Conf. 790125--10

LA-UR -79-146

TITLE: CERAMICS FOR APPLICATIONS IN FUSION SYSTEMS

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SUBMITTED TO: Talk and Publication: 1st Topical Meeting
on Fusion Reactor Materials, Miami Beach, FL
to be held Jan. 29-31, 1979

MASTER

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Form No. 836
St. No. 2629
1/75

UNITED STATES
ENERGY RESEARCH AND
DEVELOPMENT ADMINISTRATION
CONTRACT W-7405-ENG. 36

CERAMICS FOR APPLICATIONS IN FUSION SYSTEMS†

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Six critical applications for ceramics in fusion systems are reviewed, and structural and electrical problem areas discussed. Fusion neutron radiation effects in ceramics are considered in relation to fission neutron studies. A number of candidate materials are proposed for further evaluation.

1. INTRODUCTION

Ceramics are required for a number of critical applications in magnetically-confined fusion systems. Details of design, materials requirements, and operating conditions have not yet been fully identified for power reactors, where materials problems will be most severe. However, ceramic applications have been defined in sufficient detail to allow assessment of anticipated material problems [1] and development of a plan for their solution [2]. It is the intent of this paper to review these problems in the context of fusion reactor requirements.* First, ceramic applications and anticipated operating conditions are briefly described. Then major structural and electrical problem areas are discussed, and special problems related to fusion neutron effects cited. In the last section prospects for ceramics in fusion systems are summarized and candidate materials proposed for further evaluation and development.

2. CERAMIC APPLICATIONS IN FUSION SYSTEMS

Most requirements for ceramics in fusion devices arise from the need for electrical insulators, which in turn results from the characteristically large applied or induced electrical and magnetic fields of these machines. A major potential application not requiring insulating properties is that for a low-Z first wall or liner. Critical ceramic applications are described in the following section.

2.1 Insulators for RF heating systems

Plasma temperature can be raised to ignition by applications of radio frequency (RF) power. RF heating designs vary considerably with frequency and fusion system under consideration (frequencies from 10^6 to 11^{11} Hz are feasible). Proposed configurations include ceramic-filled waveguides, coaxial power leads with insulator-coated antennas inside the first wall, and hollow waveguides with ceramic windows separating pressurized and evacuated sections. Moderate electric fields (0.1 to 1 kV/mm) are anticipated; however, a very low loss tangent ($\sim 10^{-4}$) may be required. Base-line operating temperature for the ceramics is at or below that of the blanket ($\sim$800-1000 K), but heating effects from irradiation and RF energy losses may lead to high operating temperatures and thermal stresses. First-wall-like neutron fluxes (e.g., $\sim 10^{19}$ n/m^2·s for a 2 MW/m^2 reactor) will be encountered with some designs. Structural damage from neutrons can be expected, with possible additional deterioration from exposure to ions and neutral fuel gases.

2.2 Neutral beam injector insulators

Neutral beam injectors are used to fuel and heat the plasma. These devices are located outside the blanket, so that their insulators will be exposed to only moderate neutron fluxes (on the order of 10^{14}-10^{16} n/m^2·s). The insulators can be water cooled to keep their temperature below $\sim$500 K, but significant thermal stresses may result. Since injectors are high voltage devices, high electric fields (perhaps 5 kV/mm) must be withstood. A resistivity of at least $10^6 \Omega$·m is needed to control ohmic heating effects. Insulators can be shielded from hydrogenic ions; however, a gamma flux of $\sim 10^4$ Gy/h may be encountered.

2.3 Toroidal current break

Proper coupling of magnetic fields to the plasma requires that the electrical continuity of a metallic torus be interrupted by a high-resistance section (the current break). A number of ideas for the configuration of a ceramic break have been proposed; monolithic ring, plasma-sprayed layer, and segmented ceramic inserts are examples. Operating temperature will be that of the blanket, with neutron fluxes varying from near that of the first wall to perhaps 100 times lower depending on location of the insulator within the blanket. All designs specify that the ceramic be put under compression, but additional stresses may arise from differential expansion effects. Resistivity and dielectric strength requirements will be modest, so that structural integrity is the primary concern.

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†Work performed under the auspices of the U. S. Department of Energy.

*Other reviews which address various aspects of this topic are cited in references 3-8.

2.4 Magnetic coil insulators

Magnetic fields used to compress and confine the plasma are generated by large superconducting and normally-conducting coils. For some applications organic insulators will suffice, but coils operating in a significant radiation field (greater than $\sim 10^7$ Gy or $\sim 10^{22}$ n/m^2) will require ceramic insulation. The most severe environment will be encountered by the poloidal field coil, which may be located in a high neutron flux region near the first wall. Voltage across magnetic coil insulators is a few kilovolts at most, so that dielectric strength requirements are not severe. However, high stresses induced by the operation of large coils will be imposed on the ceramics, so that the likely failure mode is structural rather than electrical.

2.5 Direct converter insulators

Mirror reactor designs specify the use of a direct converter to recover the energy of ions which escape magnetic confinement. Such a device may also be used to collect the energy of charged particles in neutral beam injector systems. The direct converter operates at a high voltage (perhaps 180 kV dc), and requires electrical feedthroughs each capable of standing off a significant fraction of this voltage. Insignificant neutron flux is expected at the location of these feedthroughs, but ion impingement and ionizing radiation may be significant unless effective shielding can be developed. Operating temperatures as high as 1300 K are possible if thermal conversion as well as direct conversion is utilized. However, problems with electrical properties will probably require that this temperature be reduced by water cooling. Thermal stresses are therefore expected to be large.

2.6 Low-Z first wall/liner

An important application proposed for ceramics in fusion reactors is that of a first wall structure or liner. Advantages of low-Z ceramics, when compared with most metals, include reduced plasma impurity effects, low induced radioactivity, high operating temperatures, and low cost. Since in this application good insulating properties are not required, ceramics such as SiC can be considered. First-wall ceramics will be exposed to a high flux of neutrons, ion and neutral gas bombardment, and intense electromagnetic radiation. Large thermal stresses will result from the high heat flux. If a radiatively-cooled liner is used, operating temperatures will be very high (perhaps 2000 K), so that vapor pressure will become a consideration. A convectively-cooled wall will operate at a temperature nearer that of the blanket, but additional stresses will result from structural constraints and coolant pressures [5].

3. CERAMIC PERFORMANCE IN A FUSION ENVIRONMENT

3.1 Structural properties

3.1.1 Swelling

Neutron-induced swelling of from zero to several percent has been observed in ceramics, depending on material, dose, and irradiation temperature. Growth is attributable either to the creation of new lattice sites or to the presence of point defects which dilate the lattice. In the former case, defect aggregates such as voids and dislocation loops can typically be observed by transmission electron microscopy (TEM). Figure 1 shows aligned voids in irradiated Al_2O_3, along with dislocation tangles attributed to the growth and interaction of interstitial loops [9]. This material was irradiated in the EBR-II fission reactor at 1100 K to a dose of 2.1×10^{26} n/m^2 ($E_n > 0.1$ MeV), or ~ 20 displacements per atom (dpa).

Radiation damage processes in insulating ceramics can be complex, due to electrostatic effects and the presence of two or more sublattices. For example, in Al_2O_3 voids must be composed of a stoichiometric ratio of aluminum and oxygen vacancies [10]. If only oxygen vacancies coalesced, the result would be an inclusion of aluminum metal, whereas if aluminum vacancies condensed an oxygen fluid inclusion would be formed. The process of cation colloid formation has been observed in irradiated NaCl [11]. Dislocation loop structures can similarly be complex; electrostatic effects allow only certain anion-cation stacking sequences to be stable [12]. These considerations suggest that the ratio of dpa rates on the two sublattices can affect the nature of aggregated defects formed.

When swelling is attributable to lattice dilation by point defects (or other fine-scale

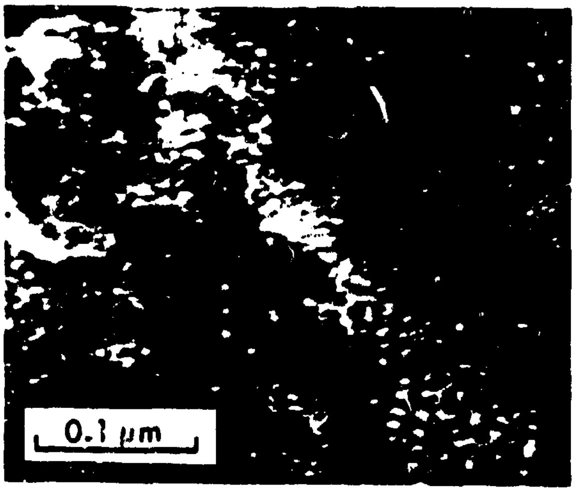

Fig. 1. 100 Å dia voids in neutron-irradiated Al_2O_3 (188,000X) [9].

defects) growth calculated from x-ray peak shifts agrees with that measured macroscopically. This is the case with SiC, where swelling of ~3% at RT is observed but no aggregated defects are seen in TEM [13]. Apparently large concentrations of point defects can be stable in this ceramic lattice.

Considering the complexity of radiation effects in ceramics, it is not surprising that temperature-dependence of swelling is unpredictable. Whereas most metals show a swelling peak in the temperature range 0.4 to 0.6 of the absolute melting temperature T_m, ceramics often do not. Figure 2 presents swelling as a function of temperature for five ceramics which show peaks ranging from 0.21 to 0.53 T_m [12]. It is clear that until damage mechanisms in ceramics are fully understood, temperature-dependence of swelling cannot be predicted but must be determined experimentally.

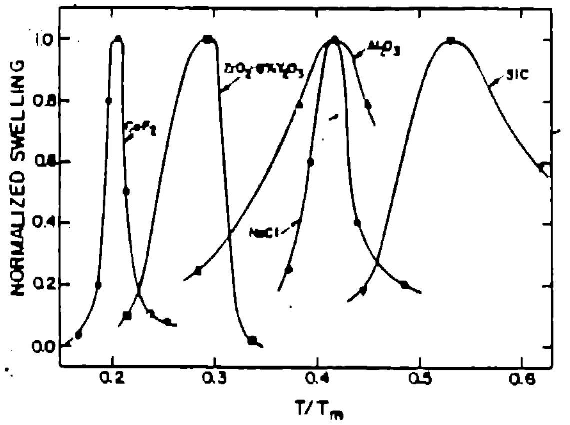

Fig. 2. Temperature-dependence of swelling for five ceramics [12].

Swelling behavior of a number of candidate ceramics for fusion applications has been evaluated in elevated-temperature fission reactor irradiation studies in EBR-II. Results after irradiation at 1015 K to 2.8 x 10^{25} n/m^2 (E_n > 0.1 MeV) show that $MgAl_2O_4$, $Y_3Al_5O_{12}$, Y_2O_3, Y_2O_3-1%ZrO_2, Si_3N_4, Si_2ON_2, and $Si_4Al_2O_2N_6$-5%Y_2O_3 all swelled less than 0.5 vol %, whereas Al_2O_3 and BeO swelled in the range of 1 to 3 vol% [14]. Irradiation at 925 and 1100 K to doses between ~1-2 x 10^{26} n/m^2 gave the following approximate results [15]:

- less than 0.5 vol % swelling for $MgAl_2O_4$, $Y_3Al_5O_{12}$, Si_2ON_2, and $Si_4Al_2O_2N_6$-5%Y_2O_3
- 0.5 to 1.5 vol % swelling for Si_3N_4, Y_2O_3, and Y_2O_3-1%ZrO_2
- 3 to 10 vol % swelling of Al_2O_3 and BeO.

Thus a number of ceramics exhibit good dimensional stability to rather high fission neutron doses at the temperatures evaluated. However, non-cubic polycrystalline ceramics are subject to internal cracking even at low swelling levels if grain growth is anisotropic. For example polycrystalline BeO, which expands preferentially along the c axis at 375 K, suffers drastic weakening at doses greater than 2 x 10^{24} n/m^2 (E_n > 1 MeV) and bulk swelling values of ~1 vol% [16]. Therefore even small dimensional changes can have serious consequences for non-cubic ceramics, unless single-crystal forms are used.*

3.1.2 Strength

Tensile fracture strength of ceramics is characterized by the relationship $\sigma_f = (2E\gamma_f/c)^{1/2}$, where E = Young's modulus, γ_f = fracture energy, and c = size of crack-initiating flaw (either surface or bulk). Cracks can grow slowly until fracture conditions are achieved, a process known as static fatigue. Room-temperature bend strengths for typical high-quality refractory materials range from ~2 to 9 x 10^8 N/m^2, but a large factor of safety must be used in design to take into account the statistical nature of flaw size distributions and static fatigue. Temperature-dependence of strength for refractory ceramics is not large, so that room-temperature strength values are often roughly applicable up to blanket temperatures and beyond. Brittle material design usually calls for ceramics to be placed in compression, but high-strength materials such as Si_3N_4 and SiC perform well under flexural stress conditions.

Progress has been made in recent years in development of ceramic microstructures that increase strength. For example, Bansal and Heuer attained an increase of ~50% for non-stoichiometric spinel (MgO·3.5 Al_2O_3) by precipitation strengthening [18]. Results such as these suggest that even stronger ceramics than those presently considered high-strength materials will be developed in the future.

The most likely sources of strength change in a fusion environment are surface damage (from ion irradiation, neutral gas erosion, or evaporation) and bulk damage (from neutron irradiation). Surface damage can alter flaw size, while bulk damage may affect either flaw size or fracture energy. Since technological ceramics typically contain significant surface and internal flaws resulting from fabrication, the fusion environment will not necessarily have a major effect on the flaw size term in the strength equation. Exceptions may be microcracking (from anisotropic grain expansion), void formation at grain boundaries, grain boundary grooving (from chemical erosion, and surface smoothing (from ion or chemical sputtering). In the last case strengthening would be expected.

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*With the advent of economical methods for continous growth of single crystals [17], ceramics made by these techniques have become candidate materials for fusion applications.

Neutron irradiation can alter fracture toughness $(2E\gamma_f)^{1/2}$ by introducing microstructural changes. Hurley [19] found that room-temperature fracture toughness (as well as hardness) of single-crystal Al_2O_3 irradiated to 2.8 x 10^{25} n/m^2 (E_n > 0.1 MeV) at 1015 K was significantly increased by the presence of voids and dislocation tangles. $MgAl_2O_4$, which showed no aggregated damage, and $Y_3Al_5O_{12}$, which contained only damage clusters, exhibited essentially no change in fracture toughness. Similarly, cubic phase polycrystalline SiC, which contained fine-scale damage unresolvable by TEM, did not show a significant strength change after irradiation to 2.8 x 10^{25} n/m^2 (E_n > 0.18 MeV) at 903 and 1293 K [13]. On the other hand, polycrystalline MgO (cubic) exhibited an enhanced tendency toward transgranular fracture after neutron irradiation, suggesting a decreased fracture toughness [20]. Damage microstructure in MgO is characterized by dislocation loops and tangles [21]. The behavior of MgO remains unexplained and may be atypical of ceramics.

The discussion to this point has dealt only with post-irradiation strength. However, ceramics used in applications that involve operating temperatures at or above those of the blanket with concurrent stress may be subject to in-situ creep. This must be taken into account by designers, both because of dimensional changes and because creep can induce flaws which lead to fracture [22]. The addition of neutron damage may cause significant changes in creep behavior. In-reactor tests of UO_2 show that creep rate below ∼1500 K is increased by fission damage, an effect attributed to the migration of interstitials to dislocations and consequent enhancement of dislocation mobility [23]. Nevertheless, a gradual increase in density of irradiation-induced damage could impede dislocation motion [24], so that creep rate may show a decrease after long time periods.

3.1.3 Thermal conductivity

Thermal conduction in electrically-insulating ceramics at low and moderate temperatures is by phonon transport (propagation of elastic waves). Phonon conduction is highest for ceramics with simple lattice structures in which the difference in atomic weight of anion and cation species is small. The importance of thermal conductivity to fusion applications is that high values result in lower operating temperatures and thermal stresses. Some typical thermal conductivity values for polycrystalline ceramics are shown in the following table:

	Thermal Conductivity (W/m.K)	
Material	300 K	1300 K
Al_2O_3	34	5.4
$MgAl_2O_4$	20	5.2
BeO	280	20
ThO_2	14	2.6
SiC	76	38
Si_3N_4	36	19

These values are generally consistent with the above guidelines. It may be seen that thermal conductivities decrease significantly at higher temperatures, where phonon-phonon scattering is increased. Conductivity of single-crystal Al_2O_3 at 1300 K is roughly a third higher than the value shown for polycrystalline material and increases at higher temperatures, due to photon conductivity. This mechanism of heat transfer is particularly important for the radiatively-cooled first-wall liner application, where single crystals can offer advantages over polycrystalline ceramics.

Irradiation-induced defects decrease thermal conductivity by increasing phonon scattering. Calculations show that point defects are more effective than are aggregates, for a given number of defect sites [25]. Thus higher irradiation temperatures, where greater defect aggregation is expected, usually result in lesser reductions [26,27]. The magnitude of reductions varies greatly with ceramic tested; Price [26] irradiated SiC to a dose ≥ 2.7 x 10^{25} n/m^2 at 825 and 1375 K, and reported subsequent room-temperature decreases of ∼37% and 67% respectively. Hurley [27] measured room-temperature thermal diffusivity of twelve oxides and nitrides after irradiation to fluences of 0.3-2.3 x 10^{26} n/m^2 at 925, 1015, and 1100 K, and found reductions of from ∼3% (for single-crystal $MgAl_2O_4$) to 93% (for Si_2ON_2). SiC showed saturation in this property below 10^{25} n/m^2 [28], while saturation or a tendency toward saturation below 2 x 10^{26} n/m^2 was seen in the twelve ceramics investigated by Hurley. The fractional reduction in thermal conductivity due to irradiation is less when measured at elevated temperatures. For example, the decrease for Al_2O_3 at 723 K is 15% compared with 45% at 300 K [15]. Thus the effect of radiation damage on thermal conductivity, while remaining significant, becomes less of a problem at higher operating temperatures.

3.1.4 Physichemical surface alterations

In several fusion applications ceramic surfaces will be exposed to some or all of the following:

- light ions (D, T, and He)
- impurity ions (e.g., from first wall)
- neutral fuel gases
- fast neutrons
- photons
- electrons.

The consequences of this exposure are of concern both because of plasma contamination problems and because of the potential for damaging the ceramic surface. Principal mechanisms for material alteration are physical sputtering, blistering, chemical erosion, and vaporization, with primary material concerns being ceramic thinning and changes in surface topology or composition.

Physical sputtering. Ions and neutrons which strike a surface can cause ejection of near-surface atoms. Sputtering yield S (atoms ejected per incoming particle) is primarily a function of surface binding energy [29]. Most sputtering measurements have been made on metals, but since ceramics and metals have similar binding energies, yields should be similar. A comparative study [30] of heavy-ion sputtering from oxides and their parent metals generally supports this prediction; for example, yields from Al_2O_3 and MgO were slightly lower than those for Al and Mg, while yields from ZrO_2 and Nb_2O_5 were slightly higher than the values for Zr and Nb. Some other ceramics showed preferential oxygen sputtering, which could lead to degradation in electrical properties of insulators. However, the phenomenon was primarily limited to ceramics in which it is energetically favorable to form lower oxides. In the absence of this effect, the likelihood of preferential sputtering of anions or cations depends on such factors as particle energy transferred to each sublattice and relative displacement energies; these may on balance favor preferential cation ejection.

For light ions (O, T, and He), sputtering yields are strongly dependent on particle energy, with S for many metals peaking at roughly 10^{-3} to 10^{-1} for energies of 1 to 10 keV [31]. Values for SiC are similar to those for metals, i.e., S = 9×10^{-3} for 5 keV hydrogen ions [32]. Yields for heavy ions can be much higher, with no apparent peak in the energy range of interest [32]. Sputtering yields for metals irradiated with 14 MeV neutrons are on the order of 10^{-4} [33], and are therefore not considered a major problem.

Surface roughening is often observed after sputtering experiments, with detailed examination showing a dependence on crystallographic orientation [32]. A roughened surface can be deleterious in some ways (for example, by increasing impurity adsorption), but may by geometrical effects reduce loss of surface material.

Blistering. When gas ions penetrate the surface of a material in which permeability is insufficient to accommodate the gas, blistering can result. Hydrogen permeability is generally low in both crystalline ceramics and glasses; that for helium is also low in ceramics, but significant helium permeability is observed in some glasses. Studies by Mattern et al. [34] illustrate the dependence of blistering on gas diffusivity. Glasses in which composition was varied to alter helium diffusivity were irradiated with 150 kV He^+ at 383 and 493 K. With a diffusivity below $\sim 3 \times 10^{-12}$ m^2/s blistering was observed, whereas above this value none was seen. Figure 3 shows the nature of blisters formed with D $\gtrsim 10^{-15}$ m^2/s. These results indicate that higher operating temperature should generally reduce blistering, and that adjustments of composition can achieve a similar effect. Dramatic increases in diffusivity may result from slight compositional changes; Fowler et al. [35] found that the addition of 0.2% MgO to Al_2O_3 increased diffusion coefficients by four to five orders of magnitude, apparently as a result of an alteration in diffusion mechanism.

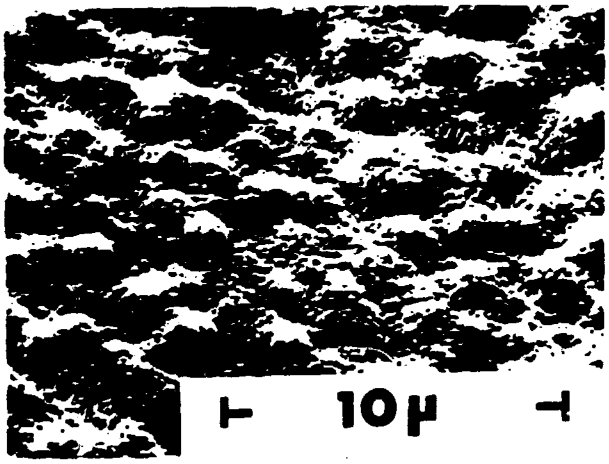

Fig. 3. Blisters in He^+-irradiated glass (35% Li_2O·65% SiO_2) (3150X) [34].

It is apparent that blistering induces surface roughness (Fig. 3). When exfoliation occurs, a material loss rate as high as ∿0.4 atoms/particle may result [36]. However, high-dose studies of SiC have shown that atom-by-atom sputtering can erode away adherent blisters [32]; in this case the blistering phenomenon becomes less important.

Chemical Erosion. Oxides, carbides, and nitrides are subject to chemical reduction by hydrogen isotopes. High-energy particles may penetrate the surface and react internally (e.g., to form OH in Al_2O_3 [37], or back-diffuse and react at the surface. Exposure to lower-energy (e.g., 2000 K) neutral atomic hydrogen, which is much more reactive than is the molecular gas, can also lead to surface attack. Particle energy, surface temperature, and surface chemical processes (sticking and recombination probabilities, thermodynamic reactions, and reaction kinetics) all play a role in determining the seriousness of chemical attack.

Studies of the response of SiC to irradiation with 1 keV hydrogen ions have been carried out by Roth et al. [38]. While sputtering was observed, it was concluded that the process was physical rather than chemical in nature because yield showed no pronounced temperature dependence from RT to 883 K and no increase in hydrocarbons was noted by mass spectrometry. These findings were in contrast to those for graphite, where chemical erosion was indicated by accelerated material loss and hydrocarbon production with heating, up to ∿900 K. Chemical attack of UO_2 and Al_2O_3 exposed to 2500 K neutral atomic hydrogen has been investigated by Dooley et al. [39]. Uranium dioxide showed a detectable reaction rate at ∿775 K; this was attributed to the relative ease with which UO_2 can be partially reduced, to UO_{2-x}. Al_2O_3 proved much more resistant to attack, with no reaction observed below ∿1600 K. Above that temperature H_2O and Al were detected spectrometrically, and an aluminum skin found to be formed on the sample. Rate of chemical attack was limited by vaporization rate of this metallic skin.

With well-chosen ceramics, chemical erosion problems should be limited to applications where operating temperatures are well above those of the blanket. For the high-temperature first-wall liner application principal material concerns are thinning and changes in near-surface structural properties, while for the insulating coating on RF antennas possible loss of insulating properties is an additional factor.

Vaporization. Only for those ceramic applications where temperatures will be well above that of the blanket is vaporization expected to be a problem. Estimated evaporative temperature limits based on a wall erosion rate of 1 mm/yr (taking into account both vaporization and chemical reaction) show that typical candidate ceramics fall within the range 1300-2100 K [5]. Some temperatures of interest are Si_3N_4, 1323 K; Al_2O_3, 2033 K; and SiC, >1793 K. Graphite has a limit of 2253 K, and has thus been proposed as a coating for lower-temperature wall materials. The limit for Si_3N_4, a material with otherwise excellent performance characteristics, is disappointingly low; however, recent results indicate that several forms of this ceramic exhibit lower vaporization rates than were predicted [40].

It is apparent that fusion-reactor ceramics may be subjected simultaneously to a variety of damage processes, so that synergistic effects are probable. For example, if chemical erosion results in surface reduction, then physical sputtering and vaporization rates will be altered. The list of possible synergistic effects is too extensive to mention here; however, the point should be made that performance limits for ceramics must be determined under conditions which simulate all relevant aspects of the operating environment.

3.2 Electrical properties

3.2.1 Dielectric strength

Bulk dielectric breakdown of insulators can occur by either the thermal or the avalanche mechanism [41]. In the former case, a small current flow within the material leads to Joule heating which in turn accelerates conduction until breakdown occurs. Low resistivity and long times of voltage application enhance the likelihood of breakdown, so that this mechanism is more likely to operate at high temperatures and under ac or dc (as opposed to short-pulse) conditions. If thermal breakdown does not occur, then at higher voltages electron collision ionization and multiplication can cause avalanche breakdown. This mechanism is not strongly temperature-dependent.

Temperature- and time-dependence of breakdown vary with the insulator under consideration. For Al_2O_3, the thermal mechanism operates above 450 K under dc conditions [42], while breakdown under voltage pulses of 10^{-2} sec or less at 973 K appears to be by the avalanche mechanism [43]. Dielectric breakdown strength (dbs) for one form of alumina tested with short pulses was roughly constant at $\sim$40 kV/mm from RT to 1073 K, whereas dbs under dc conditions dropped from this value to $\sim$4 kV/mm at 1073 K [43]. It is apparent that in this work thermally-induced increases in conductivity did not degrade dielectric strength under avalanche breakdown conditions.

There is some evidence that repeated application of sub-breakdown voltage pulses to insulators can lead to a decrease in breakdown strength. Bunch [44] found that the room-temperature dielectric strength of single-crystal Al_2O_3 was decreased roughly 20% by prior application of $\sim 10^4$ high-voltage pulses. However, some samples did not show this "electrical fatigue" effect even up to $\sim 10^5$ pulses. Statistical calculations based on dislocation density indicated that the likelihood of a sample not showing fatigue correlated roughly with the likelihood of dislocations being absent from the breakdown area of a dimpled sample. It should be noted that the insulator tested had a high dbs ($\sim$180 KV/mm for a low number of pulses), so that it is not clear whether a fatigue effect is to be expected in lower-grade ceramics.

Insulators used in most critical fusion applications will be exposed to displacive and/or ionizing radiation. Structural damage can supply internal sites for electron emission or scattering as well as defect states within the band gap. The latter can alter charge carrier concentration by supplying electrons and holes to conducting states, or by serving astrapping and recombination sites. Bunch [44] measured the short-pulse, room-temperature dbs of single-crystal Al_2O_3 after elevated-temperature irradiation to $1\text{-}2 \times 10^{26}$ n/m^2, and found no degradation. It is apparent that the concentration of small voids and dislocations present (Fig. 1) did not have a major effect on dielectric strength of this material, at least under avalanche breakdown conditions. However, it has been pointed out [45] that larger pores ($\sim 10^{-4}$ mm) could support an electron avalanche sufficiently energetic to produce ionization events at the pore wall in a 10^5 V/mm field.

The effect of ionizing radiation is to excite charge carriers into conducting states and into and out of traps and recombination centers. Since charge carrier concentration at any one time is a balance of several kinetic processes, rate of ionizing radiation is considered more important than total dose. (However, the effect of lengthy exposure to ionizing radiation needs further investigation.) Britt and Davis [42] investigated the effect of an x-ray dose rate of 2×10^4 Gy/h on dbs of Al_2O_3. A slight reduction in dielectric strength was observed in the avalanche temperature range (up to 450 K). This was attributed to radiation-induced filling of traps by charge carriers which were then available as avalanche collision targets. No significant reduction was seen in the thermal breakdown region (> 450 K), where field-enhanced thermal emission exceeded radiation-induced carrier generation. These findings are relevant to the neutral beam injector insulator application, where a gamma flux of $\sim 10^4$ Gy/h may be encountered, but such a dose is far too low to simulate first-wall conditions.*

Dielectric breakdown of insulators often occurs across the surface rather than through the bulk. Surface breakdown is postulated [46] to take place in two steps: electric field accentuation, followed by flashover. In the first step, surface irregularities cause field enhancement and consequent emission of electrons. When these strike the insulator surface, secondary electrons are emitted. The surface is left positively charged by loss of electrons so that the field is further enhanced near the cathode. When the electron avalanche desorbs surface impurities and these are ionized by the electric field, flashover occurs. Secondary electron yield is an important material parameter, but insulator configuration and cleanliness may be more significant factors in determining surface dbs. Dielectric strength can range from almost arbitrarily low values under unfavorable conditions up to ~ 10 kV/mm or higher with controlled geometry and environment [47]. Conditioning (i.e., a gradual increase in voltage) aids in attaining a high dielectric strength, apparently by removing surface irregularities and/or impurities. Coatings which offer controlled surface resistivity, low secondary emission yield, or improved surface cleanliness are often used to increase dbs.

To summarize, insulators for technological applications do not possess a unique bulk or surface dielectric strength. Insulator size, geometry, electrode configuration, temperature, environment, and mode of voltage application can be as important as material characteristics in determining dbs. Thus both reactor designers and materials scientists have important roles in the successful development of high-voltage insulators for fusion devices.

3.2.2 Electrical Resistivity

Electronic conduction in insulators takes place by motion of electrons or holes in an electric field. At room temperature a good technological insulator exhibits a resistivity so high ($>10^{12}$ $\Omega\cdot$m) that measurement of the exact value is difficult. However, a steep temperature dependence is observed as charge carriers are thermally stimulated into conduction states. Typical values for ceramics such as Al_2O_3, BeO, and ThO_2 are in the range 10^4-10^7 $\Omega\cdot$m at 1000 K [48]. Silicon carbide, a prime candidate for low-Z first wall applications, is a semiconductor [49] and is not suitable for most insulator applications.

Irradiation can affect resistivity by inducing structural damage and depositing ionizing energy. Structural changes may alter electronic properties (e.g., introduce trapping, recombination, or scattering centers) in such a way that resistivity could either increase or decrease. Of greater concern, however, is absorption of ionizing energy, which can generate a density of charge carriers beyond that induced by thermal effects. Davis [48] measured conductivity of a number of ceramics in a gamma field of $\sim 5 \times 10^4$ Gy/h and found that below temperatures of 400 to 700 K (depending on the material) a significant increase was noted (e.g., from $\sim 10^{-12}$ to 10^{-9} $(\Omega\cdot m)^{-1}$) at room temperature. Above these temperatures the thermal contribution dominated. These results show that neutral beam injector insulators operating near room temperature in a gamma flux of $\sim 10^4$ Gy/h should easily meet a design resistivity requirement of 10^6 $\Omega\cdot$m.

Much higher rates of absorption of ionizing energy may be encountered in RF, current break, and some magnet insulator applications. Van Lint et al. [45] calculated increases in conductivity at high ionizing fluxes under fusion reactor conditions. The relationship $\Delta\sigma(\Omega\cdot m)^{-1} = k\dot{\gamma}$ (Gy/s) was used, i.e., proportionality between conductivity changes $\Delta\sigma$ and ionizing dose rate $\dot{\gamma}$ was assumed. This assumption is most appropriate at lower temperatures [50]. Values for k, a material parameter, range from $\sim 10^{-3}$ to 10^{-13} for typical insulators but may increase or decrease with accumulated irradiation dose. Applying the abovr relationship to a case where $\dot{\gamma} = 10^4$ Gy/s and $K = 10^{-9}$-10^{-13}, the resulting change in conductivity is 10^{-5}-10^{-9} $(\Omega\cdot m)^{-1}$. Insulators with a negligible initial conductivity will under this ionizing flux exhibit resistivities of 10^5-10^9 $\Omega\cdot$m. Such resistivities are on the order of those expected at blanket temperatures considering only thermal effects, but represent a significant degradation at lower temperatures.

Klaffky et al. [50] evaluated radiation-induced conductivity in single-crystal Al_2O_3 as a function of electron bombardment-induced-ionization rate, temperature, and crystal perfection. The dependence of conductivity of Cr-doped and neutron-irradiated Al_2O_3 on ionization rate was found to change from exponent one to less than one with increasing temperature. This effect is consistent with a model involving capture of electrons at a recombination center and their subsequent recombination with holes thermally released from a hole trap at intermediate temperatures. Change of conductivity with temperature at constant ionization rates for the same two forms of Al_2O_3 is shown in Fig. 4; the initial decrease can be attributed to reduced mobility (from longitudinal optic mode phonon scattering) or hole release and subsequent recombination

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*At the first wall of a 2 MW/m^2 fusion reactor the neutron flux of $\sim 10^{19}$ n/m$\cdot$s will alone deposit $\sim 10^4$ Gy/s of ionizing energy in an insulator.

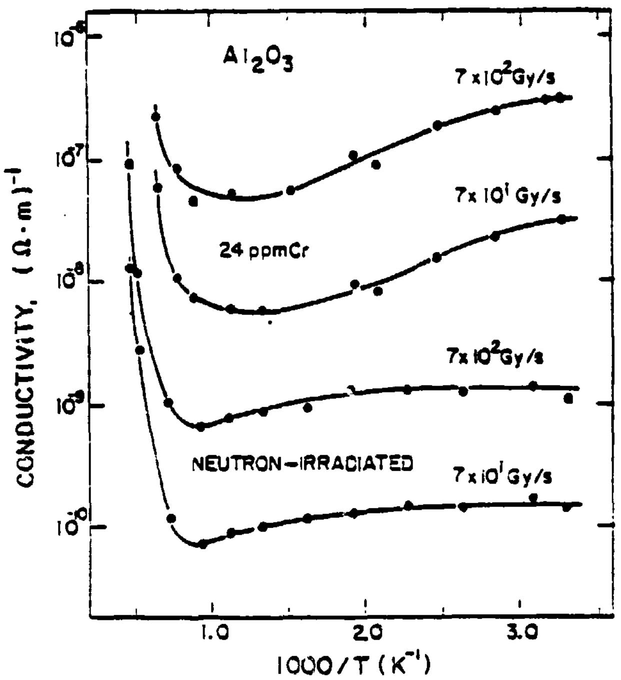

Fig. 4. Conductivity of Al_2O_3 as a function of temperature and intensity of ionizing radiation [50].

with electrons at a shallow trap, and the increase to detrapping from deep electron traps. It is apparent from these results that elevated-temperature resistivity behavior under ionizing radiation can be complex, and must be determined experimentally.

Surface resistivity is strongly dependent on condition of the insulator surface. Where contamination is expected, glassy insulators or glazed ceramics are often specified because of their resistance to impurity pickup and ease of cleaning. Injection of energetic hydrogenic ions and deposition of sputtered or vaporized metal can degrade surface resistivity, thus presenting a problem for insulators in some applications (e.g, neutral beam injectors and direct converters). Design accommodations such as insulator shielding may be required to minimize surface degradation.

Bulk conductivity in insulators can take place by motion of ions as well as electrons and holes. This topic is discussed in the next section.

3.2.3 Electrolysis

Ionic conduction involves the motion of matrix or impurity ions toward the electrode of opposite polarity. There is considerable uncertainty as to whether current in insulators is carried primarily by ionic or electronic charge carriers, but many materials (e.g., Al_2O_3, BeO, ThO_2, ZrO_2, and Y_2O_3) have been shown to exhibit at least partial ionic conduction [51]. It is generally believed that the fraction of current carried by ions increases as temperature rises, but there is some evidence to the contrary [51].

Under ac fields ionic conduction does not alter distribution of ions. However, ionic current flow under dc voltages results in the separation of cationic and anionic species. The consequence of this process of electrolysis can be serious degradation in electrical and structural integrity of an insulator. Gibson and Ballard [52] evaluated this phenomenon in a porcelain insulator of mixed crystalline and amorphous phases. When a rather low dc electric field ($\sim$100 V/mm) was applied at 295 K no effects were observed. However, at 355 K significant structural and electrical changes took place and at 425 K serious damage occurred. The dominant charge carriers were Na^+ and K^+ ions in the vitreous phase, although oxygen migration was also detected. In one test of 47 hrs at 475 K, sufficient alkali deposits formed at the cathode to change the dimensions of the insulator at that point and cause structural failure of the porcelain-to-metal seal. A depletion layer was formed near the anode, resulting in steep voltage gradients and increased likelihood of dielectric breakdown.

It is clear that the porcelain described above would not be adequate for such fusion applications as neutral beam injector and direct converter insulators, where elevated temperatures and high dc electric fields are typical. Refractory ceramics exhibit better resistance to prolonged usage under dc conditions, but these also have their limits. Weeks et al. [53] found that single-crystal MgO and $MgAl_2O_4$ became conducting and broke down when subjected to electric fields of 10-100 V/mm for 5 to 150 hrs at 1273 K. Since diffusivity is typically enhanced by irradiation, it must be anticipated that a significant neutron flux will accelerate electrolysis. Where this process is found to limit insulator lifetimes, electric fields and/or operating temperatures must be reduced.

3.2.4 Loss Tangent

A low loss tangent ($\tan\delta$) is required for insulators used in RF energy transmission, to reduce power losses and avoid overheating and thermal stresses in the dielectric. Energy loss* in an insulator can result from three frequency-dependent processes [54]:

- ion migration losses (dc conductivity, ion jump, and dipole relaxation)
- ion vibration and deformation losses
- electron polarization losses.

At room temperature the first process operates primarily at frequencies below $\sim10^6$ Hz, the second above $\sim10^{10}$ Hz, and the third at optical frequencies ($\sim10^{15}$ Hz). Losses are thus at a minimum over most of the frequency range of interest for RF heating applications. Ion

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*Power loss is proportional to the product of loss tangent and dielectric constant, but the former usually shows the wider variability.

migration losses are strongly temperature-dependent, while those from other sources are not.

A loss tangent of $\sim 10^{-4}$ is often mentioned in conceptual studies of RF heating [55,56]. Such a value is attained only in high-quality low-loss insulators at low temperature, with tan δ = 10^{-} being more typical of commercial insulators. The extent to which a given insulator suffers an increase in loss tangent with increasing temperature depends on test frequency. Results for a commercial steatite are shown below [57]:

frequency, Hz	tan δ	
	300 K	775 K
10^5	1×10^{-3}	60×10^{-3}
10^7	1×10^{-3}	30×10^{-3}
10^{10}	3×10^{-3}	5×10^{-3}

In general, high frequencies are correlated with good high-temperature performance.

Since the mechanism of energy loss is motion of charge carriers (ions and electrons), it is not surprising that irradiation-induced lattice disruption leads to an increase in loss tangent. MacChesney and Johnson [58] found that low doses of fast fission neutrons (6×10^{21} to 5×10^{23} n/m^2) caused an increase of tan δ in Al_2O_3 by factor of 13 and 3 respectively, at room temperature and 10^6 Hz. It was suggested that radiation annealing was responsible for the decrease at the higher dose. Budnikov et al. [59] irradiated Al_2O_3 with 8×10^{25} thermal n/m^2 and observed an increase in tan δ by 25 times at 775 K and 10^6 Hz. The degradation in this study was attributed to the formation of 0.1 wt.% silicon by transmutation of ^{28}Al.

Perhaps nowhere in the fusion reactor development program is there a clearer relationship between reactor design and material performance than in the area of insulators for RF heating. The effects of operating temperature, frequency, and radiation damage level on insulators *must* be taken into account in design and specification of RF power systems, if these systems are to meet performance requirements.

3.3 Fabrication

Many applications for ceramics in fusion systems call for parts of large size (perhaps joined or coated), complex shape, and high quality. Further, these requirements will in many cases have to be met with materials specially developed for the severe fusion environment, where fabrication techniques have not been optimized. The following is a partial list of requirements which may be imposed on fabricators, for the critical applications identified earlier:

- insulators for RF heating systems--large size, low porosity, high purity, special material
- neutral beam injector insulators--large size, complex shape, low porosity
- toroidal current break--large size, special material
- magnetic coil insulators--special material for the poloidal coil application
- direct converter insulators--low porosity, special material if thermal conversion is used
- low-Z first wall/liner--large size, complex shape, high purity, special material.

Fabrication costs may also be an important consideration in cases where unusual requirements or large quantities of material are involved.

A recent survey of the state of the art for fabrication of alumina (and, by implication, other) ceramics have shown limited industrial capabilities in some area important to development of fusion power [60]. It is apparent that further fabrication activities, carried out in conjunction with reactor designers and material property investigators, will be required to develop ceramics for advanced fusion systems.

4. FUSION NEUTRON IRRADIATION EFFECTS

Until high-intensity fusion neutron sources become available, irradiation studies must principally be conducted in fission reactors and low-intensity fusion neutron sources.* In order to make maximum use of fission neutron data and allow estimates of differences in damage effects induced by fission and fusion neutrons, correlations are required in the following areas:

- nature of damage production events
- effect of differing ionizing/displacive energy loss ratios
- effect of differing dpa/gas atom generation ratios.

Low-dose studies have been carried out in two ceramics to determine the relative number of surviving optically-active point defects after near-room-temperature irradiation in fission reactors and the Rotating Target 14 MeV Neutron Source (RTNS). Chen et al. [61] found roughly twice as much damage from fusion neutrons as from fission neutrons in MgO, as did Evans and Bunch [62] in Al_2O_3. A ratio of 2 is in reasonable agreement with estimates from damage energy calculations [61,62]. This indicates that point defect survival fractions are similar after damage by fission and fusion neutrons, and thus suggests that damage production processes may not be vastly different in the two cases. On the other hand, a comparison of aggregated defects in metals after fission and 14 MeV neutron irradiation has shown a significant difference in cluster size and number density; this is attributed to differences in probability of vacancy-interstitial recombination [63]. A comparison of cluster sizes has not been carried out for a ceramic.

When calculated damage per neutron in a fast fission reactor spectrum is compared with that at the first wall of a fusion reactor, it is

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*Certain topics in radiation damage (such as the significance of rate effects) can best be addressed by ion and electon irradiation studies.

found that in low-Z ceramics roughly equal damage is induced. Since somewhat higher flux levels can be obtained in the core of fission reactors than at the first wall of a 2 MW/m^2 fusion reactor, real-time or slightly accelerated damage rates can be simulated in fission reactors. However, damage simulation in compounds must be evaluated not only as a function of total dpa rate, but also as a function of the ratio of dpa rates on cation and anion sublattices. This is necessary because the nature of stable damage formed (both point defects and aggregates) can depend on this ratio. An example is the phenomenon of void formation in Al_2O_3, where a void must contain a ratio of two aluminum vacancies to three oxygen vacancies. If the dpa ratio varies with neutron spectrum, the likelihood of formation of voids (perhaps in competition with other aggregates such as aluminum precipitates) may differ between fission and fusion neutron irradiation studies. Calculations of dpa ratios as a function of neutron spectrum for typical ceramics are needed to allow the importance of this effect to be evaluated.

The nature of permanent damage in ceramics has been found to vary with the amount of ionizing energy absorbed, for a fixed amount of displacive energy deposited. Ion irradiation studies by Arnold et al. [64] have shown that optical absorption and volume expansion in Al_2O_3 vary in an opposite sense as large changes are made in ionizing energy component; these effects are attributed to variations in charge state of the predominant point defect. It is thus apparent that the ratio of ionizing to displacive energy deposited must be considered when conducting simulated fusion reactor irradiations. Ionizing energy can be deposited in the lattice from absorption of electromagnetic radiation or from the electronic component of particulate energy loss Parkin [65] has calculated the ratio (energy partitioned into electronic processes/energy into atomic processes) for neutron irradiation of aluminum to be 0.81 for row 7 of the EBR-II fission reactor, 2.71 at the first wall of a fusion reactor, and 4.51 for a pure 14 MeV spectrum. However, a comparison of optical absorption spectra from Al_2O_3 irradiated with fission neutrons [66] and 14 MeV neutrons [62] shows that the ratio of number of defects of each charge state (F/F$^+$ centers) does not vary greatly in the two studies. If this observation holds true at higher damage levels characteristic of fusion applications, then the rather small differences in ratio of ionizing-to-displacive energy for fission and fusion neutron spectra will have been shown not to be important. Nevertheless, differences in rate of electromagnetic energy deposition must still be considered.

Perhaps the most significant damage-related distinction between fission and fusion neutrons is their different abilities to induce transmutations. Cross-sections for nuclear reactions typically rise sharply between 1 and 14 MeV, so that much higher concentrations of transmutation products are generated in a fusion environment. For example, Parkin and Goland have shown [67] that the spectrum averaged (n,α) cross-section for production of helium from aluminum is 443 times higher for a first-wall spectrum than for Row 7 of EBR-II. Muir [68] and Rovner and Hopkins [5] have calculated the rate of generation of gaseous and metallic transmutation products for typical ceramic elements and compounds in a first-wall neutron flux. After a year's exposure in a 2 MW/m^2 fusion reactor, ceramics will typically contain 1000-2000 appm each of hydrogen, helium, and metallic impurities. The elements beryllium, boron, and carbon (e.g., in BeO and B_4C) have higher-than-average (n,α) cross-sections, so that estimates of helium content in ceramics containing these species will be higher than the above values.

The presence of gaseous impurities along with displacive damage is usually considered deleterious. Hydrogen and helium generally exhibit low solubilities in ceramics and will thus precipitate, given sufficient mobility (i.e., at elevated temperatures). Lattice energetics may favor their deposition as gas bubbles at grain boundaries, as has been observed with helium in ThO_2 [69] and PuO_2 [70]. This can lead to a weakening of the structure and ultimately to grain boundary separation. Intergranular void formation can also be assisted by the presence of insoluble gases, which stabilize sub-critically-sized void nuclei (vacancy aggregates) against re-solution. This is not necessarily disadvantageous; if a ceramic which would otherwise contain a relatively small number of large voids has the concentration of void nuclei increased by the presence of gas, the result may be a finer void distribtuion and less degradation of physical properties. However, an overall increase in void volume could result from more effective scavenging of vacancies into voids.

Helium is soluble and has a high diffusivity in some glasses, so that problems with this gas can be reduced by using glass in bulk form or as a grain boundary phase in polycrystalline ceramics. Nevertheless, the presence of hydrogen will remain a problem.

Metallic transmutation products must be accommodated electrochemically and dimensionally in the ceramic, with their ultimate disposition being dependent on the nature of the lattice and transmutation product in question. Impurities might be accommodated by solution, precipitation of a second phase, or formation of planar defects, perhaps assisted by valence changes. The effect of such defects on physical properties will not necessarily be large, and will in some cases be similar to those observed when normal impurities are present. However, the fact that transmutation products are intially injected forcibly into solution may mean that their behavior will be quite different from that of the same impurities introducted chemically.

The effect of transmutation-induced hydrogen and helium (as well as metallic products) could be simulated by irradiating an isotopically-adjusted ceramic in a mixed-spectrum reactor. The use of ^{17}O would allow generation of helium by the thermal neutron reaction $^{17}O\ (n_{th},\alpha)^{14}C$, and of hydrogen by the $^{14}N\ (n_{th},p)^{14}C$ reaction, (In the latter case, ^{15}N must be used to suppress hydrogen formation.) The fast component of the neutron spectrum would generate displacements simultaneously with gas production.

Sialon is an appropriate model ceramic for such a study. It may be shown [71] that $Si_3Al_3O_3N_5$ containing oxygen of 50% ^{17}O content and nitrogen of 7.1% ^{14}N content will after a year of exposure in the Oak Ridge Reactor have experienced a damage level of ∿6 dpa, ∿400 appm of hydrogen generation, and ∿270 appm of helium generation, thus roughly matching the dpa/gas atom ratios expected under first-wall irradiation conditions. By adjusting the isotopic content of a series of sialon samples, the effects of displacement only, displacement plus helium production, and displacement plus hydrogen production can be studied separately.

5. CONCLUSIONS

Ceramics are needed for a number of critical applications in fusion systems. Despite the severe environments characteristic of these applications, it appears that in most cases materials can be specified which will intially satisfy reactor requirements. (The possible exception is insulators for RF heating systems, where both electrical and structural demands can be very stringent). However, ceramic degradation during reactor operation may result in significant shortening of component lifetimes. Consideration of structural and electrical properties of ceramics in the context of anticipated operating conditions suggests that long-term degradation effects resulting from neutron irradiation, ion bombardment, and electrolysis are most likely to prove lifetime-limiting. Of these, neutron irradiation problems appear to be the most difficult because (1) major property changes can result, (2) these problems are not easily alleviated by changes in reactor design and mode of operation, and (3) fusion neutron damage is difficult to simulate.

A number of ceramics can be identified which exhibit good physical properties and show reasonable resistance to fission neutron damage. Included are $MgAl_2O_4$, $Y_3Al_5O_{12}$, Si_3N_4, sialons, Al_2O_3 (in some forms), and SiC. Since a high percentage of ceramics tested to date have shown adequate performance after fission neutron irradiation, it seems likely that as further tests are conducted other materials will be added to the above list. On the other hand, results from high-flux fusion neutron studies, when available, are likely to reduce the number of candidate materials. It is anticipated that materials development efforts will be required for prime candidate ceramics in order to optimize their structural and electrical properties for a fusion environment.

ACKNOWLEDGEMENTS

The author wishes to thank R. W. Klaffky and co-workers at Brookhaven National Laboratory for permission to cite unpublished results in Fig. 4.

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