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BERYLLIUM – A UNIQUE MATERIAL IN NUCLEAR APPLICATIONS

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ABSTRACT

Beryllium, due to its unique combination of structural, chemical, atomic number, and neutron absorption cross section characteristics, has been used successfully as a neutron reflector for three generations of nuclear test reactors at the Idaho National Engineering and Environmental Laboratory (INEEL). The Advanced Test Reactor (ATR), the largest test reactor in the world, has utilized five successive beryllium neutron reflectors and is scheduled for continued operation with a sixth beryllium reflector. A high radiation environment in a test reactor produces radiation damage and other changes in beryllium. These changes necessitate safety analysis of the beryllium, methods to predict performance, and appropriate surveillances.

Other nuclear applications also utilize beryllium. Beryllium, given its unique atomic, physical, and chemical characteristics, is widely used as a “window” for x-rays and gamma rays. Beryllium, intimately mixed with high-energy alpha radiation emitters has been successfully used to produce neutron sources.

This paper addresses operational experience and methodologies associated with the use of beryllium in nuclear test reactors and in “windows” for x-rays and gamma rays. Other nuclear applications utilizing beryllium are also discussed.

KEY WORDS: Metals, Modeling Methodology, Structural Analysis

1. INTRODUCTION

Beryllium is a light weight metal with unique properties that make it very desirable for certain nuclear applications. Being one of the lightest known structural metals has contributed to beryllium being used in a wide variety of both nuclear and non-nuclear applications. Its light weight makes it an obvious candidate for consideration in aerospace components, especially if certain nuclear characteristics are also desired. It is not the relatively low density of beryllium, however, that causes it to be of interest in nuclear reactor applications. It is the combination of properties exhibited by beryllium that result in it being a very attractive material for use as a neutron reflector. Physically small nuclear reactors, such as test/research reactors and those used in space applications, typically include neutron reflectors to more efficiently utilize the neutrons

that are produced during reactor operation. In addition to being an excellent neutron reflector material, beryllium is also an attractive material as a neutron moderator, i.e., it effectively “moderates” or reduces the energy of neutrons. In many nuclear reactor designs, it is desirable not only to retain the neutrons within the reactor core, but also to reduce the energy of the neutrons so that they more effectively sustain the fission process. Beryllium has been the material of choice for the neutron reflectors (and/or for some neutron moderation) for a number of nuclear reactors. For example, the BR2 Belgian Engineering Test Reactor in Mol, Belgium utilizes beryllium. Other examples of nuclear reactors that utilize beryllium are the MARIA research reactor of the Institute of Atomic Energy in Poland¹ and the High Flux Isotope Reactor near Oak Ridge, Tennessee in the United States.

The success of beryllium as a test reactor neutron reflector is especially evident in that it has been used in three generations of test reactors at the Idaho National Engineering and Environmental Laboratory (INEEL) in the United States. Beryllium reflectors were used in both the Materials Test Reactor (MTR), that operated from 1952 to 1970, and the Engineering Test Reactor (ETR), that operated from 1957 to 1981. Startup of the Advanced Test Reactor (ATR) occurred in 1967; the ATR has used five successive beryllium reflectors and will continue operation with a sixth beryllium reflector beginning in 2005.

Table 1 lists a variety of basic material properties to demonstrate how beryllium compares with several materials that are used in nuclear reactors. Of the materials shown in Table 1, beryllium and graphite are utilized in the most similar manner, namely as neutron moderators and reflectors. Although graphite has a lower thermal neutron macroscopic absorption cross section than beryllium, it is not as strong (in block form) as beryllium. Hot-pressed beryllium block is more suitable for structural purposes than is graphite block, which is relatively soft and permeable. Graphite is more suitable for applications at high temperatures and is used extensively in many of the British gas-cooled power reactors. Test reactors, however, are significantly smaller than power reactors and often operate at significantly lower temperatures. Test reactors, such as the MTR, ETR, and ATR, were designed to operate at relatively low temperatures and to use purified water as the primary coolant. Successful operation of these three reactors has occurred with beryllium in direct contact with the primary coolant water; graphite, however, would not be expected to perform adequately in the same environment.

As seen in Table 1, the atomic weight of beryllium is significantly less than the other materials. The smaller atomic weight (and atomic number) of beryllium is a unique property relative to the atomic weights of other structural reactor materials in that it causes greater neutron moderation and reflection in beryllium. For a given reactor power, this unique feature allows for smaller reactor core volumes when beryllium is used as the reflector material. The spatial and weight constraints for small nuclear reactors to be launched outside the earth’s atmosphere make beryllium an attractive candidate for reflectors for such reactors.

Table 1 also shows that beryllium has a significantly higher modulus of elasticity than the other materials listed. In connection with this characteristic, beryllium is sometimes viewed with suspicion as a “brittle” material. Current engineering analytical tools and beryllium manufacturing techniques can be used to alleviate this concern.

Table 1. Comparison of Beryllium with Several Materials Used in Nuclear Applications

Properties (typical – unless noted otherwise)	Beryllium	Al Alloy (6061-O alloy)	Graphite (nuclear grade)	Stainless Steel (347)	Zircaloy (-2 alloy)
Density (g/cm ³) - at room temperature (RT)	1.85 ^a	2.70 ^b	1.65-1.75 ^e	8.027 ^f	6.55 ^c
Melting Point/Range (K) (sublimation temp. for graphite)	1558 ^a	855-925 ^b	~3923 ^c	1700 ^f	2093 ^f
Specific Heat (J/kg·K) – at RT	1925 ^a	896 ^d	711 ^c	502 ^f	297 ^c
Thermal Conductivity (W/m·K) – at RT	216 ^a	180 ^b	156 ^c	15.6 ^c	11.6 ^c
Coefficient of Thermal Expansion (x 10 ⁻⁶ cm/cm/°C) – at RT	11.4 ^a	23.6 ^b	1.8-3.6 ^e	16.5 ^f	5.2 ^f
Modulus of Elasticity (x 10 ⁵ MPa) - at RT	3.03 ^a	0.696 ^d	0.0689-0.103 ^e	2.00 ^f	0.958 ^c
Yield Strength (MPa) – at RT	186-262 ^b (hot-pressed block)	55 ^b	57.9 ^c (compressive)	207 ^c	296 ^c
Ultimate Strength (MPa) – at RT	228-352 ^b (hot-pressed block)	125 ^b	13.8 ^c	517 ^c	490 ^c
Atomic Weight	9.01 (Be)	26.98 (Al)	12.01 (C)	55.85 (Fe)	91.22 (Zr)
Thermal neutron macroscopic absorption cross section (cm ⁻¹) - at RT	0.00123 ^c (Be)	0.015 ^c (Al)	0.00032 ^c (C)	0.271 ^g	0.008 ^c (Zr)

a. From data published by Brush Wellman Inc.

b. H.E.Boyer and T.L.Gall, Editors, Metals Handbook, Desk Edition, American Society for Metals, 1985.

c. S. Glasstone and A. Sesonske, Nuclear Reactor Engineering, D. Van Nostrand Company, Inc., 1967.

d. S. T. Polkinghorne and J. M. Lacy, “Thermophysical and Mechanical Properties of ATR Core Materials,” Idaho National Engineering and Environmental Laboratory Report, PG-T-91-031, August 1991.

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f. H. Etherington, Editor, Nuclear Engineering Handbook, New York, McGraw-Hill Book Company, 1958.

g. Calculated value based on nominal major constituents in 347 SST and atomic weights and microscopic cross sections from S. Glasstone and A. Sesonske, Nuclear Reactor Engineering, D. Van Nostrand Company, Inc., 1967.

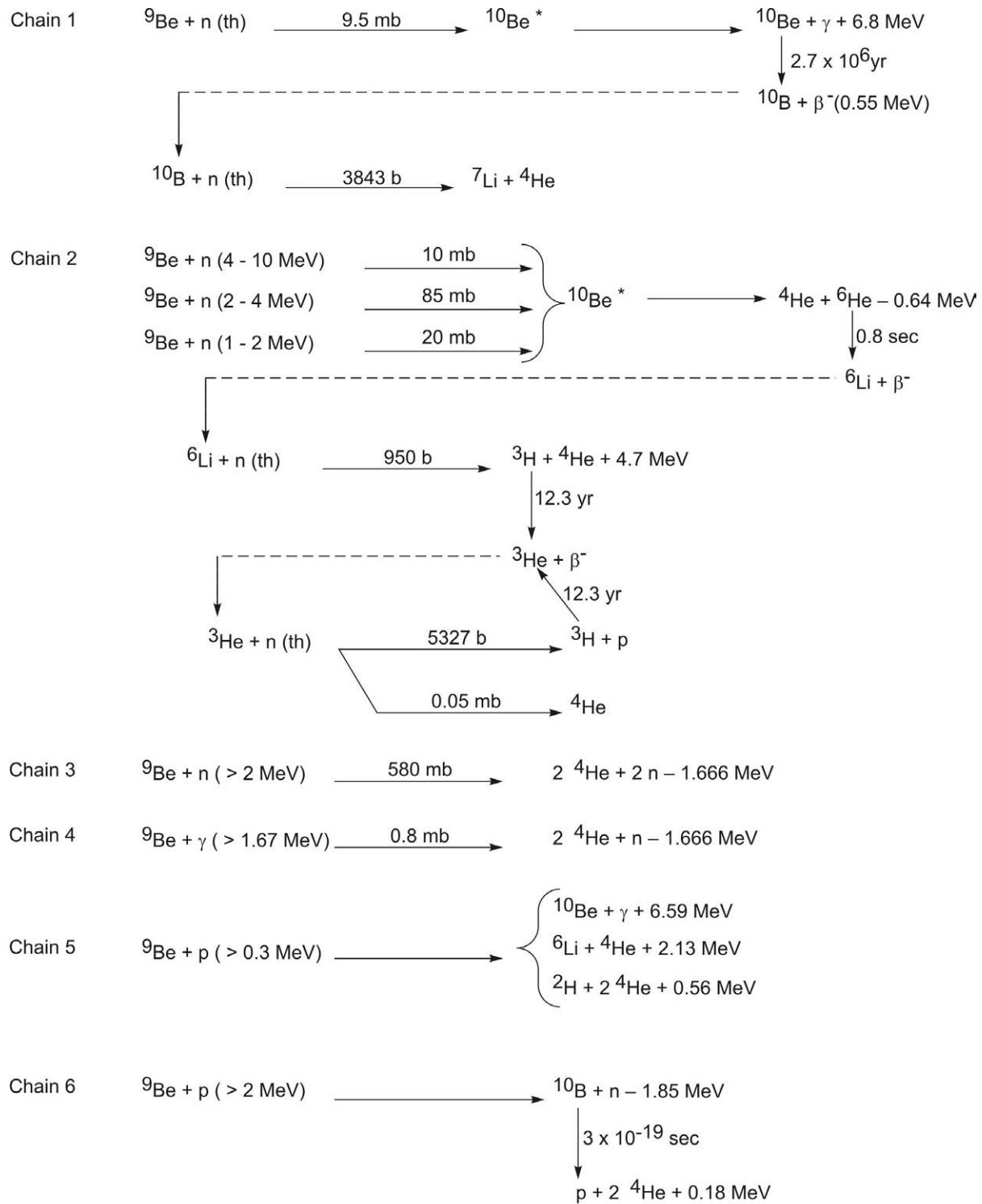
2. BERYLLIUM NUCLEAR REACTIONS

As shown in Table 1, beryllium is characterized by a relatively low thermal neutron absorption cross section. Although this feature, along with several others, makes beryllium the material of choice for reactor reflectors in many cases, it cannot be dismissed in the actual performance of a beryllium reflector. The nuclear reactions involving beryllium are of significance in nuclear reactor operations and analyses. Figure 1 lists a number of nuclear reactions involving beryllium. Examination of the six reaction chains in Figure 1 reveals that helium is a reaction product in every chain. Reaction chains 2, 3, and 4 are especially of interest relative to the intense neutron and gamma radiation environment of an operating reactor. Long term exposure of beryllium structures to these reactions results in the gradual buildup of helium within the beryllium. It has been long recognized that helium buildup in irradiated beryllium is associated with swelling and changes in mechanical properties of the beryllium. References 2, 3, and 4 are examples of investigations pertaining to irradiation induced changes in beryllium. The swelling and resultant internal stresses can limit the useful operating life of beryllium, and it is for this reason that beryllium reflectors have required periodic replacement, e.g., in the Advanced Test Reactor.

The beryllium nuclear reactions depicted in Chain 2 of Figure 1 also reveal two additional aspects of reactor operations when beryllium reflectors are involved. One of the significant reaction products in Chain 2 is tritium (^3H), a radioisotope of concern in nuclear reactor waste streams. Extended exposure of beryllium in a test/research reactor can result in significant buildup of tritium in the beryllium. Concerns related to the potential release of tritium from irradiated beryllium have been recognized and documented. Although test/research reactors do not typically use low-density beryllium, tests have revealed that structural failures of highly irradiated low-density beryllium can result in significant releases of tritium.⁵ A second aspect of beryllium nuclear reactions that can directly affect reactor operation is related to the gradual buildup of reaction products that have relatively high thermal neutron absorption cross sections, particularly lithium-6 (^6Li) and helium-3 (^3He). Helium-3, as indicated in Chain 2 of Figure 1, has a very large absorption cross section. Helium-3 is being both generated and depleted during reactor operation, but during reactor outage conditions the depletion ceases. Therefore, during reactor outages, especially extended outages, a highly irradiated beryllium reflector can experience significant increases in helium-3 concentrations due to the radioactive decay of tritium. Methods have been developed to account for these aspects of changes in beryllium reflectors; one approach is reported in Reference 1.

3. A METHODOLOGY FOR ASSESSING IRRADIATED BERYLLIUM

Beryllium, used in a high neutron radiation environment such as in the Advanced Test Reactor (ATR), experiences swelling. The stresses associated with this swelling have been successfully assessed assuming that the induced swelling can be modeled in a manner similar to the approach used for thermal expansion (or swelling). This approach, although an oversimplification of the microscopic material response, has been found to provide adequate results relative to actual performance of beryllium and predicted failures in critical regions. Applying a thermal expansion analogy allows the use of computer codes developed for analyzing thermal stresses and deformations.



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Figure 1. Beryllium Nuclear Reaction Chains.⁶

The neutron irradiation damage in materials is largely dependent on the neutron flux (number of neutrons crossing a unit area in a unit of time), the energy of the neutrons, duration of the neutron exposure, and the temperature during the irradiation. Nuclear test reactors that include beryllium as a neutron reflector are generally based on utilization of “thermal” neutrons for sustaining the fission process in the reactor fuel. “Thermal” or “slow” neutrons are those with a kinetic energy that is in thermal equilibrium with their environment. Neutrons released during the fission process are primarily high energy or “fast” neutrons and it is the high energy neutrons that are generally responsible for causing swelling in beryllium. A test reactor that utilizes beryllium is generally designed to include a “moderator” material to reduce the energy of “fast” neutrons to the extent that they become “thermal” neutrons that are desirable for sustaining the fission process. Many high energy neutrons experience significant interactions with surrounding materials, including beryllium, prior to their becoming “thermal” neutrons. The combination of the high energy neutron flux, ϕ , and duration of neutron exposure is represented as the fluence, Φ , where

$$\Phi(x,y,z,t) = \int_0^t \phi(x,y,z,t) dt \quad (1)$$

The irradiation swelling response of typical materials, including beryllium, is generally assumed to be an exponential relationship of the form

$$\varepsilon = a\Phi^m(x,y,z,t) , \quad (2)$$

where ε is the one-dimensional swelling strain and constants “a” and “m” are dependent on the material.⁷ In applying the thermal analogy, a thermal strain equivalence has been expressed as,

$$\varepsilon = \alpha(t)T(x,y,z) = a\Phi^m(x,y,z,t) , \quad (3)$$

where the expansion coefficient, α , is arbitrarily assumed to be a function of time only and the temperature distribution, T , is assumed to be dependent only on position.⁷ If the neutron flux is assumed to be constant over time and an expansion coefficient at a time t_1 is assumed, then a corresponding “pseudo-temperature” field can be expressed⁷ as

$$T_1(x,y,z) = [a/\alpha(t_1)] \phi^m(x,y,z)t_1^m . \quad (4)$$

It then follows that the expansion coefficient for any other point in time, t_2 , would be⁷

$$\alpha(t_2) = \alpha(t_1)[(t_2)/(t_1)]^m , \quad (5)$$

and the irradiation induced stress at time t_2 , being proportional to the expansion coefficient, would be $\sigma(t_2)$, where (per Reference 8)

$$\sigma(t_2) = \sigma(t_1)[(t_2)/(t_1)]^m . \quad (6)$$

Knowing constants “a” and “m” and the neutron fluence therefore allows the determination of the irradiation-induced stress in the beryllium.

Assessment of Advanced Test Reactor beryllium reflector components subjected to severe irradiation-induced stresses has been based on the maximum stress theory of failure.⁸ As indicated in Reference 8, “This theory assumes failure of a member subjected to combined stresses occurs when one of the principal stresses becomes equal to the strength (yield or fracture) in a uniaxial tension or compression test.” Therefore, in a finite element stress analysis (performed when actual fluence reaches a given value A in Figure 2), the predicted failure point (intersection B in Figure 2) occurs when the irradiated beryllium strength and the induced stress become equal.

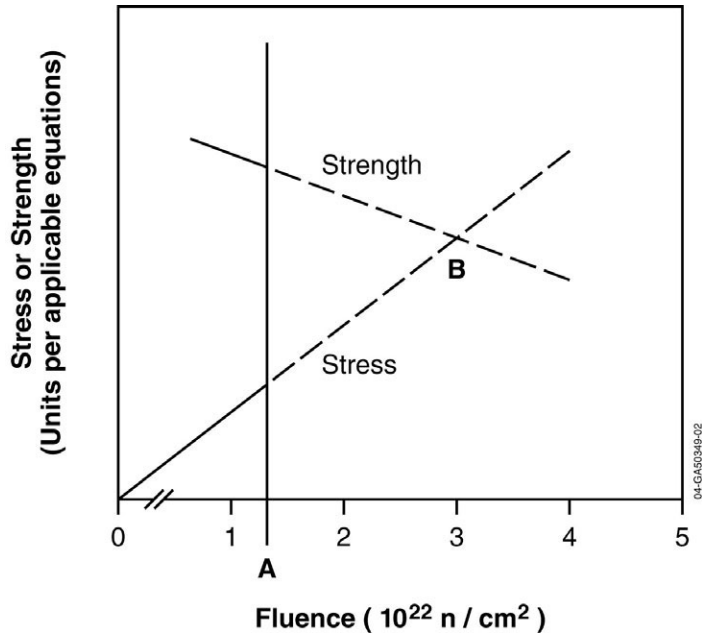


Figure 2. Irradiated Beryllium Stress and Strength Relative to Neutron Fluence.

Measurements of irradiated beryllium specimens have made it possible to determine the constants “a” and “m” for fluence levels up to $3 \times 10^{22} \text{ n/cm}^2$ (for neutron energies greater than 1 MeV).⁹ Reference 9 indicates that the geometric regression curve for beryllium growth data can be represented as

$$\Delta L/L = 1.829 \times 10^{-3} (\phi t)^{1.035} \quad (7)$$

where the fluence, ϕt , is in units of 10^{22} n/cm^2 ($E > 1 \text{ MeV}$).

Flexure tests and splitting tensile tests of irradiated beryllium have been performed with linear regression analysis of the results indicating that the flexure strength, S_f , can be represented by

$$S_f = 530 - 95.1(\phi t) \quad \text{MPa} \quad (8)$$

where, ϕt is the fluence in units of 10^{22} n/cm^2 ($E > 1 \text{ MeV}$).⁹

Likewise, the linear regression analysis strength relationship for combined data of both flexure and splitting tensile tests was found to be S_s , represented by

$$S_s = 367 - 51.6 (\phi t) \quad \text{MPa} \quad (9)$$

where, ϕt is the fluence in units of 10^{22} n/cm^2 ($E > 1 \text{ MeV}$).⁹

The strength of ATR irradiated beryllium at a given location is obtained by interpolating between Equations 8 and 9, with the interpolation based on the percentage of bending stress at the location.¹⁰

The irradiation limit for ATR beryllium is essentially based on using the above relationships to predict when the induced stress at a given location becomes equal to the irradiated strength of the beryllium at that location. Some additional modifying factors, not addressed here, are incorporated into the assessment to account for previous performance of the same type of beryllium components and to assure adequate confidence (95% confidence level) that beryllium cracking will not occur in certain critical locations. To achieve the longest practical and safe life of beryllium components in the ATR, some limited cracking at non-critical locations is allowed. To predict ATR beryllium performance, the number of time 'periods' to cracking, N , is incorporated into the above relationships. The value of N for a given location is determined by solution of the following equation:¹⁰

$$\sigma N^{1.035} = A + BN(\phi t) \quad (10)$$

where

- σ = maximum calculated stress after one 'period' of time
- N = number of time 'periods' to cracking
- A = initial strength
- B = slope of strength equation
- ϕt = fluence after time 't' .

Knowing the associated nodes in the finite element analysis, the fluence values can be calculated from the pseudo-temperatures. The fluence values can then be used in the interpolated beryllium strength equation (in which 'B' is a negative number) to predict the mean strength at the times under consideration.

This basic methodology has been used to predict when ATR beryllium cracking will occur in non-critical regions and to prescribe the end-of-life of beryllium components to assure that cracking will not occur in critical regions. As part of the safety basis for continued use of cracked beryllium, the surveillances of the beryllium and associated components are increased after cracking is first detected. Application of this methodology provides a basis to support safe use of beryllium reflectors in the severe radiation environment of the Advanced Test Reactor.

Figure 3 shows side and end views of an ATR beryllium reflector block. The end view provides an indication of the relative sizes and number of holes that penetrate the full length of a block and also identifies the ligament location where neutron radiation induced stresses are greatest following extended reactor operation. The ligament identified in the figure is in a non-critical

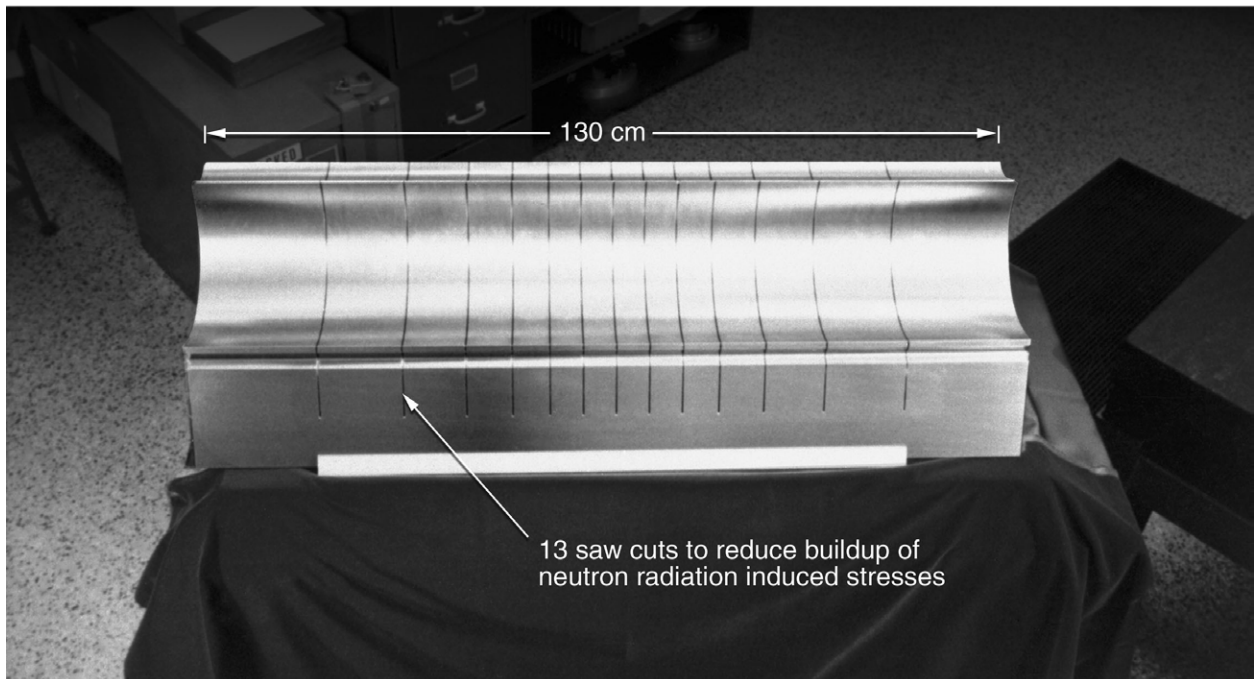
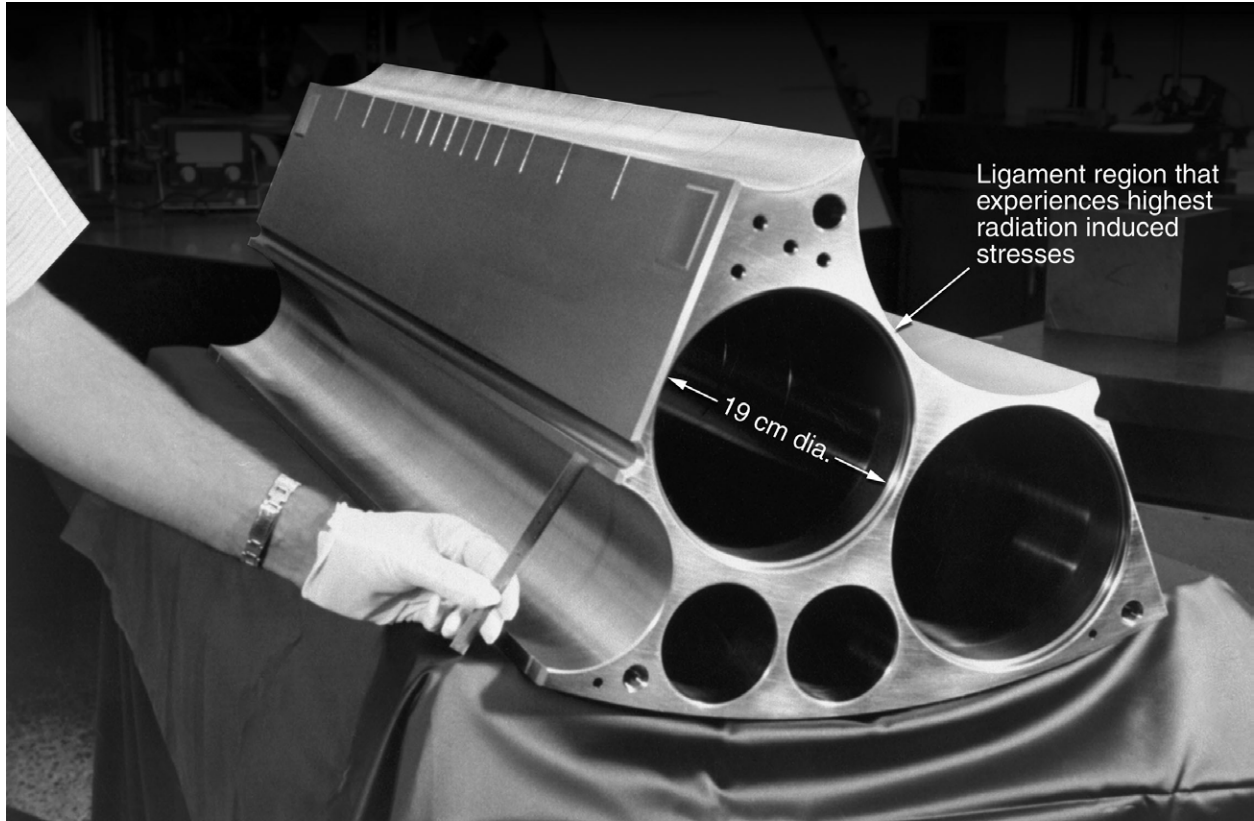


Figure 3. End and Side View Photos of an ATR Beryllium Reflector Block.

region where carefully monitored cracking is permitted. The side view in Figure 3 gives an indication of the length of a reflector block and also shows the saw cuts that have become a standard design feature to assist in reducing neutron radiation induced stresses.

4. BERYLLIUM IN OTHER NUCLEAR APPLICATIONS

4.1 Neutron Sources and Multipliers The binding energy per nucleon in the beryllium nucleus is sufficiently low that a nuclear reaction with an energetic particle can result in a neutron being emitted as a reaction product. This characteristic is shown, for example, in reactions depicted in Chain 4 and Chain 6 in Figure 1. The reaction shown in Chain 4 can be utilized to produce a neutron source by subjecting beryllium atoms to high energy gamma rays from a radioactive material. Radioactive antimony, for example, can be placed in close proximity to beryllium to produce neutrons. This technique has been used to produce neutron sources to assist in the startup of nuclear reactors. Radioactive materials that emit gamma rays of sufficient energy (> 1.67 MeV - as shown in Chain 4 of Figure 1) to produce neutrons from beryllium typically have relatively short half-lives. The neutron production rate decreases as the gamma ray emitter decays. To achieve a more steady state neutron source, a different nuclear reaction has been used to produce neutrons from beryllium. Radioactive materials that emit high-energy alpha particles can also be used to produce neutrons from beryllium. The relatively short range of alpha particles requires that these types of materials be placed in intimate mixtures with the beryllium or be chemically reacted to form compounds with the beryllium. Using this technique, a compound of plutonium (Pu-239) with beryllium has been found useful as a neutron source.

A more futuristic potential nuclear application of beryllium is to use it as a neutron multiplier. The reaction shown in Chain 3 of Figure 1 indicates the mechanism by which beryllium can act as a neutron multiplier. Beryllium has received much attention as a potential neutron multiplier material for fusion reactors. For example, a 1998 report on irradiated beryllium indicated that large quantities of beryllium were expected, at that time, to be used as a “neutron multiplier and plasma facing material in the International Thermonuclear Experimental Reactor (ITER).”¹¹

4.2 “Windows” for X-rays and Gamma Rays The combination of a low atomic number, low density, and other suitable mechanical properties of beryllium make it an excellent candidate for “windows” especially for low energy x-rays and gamma rays. Beryllium has been used in numerous devices in this manner to maximize the transmission of x-rays and gamma rays from x-ray generating devices and sealed sources and the transmission into radiation detectors. In applications involving significant pressures and temperatures there has been repeated success in joining beryllium components to Monel-400 alloy using a brazing process. There are a variety of surface treatments that can be applied to beryllium for corrosion protection. Anodized beryllium surfaces, for example, of gamma ray densitometer devices have been found to provide adequate corrosion protection for exposures to steam and purified water at elevated temperatures.

5. CONCLUSIONS

Beryllium has unique characteristics that contribute to its usefulness in a variety of nuclear applications. Beryllium can be used successfully for extended periods of time in high radiation, nuclear reactor environments by applying a thermal expansion analogy to predict swelling and

concomitant stresses. Nuclear reaction products in irradiated beryllium are recognized and can be accommodated in nuclear reactor performance predictions. Beryllium has proved to be an excellent “window” for optimal transmission of x-rays and low-energy gamma rays. Beryllium can be used in combination with selected radioactive materials to produce useful sources of neutrons. Beryllium is an attractive candidate as a “neutron multiplier” material for future fusion reactor facilities.

6. ACKNOWLEDGEMENTS

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