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Effect of Neutron Radiation on the Dielectric, Mechanical and Thermal Properties of Ceramics for RF Transmission Windows

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Abstract

The behavior of electrically insulating ceramics was investigated before and after exposure to neutron radiation. Mechanical, thermal and dielectric specimens were studied after exposure to a fast neutron dose of 0.1 displacements per atom (dpa) at Oak Ridge National Laboratory (ORNL). Four materials were compared to alumina: polycrystalline spinel, aluminum nitride, sialon and silicon nitride. Mechanical bend tests were performed before and after irradiation. Thermal diffusivity was measured using a room temperature laser flash technique. Dielectric loss factor was measured at 105 MHz with a special high resolution resonance cavity. The materials exhibited a significant degradation of thermal diffusivity and an increase in dielectric loss tangent. The flexural strength and physical dimensions were not significantly affected by the 0.1 dpa level of neutron radiation. The aluminum nitride and S silicon nitride showed superior RF window performance over the sialon and the alumina. The results are compared to radiation studies on similar materials.

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DISCLAIMER

1. Introduction:

Monolithic ceramics have been used extensively as radio frequency (RF) feed-through standoff windows [1]. Environmental standoff, or transmission windows require fully dense ceramics with a very high thermal shock resistance, and a very low dielectric loss factor. Neutron radiation severely degrades both the dielectric and thermal performance. The critical requirement of ceramics is to maintain acceptable thermal shock resistance and dielectric loss factor after neutron irradiation.

This study addresses windows that will be located in a neutron flux zone that will typically experience lifetime neutron doses of less than one dpa. The dose of 0.1 dpa (one atom in ten is displaced by the neutron radiation) is a design limit that was set for the RF systems for the International Thermonuclear Experimental Reactor (ITER). In terms of radiation induced degradation of mechanical and physical properties these are very low doses. However, a significant decrease in thermal shock resistance due to neutron damage, at doses as low as 0.1 dpa, can cause severe failure conditions in the window. Neutron induced increases in dielectric losses cause higher rates of energy absorption leading to catastrophic failure. Decreased RF transparency causes an increase in operation temperature and severe thermal gradients. This effect couples with decreased thermal shock resistance to cause window failures.

Irradiation studies have been performed on similar materials to those in this study [2-9]. Differences in fabrication and sintering formulations can result in widely different material performance. Testing a variety of materials is therefore an advantage in developing and finding a successful material for RF feed-through windows.

Subject index codes: N01
R02
R03
R04

1.1. Thermal Shock Resistance

The thermal shock resistance figure of merit, R, can be represented by an expression containing relevant material properties as follows [10]:

$$R = k\sigma(1-\nu)/\alpha E$$

where, k, σ, ν, α, and E are thermal conductivity, flexural strength, Poisson's ratio, coefficient of thermal expansion, and Young's modulus, respectively.

The term most affected by neutrons is the thermal conductivity. It is therefore necessary to measure the reduction of thermal conductivity from neutron radiation effects. Neutrons can drastically lower thermal conductivity due to the scattering of phonons by neutron produced point defects (displaced ions) even at very low doses [11]. The thermal conductivity can be degraded very quickly to 50%, or less, of its original value. It is important to use materials with a high initial thermal conductivity as well as radiation resistance. Aluminum nitride (AlN) has an initial thermal conductivity of over 150 W/mK making it a good candidate for a window. Silicon nitride (Si_3N_4) and sialon have lower thermal conductivities than AlN, but they have much higher flexural strengths, thereby enhancing thermal shock resistance. This warrants testing their performance in a neutron environment. Materials with a high R value from high thermal conductivity, but low strength are more sensitive to low-dose neutron degradation of thermal shock resistance.

1.2. Dielectric Properties

The absorbance of energy in the window is governed by the dielectric loss factor, and the frequency of the RF energy passing through the window. The loss factor is the product of the dielectric constant (ε), and the loss tangent ($\tan\delta$) of the ceramic at the frequency of interest. In magnetic fusion reactors, typical RF heating system operation frequencies are from 50 MHz to 200 MHz. At these frequencies, the primary material response to the RF waves is orientation of dipoles and electronic distortion of atoms. The other contributions to loss are reflectance by defects in the material such as porosity, flaws and phase interfaces introduced during fabrication.

The dielectric constants of alumina and beryllia are fairly resistant to the effects of neutrons, however, the loss tangents are significantly increased [12]. Therefore, neutron radiation will generally decrease the transparency of most ceramics to RF energy due to changes in the loss tangent and not in the dielectric constant. Increases in the loss tangent of up to 200% have been previously observed [13, 14].

2. Experimental Details

2.1. Candidate Materials

Traditionally, single crystal alumina (sapphire) and polycrystalline alumina have been the materials of choice for RF power window applications. Several promising commercial polycrystalline ceramics are investigated in this study including: alumina, spinel, silicon nitride, and sialon. Extensive work was performed to develop new window materials, and to locate and evaluate the most promising ceramics from industry not yet studied. Initially a ceramic matrix composite (CMC) was also considered for this investigation. Fiber-reinforced CMCs were considered due to their ability for graceful failure which could prevent catastrophic system failure. However, fiber reinforced CMCs are hard to optimize for strength and dielectric performance, because it is difficult to obtain high strength and low dielectric loss in the same material. At the time of the radiation exposure tests, the CMC was not developed sufficiently to justify inclusion in the program.

Irradiations were performed in the high flux isotope reactor (HFIR) at Oak Ridge National Laboratory (ORNL). The specimens were irradiated using the hydraulic rabbit system in HT-3 position of the reactor core. The size of the rabbit required the use of scaled down specimens shown in Figure 1. Specimens were exposed at 150°C to a fast neutron fluence of 1.0×10^{24} n/m^2. Assuming the ceramics possess average sublattice displacement energies of 40 eV, a damage level of 0.1 dpa is produced by the fluence of 10^{24} n/m^2 in 36 hours.

2.2. Thermal Tests

Thermal conductivity was measured using the xenon thermal flash method. Two thicknesses of specimens were required to accurately determine the thermal conductivity, as the optional thickness of the specimen is dependent on the thermal conductivity. The thin, 1 mm, specimen, is required for materials with low thermal conductivity of less than 20 W/mK, and the thick specimen, 3 mm, for materials greater than 20 W/mK. To account for the unknown amount of degradation of the thermal conductivity from neutron irradiation, both thicknesses were irradiated. The Clark and Taylor method for calculating the thermal diffusivity based on the half-rise time of the back-surface infrared signal was used. For each specimen a thin layer of graphite was applied to both sides of the specimen to minimize sample shine-through. A series of samples were measured with varying graphite thickness to ensure the diffusivity was unchanged by this procedure.

2.3. Bend Tests

Flexural strength measurements were made using a four-point bend fixture with outer and inner spans of 19 mm and 9.5 mm. All specimens, with the exception of two which were rejected, failed within the inner span. The cross-head speed was 0.0085 mm/s. The statistical program SELECT was used to determine the maximum likelihood ratio for each set of data to determine whether the data was best described by a Log-Normal, Exponential, Weibull, Uniform, or Gamma distribution. As expected, and with the exception of one data set, the Weibull distribution was selected as the best representation of the data. Ten specimens of each material were tested, and the results were statistically analyzed using the Weibull distribution.

2.4. Dielectric Tests

Dielectric measurements were performed in a high resolution loss tangent measurement coaxial resonance cavity shown in Figure 2. This approach ensures very high accuracy in the values of dielectric loss tangent. The ceramic sample was part of a capacitor which terminated the center of the coaxial cavity. The cavity was designed to resonate at a nominal frequency of 120 MHz. The cavity was monitored with a quality factor circuit by using an HP 8753C network analyzer. The calculation methods are described elsewhere [14].

3. Results and Discussion:

As can be seen in Table 1, nitride based ceramics offer a significant improvement in thermal shock resistance over alumina and spinel. The flexural strength of these nitride ceramics is a significant contributor to high thermal shock resistance, which is an advantage since neutrons have a minor effect on strength at these relatively low doses. The first five materials in Table 1 were irradiated, and the results of initial and post-irradiation tests follow.

Post-irradiation dimensional measurements indicate only slight volumetric changes (<0.4%), as expected. Numerous studies, mentioned earlier, have been performed on alumina under neutron irradiation conditions that are comparable to the present work. These studies have reported volumetric swelling levels of 0.07 to 0.85% in alumina following irradiation to fluences of 0.5 to 5×10^{24} n/m^2 (E>0.1 MeV) at temperatures of

60-200°C, which agrees with the swelling of 0.15% obtained in this study for the HFIR-irradiated alumina specimen. The reported swelling for neutron-irradiated AlN ranges from 0.09 to 0.3% for fluences of 0.2 to 5×10^{24} n/m^2 (E>0.1 MeV) at temperatures of 80-300°C, with the highest amounts of swelling occurring at the lowest temperatures in this fluence range. There have been only a limited number of studies on neutron irradiated spinel at temperatures below 300°C [15, 16, 17]. The reported volumetric swelling ranges from 0.01 to 0.2% for damage levels between 0.02 and 0.5 dpa. A swelling level of 0.8% was measured for spinel irradiated to a high fluence of 2.1×10^{26} n/m^2 (E>0.1 MeV) at ~150°C [17]. We are unaware of any previous published neutron irradiation studies on sialon or Si_3N_4 at temperatures below 300°C and fluences above 0.1×10^{24} n/m^2 (E>0.1 MeV).

Figure 3 shows the effect of the neutron dose on flexural strength. The figure shows minimal degradation of the strength at this low dose. Typically the strength of ceramics decreases monotonically with dose. The activation of this particular grade of Si_3N_4 was too high for mechanical testing in the facility at ORNL. This grade is similar to most commercial Si_3N_4 in that it uses glassy phase sintering aids for ease of processing. Many low to moderate performance grades use alumina or magnesia. However the high performance materials add transition elements to modify the microstructure. These improved compositions have increased toughness, high temperature resistance and thermal conductivity. The particular grade chosen for this study does have a small amount of an element with a very large neutron cross section. This fact was not noticed until after the irradiations were complete. Re-formulation of this material with a different sintering aid composition would dramatically lower the specimen activation while maintaining superior window performance parameters.

Previous studies on Al_2O_3, AlN and $MgAl_2O_4$ specimens exposed to comparable low-temperature, low-dose neutron irradiation conditions have reported similar minor changes in flexural strength [17, 18]. The threshold fluence for significant decreases in the flexural strength of AlN and Al_2O_3 has been reported to be about 3 to 5×10^{24} n/m^2 (E>0.1 MeV), 3 to 5 times higher than the neutron fluence in the present study. A slight amount of strengthening (<20% increase in strength) has been reported for spinel specimens irradiated to fluences of 20 to 200×10^{24} n/m^2 (E>0.1 MeV) at 100 to 150°C . On the other hand, a slight decrease (<20% change) in flexural strength has been reported for AlN following irradiation to a fluence of only 0.18×10^{24} n/m^2 (E>0.1 MeV) at 100°C [19]. Decreases in flexural strength of 20 to 60% and a corresponding decrease in the Weibull modulus (i.e., an increase in the scatter of the measurements) have been reported for AlN and Al_2O_3 ceramics irradiated to fluences above 10×10^{24} n/m^2 (E>0.1 MeV) at temperatures below 400°C.

The nitride ceramics exhibit the highest net drops in thermal diffusivity as seen in Figure 4. In the case of Si_3N_4 and AlN the final values are still reasonably higher than the other materials. Spinel shows the smallest change of all the materials. The horizontal line at the post-irradiation value of the alumina is the baseline for comparison.

The results from the present HFIR irradiation are in fair agreement with previous thermal diffusivity and thermal conductivity studies on Al_2O_3, $MgAl_2O_4$ and AlN. We are not aware of any previous thermal diffusivity measurements on irradiated Si_3N_4 or sialon ceramics. Previous studies on irradiated Al_2O_3 have reported decreases in the thermal diffusivity ranging from ~20 to 65% following irradiation to fluences of 1 to 5×10^{24} n/m^2 (E>0.1 MeV) at temperatures of 60 to 300°C [20, 21], which is somewhat larger than the change of ~15% observed for the Wesgo AL995 alumina specimen in the present study. The reported decrease in thermal diffusivity of $MgAl_2O_4$ irradiated at 100-400°C to damage levels of 0.02 to 0.5 dpa in two different studies ranged from ~25 to 50%, which is also larger than the ~5% degradation observed in the present 0.1 dpa irradiation. However, one of these studies was performed on nonstoichiometric spinel, and the dose in the other study as five times higher than the present work. Two studies on the same grade of AlN (Tokuyama Soda Co. Shapal grade) have reported a degradation in the thermal diffusivity of 15 to 20% following neutron irradiation to fluences of 0.18 to 0.5×10^{24} n/m^2 (E>0.1 MeV) at ~100°C, and ~75% following high energy proton irradiation at ~300°C to a dose of 0.5 dpa. The result for AlN from the present study falls between these two sets of data.

The change in dielectric loss factors are listed in Table 2. There is very limited previous work on the effects of neutron irradiation on the dielectric properties of ceramic insulators in the ion cyclotron range of frequencies (~50 to 150 MHz) [13]. The loss tangent of a 97.5% purity alumina was observed to increase from its unirradiated value of $\sim 1.5 \times 10^{-4}$ at 65 MHz following neutron irradiation at 60°C, reaching a value of $\sim 4 \times 10^{-4}$ at a fluence of 1×10^{24} n/m2 (E>1 MeV) [22, 23]. A factor of ~20 increase in loss tangent at 65 MHz was observed for AlON irradiated under the same conditions [22]. The loss tangent of a very lossy grade of alumina (Vitox) first increased and then decreased with increasing neutron fluence in the same irradiation experiment [22,23].

The effect of neutron radiation on the window performance parameters of thermal shock resistance and loss factor are shown in Table 2.

To best evaluate the effect of neutron damage on window performance a plot of dielectric loss factor verses thermal shock resistance, as in Figure 5 is useful. Proximity to the upper right section of the graph (low loss factor and high thermal conductivity) represents excellent window performance. As shown in the graph silicon nitride has the best performance after radiation exposure.

Conclusions:

Silicon nitride was the best performing material in the group of materials tested. The bend strength of the Si_3N_4 was not measured due to high activation, however the drop in strength is assumed to be very small. The thermal shock resistance was based on this assumption. Slight modification of the Si_3N_4 composition could be used to eliminate the activation problem and take advantage of the performance demonstrated here. The irradiated sialon had the next best window performance parameters. The alumina and spinel both showed considerable increases in dielectric loss factor. This would indicate that these are less desirable in comparison to the silicon nitride.

- AlN maintained good performance in thermal shock , but was the worst performer in dielectric loss factor.
- A modified Si_3N_4 and Sialon outperformed alumina and spinel.
- Silicon nitride appears to be an excellent option for RF transmission window.

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Table 1. Pre-irradiation window performance data.

	Material	Thermal Shock Resistance Factor[a] (W/m)	Loss Factor [b] (1 MHz)
1	Wesgo Alumina	2123	0.00047
2	Aluminum Nitride	31288	0.0035
3	Sialon	17485	0.0025
4	Silicon Nitride	15819	0.0014
5	Spinel	813	0.00061

a) $R = K\sigma(1-\nu)/(\alpha E)$; estimated using *Composite Technology Development* (CTD) test data and literature data.

b) CTD test data.

Table 2. Neutron effect on window performance parameters.

Material	Irradiation Condition	Loss Factor	Figure of Merit (W/m)
Alumina 99.5%	Pre	0.0005	2123
	Post	0.0188	1754
Aluminum Nitride	Pre	0.0035	31288
	Post	0.0284	12393
SiAlON	Pre	0.0025	17485
	Post	0.0161	12798
Silicon Nitride	Pre	0.0014	15819
	Post	0.0070	9650
Spinel $MgAl_2O_4$	Pre	0.0006	813
	Post	0.0260	752

Figure Captions

Figure 1: Thermal/Dielectric and mechanical specimens used in irradiation of ceramic materials (dimensions in millimeters).

Figure 2. High resolution loss tangent measurement cavity.

Figure 3. Effect of 0.1 dpa fast neutron dose of on flexural strength.

Figure 4. Effect of a 0.1 dpa fast neutron dose of on thermal diffusivity.

Figure 5. Window performance parameters before and after a fast neutron dose of 0.1 dpa.

Fig. 1

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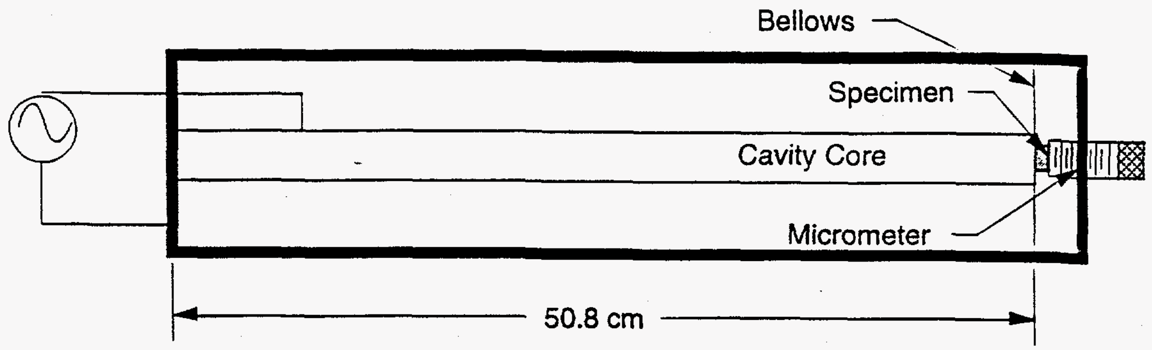


Fig. 2

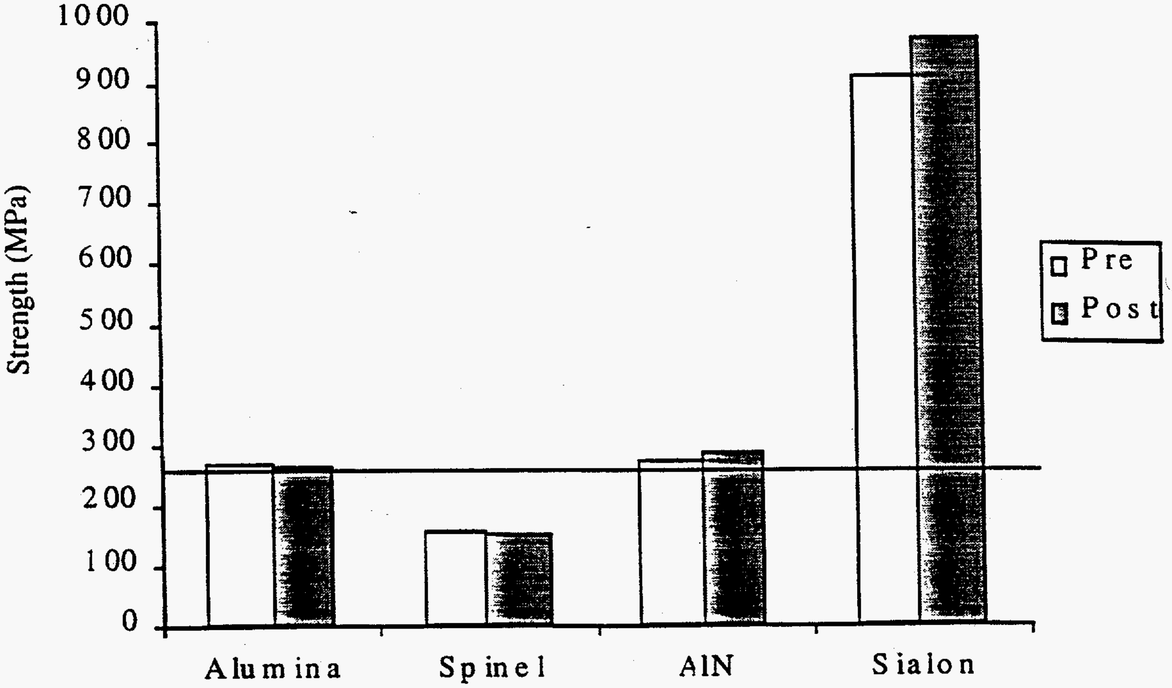
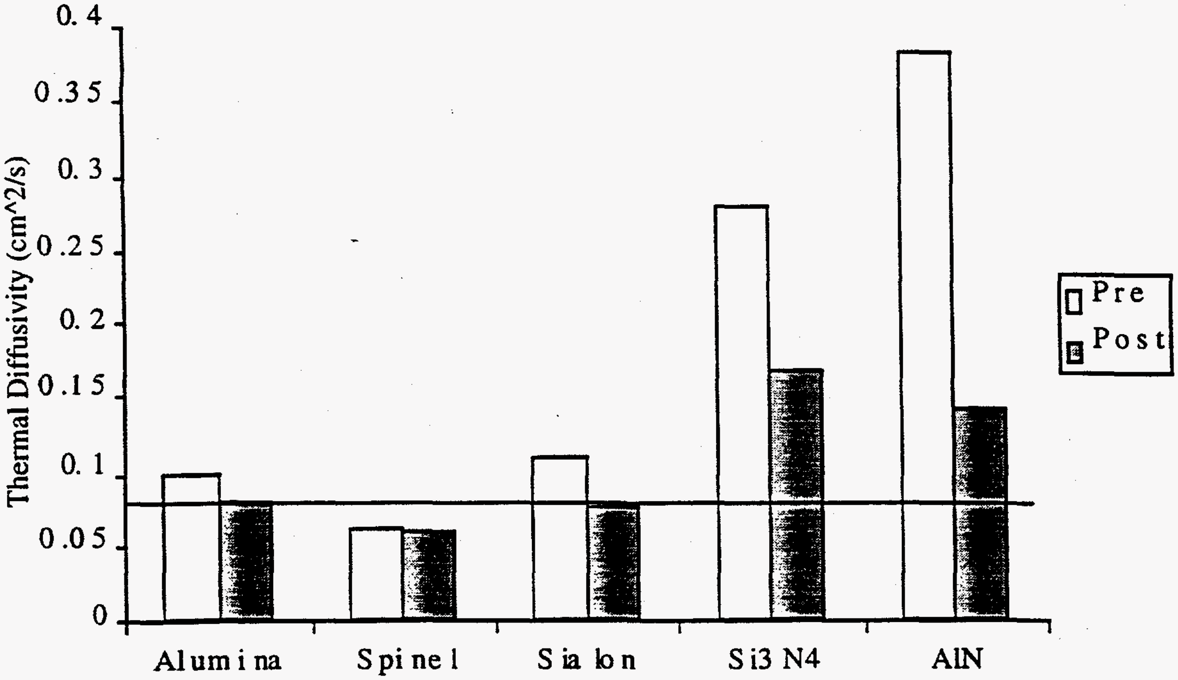
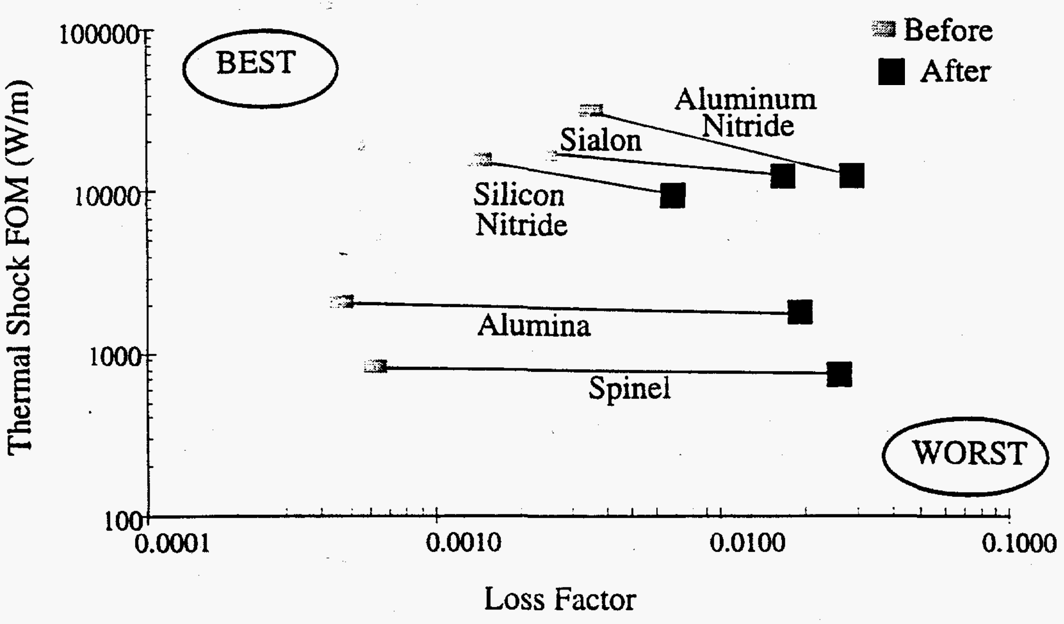

1000
900
800
700
600
500
400
300
200
100
0
Strength (MPa)
Pre
Post
Alumina
Spinel
AlN
Sialon

Fig. 3.



0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Thermal Diffusivity (cm^2/s)
Alumina
Spinel
Sialon
Si3 N4
AlN
Pre
Post

Fig. 4.



100000
10000
1000
100
0.0001
0.0010
0.0100
0.1000
Thermal Shock FOM (W/m)
Loss Factor
BEST
WORST
▨ Before
■ After
Aluminum Nitride
Sialon
Silicon Nitride
Alumina
Spinel

Fig. 5.