Use of Beryllium and Beryllium Oxide in Space Reactors

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Use of Beryllium and Beryllium Oxide in Space Reactors

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Abstract. Beryllium and beryllium oxide are attractive candidate materials for neutron reflector application in space reactors due to their beneficial combination of low density and high neutron moderation and reflection capabilities. Drawbacks to their use include the expense of working with toxic materials, a limited industrial infrastructure, and material properties that are challenging in the non-irradiated state and seriously degrade under neutron irradiation. As an example of neutron effects, mechanical properties degrade under relevant conditions to the point where encasement in structural alloys is necessary. Such measures are required if neutron fluence exceeds ~1×10^{24} n/m^2 (E>0.1 MeV). At high temperatures (>500˚C for Be and >600˚C for BeO), irradiation-induced swelling may also limit the maximum allowable dose without additional engineering measures. Significant volumetric swelling (>5%) can occur in these materials during neutron irradiation at elevated temperatures for neutron fluences above 1×10^{25} n/m^2. This paper will review Be and BeO fabrication considerations, and summarize the effects of neutron irradiation on material properties.

INTRODUCTION

Due to their low density and good neutron scattering properties, nuclear grade beryllium metal and ceramic beryllium oxide have been proposed for a wide range of space nuclear power systems. In particular, they are leading candidates for axial (BeO) and radial (Be) neutron reflectors. This paper briefly reviews the domestic infrastructure for manufacture and fabrication of these materials for near-term space reactor application. Additionally, the effects of neutron irradiation on properties of the beryllium (Barabash et al. 2000; Khomutov, 2002; Snead, 2004) and berylium oxide (Elston and Caillat 1958; Keilholtz et al., 1964; Kelly, 1994; McDonald, 1963) are discussed with particular attention given to application temperature and maximum neutron doses. As discussed in the following, both of these materials can become very brittle following neutron irradiation. Therefore, they must be encased in a structural material cladding.

BERYLLIUM : FABRICATION OVERVIEW

Working with beryllium containing materials has become increasingly difficult due to the discovery of Chronic Beryllium Disease (CBD) more than 50 years ago. The combination of litigation surrounding CBD and the safeguards now standard for handling beryllium have severely reduced the domestic manufacturing and fabrication infrastructure, as well as the demand, for beryllium. Currently, Brush-Wellman (BW) is the principal domestic manufacture of beryllium supplying high-purity structural and optical grade materials for applications such as the space-shuttle door and window frames, structural members and optics on satellites, and some domestic applications such as structure of the Webb telescope. Recently, BW negotiated with the Defense Logistics Agency to procure high-purity beryllium (previously mined and purified by BW) from the National Stockpile ensuring a stable supply of beryllium raw material. Moreover, the BW Topaz Mountain Range Mine in Utah is still in operation. Currently, BW is still investing in new facilities for processing of beryllium and is capable of providing nuclear grade vacuum hot pressed material (such as S65). The standard billet size in current production is 46” in diameter and 42” in length. Additionally, BW has current capability to produce a range of tube products up to 70” in diameter. There are currently about 15 beryllium commercial machining outfits domestically. Axis Technologies (formerly Speedring) is the largest machining company and has the capability for the largest finish machining. Additionally, a

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California based BW subsidiary, Electrofusion Products, has a wide range of capability for joining of beryllium (brazing, electron beam welding, etc) and the ability to form fairly complex structures.

BERYLLIUM : PROPERTIES AND IRRADIATION EFFECTS

Beryllium has been considered for nuclear fuel cladding, nuclear fuel compacts and as a neutron moderator for fission power plants dating back to the early 1950's. Other than the non-structural application as a core moderator/reflector, this material has found limited use in nuclear applications due to its low ductility even in the absence of irradiation. Beryllium is found in nature as beryl $Be_3Al_2Si_6O_{18}$ and chrystoberyl $BeAl_3O_4$. In its pure metallic form it has a highly anisotropic hexagonal crystal structure with very interesting anisotropic properties. The use of beryllium has been reviewed for use in various fusion devices such as International Thermonuclear Experimental Reactor (ITER) and therefore nuclear performance has been a focus. The ITER program in particular has assembled properties in a handbook form. Additionally, a comprehensive report on ITER-grade beryllium has been published as part of the Atomic and Plasma-Material Interaction Data for Fusion (Dombowski et al., 1994). Another excellent recent source of information on beryllium in nuclear applications is the biannual International Energy Agency workshop proceedings on the subject, for example (Longhurst, 1995).

The limited ductility of the various types of beryllium is a function of many factors including temperature, chemical purity, grain size and to some extent the rate at which the material is strained. Moreover, the hexagonally close packed beryllium crystal itself is resistant to slip, severely limiting ductility potential of the material. The beryllium hcp crystal has only two operating slip modes (at least at low temperatures) being basal slip on {0001} planes and prismatic slip on {1010} planes. For high purity beryllium, ductility can be categorized into three temperature-dependent regimes. In the low temperature regime (T<200 °C) the shear stress required to activate prismatic slip is quite high and failure typically occurs by transgranular fracture of the basal plane. Total elongation in this temperature range for high quality vacuum hot pressed material (e.g. Brush Wellman S-65C at 150 °C) is ~5% in the direction parallel to the pressing direction and ~20% in the transverse direction. As the temperature is increased above this lower temperature regime the critical stress for prismatic slip decreases and both slip modes combine to yield peak total elongations of about 50% both parallel and transverse to the forming direction due to anisotropic texturing of the grains. In this intermediate temperature regime (~200-500 °C) the failure is primarily ductile/fibrous tearing. As the temperature is further increased, intergranular failure begins to occur returning the total elongation to below 20%.

Irradiation of beryllium with high-energy neutrons has the effect of producing small dislocation loops or "black spots" for temperatures <400 °C (Barnes, 1961; Beeston et al., 1973; Gelles and Heinisch, 1992; Rich and Walters, 1961; Walters, 1966). Helium, formed by interaction of beryllium with fast neutrons, forms visible bubbles at temperatures above 325-400°C(Beeston et al., 1984; Gelles et al., 1994). The helium bubbles tend to form at grain boundaries from 325-600°C(Beeston, 1967; Gelles et al., 1994; Hickman and Bannister, 1963; Hickman et al., 1963; Hyam and Sumner, 1962; Rich and Walters, 1961; Stevens and Hickman, 1965; Sumerling and Hyam, 1961; Weir, 1961) and are also reported to decorate dislocations within grains in the temperature range of 450-550°C(Gelles et al., 1994; Rich and Walters, 1961). In a recent work(Chakin and Ostrovsky, 2002) at high dose (1 x 10^{26} n/m^2, E>0.1 MeV) at an irradiation temperature of ~100 °C Chakin and Ostrovsky observed a high density of ~ 20 nm interstitial dislocation loops. No helium bubbles were resolvable until the sample was annealed to greater than 500 °C at which point evenly distributed 1-4 nm bubbles were seen.

The properties of nuclear-grade beryllium have been studied before and after neutron irradiation at conditions relevant for space reactor application (Barabash et al., 2000; Gelles and Heinisch, 1992; Scaffidi-Argentina et al., 2000). The impact of neutron irradiation fluences up to 1x10^{21} n/cm^2 on the cavity swelling behavior of Be is small for temperatures below 500°C, whereas significant swelling (>>1%) occurs at higher temperatures. The amount of swelling in Be increases with increasing temperature above 500°C. Figure 1 summarizes some of the swelling data obtained on nuclear-grade Be (Barabash et al., 2000). The amount of swelling at high temperatures (>500 °C) can be reduced by using fine-grained Be (Barabash et al., 2000; Gelles and Heinisch, 1992). Be has a high fast neutron cross-section for He production, and therefore high-temperature He embrittlement (intergranular cracking) is a concern for Be components exposed to neutron fluences greater than ~1x10^{21} n/cm^2 at temperatures above ~500 °C (corresponding to 0.5 T_M for Be, where T_M is the melting temperature). Neutron irradiation studies have observed that the strength of irradiated Be decreases rapidly at temperatures above 600 °C in specimens irradiated to neutron fluences >1x10^{21} n/cm^2 (Barabash et al.; 2000, Gelles and Heinisch, 1992). The high temperature embrittlement of irradiated Be is less severe in fine-grained material compared to large-grain material due to distribution of helium over more grain boundary area in fine-grained specimens. Temperature excursions above 600 °C in space fission reactor components would likely produce high swelling (due to growth of He bubbles) and cause cracking within the Be (with resultant decreases in thermal conductivity) and significant stress on the surrounding structural casing. The helium production in the Be reflector needs to be calculated to determine if He bubble swelling is an important lifetime-limiting consideration. The probability of temperature excursions above 600 °C in the radial reflector also needs to be determined. Operation of the radial reflector at temperatures below ~550 °C should produce acceptable behavior in Be encased in structural cladding for moderate-power (10-100 kW_{th}) space reactors with operating lifetimes of a few years.

Both point defect clusters and helium bubbles adversely affect the mechanical properties of beryllium, regardless of its metallurgical form. Point defect clusters and bubbles impede dislocation motion resulting in a severe reduction in elongation and increased strength (Bartz, 1958; Beeston 1967, Beeston, 1970; Beeston et al., 1992; Beeston et al., 1973; Hickman, 1961; Hickman and Bannister, 1963; Hyam and Sumner, 1962; Morozumi et al., 1977; Rich and Walters, 1961; Stevens and Hickman, 1965; Walters, 1966) and hardness (Beeston, 1970; Hickman, 1961; Hickman and Bannister, 1963; Rich and Walters, 1961; Weir, 1961). The irradiation effects database has been reviewed in the past (Beeston, 1970; Dalle-Donne et al., 1998; Gelles and Heinisch, 1992; Scaffidi-Argentina et al., 2000) mainly including work conducted prior to 1970. More recently a number of research groups have been studying the thermomechanical properties of state-of-the-art forms of beryllium for fusion reactor applications (Chakin et al., 2002; Chaouadi et al., 1997; Fabritsiev et al., 1999; Ishitsuka and Kawamura, 1998; Kupriyanov et al., 1996; Moons et al., 1998; Moons et al., 1996; Pokrovsky et al., 1996; Sernyaev et al., 1999). Good reviews of the more recent work can be found in the literature (Barabash et al., 2000; Khomutov 2002). A recent paper by Snead (Snead 2004) reviews the thermomechanical behavior of high-quality beryllium under low-temperature (<300 °C) space-reactor relevant neutron fluence conditions. Moreover, this paper provides data on Brush-Wellman S-65 grade beryllium, which would likely be a prime candidate space reactor application. Of importance is that for S-65 grade beryllium significant tensile embrittlement occurs for a fast neutron dose as low as 1 x 10^{24} n/cm^2 (E>0.1 MeV.) Thermal conductivity exhibits no change at these low temperatures to doses an order of magnitude higher.

BERYLLIUM OXIDE : FABRICATION AND OVERVIEW

Beryllium oxide (BeO) was studied extensively as a fission reactor moderator throughout the 1950's and 1960's. The primary attraction to this material (in addition to its moderating power-it has essentially the same number density of beryllium atoms as beryllium metal) is its high-temperature capability (melts at 2550 °C) and good thermal conductivity. Moreover, it is relatively radiation resistant.

Beryllium oxide powder is processed either through the fluoride route or by the sulfuric acid route, producing beryllium hydroxide which can be reduced to the oxide form by heat-treating to 1800°C. Purification of the powder is typically carried out for reactor use by dissolution in sulfuric acid, and removal of aluminum by precipitation with ammonium sulfate. The sulfate powder is then crystallized and calcined to oxide at 1150°C. Monolithic beryllium oxide ceramic can be formed using a variety of conventional ceramic processing routes. A variety of binder materials such as resins or starches may be used as pre-firing binders. Where machining of sintered products is required, a pre-firing of the article is carried out at temperatures from 1200-1500°C, after which machining is possible with carbide tipped tools, followed by a final heat treatment at 1700-2000°C.

The densities of cold pressed and sintered materials may vary considerably, but high densities may be produced using high purity oxide and careful control of the pressing and heating conditions. Hot pressing powders at temperature of ~ 1700 °C achieves densities close to the theoretical density of 3.03 g/cc. At temperatures above ~ 1800 °C crystal growth becomes rapid and the ceramics suffer significant loss in strength. Optimum hot pressing conditions are generally in the 1700-1800 °C range. As with density, the thermophysical properties of beryllium oxide properties are a strong function of manufacturing conditions. The leading supplier of BeO in the US is Brush-Wellman. As with metallic beryllium the domestic infrastructure has been severely limited. However, Brush Technical Ceramic and Axis Technologies have the capability of pressing and machining BeO in large plate, rod and tubular geometries.

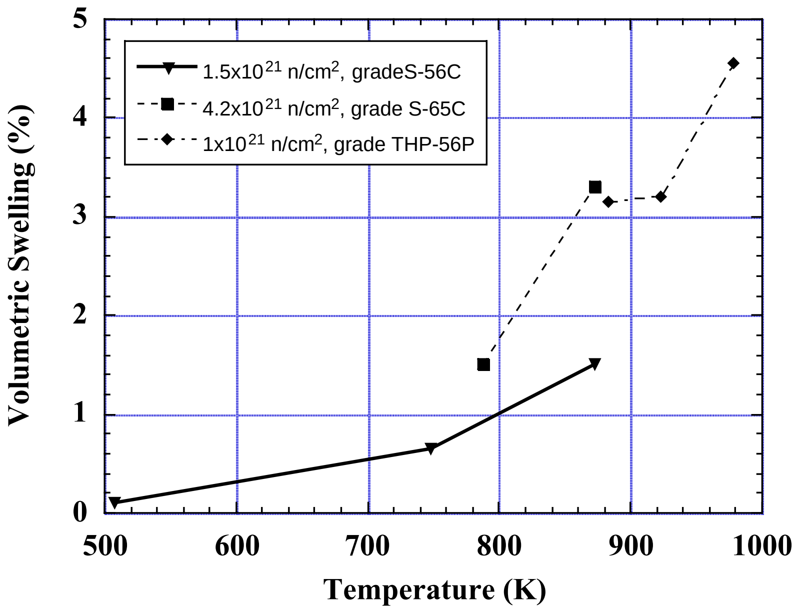


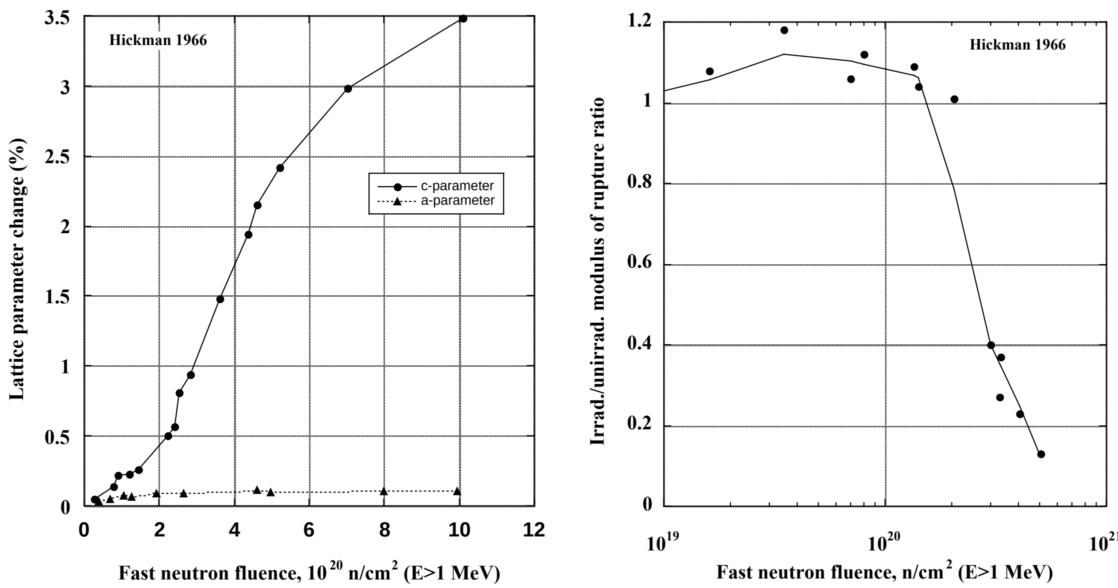
FIGURE 1. Summary of Swelling Behavior of Nuclear-grade Beryllium After Neutron Irradiation at Various Temperatures (Barabash et al., 2000).

BERYLLIUM OXIDE : PROPERTIES AND IRRADIATION EFFECTS

There are a number of good references on the irradiation performance found in the early irradiation-effects literature (Elston and Caillat, 1958; Elston and Labbe, 1961; Gilbreath and Simpson, 1958; Keilholtz et al., 1964; McDonald, 1963; Wilks, 1968) including the proceeding of a conference on the use of BeO in nuclear applications (Beeston et al., 1973) and a general review (Simnad et al., 1966), though these are limited to pre-1970 materials. Irradiation-induced swelling of BeO was studied in the early 1960's (Clark et al., 1961, Elston and Labbe, 1961) and indicated that bulk material swelled isotropically at a rate proportional to fast neutron fluence for irradiation temperatures of ~ 100°C. The dimensional changes were consistent with the lattice dilations when the anisotropic swelling of the hexagonal close packed crystal basal "c" and prism "a" axes is taken into consideration. Early work showed that the changes in the "c" axis dilation are about ten times greater than those in the "a" axis (Elston and Labbe, 1961). Hickman and Pryor (Hickman and Bannister, 1963) have developed a detailed model for the macroscopic irradiation-induced volume change for BeO in the absence of micro-cracking, which fits the low-dose data well.

At low doses, pinning of dislocations leads to material strengthening (and increased elastic modulus) as observed by Clarke (Clarke, 1963). For intermediate dose (> $1x10^{24}$ n/m^2, E>1 MeV) anisotropic swelling of the BeO crystallites lead to serious microscopic strain and eventual disintegration of the ceramic. Microcracking in polycrystalline samples can be observed by an increase in open porosity, a decrease of the strain component of X-ray line broadening and a sharp decrease in bend strength. As reviewed by Wilks (Wilks, 1967; Wilks, 1968), the irradiation induced microcracking in BeO depends upon the grain size, density, fabrication method and the irradiation temperature. At a given irradiation temperature microcracking and powdering occur at decreasing dose with increasing grain size, whereas for a given grain size the doses required increase with irradiation temperature. The dose for onset of microcracking increases from ~0.5 to $4x10^{24}$ n/m^2, E>1 MeV as the BeO grain size is decreased from 100 to ~ 4 μm (Hickman, 1966). Cold pressed and sintered material is more resistant to microcracking than hot-pressed material, probably because the latter tends to already contain nascent microcracks prior to irradiation.

Figure 2 gives example data showing the expansion of the "c" and "a" axis lattice parameters as a function of neutron dose (Hickman, 1966). The degradation in strength caused by the onset of microcracking is also shown in this figure. The swelling-induced microcracking is a function of temperature, with loss of mechanical properties occurring at higher dose for higher temperature irradiation. For example, a dose of ~ 2 x 10^{25} n/m^2 is the approximate lifetime for materials irradiated to ~100 °C. For temperatures above ~ 350°C, a fluence limit of > 4 x 10^{25} n/m^2 can be set for early grades of BeO (Keilholtz et al., 1966; McDonald, 1963). It should be noted that the degradation in mechanical properties cannot be simply explained by anisotropic point defect swelling (lattice parameter change) of crystallites which occurs at low temperature. Whereas the lifetime dose of BeO does increase with temperature, the lattice parameter change at elevated temperatures is significantly reduced and approaches zero,



(a) BeO Lattice Parameter Change. **(b)** Change in BeO Strength.

FIGURE 2. Effect of 75-100 °C Neutron Irradiation on Beryllium Oxide (Hickman, 1966).

yet the mechanical properties still degrade. The reason for the higher temperature degradation in mechanical properties is unclear, but may be caused by either an intrinsic material response (such as loop swelling or helium effects) or an artifact of the processing (such as grain boundary impurities).

The thermal conductivity of BeO degrades with neutron irradiation, due to phonon scattering associated with creation of lattice vacancies and internally generated voids and microcracks. This thermal conductivity degradation has been studied by Elston and Labbe (Elston and Labbe, 1961) and others (Gilbreath and Simpson, 1958; McGill and Smith, 1959) For the case of Elston and Labbe (Elston and Labbe, 1961) thermal conductivity was reduced by 80% following 100°C fast neutron irradiation to 6 x 10^{19} n/m^2. However, the thermal conductivity degradation due to vacancy creation in BeO is less on a normalized dose basis as compared to other high-temperature ceramics such as aluminum nitride and silicon carbide. The effect of neutron irradiation at 60 °C on the thermal conductivity of Brush-Wellman BeO-995 and other high thermal conductivity ceramics is given in Figure 3. From this figure it is apparent that BeO does not suffer thermal conductivity degradation to fast neutron doses up to ~1 x 10^{22} n/m^2 (E>0.1 MeV) at 60 °C, whereas other moderators such as graphite would suffer substantial thermal conductivity degradation. For higher irradiation temperatures, the defect mobility of BeO is considerably higher than other ceramics such that the (crystal) thermal conductivity degradation is very low. However, as discussed earlier, strain-induced microcracking would significantly degrade the thermal conductivity at high irradiation temperatures.

It is interesting to note that stored energy, or Wigner energy, can be significant for neutron irradiated beryllium oxide and must be considered for applications where the irradiation temperature is less than 100 °C in order to prevent a runaway thermal excursion during a positive temperature transient. Store energy values of ~ 380 J/g for fluences of 4 x 10^{24} n/m^2 (E>1 MeV) have been reported (Elston, 1963; Heuer and Stolarski, 1966; Roux et al., 1964). The stored energy is much reduced above irradiation temperatures of 100°C and can be ignored in most cases.

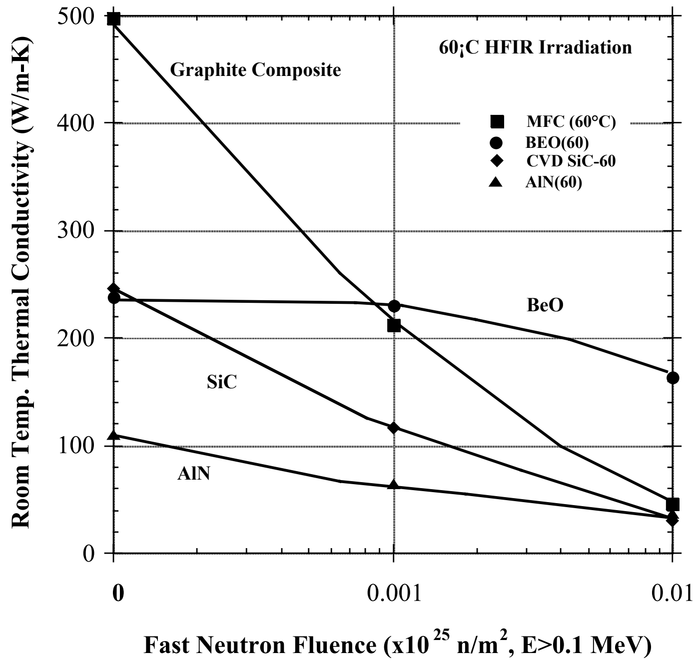


FIGURE 3. Irradiation-Induced Thermal Conductivity Degradation of High Conductivity Ceramics(Snead et al., 2005).

CONCLUSIONS

The manufacturing, fabrication, and finish-machining infrastructure currently exist, and appears to be stable in the short-term, for both nuclear grade beryllium and beryllium oxide. While the currently produced bulk material size appear to be adequate for generic space reactor application, significant cost and delays would be required if larger sizes are demanded. The data-base for both irradiated and non-irradiated nuclear-grade beryllium exists. Vacuum hot-pressed S-65 grade material manufactured by Brush Wellman may be a good reference material due to overall good material quality and the maturity of its data-base. This material is also in routine production. Much less is known about beryllium oxide, and due to the sensitivity of materials properties (such as thermal conductivity and a lesser degree strength) a complete evaluation would be required of candidate BeO grades currently being produced. Beryllium undergoes significant embrittlement at fairly low doses, with near complete embrittlement occurring for fast neutron fluxes > $1x10^{24}$ n/m^2. Volumetric expansion occurs in beryllium, especially for the higher temperature considered for space-fission applications. By limiting the operating temperature to < 500 °C and fast neutron fluences less than $1x10^{25}$ n/m^2 the volumetric swelling can be avoided. Thermal conductivity reduction for beryllium can be ignored for materials which have not undergone significant swelling.

A severe reduction in the strength and thermal conductivity of BeO is anticipated for fluences above ~$2x10^{20}$ n/cm^2, due to radiation-induced microcracking. For fluences below ~$1x10^{25}$ n/m^2, the radiation-induced microcracking in BeO is most severe at temperatures below 300°C (Wilks, 1968). The dose for the onset of cracking can be increased by utilizing fine-grained (<10 μm) material (Wilks 1968), but there is no known solution to avoid pronounced microcracking in BeO at fluences above ~1 $x10^{21}$ n/cm^2 and temperatures below 600 °C. Radiation induced swelling of BeO becomes a concern at temperatures >600 °C and fluences >$1x10^{21}$ n/cm^2. Volumetric swelling levels of 3-4% have been observed in BeO after irradiation to a fluence of $1x10^{21}$ n/cm^2 (Wilks, 1968). Higher swelling levels are expected at higher doses, although this needs to be confirmed. In summary, fine-grained BeO encased in stainless steel or another structural cladding appears to be a suitable reference design option for moderate-power (10-100 kW_{th}) space reactors (assuming the temperature will be maintained above 300 °C), although some additional analysis is needed to accurately estimate its design lifetime.

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