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COMPARISON OF BeO VERSUS GRAPHITE AS A MODERATOR FOR MGCR

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Properties Desired in a Moderator

The major requirement for a moderator is that of good neutron economy; this can be expressed by these factors: high slowing down power for neutrons, and low thermal absorption cross-section. The result is that BeO is a better moderator (for a core of 5-7 ft) than graphite. For example, the slowing down power of BeO is 2.5 times greater than that of graphite (see Table I); the conversion ratio of a BeO moderated core is 2.0 times better than for a graphite core; the burn-up lifetime of a BeO core is 1.5 times greater than that of graphite. Also, the (n, 2n) reaction in BeO helps the reactivity of the reactor.

In addition to the nuclear requirements for the moderator, mechanical, physical and chemical properties are important, particularly the manner in which these properties are affected by radiation. From the data in Table I, one can conclude that BeO is a better moderator than graphite for the 5-7 ft core.

Of primary importance, of course, is the cost of the moderator. In this case, graphite is less expensive than BeO. However, when the nuclear properties are factored in, this lower cost of graphite is offset by the greater neutron economy resulting from the use of BeO in the MGCR.

As noted above, the moderator must perform satisfactorily under irradiation. This subject is discussed in the next section.

Table I

Properties of Moderators and MGCR Cores

Nuclear Properties	BeO Core Un-Irr.	BeO Core Irrad.	Graphite Core Un-Irr.	Graphite Core Irrad.
Slowing down power	0.16		0.063	
Fermi Age, cm^2	103		320	
Microscopic cross-section, mb	11.0		3.7	
Over-all rating for MGCR	Excellent		Good	
Physical Properties				
Thermal conduct, cal/sec cm °C (200°)	0.25	0.15	0.30	0.12
Thermal expan., 10^{-6}/°C (20-1800)	9.8	Slight effect	3.0	No change
Thermal shock resistance	Good		Excellent	
Change in length after irrad,% (>600°C irr.)		+0.01		-0.05
F. P. release from diluted fuel	Low		High	
Over-all rating for MGCR	Good		Good	
Mechanical Properties				
Tensile Strength, psi RT	21,000	21,000	1,300	
Compressive strength, psi RT	175,000	200,000	6,000	7,000
Young's modulus, psi RT	52×10^6	20×10^6	1.2×10^6	3×10^6
Creep, %/hr, 2300°F, 100 psi	5×10^{-5}		$<5 \times 10^{-5}$	
Over-all rating for MGCR	Excellent		Fair	
Chemical Stability, threshold reaction temperature, °C				
With O_2	No React.		420	<420
With H_2O (10^{-4} atm)	1250°C		700	
With CO_2	No React.		900	
Over-all rating for MGCR	Excellent		Fair	
Economics				
Cost of fabricated mod. blocks	$22/lb.		$2/lb.	
Over-all reactor economics	Excellent		Fair	

Irradiation Effects on Beryllium Oxide

We have reviewed the results of published work on irradiation effects on BeO and on BeO containing UO_2 dispersions.[1-12] The conclusions may be summarized as follows.

(1) U.S. Work.[1-8]

(a) Work at Argonne[1] included irradiation of pure BeO and of BeO-UO_2 compacts containing 2% and 10% UO_2 (30% enriched) by exposure to 3.6×10^{19} nvt (fast) and 1.3×10^{20} nvt (thermal). The only measured property of the pure BeO that was affected appreciably by the irradiation was thermal conductivity which decreased to about 60% of the original value and appeared to be approaching saturation value. After annealing the irradiated samples at 980°C the conductivity values were found to be almost equal to the values before irradiation. Considerably larger effects were observed in the BeO-UO_2 samples. Linear expansions up to 1% occurred after the longest irradiation, which could be annealed out at 900°C. The compressive strength decreased from 20 to 30% and appeared to have reached a saturation value. The thermal conductivity decreased by a factor of six for both the 2% and the 10% UO_2 compacts. The temperature of irradiation has a marked effect since the 10% UO_2 compacts were irradiated at a higher temperature (650°C). Several experiments were carried out to determine the effect of post-irradiation annealing on thermal conductivity of the fueled samples. Annealing at 1000°C for 7 hours increased the conductivity by a factor of three, and further heating at temperatures as high as 1200°C produced no additional change in thermal conductivity. The first sign of annealing was observed at about 600°C.

(b) Preliminary data from ORNL on pure BeO exposed to 7×10^{19} nvt (fast) indicates that the thermal conductivity decreased by 30%, with no change in density.[2]

(c) Results of the GE-ANP irradiation work on fueled BeO samples was presented at the classified Fuel Conference held in Gatlinburg last year. We attended this meeting and detailed notes are available (M. Simnad, classified files).[3]

(d) Recent irradiation studies at Battelle for our MGCR project included irradiations of BeO-UO_2 fuel pellets.[4] These samples contained 20% UO_2 by volume and were exposed to about 1 x 10^{19} nvt*, with axial temperature of 2350°F and ambient gas temperature of 1500°F. This exposure resulted in a 1.5% burnup of the U-235 atoms. No dimensional changes were observed after irradiation. Similar samples are being irradiated in the MTR.

(e) The creep properties of BeO have been measured recently at temperatures above 1500°C.[5] No work has been reported on the creep properties of irradiated BeO. Swelling of BeO at 1300°C probably would be appreciable.

(2) British Work

(a) The review paper on ceramic fuels presented at Geneva[6] discusses BeO. It is stated that the thermal shock resistance is high so that there is the possibility that cracking may not occur under irradiation. The effect of irradiation is reported to be a 0.3% expansion in the C-spacing with no change either in the A-spacing or the thermal conductivity. The swelling temperature (due to bubble formation) is considered to be above 1000°C.

(b) Two recent publications from Harwell[7,8] deal with the stability of BeO and BeO-UO_2 fuels under reactor conditions. In the introductory review attention is drawn to the fact that the results from the U.S. and France show a considerable variation in the magnitude of the irradiation-induced changes and that important parameters such as irradiation temperature are sometimes

*thermal neutrons, effective flux

missing. (1) BeO: Mechanical properties are affected by only a small amount for doses up to 10^{21} nvt. The compressive strength shows a 9% increase for a dose of 1.3×10^{20} nvt. On annealing, changes in mechanical properties take place at 850°C (annealing commences) and 1500°C (where the anneal is complete). (2) BeO-2%UO_2: Thermal conductivity decreased by about 80% for a dose of up to 3×10^{20} nvt and the decrease does not appear to be saturating at this dose. Compressive strength and Young's Modulus decreased by about 30%, and these changes appear to saturate between 5×10^{19} and 5×10^{20} nvt. No British data on creep of BeO are available.

(c) The swelling of irradiated Be metal upon annealing at various temperatures has been studied in detail.[9] Bubble formation commences at 600°C and eventually results in a swelling of 30% at 1000°C.

(3) <u>French Work</u>

(a) A 20 page paper was presented at the 2nd Geneva Conference on the physical and mechanical properties of sintered beryllia.[10] The conclusion in this paper is that "the physical and mechanical properties of beryllia are judged to be sufficient, in first approximation, to justify the use of this material as moderator in a high temperature reactor." Samples of beryllia were irradiated to 10^{21} nvt (fast) and 7×10^{19} nvt (thermal). Length changes up to 0.25% were observed. The thermal conductivity decreased by 80%. Annealing of the irradiated samples indicates that above 500°C the effect of irradiation is relatively less important since recovery appears to take place. Recovery is complete at 1200°C, while at 1000°C recovery is 50%. The compressive strength falls to about 20% of the original value. The elastic modulus is markedly lowered, but recovers upon annealing.

(b) A recent paper[11] presents electron-microscope studies of irradiated BeO specimens annealed at temperature up to 1500°C after exposure to 5.5×10^{19} nvt (fast), 7×10^{20} (thermal). The irradiated samples showed fewer striations in fractured sections than the original material. The heating was carried out for 24 hours. The results were as follows:

800°C: no change in structure.

1000°C: two types of bubbles were evident: a large number of small (a few hundred Angstrom units in diameter) and a few large bubbles (5 to 10 times larger).

1200°C: marked change in structure. Elongated and oriented bubbles, several microns in size.

1500°C: large bubbles (tens of microns in diameter), fewer in number and much less elongated. Random distribution.

(c) Telegram reply from Salesse[12] (head of metallurgy at Saclay) received 9-8-59. Little information, not already known, is reported.

Proposed Research on Moderator Ceramics for MGCR

A. One of the most important technical problems in the use of BeO or graphite as MGCR moderator blocks is to determine the effects of structure (macro, micro, and sub-grain) and chemical impurities on:

(1) Dimensional stability under simultaneous temperature, radiation, and mechanical stresses. Specifically, the effects of structure on creep and tensile strength, and their change on irradiation is not precisely known and should be determined. This is true for graphite and BeO.

(2) Chemical reactivity under simultaneous temperature and radiation effects. Specifically, the effects of "catalytic" impurities on

reaction with impurities in helium, and with metal claddings at high temperature under irradiation should be investigated, e.g., the rate of reactions:

$$BeO + H_2O \longrightarrow \underset{\text{Volatile}}{\overline{Be(OH)_2}}$$

$$C + H_2O \longrightarrow H_2 + CO$$

may be accelerated or inhibited by chemical impurites in the solid.

It should be recognized that analytical methods for characterization of BeO and graphite ceramics are inadequate in the present state of the art. A considerable effort (money) will be necessary to utilize the best analytical facilities now available,and where necessary to develop improved analytical methods, e.g.

(1) Electron and field emission microscopy.

(2) X-ray low angle scattering studies.

(3) Gas-adsorption studies.

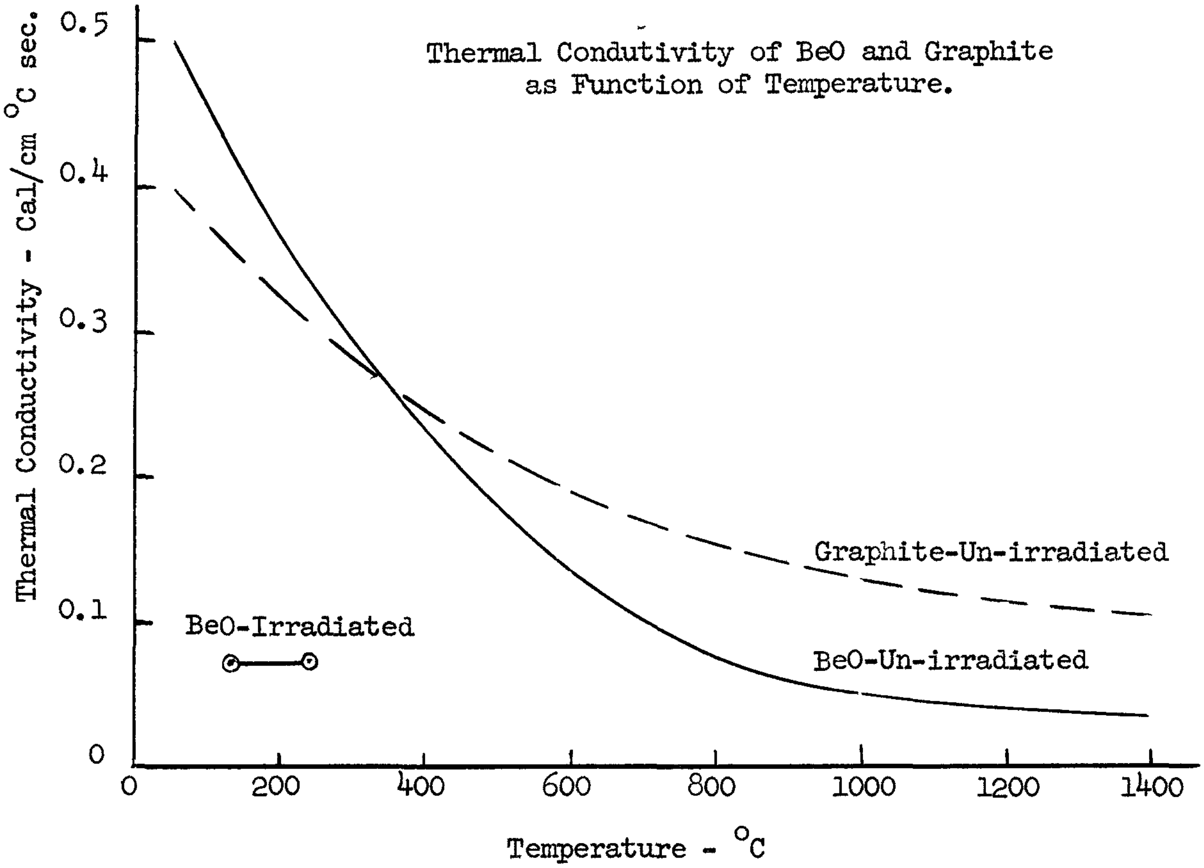
(4) Precise spectrographic analytical techniques.

B. Another important problem is that of gas generation and the effect of this gas on the mechanical and physical properties of the moderators. For example, it has been reported that gas in beryllium oxide is formed from the (n, 2n) and (n, α) capture reactions. This gas, coupled with lower creep strengths of beryllium oxide at temperatures above 1100°C, could result in swelling of the oxide. As described in the previous section, such swelling has not been observed to any extent in a few irradiation tests that have been conducted. However, these tests were run at low temperatures. Hence, it is proposed to test BeO at high temperatures in a reactor to high burn-ups and determine, as a function of temperature, neutron flux and total exposure time, the swelling characteristics, if any, of BeO.

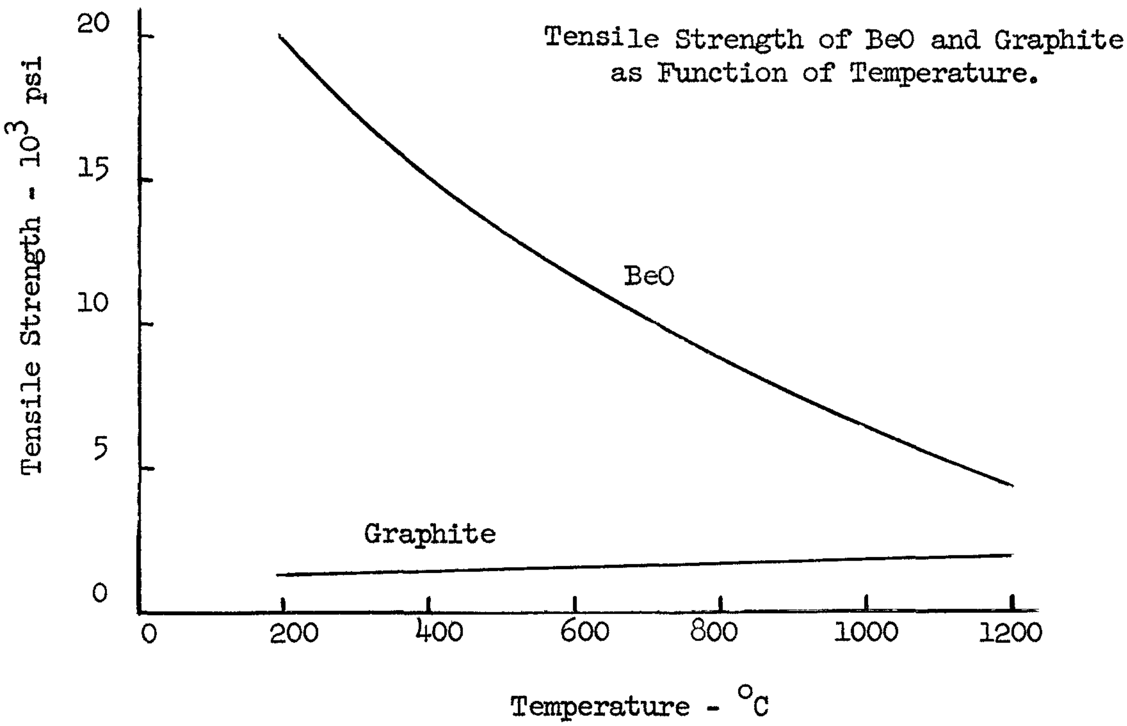
C. Thermal shock resistance of graphite and BeO are reported to be good. For the MGCR design, it would be desirable to check this feature by subjecting the full size moderator blocks to design temperature gradients and establish the performance characteristics of these blocks.

D. Moderator blocks of MGCR design have not been produced from BeO. This is not considered to be a real hard job; however, some development work will be required.

E. As mentioned earlier, carbon transport by impurities in helium is a real problem for the graphite core. For an all-beryllia core, this problem does not exist. Also, carburization of metallic surfaces by CO (O_2 + graphite reaction) disappears with an all-beryllia core.



Thermal Condutivity of BeO and Graphite
as Function of Temperature.
Thermal Conductivity - Cal/cm °C sec.
0.5
0.4
0.3
0.2
0.1
0
Graphite-Un-irradiated
BeO-Irradiated
BeO-Un-irradiated
0
200
400
600
800
1000
1200
1400
Temperature - °C



Tensile Strength of BeO and Graphite
as Function of Temperature.
Tensile Strength - 10³ psi
20
15
10
5
0
BeO
Graphite
0
200
400
600
800
1000
1200
Temperature - °C

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3. Notes on AEC Gatlinburg Meeting on Reactor Fuels, 1958 (M. Simnad, classified notes).

4. D. Johnson, MGCR- "Experimental Data Reported by Battelle Memorial Institute for MGCR Fuel Material", GAMD-931.

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6. Murray and Williams, "Ceramic and Cermet Fuels", 2nd Geneva Conference, Paper 318.

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8. Ghosh and Clarke, "An Experiment Concerning the Presence of Free Gas in Irradiated Beryllia", AERE-R-2889 (1959).

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10. Elston and Caillat, "Physical and Mechanical Properties of Sintered Beryllia under Irradiation", 2nd Geneva Conference, Paper 1159.

11. Frisby, Bisson, Caillat, "The Precipitation of Helium in Irradiated BeO", J. Nuclear Materials, Vol. 1 (1959) No. 1 page 106.

12. Telegram from Salesse (Head, Metallurgy Department, Saclay), 9-8-59.