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TITLE CERAMICS FOR FUSION DEVICES

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CERAMICS FOR FUSION DEVICES

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ABSTRACT

CERAMICS ARE REQUIRED for a number of applications in fusion devices, among the most critical of which are magnetic coil insulators, windows for RF heating systems, and structural uses. Radiation effects dominate consideration of candidate materials, although good pre-irradiation properties are a requisite. Materials and components can be optimized by careful control of chemical and microstructural content, and application of brittle material design and testing techniques. Future directions for research and development should include further extension of the data base in the areas of electrical, structural, and thermal properties; establishment of a fission neutron/fusion neutron correlation including transmutation gas effects; and development of new materials tailored to meet the specific needs of fusion reactors.

INTRODUCTION

Ceramics are required for a variety of applications in magnetically-confined fusion devices. These include:

- insulators for lightly-shielded magnetic coils
- windows for RF heating systems
- ceramics for structural applications
- insulators for neutral beam injectors
- current breaks
- direct converter insulators.

Operating conditions in these devices can be severe, involving intense radiation fields, high mechanical and electrical stresses, and large thermal fluxes. Materials properties of special concern fall into three major categories, which can be subdivided into several more specific topics. They are:

Structural
- swelling
- strength

Thermal
- thermal conductivity

Electrical
- resistivity
- dielectric breakdown strength
- loss tangent.

In any examination of the subject of ceramics for fusion applications, including the present review, materials problems must be evaluated in the context of these potential problem areas.

The subject of this paper has been reviewed earlier by Holmes-Siedle (1), Rovner and Hopkins (2), Clinard (3,4), Clinard and Hurley (5), Suzuki et al. (6), Clinard et al. (7), and Pells (8). These works give much of the background needed for a full understanding of this subject, and are valuable supplements to the present discussion. Nevertheless, since fusion reactor designs, materials requirements, and available ceramic data are continually evolving, a re-examination of this topic seems appropriate. In the present review the three most critical ceramic applications* (the first three in the list above) are described, and performance of several

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*Selection of the most critical applications for ceramics in fusion devices is a difficult and inexact process, since reactor needs and operating conditions are subject to frequent change. However, it appears that these three present the greatest difficulties, and are therefore the focus of this paper. All have in common a severe radiation environment, and their materials requirements taken together are representative of most major ceramics applications.

candidate materials is considered. This is followed by a discussion of opportunities for materials optimization, and the paper concludes with recommendations for future directions in research and development.

MAGNETIC COIL INSULATORS

REQUIREMENTS - Fusion reactor designs call for large normal (i.e., non-superconducting) magnets to serve as choke coils for mirror machines, divertors and plasma shaping coils for tokamaks, and to meet a number of other requirements. Choke and divertor coils must be placed close to the first wall, and will therefore be exposed to lifetime neutron irradiation doses of $\sim 10^{27}$ n/m^2 and ionizing fluxes of 10^4 Gy/s. The ceramic insulators for these magnets will operate near room temperature under high mechanical stresses.

The most detailed design study to date from the standpoint of the insulator has been carried out for the choke coil (9). It is estimated that lifetime swelling cannot exceed 3 vol %, and retention of adequate strength is a requirement. Degradation of thermal conductivity will have little effect in this application. Dielectric strength requirements are modest, but electrical resistivity cannot fall below a calculable (design-dependent) level without causing significant shunting of current to the copper conductor.

CANDIDATE MATERIALS - In the following paragraphs anticipated performance of candidate materials is discussed in terms of the currently available data base. Magnesium aluminate spinel ($MgAl_2O_4$) is the prime candidate ceramic for this application, with MgO and Al_2O_3 also under consideration. Spinel is a low-swelling ceramic: after irradiation to 2.1×10^{26} n/m^2 at 430 K, a polycrystalline form of this material was found to swell only 0.8 vol % (10). If it is assumed that fast fission and fusion neutron damage effects are similar (an assumption implied in this paper unless otherwise stated, but discussed in a later section), a swelling rate linear with dose would indicate a lifetime of 4.3 y at the first wall of a 2 MW/m^2 machine. Strength of the spinel ceramic described above was increased 20 % by that irradiation exposure (10), perhaps as a result of impedance of crack initiation or propagation.

MgO showed less dimensional stability under these irradiation conditions, exhibiting a swelling of 2.6-3.0 % (10); however, strength also increased in this material, by 12-24 %. An equivalent high-dose study has not been carried out for Al_2O_3, but anisotropic swelling of grains within a polycrystalline insulator may lead to premature structural failure.* It may be possible, with the advent of economical techniques for growth of single crystals in technologically-useful shapes (11), to produce Al_2O_3 parts immune to internal damage from anisotropic growth.

It is not anticipated that electrical resistivity of these candidate ceramics, measured after irradiation, will show a significant decrease unless major structural damage such as severe cracking has occurred. During irradiation, however, significant degradation may accompany absorption of ionizing energy as charge carriers are temporarily excited into conducting states.

The most extensive investigation of radiation-induced conductivity (RIC) has been carried out by Klaffky et al. (12,13), in Al_2O_3. Several forms of single-crystal Al_2O_3 differing in purity (12) and in one case with prior radiation damage (13) were irradiated with 1.5 MeV electrons of three different beam intensities. Representative results at 300 K are shown in Fig. 1. It may be seen that RIC increases dramatically with increasing rate of absorption of ionizing energy, but absolute value of conductivity is reduced by the presence of Cr_2O_3 impurity or structural irradiation damage. This behavior was interpreted to be the result of a combination of effects such as creation of electron-hole pairs, trapping and detrapping, scattering, and carrier recombination. With respect to the choke coil application, calculations (9) have shown that moderate increases in conductivity should be tolerable with careful coil design if the degradation found in Al_2O_3 is representative of that in other insulators, can be linearly extrapolated to higher ionizing fluxes, and accurately reflects behavior under other (more fusion-like) irradiation conditions.

Some coil designs specify powdered rather than solid insulator material. In this case swelling of several percent is tolerable, and strength is not a factor. On the other hand, gases interspersed with the ceramic particles may suffer greater loss of insulating properties under irradiation than does the ceramic itself: one estimate puts the degradation of resistivity at ten times that of the ceramic (9). For this reason, and a related concern about thermal runaway in powdered ceramic (9), the most recent choke coil design (14) specifies solid insulating material.

WINDOWS FOR RF HEATING SYSTEMS

REQUIREMENTS - Radio frequency (microwave) plasma heating is required for most fusion reactor concepts. RF energy can be transmitted to the plasma by several techniques: at lower frequencies ($\sim 10^8$ Hz) coaxial cables may be used, while at intermediate (10^9 -10^{10} Hz) and high (10^{11} -10^{12} Hz) frequencies various types of waveguide systems are appropriate. Waveguides will probably require windows to separate the environment of the plasma chamber from that of the RF source, which may include a pressurized

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*$MgAl_2O_4$ and MgO have cubic crystal structures, while Al_2O_3 has a hexagonal structure.

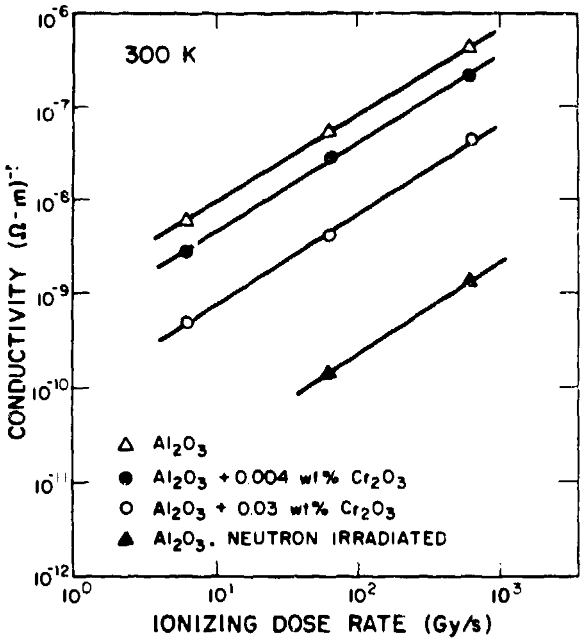

Fig. 1 - Radiation-induced conductivity as a function of rate of absorption of ionizing energy for three forms of Al_2O_3 (taken from the work of Klaffky et al.) (12, 13).

dielectric gas such as SF_6. These windows will absorb energy from the beam, and will be actively cooled to near room temperature by a fluid passed between two laminar plates. Thermal stresses will be high, and since the consequences of structural failure are severe (e.g., contamination of the plasma chamber with coolant), structural reliability is a prime requisite. With respect to anticipated radiation environment, it is not clear at this time whether windows can be located well away from the first wall (perhaps behind shielding) or whether waveguide transmission problems will dictate placement at or near the first wall.

CANDIDATE MATERIALS - Materials requirements for this application include low loss tangent* (to minimize absorption of microwave energy), high strength, and high thermal conductivity. If the windows are remote or heavily shielded, lifetime radiation dose may be no higher than 10^{22} n/m^2. In that case, only loss tangent is likely to be of concern. Fowler (15) has measured the effect of low neutron doses on loss tangent over the range ~10^4 -10^6 Hz for single-crystal Al_2O_3, a candidate material for this application. An increase by a factor of ten was observed at 2 x 10^4 Hz after a dose of 10^{22} n/m^2, while no change was detected at 2 x 10^6 Hz. More data are needed to determine the effect of irradiation at higher frequencies, where energy absorption is greater at a given value of loss tangent (16).

At slightly higher neutron doses, degradation of thermal conductivity begins to be significant. BeO is a promising window material because of its good thermal conductivity. However, this parameter is reduced by a factor of two after irradiation to 4 x 10^{23} n/m^2 (17). The extent to which window stresses in BeO are increased by increases in loss tangent and thermal conductivity at 10^{11} Hz have been calculated by Fowler using finite element analysis techniques, and are shown in Table 1: clearly, a significant increase in either parameter could threaten window survival.

The insulating ceramics spinel and silicon nitride are also considered candidate materials for this application. $MgAl_2O_4$ is known to exhibit a propensity for recombination of its irradiation-induced defects, at least at high temperatures (19), and might therefore show less degradation than does Al_2O_3. Si_3N_4 is considered a candidate because of its excellent strength and thermal shock resistance, properties of great importance for window applications. Silicon carbide, also a strong, thermal shock-resistant ceramic, is a semiconductor; nevertheless, this material is under consideration for low-frequency applications where heating problems are less severe. Fowler (20) has calculated that at 10^8 Hz (an ion cyclotron resonant heating frequency) the maximum tolerable loss tangent is ~0.1. However, measurements showed that loss tangent for a chemically vapor-deposited (CVD) form of SiC was significantly greater than that value. It is possible that other forms of this material would exhibit better performance.

If it is necessary to locate RF windows near the first wall, neutron doses will be severe (~10^{27} n/m^2), and it is not clear whether any ceramic can function satisfactorily under these conditions, at least at high frequencies. Direct exposure to the plasma will further worsen the situation, since thermal loading from the plasma will additionally reach the window. It is imperative that designers and materials researchers work together to optimize both window performance and location.

CERAMICS FOR STRUCTURAL APPLICATIONS

REQUIREMENTS - Ceramics have been proposed for applications such as first wall and blanket components, armor, limiters, and heat sinks. These materials can offer a number of advantages over metals for structural uses:

- low nuclear activation, which can reduce service and disposal problems
- low cost of starting materials

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*Absorptivity is a function of both loss tangent and dielectric constant, but the latter property does not vary greatly with material or irradiation dose.

Table 1 - Percent Increases in Maximum Tensile Stress in a BeO Window Over the Starting Value at 10^{11} Hz, as a Result of the Indicated Changes in Loss Tangent (18) and Thermal Conductivity (15).

Loss Tangent	Thermal Conductivity, W/cm K	% Change in stress
10^{-4}	3	-
10^{-4}	1.5	+50
10^{-3}	3	+900

- low atomic number of constituents, desirable for first-wall applications to reduce plasma contamination effects
- high operating temperatures, useful for first-wall and other applications involving high thermal loads.

Properties of importance here are strength, thermal conductivity, and dimensional stability, all over a wide range of temperatures.

CANDIDATE MATERIALS - Silicon carbide has been extensively studied for structural applications in nuclear systems, and in the high-purity, cubic β-phase CVD form has been shown to exhibit good strength retention after irradiation to 2 x 10^{26} n/m^2 at 1013 K (21). Swelling below $\sim$1300 K appears to reach a temperature-dependent saturation value ranging between 3 % (at 300 K) and zero (at 1300 K) (22,23), but above that temperature enters a void swelling regime (24) where high doses could lead to extensive swelling. Thermal conductivity is severely degraded by neutron irradiation, even at relatively low doses (25), so that use in high thermal flux applications is likely to be somewhat compromised. Silicon nitride has a non-cubic (trigonal) crystal structure, suggesting that stresses from differential swelling could limit high-dose performance. Nevertheless, strength is little degraded after irradiation to 2 x 10^{26} n/m^2 at 680 and 815 K (23). This good behavior may be attributable either to the low observed swelling ($\sim$1%), or to low swelling anisotropy.

Radiation-induced changes in thermal diffusivity (approximately proportional to thermal conductivity) for a number of ceramics, including Si_3N_4, are shown in Fig. 2. Certain general observations can be drawn from these results:

- lower irradiation temperatures cause greater degradation
- a saturation effect is evident above $\sim$1 x 10^{26} n/m^2
- ceramics can vary greatly in their response to damage.

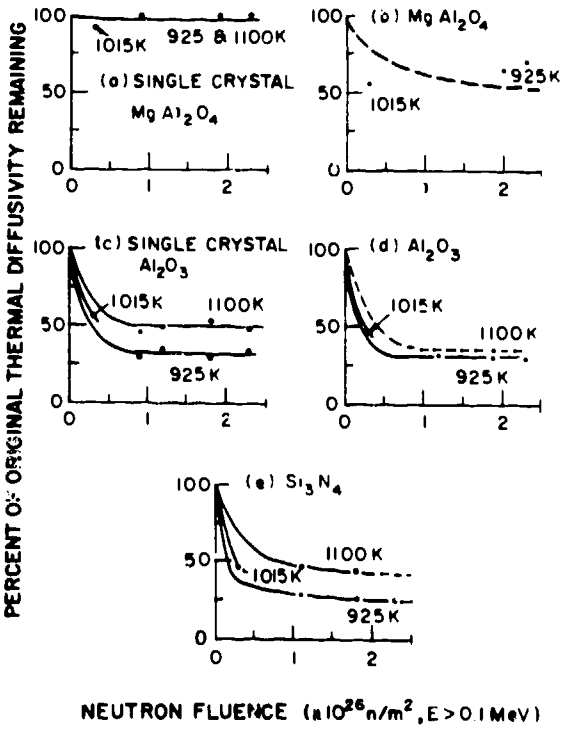

Fig. 2 - Reduction of room-temperature thermal diffusivity in five ceramics after neutron irradiation at the indicated temperatures (26).

These observations are consistent with a model involving scattering of phonons by radiation-induced lattice defects (26). With respect to Si_3N_4, its performance in a high thermal flux environment, like that of SiC, will be reduced by degradation of thermal conductivity.

Spinel ($MgAl_2O_4$) is not intrinsically a high strength, high thermal conductivity ceramic, and is therefore not a prime candidate material for structural applications. However, its good behavior under irradiation is noteworthy, and could result in its use where radiation conditions are particularly severe. Volume and strength changes for three forms of this ceramic after irradiation to 2 x 10^{26} n/m^2 at 680 and 815 K are shown in Table 2. It may be seen that volume changes are minimal, and that strength of this cubic material is actually increased by neutron damage. The low swelling is explained by the observation that the vast majority of displaced atoms recombine rather than form aggregates or remain isolated after high-temperature irradiation (19,23). Strength increases are attributable to the suppression of crack nucleation or propagation by the damage microstructure (23).*

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*Crack propagation in irradiated Al_2O_3 is similarly impeded, leading to increased fracture toughness (27).

Table 2 - Swelling and Strength Changes in Three Forms of $MgAl_2O_4$ after High-Dose Neutron Irradiation at Elevated Temperatures (23).

Material	Temperature, K	Volume Swelling, % *	Strength Change, %
Single-Crystal $MgAl_2O_4$	680	0.05	+92
	815	-0.11	+75
Polycrystalline $MgAl_2O_4$	680	-0.19	+38
	815	-0.35	+34
Polycrystalline $MgAl_2O_4$	680	-0.39	+39
	815	-0.31	+22

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* The negative sign denotes densification.

It has been suggested that in some cases properties of structural ceramics could be improved by employing laminated materials systems. However, these are subject to differential swelling, which can lead to delamination, cracking, and exposure of the substrate. Deleterious consequences of differential swelling have been demonstrated in a study of a SiC-graphite laminate irradiated to 2×10^{26} n/m^2 at 680 K, where the former material swelled 1.5 % and the latter densified ~7 %: the result was almost-complete separation (23). It appears that monolithic materials will be required for most high-dose applications.

OPTIMIZATION OF CANDIDATE CERAMICS

There are two broad approaches to the optimization of ceramic components for fusion applications:

- o modifications in chemistry and microstructure
- o application of brittle material design and testing techniques.

The extent to which these approaches are appropriate depends on the specific application, but both should be considered as part of any materials development effort.

CHEMICAL AND MICROSTRUCTURAL MODIFICATIONS- In most cases this approach to optimization will be invoked to enhance material performance and lifetime in a radiation environment. Chemical modifications might include exclusion of elements (or isotopes) likely to transmute to form gaseous species. As an example, it can be noted from the data presented in Table 2 that polycrystalline $MgAl_2O_4$ did not exhibit the same increase in strength upon irradiation as did single-crystal material: The reason may be that both polycrystalline forms contained Li, which transmutes to form He (23). It may be possible to increase the extent of strength enhancement in polycrystalline $MgAl_2O_4$ by control of this impurity. Other areas where chemical modifications could be helpful include introduction of electronic trapping or recombination centers to improve electrical properties (Fig. 1), and removal of impurities to enhance transmissivity of RF window materials.

Microstructural alterations offer potential benefits such as reducing swelling or its deleterious consequences, retaining or enhancing strength, and minimizing degradation of thermal conductivity. Fine dispersants could be added to serve as recombination centers for structural defects, resulting in reduced swelling and improved retention of initial thermal conductivity. Small grain size, which can be attained by a number of fabrication techniques, would allow non-cubic ceramics such as Al_2O_3 to retain usable strength to higher fluences (28). Composite strengthening, a technique increasingly used for applications not involving radiation, offers benefits for fusion applications if the microstructure is sufficiently fine to avoid problems with differential swelling.

One area where chemical and microstructural modifications offer great potential for improved performance is elimination of damage sensitive grain boundary phases. Many ceramics, including those specified for high stress, high temperature applications, contain additives to enhance densification

Fig. 3. Bright-field transmission electron microscope image of damaged grain boundary phase in Si_3N_4 after irradiation to 2×10^{26} n/m^2 at 1100 K (32) (bar = 0.4 μm).

during fabrication. As a result impurity phases may appear at grain boundaries, and these often exhibit poor radiation resistance. Figure 3 shows such a phase, partly glassy, that has obviously suffered significantly more damage than has the Si_3N_4 matrix. Grain boundary damage, rather than anisotropic swelling, may be responsible for the slight strength loss observed in this ceramic (23). It is apparent that in many cases new or improved fabrication techniques should be employed to produce materials optimized for fusion applications.

BRITTLE MATERIAL DESIGN AND TESTING TECHNIQUES - The major drawback to the use of ceramics for structural applications is their characteristic brittleness. Traditional design approaches involve loading ceramics in compression, but this is often not possible. In order to obtain maximum service strength and reliability from ceramics, an accurate data base must be established for the intended operating conditions and these results incorporated into brittle material design analyses.

The realization that ceramics fail as a result of propagation of flaws whose sizes have a statistical distribution and whose propagation characteristics are environment-dependent, has led to techniques for establishing mechanical reliability of components. This approach has been used (29) to predict the behavior of microwave windows in the absence of irradiation. Materials tests include:

- o strength measurements, including effect of surface finish, test environment, and rate of stress application
- o static fatigue (time-to-failure at constant load) tests,

followed by construction of a fatigue reliability/probability of failure diagram.

Proof testing can effectively by used to eliminate sub-standard components and thereby enhance reliability. Price and Hopkins (21) combined proof testing and neutron irradiation testing of SiC to show that the benefits of proof testing are retained after high neutron exposures for CVD and sintered SiC, but not for reaction-bonded material. These results were interpreted in terms of the effect of irradiation on flaw size and distribution. Materials tests such as these must be combined with finite element design analyses and sophisticated design methodologies (30) to develop an optimized product.

RECOMMENDATIONS FOR FUTURE WORK

This review has directly or indirectly identified a number of areas where further work is needed. Many of these can be assembled into component-specific categories, whereas others are more general in nature.

MAGNETIC COIL INSULATORS - The most urgent needs here are for more results on radiation-induced conductivity, and on the effect of near-room-temperature, high-dose neutron irradiation on structural properties.

WINDOWS FOR RF HEATING SYSTEMS - Data are especially needed on radiation-induced loss tangent degradation, especially at high frequences. Although preliminary calculations (15) suggest that measurements concurrent with radiation may not be needed, this question needs further attention.

CERAMICS FOR STRUCTURAL APPLICATIONS - The present data base on temperature and fluence-dependence of changes in density, strength, and thermal conductivity is incomplete, and an extensive radiation testing program is needed to fill the gaps. Strength tests to date have for the most part been conducted rapidly, under simple stress conditions; more realistic tests should be incorporated into the program.

GENERAL NEEDS - Until intense sources of fusion neutrons become available, it will be necessary to make use of fission reactors for high-dose irradiation tests. Correlation of damage events can be established by appropriate calculations, and by comparison of low-dose damage effects such as swelling utilizing present-day 14 MeV neutron sources. The effect of transmutation gases can be simulated by incorporating into test materials selected isotopes transmutable by thermal neutrons, and irradiating in a mixed-spectrum fission reactor. In this way both displacement events and gases of interest

Table 3 - Displacements per Atom (dpa) and He and H Concentrations Attainable in $Si_3Al_3O_3N_5$ by ^{17}O and ^{14}N Doping and Irradiation to 5×10^{25} fast n/m^2 plus 8×10^{25} thermal n/m^2 (One Year in ORR).

Isotopic Percent Dopant	Damage Level, dpa	Gas Content, appm
99.5 ^{15}N 0 ^{17}O	6	-
7.1 ^{14}N, 0 ^{17}O	6	400 H
99.5 ^{15}N 50 ^{17}O	6	270 He
7.1 ^{14}N 50 ^{17}O	6	400 H, 270 He

can be simulated. A plan has been proposed for such a study, utilizing controlled amounts of ^{14}N and ^{17}O additives to $Si_3Al_3O_3N_5$ (31). The thermal neutron reactions to be exploited are $^{17}O(n, \alpha)$ ^{14}C and $^{14}N(n,p)$ ^{14}C. Table 3 describes the four damage states that can be attained, using the Oak Ridge Reactor. The dpa/gas ratios attained are roughly those expected at the first wall of a fusion reactor.

Finally, as was described in the previous section, materials optimization by control of chemical and microstructural makeup, as well as utilization of brittle materials design and testing techniques, should be an integral part of any effort to develop advanced ceramics for fusion applications.

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