Materials Problems for Highly Irradiated ICRH Launchers in Fusion Reactors

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Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

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MATERIALS PROBLEMS FOR HIGHLY IRRADIATED ICRH LAUNCHERS IN FUSION REACTORS

LAILA A. EL-GUEBALY, Fusion Technology Institute
1500 Johnson Drive, University of Wisconsin
Madison, Wisconsin 53706
(608) 263-1623

ABSTRACT

The recent advances in radiofrequency (RF) heating have led to the use of substantial amounts of RF power for startup and heating. The RF heating system requires a number of ceramics for coaxial feedthrough insulation, antenna radomes, windows, and ceramic-filled waveguides. These dielectric materials should maintain electrical and structural integrity in a severe radiation and thermal environment. It is becoming more evident that the ceramic materials could be the weak point in the performance of the RF system, unless they are properly chosen.

INTRODUCTION

The intent of this paper is to review the performance of ceramics in a fusion reactor operating environment, evaluate mechanisms of radiation damage, summarize briefly the work which was done with emphasis on recent developments, establish limits for the use of the ceramics in fusion reactors, and propose a number of ceramics for the ICRH systems.

The RF launchers are located close enough to the plasma and required to operate in a first wall-like neutron flux level, i.e. fluxes of ~ 3 x 10^{14} n/cm^2-s and 1.5 x 10^{14} γ/cm^2-s, a fast neutron fluence (E_n > 0.1 MeV) of ~ 6 x 10^{21} n/cm^2 per full power year (FPY), and an ionizing dose rate of 10^5-10^6 rad/s for a unit neutron wall loading.[1] Current power reactors are designed for 3-10 MW/m^2 and operate for 20-30 FPY's. Moderate electric fields (< 1 kV/mm) are expected and the operating temperature is likely to be 100-300°C. It is of importance to study the performance of ceramics in such fusion reactor-relevant environments and to assess the combined effects of radiation damage, pressure and temperature gradients with accompanying stresses on the properties for which they are selected.

Candidate ceramics for the ICRH launcher are alumina, beryllia, magnesia, spinel, silicon carbide, silicon nitride, silicon oxide, MACOR, titania, and yttria. Some of the basic physical properties of these ceramics are given in Table I. Required electrical properties for ICRH systems include low loss of transmitted RF power in the frequency range of 10-300 MHz, high dielectric constant, low electrical conductivity, and high dielectric strength. Additional requirements include adequate thermal conductivity, high radiation resistance, mechanical integrity, thermal stress-strain cycling resistance, and system integrability. As will be seen later, even a moderate radiation environment can cause significant changes in the data presented in Table I. Since most ceramics have adequate electrical properties for ICRH systems, it seems that irradiation tests are likely to eliminate some candidate materials from the above list. On the other hand, further studies may add high performance, radiation resistant materials for fusion reactor applications.

Yet, the radiation effects on ceramics have not been widely studied over the wide range of fluence and temperature. Some excellent data have been gathered by a few investigators.[2-4] However, much of the work has been confined to relatively low irradiation levels, basic classes of ceramic materials (magnesia, alumina, beryllia), simulation irradiation techniques (electron and fission neutron spectra), a specific irradiation temperature range, particular crystal structure, and/or tests of certain responses under limited irradiation conditions. In the following, the effects of radiation on the electrical properties, thermal conductivity, and structural properties are discussed. Due to space limitations here only the last property will be discussed in detail in that the mechanical rather than the electrical degradation appears to be lifetime limiting for most ceramics and the reader should refer to the main report[5] for a detailed discussion of all properties.

ELECTRICAL PROPERTIES

A. Loss Tangent

Power losses in any material depend mainly on the frequency of the wave, the loss tangent (tan δ) and the dielectric constant of the material. The RF heating is most severe in the higher GHz frequency range of the LHRH and

Table I. Characteristics of Ceramics‡

Material	Type of Conductor	DC Electrical Resistivity* (Ω-cm)	Dielectric Constant*	Dielectric Strength (kV/mm)	Loss Tangent (at 100 MHz)	Thermal Conductivity (cal/cm-s°C) at 100°C	Thermal Stress Resistance
Al_2O_3	Non-cond.	5×10^{13}	8.8	40-160	0.0003	0.069	Very Good
BeO	Non-cond.	10^{16}	6.6	10		0.5	Excellent
MgO	Non-cond.	10^{15}	9.65		< 0.0003	0.082	Fair-Poor
$MgO \cdot Al_2O_3$	Non-cond.	$> 10^{14}$				0.033	Fair
SiC	Semicond.	10^5				0.133	Excellent
Si_3N_4	Non-cond.	10^{13}	9.4			0.045*	Excellent
SiO_2	Non-cond.	10^{15}	3.8	0.35	0.0002	0.0033	Excellent
MACOR	Non-cond.	$> 10^{14}$	6.0	1	0.003+	0.0031	Excellent
TiO_2	Semicond.	10^{11}	100	100-210	0.00025	0.015	Excellent
Y_2O_3	Non-cond.					0.02	Fair-Poor

‡ See Ref. 5 for references. * At room temperature. + At 0.1 MHz.

ECRH systems. Low loss tangent materials are required for the RF systems to minimize the power losses. Insulators (nonconductors) tend to have low loss tangents (10^{-4}-10^{-3}) in the ICRH frequency range and low AC electrical conductivity. On this basis, semiconductors must be excluded from the candidate material list for ICRH heating systems to avoid excessive heating losses in the ceramics.

The small amount of data available demonstrates that the loss tangent is highly degraded by fission neutron irradiation,[6] due to the marked increase in the AC electrical conductivity, but the degradation still remains unknown in the MHz frequency range of interest. Such changes in tan δ could create serious effects in RF ceramics as thermal gradients for the increased loss tangent exceed those of the unirradiated values by about an order of magnitude and that raise the operating temperature and thus consequently accelerate the structural failure due to thermal stresses.

B. Electrical Conductivity

In addition to the conductivity increase by thermal effects, irradiation degrades the electrical resistivity of the materials by altering the electronic properties and inducing chemical and structural defects. At low temperatures, the change in DC conductivity goes as the ionizing dose·rate.[4] For instance, a typical first wall dose is 10^4 Gy/s for a unit wall loading reactor. This results in a conductivity change of 10^{-4}-10^{-9} $(\Omega\ m)^{-1}$ which is comparable to that induced by thermal effects at ~ 1000°C. Work to date[7-9] has identified up to three orders of magnitude increase in the <u>DC</u> electrical conductivity of ceramics exposed to moderate ionizing doses. No data are available on the radiation-induced <u>AC</u> conductivity of ceramics at elevated doses in the MHz frequency range of interest for the ICRH heating systems.

C. Electrolysis

Electrolysis can destroy the ceramics particularly at elevated temperatures and electric fields. Since diffusivity is enhanced by irradiation, it is anticipated that intense flux will accelerate electrolysis and limit insulator lifetimes. Therefore, ceramics must be avoided in high temperature and electric field applications where electrolysis might be a problem. In addition, the insulator temperature should be kept low during operation, by active cooling if necessary, to avoid thermal stress-induced failure.

D. Dielectric Breakdown

Dielectric breakdown of ceramics often occurs across the surface rather than through the bulk of the material. Insulators do not possess a unique dielectric strength. There is a strong relation between the design and the material performance of ceramics in RF systems. Size, geometry, surface preparation and temperature environment are as important as material properties in determining the dielectric breakdown strength of the ceramics.[2] Coatings or glossy insulators are often used in the RF launchers to increase the dielectric breakdown strength by offering low secondary electron yield, controlled surface resistivity, and/or improved surface cleanliness. Also, the gaps are filled with inert gases (such as SF_6, argon, and freon) to further enhance the voltage standoff characteristics of the system.

THERMAL CONDUCTIVITY

The importance of the thermal conductivity of the insulators is that high values result in lower operating temperatures and thermal stresses. Thermal conductivities of some ceramics are listed in Table I and these values are significantly reduced at higher temperatures. Due to the superior thermal conductivity of BeO (room temperature value is ~ an order of magnitude higher than most oxides and ~ equal to that of Al metal), it is usually selected as a candidate material for RF systems. The value of this feature is diminished in a neutron environment as will be shown below.

Thermal conductivity is significantly reduced by irradiation induced damage, as defects scatter the phonons by which heat is conducted in insulators. The magnitude of the reduction varies greatly with the ceramic and depends on measurement temperature and irradiation dose. However, all tested specimens showed the same general features:

- a decrease of thermal conductivity with temperature and fluence.
- a more pronounced reduction at lower irradiation temperatures.
- a tendency to saturate at the higher irradiation fluences.

Hurley and Clinard[10] measured the thermal diffusivity, which is proportional to the thermal conductivity, of fourteen oxides and nitrides after irradiation to fission fluences as high as 2.3 x 10^{22} n/cm^2 (E_n > 0.1 MeV) at 925, 1015 and 1100 K. They found decreases of ~ 45, 50, 70, 75 and 95% in polycrystalline $MgO \cdot Al_2O_3$, Y_2O_3, Al_2O_3, Si_3N_4, and BeO, respectively, at 925 K and 2.3 x 10^{22} n/cm^2. Single crystal spinel showed the least reduction (< 1%) at every temperature and fluence, and beryllia had the greatest reduction (factor of ~ 20). Accordingly, ceramics in RF systems of fusion reactors exposed to the more damaging 14 MeV neutrons will encounter drastic reduction in the thermal conductivity. However, it should be pointed out that radiation-induced effects anneal out in the high temperature range (900-1100°C).

STRUCTURAL PROPERTIES

Swelling with accompanying stresses and dimensional changes is the major structural problem in insulators. There have been few studies of the more complicated irradiation-induced swelling in ceramics as compared with metals. The structural aspects and mechanism of damage have been discussed in detail elsewhere; a brief summary is given here. Specimens were irradiated to neutron fluences up to ~ 10^{22} n/cm^2 at a variety of temperatures (25-1000°C) and subsequent examinations included volume expansion measurements and structural change observations. In general, the degree of swelling increases with fluence and peaks in the temperature range 0.2 to 0.4 of the melting temperature of ceramics; the non-cubic structure of some ceramics reduces the tolerance for radiation damage because of anisotropic swelling.

Irradiation-induced swelling of some ceramics and the experimental conditions are summarized in Table II and some measured results are shown in Figs. 1-3. Only scattered data points are available for Si_3N_4, MACOR, Y_2O_3, and SiO_2. The non-cubic polycrystalline ceramics swell anisotropically and suffer drastic weakening at fluences > 10^{20} n/cm^2 even at low swelling levels. The data presented in Table II for non-cubic samples include the volume expansion of some which were definitely fractured, but on which dimensional measurements could be obtained.

Alumina has been irradiated over a wide range of fluences and temperatures. It is the most fully-developed ceramic and has a fairly complete data base. The single crystal form shows more resistance to structural changes than polycrystals. At high fission neutron fluences > 10^{22} n/cm^2 (E_n > 0.1 MeV), it exhibits several percent swelling, with anisotropic growth of the non-cubic material leading to microcracking.[3] Swelling occurs to a lesser degree near room temperature, and is more isotropic.

Beryllia showed large swelling even at moderate fluences. The primary mode of damage are the grain-boundary cracks, which are caused by anisotropic crystal expansion. Hickman and Pryor[13] suggested 4 vol.% swelling as the onset of the microcracking (Fig. 3). This corresponds to a fast fission neutron fluence of ~ 10^{21} n/cm^2 at the temperature range of interest for the ICRH applications (100-300°C).

The grain boundary separation does not account for all of the observed volume increase in BeO. Helium bubbles, lattice expansion, and defect clusters account for the remainder of the expansion. Appreciable quantities of He are formed in BeO material used in fusion reactors (14 MeV neutron source), since Be has a high (n,2n) reaction cross section above 2 MeV and this reaction is immediately followed by a disintegration process in which two alpha particles are emitted. This contributes significantly to the expansion of BeO and promotes microcracking by reducing the contact area between grains. On this basis, the 4 vol.% swelling onset for microcracking could be induced by a fast fusion fluence of ~ 10^{20} n/cm^2. This undoubtedly limits the usefulness of this ceramic in high fluence applications.

Magnesia is a well-developed ceramic with reasonably low irradiation-induced swelling. It exhibits good dimensional stability (3 vol.% swelling) to high fission neutron fluences (2 x 10^{22} n/cm^2; E_n > 0.1 MeV). It is a candidate for those applications where Al_2O_3 is appropriate, whereas for high irradiation environments MgO is preferable. MgO is the most popular form

Table II. Parameters and Measured Swelling in Ceramics Irradiated with Fast Fission Neutrons (E$_n$ > 0.1 MeV)

Material[a]	Form[b]	Temp. (K)	Fluence 10^{22}n/cm^2	Swelling[c] (vol %)
Al_2O_3 (NC)		425	0.1[d]	1.25[g]
	SC	430	0.03[e]	---
	SC	680	2	3.5
	SC	815	2	3.3
	SC	925	2.3	4.1[h]
	SC	1015	0.28	1.6
	SC	1015	0.3	1.7
	SC	1100	2.3	4.4[h]
		650	0.56	2.2
	PC	925	1.2	3
	PC	925	1.9	3.5
	PC	925	2.3	3.5
	PC	1015	0.28	1.9
	PC	1015	0.3	1.9
	PC	1100	1.2	6.0
	PC	1100	1.9	6.5
	PC	1100	2.3	6.5
BeO (NC)		375	0.07[d]	4[i]
		385	0.1[d]	5.8[g]
	PC	1015	0.28	3.3
MgO (C)		300	0.05[d]	1
		425	0.1[d]	1[g]
	PC	430	2.1[e]	2.6-3
		1075	0.5[d]	< 1.8
		1375	0.48[d]	< 2.1
$MgO \cdot Al_2O_3$ (C)	SC	680	2	0.05
	SC	815	2	-0.11
	SC	925	0.8	---[h]
	SC	1015	0.28	0.1
	SC	1100	2.3	---
	PC	430	2.1[e]	0.8
	PC	680	2	-0.2
	PC	815	2	-0.35
	PC	925	2.3	0.2
	PC	1015	0.28	0.3
	PC	1015	~ 0.3	0.4
	PC	1100	2.3	1.6
Si_3N_4 (NC)	PC	680	2	~ 1.1
	PC	815	2	~ 1.0
		1015	0.28	0.4
SiO_2 (NC)		675	2.4	-1.4
		825	2.5	-1.1
MACOR (NC)		825	2.7	1.1
		300	10^{-4} [f]	---
Y_2O_3 (C)	PC	650	0.6	0.2
	PC	1015	0.28	0.1

[a] NC = non-cubic, C = cubic crystal structure

[b] SC = single crystal, PC = polycrystal

[c] Negative means densification

[d] (E$_n$ > 1 MeV)

[e] E$_n$ > 0.2 MeV and 4.6 x 10^{22} thermal n/cm^2

[f] E$_n$ = 14 MeV

[g] Also see Fig. 1

[h] Also see Fig. 2

[i] Also see Fig. 3

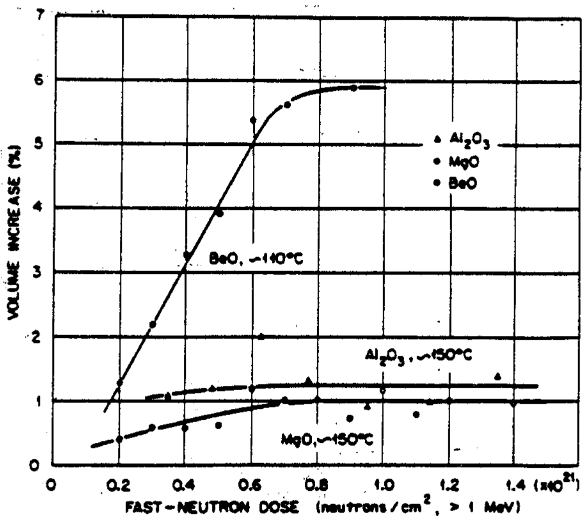

Fig. 1. Volume increases of sintered BeO, MgO, and Al_2O_3 irradiated with fast fission neutrons (Ref. 11).

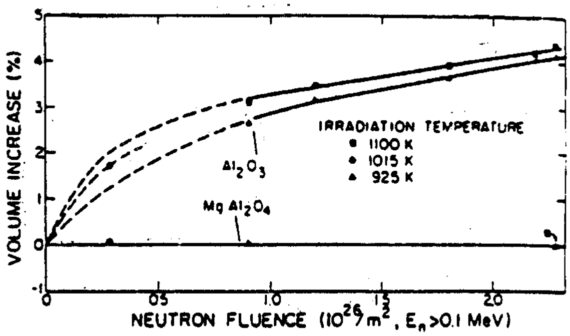

Fig. 2. Swelling in single crystals Al_2O_3 and spinel as a function of fission neutron fluence for three irradiation temperatures (Ref. 12).

of powdered insulator being used in high irradiation fields. MgO powder has been used successfully in highly irradiated normal magnets.[14] Available evidence suggests that there are probably no life-limiting mechanisms associated with neutron damage for the powdered insulators with the possible exception of degradation of the electrical properties.[14]

Spinel is the most desirable material for high irradiation applications. A good deal of research has been carried out on spinel, much of it in the area of structural changes under irradiation. It has a remarkably good radiation resistance to swelling, in particular the single

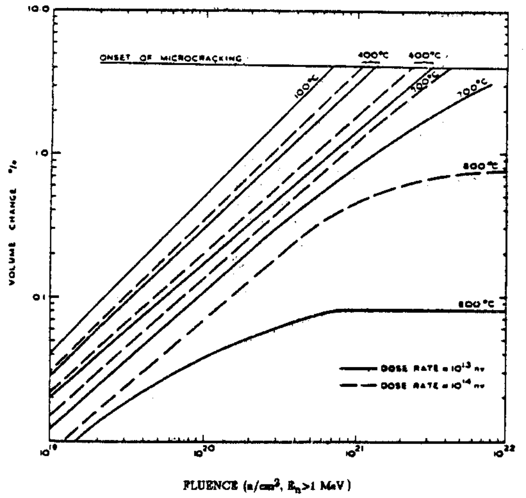

Fig. 3. Predicted volume expansion as a function of fission neutron fluence and irradiation temperature (Ref. 13).

crystal form which exhibits essentially no change in volume after irradiation at elevated temperatures to 2 x 10^{22} n/cm² (E_n > 0.1 MeV) while polycrystalline spinel shows slight swelling (0.8 vol.%) at 430 K.

Other ceramics such as MACOR, Si_3N_4, and SiO_2 are still under consideration for irradiation tests. However, they are less radiation resistant to swelling because of their non-cubic structure which leads to anisotropic expansion and, eventually, structural failure. Hence, many silicates are subject to radiolysis even at low radiation levels and should be placed in well-shielded locations. MACOR can easily be machined into intricate shapes, a very useful property for ceramics. It has been irradiated[9] to 10^{18} 14 MeV n/cm² at room temperature with no evidence of structural changes. It is estimated that a factor of five higher neutron fluence, to account for the degraded fusion spectrum (E_n > 0.1 MeV), is the limit for using the MACOR in fusion devices without any structural problems. The irradiated samples of yttria were notable in displaying insignificant swelling due to their cubic structure and more high-flux tests are required. Titania had not, to our knowledge, been irradiated previously and its behavior under irradiation is still unknown; even though there has recently been more interest in using it to fill the ICRH waveguides because of the notably high dielectric constant.

Directly following from the above, the global conclusions are:

- Single crystals are more radiation resistant than polycrystalline forms of most ceramics.
- The cubic structure of ceramics precludes anisotropic expansion.
- Non-cubic ceramics are subject to internal cracking even at low swelling levels.
- At elevated fluences, Al_2O_3 and BeO swelled the most while spinel offered the lowest degree of swelling among its class of cubic ceramics.
- In most cases, the swelling appears to rise proportionately to the fluence. Hence, the swelling can be extrapolated to lower or higher fluences, if no experimental data are available.

Since no tests are presently conducted in a degraded fusion spectrum, the question of the differences between fast fission and fusion neutron damage remains unanswered. Ultimately, fusion neutron irradiation data will be obtained from tests in the planned fusion material test facilities. At this time, it is assumed that fast fusion neutrons (E_n > 0.1 MeV) produce roughly twice the damage from fast fission neutrons.[14] If this holds true at all fluence levels, then the fluence limits for the use of the polycrystalline spinel, magnesia, alumina and beryllia in fusion reactors are 4 x 10^{22}, 10^{22}, 5 x 10^{20}, and 5 x 10^{18} n/cm² (E_n > 0.1 MeV), respectively, in the temperature range 100-300°C. These are based on 3 vol.% radiation-induced swelling (that can be accommodated without causing stress problems) in spinel and MgO, and one tenth (factor of safety) of the fluence for observed microcracking in the Al_2O_3 and BeO.

RADIATION EFFECTS ON WATER

The ceramics-filled waveguides will most probably be water-cooled in order to remove the nuclear heating as well as the heating from RF energy absorption. The losses in the water are undoubtedly larger than those in the ceramic. This mandates the use of high coolant velocity and pressure which generate more problems by creating pressure gradients across the ceramic. A pure water of high resistivity is recommended to avoid excessive RF losses. An attractive property of water is that it possesses a relatively high dielectric constant (~ 80 at room temperature) compared to ceramics. This led recently to the feasibility study of water-filled waveguides.

Water, used to fill or cool the waveguide systems, is subject to radiolytic dissociation under irradiation.[14] The major problem stems from the fact that the presence of radiolytic products in water, particularly hydrogen peroxide (H_2O_2), accelerates the erosion of the container. As a result, the container ionic species increase the conductivity of the water and, consequently, higher RF losses are anticipated. In addition, the natural contact between the container and the water causes chemical corrosion and forms an oxide layer at the surface of the container. The principal oxidizing agent for corrosion is the oxygen which is formed by the decomposition of hydrogen peroxide and some other reactions. Also, oxygen can enter the cooling circuit through whatever air is absorbed by surfaces in the system and without some sort of degassing apparatus, the corrosion rate of initially gas-free pure water will rise rapidly. If the corrosion is too severe, a stainless steel cladding, as thin as 5 mils, might be used. Another potential problem is the water activation through the water neutron-induced transmutations (^{16}N and ^{17}N formed from ^{16}O and ^{17}O in H_2O, respectively) and the presence of the longer lived radioactive corrosion products. The transport of these products through the cooling system is probably the greatest environmental hazard of the highly irradiated water-cooled/filled waveguide systems.

CONCLUSIONS

The problems associated with the use of ceramics in radiation environments are now well-recognized. The structural degradation appears to be lifetime limiting for most ceramics. Therefore, the RF system must be designed to permit periodic replacement of the ceramic components, especially those adjacent to the first wall where the radiation level is high. Spinel, magnesia, and alumina are the leading candidate ceramics for fusion applications from the structural standpoint and could be used in RF launchers up to their limits of radiation stability. Spinel is of particular interest because of the superior absence of swelling, particularly in the single crystal form. In contrast with Al_2O_3 and MgO, hardly any degradation was observed in the structural properties. Unfortunately, no data are available on the irradiation effects on the electrical properties of spinel.

Several problems have been cited in the water-cooled or filled waveguide system. Among these are radiolysis, transmutations, and decomposition of water under irradiation, and consequently the buildup of erosion/corrosion products. Initially gas-free relatively pure water with high resistivity is recommended to reduce the RF power losses.

Fusion reactor ceramics are simultaneously subjected to a variety of radiation damage processes. Further studies are required to determine the concurrent effect of ionizing and displacive radiation on candidate materials under fusion reactor-relevant environments (high fluence, degraded fusion spectrum, high ionizing dose, moderate temperature, stress conditions, and MHz frequency range). The effect the structural damage has on the electrical properties of ceramics needs also to be evaluated. Some economical methods should be developed for continuous growth of single crystal ceramics as they offer advantages over polycrystals.

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