specimen temperature of 408 K was predicted for the irradiation. The irradiation assembly included six separate tubes, each 152 mm long, which were welded to two aluminum rings 22.2 mm in diameter to form a cylindrical array. The assembly was inserted in the Advanced Test Reactor hole I-8

during cycle 48A-1, shutdown and irradiated for a total exposure period of twenty-nine effective full power days. The neutron fluence obtained is as follows:

Group Energy	Fluence
0.821-10 MeV	2.8 x 10 ²² n/m ²
5.53 keV-0.821 MeV	6.3 x 10 ²² n/m ²
0.532 eV-5.53 keV	1.1 x 10 ²³ n/m ²
0-0.532 eV	4.9 x 10 ²³ n/m ²

The fluence values reported above are based on a reactor flux calculation and the results obtained from the capsule irradiation. De-encapsulation of specimens after irradiation was performed in a hot cell using conventional techniques.

3.4 Mechanical Testing Procedure

Coupon specimens were tested to failure in three-point bending. Testing was performed on a self-aligning fixture using an Instron (model TTD) Tensile machine at a cross head speed of 0.021 mm/s with load and displacement recorded.

Since six of these coupons had been irradiated and required remote handling, all specimens, i.e., both irradiated and nonirradiated, were tested remotely in a hot cell facility at INEL. Additionally, upon fracture of the irradiated specimens there was a potential for tritium release. To provide tritium containment, all specimens, both irradiated and non-irradiated, were encapsulated in individual 0.05-mm thick polyethylene tubes.

All specimens exhibited a small amount of curvature in the plane of bending which varied from specimen to specimen. Testing was performed with the concave side in compression such that the minimum strength was measured.

Following testing, the maximum stress at failure was calculated using standard three-point bending analyses which also yielded the modulus of elasticity.

3.5 Gas Analysis

Since beta-spodumene contains ten atom percent lithium, both tritium and helium will be produced in the material during any neutron irradiation. The transmutation reaction proceeds with thermal neutrons for ⁶Li and with neutrons having energies greater than 2.5 MeV for ⁷Li. In order to quantify the amount of gas produced during irradiation and determine its disposition, miniature capsules containing beta-spodumene were prepared by Rockwell International for inclusion in the irradiation

assembly. These mini-capsules contained fractional milligram quantities of beta-spodumene sealed in 0.24 mm walled vanadium cladding. Six mini-capsules were prepared, four containing known amounts of ceramic and two which were empty. A mini-capsule was included inside the aluminum tube with each primary specimen for irradiation and later recovered when the assembly was opened in the hot cell.

Three irradiated mini-capsules, two containing beta-spodumene and one blank, were returned to Rockwell International for gas analysis along with a fragment which was taken from one of the primary specimens. Prior to performing the gas analysis, the vanadium capsules were etched in dilute nitric acid to remove any material that could have been contaminated by helium-generated surface impurities or cover gas. The capsules were analyzed for both ^4He and ^3He , the latter resulting from the decay of ^3H which was formed simultaneously with the ^4He during the irradiation. Additionally, the analysis differentiated between the gas which was released from the material and that which was retained by the beta-spodumene. This was accomplished by first recovering the gas released when the capsules were opened and then recovering separately the gas released when the samples were melted.

4. RESULTS AND DISCUSSION

4.1 Gas Analysis

The two encapsulated specimens which were evaluated had produced 328 and 280 atomic parts per million ^4He , respectively. Ninety-two and eighty-two percent of the ^4He and ^3He , respectively, had been released from the ceramic into the capsule atmosphere. Since the ^3He depends on the behavior of the tritium, it is not surprising that the release fractions differ.

The amount of He released is surprising. However, the open crystalline structure of beta-spodumene apparently allows He with an atomic radius of 0.8A [7] to pass through the structure.

4.2 Mechanical Behavior

The results of the three-point bending tests are given in Table 2. The failure stress was lowered only slightly (2.75%) from a mean failure stress of 40.25 MPa to 39.14 MPa. The elastic moduli, however, increased 12% from a mean of 1.86×10^4 MPa unirradiated to 2.09×10^4 MPa for the irradiated samples. The gain in modulus is probably greater than

TABLE 2. MECHANICAL STRENGTH AND ELASTIC MODULI OF BETA-SPODUMENE BEFORE AND AFTER IRRADIATION

Specimen	Deflection (mm)	Failure Stress (MPa)	Mean Failure Stress (MPa)	Modulus of Elasticity $\times 10^4$ (MPa)	Mean Modulus/Standard Deviation
1	0.625	40.86		1.94	
2	0.472	34.54		2.18	
3	0.584	40.42	39.14/2.35	2.06	2.09/0.16
4	0.564	38.95		2.05	
5	0.497	39.68		2.37	
6	0.625	40.42		1.92	
1a	0.731	39.7		1.64	
2a	0.803	42.74		1.59	
3a	0.762	40.71	40.25/1.67	1.59	1.86/0.59
4a	0.756	39.95		1.57	
5a	0.391	38.17		2.91	

a. Indicates nonirradiated specimens.

the calculated value since the mean value is affected by the 2.91 MPa anomalous data point.

The mechanical strength of beta-spodumene is virtually unaffected by irradiation. This is somewhat surprising considering the amount of gas generated via the $n-\alpha$ reactions. The gain in elastic modulus at the fluence level tested, while substantial, is not a critical factor since the elastic modulus of beta-spodumene is relatively low. However, this increase in elastic modulus does indicate that some damage has occurred, but not enough to generate flaws of sufficient size to lower the stress necessary for failure.

5. CONCLUSIONS AND RECOMMENDATIONS

The effects of irradiation on the mechanical behavior of beta-spodumene are minimal. The increase in elastic modulus after irradiation, which is indicative of some alteration of structure, does not appear to be prohibitive with regard to potential fusion reactor use. Although the data presented are limited in scope, the relatively unaffected behavior of beta-spodumene, combined with its unique thermal expansion behavior warrants further investigation of the material for use in fusion environments. Specific data are needed to quantify electrical and thermo-mechanical behavior as a function of neutron fluence.

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