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The effect of ionizing and displacive radiation on the thermal conductivity of alumina

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The effects of ionizing and displacive radiation on the thermal conductivity of alumina at high temperatures have been studied. The phonon scattering relaxation times for several scattering mechanisms have been used to determine the effect on the thermal conductivity. The scattering mechanisms considered are scattering by electrons excited into the conduction band, vacancies, aluminum precipitates, and voids. It is found that under irradiation conditions where the electrical conductivity and dielectric loss tangent are greatly increased there is not a significant decrease in the thermal conductivity due to phonon-electron scattering. The conditions under which the scattering due to vacancies, aluminum precipitates, and voids each produces a significant reduction in the thermal conductivity are discussed.

I. INTRODUCTION

Ion-cyclotron-resonance heating of plasmas in fusion reactors requires the development of microwave windows through which the microwaves can pass without great losses, and which can operate reliably with the thermal stresses they will be subject to. The degradation of the thermal conductivity of alumina in a radiation environment is an important consideration in studies of the mechanical reliability of these microwave windows.[1] Early studies[2,3] of the mechanical reliability did not take this degradation into account. A more recent analysis[4] gives analytical expressions for the thermal stresses so that this degradation can more easily be accounted for.

The scattering of phonons by electrons may be neglected in nonradiation environments.[5] This can be seen from Fig. 1, which is a plot of the expected reduction in the lattice thermal conductivity due to phonon-electron scattering versus electrical conductivity explained in Secs. II A and III. In nonradiation environments the electrical conductivity is on the order of $10^{-15}\ \Omega^{-1}\,\mathrm{m}^{-1}$ at room temperature and this does not lead to a significant reduction in the lattice thermal conductivity. However, in a radiation environment the electrical conductivity is greatly increased[6-9] and this increase has a great and prompt effect on the dielectric loss tangent even at frequencies as high as 100 MHz.[6,10,11] This large increase observed during *in situ* measurements raises the question as to whether there is a corresponding prompt decrease in the thermal conductivity which would not be observed in postirradiation measurements. This question if of great importance to the design of microwave windows for plasma heating devices because the conditions for catastrophic window failure must be avoided and an increase in the microwave absorption coupled with a decrease in the thermal conductivity make these conditions more likely.

In this article the decrease in the thermal conductivity expected for different irradiation conditions will be presented. The prompt reduction due to phonon scattering by electrons excited into the conduction band by radiation will be investigated, along with the reduction due to scattering by vacancies, voids, and aluminum precipitates.

II. THEORY

A. Phonon-electron scattering

The transport of heat in ceramics is dominated by phonons and the scattering of phonons by electrons may be neglected in the study of the thermal conductivity of alumina in nonradiation environments; however, in a radiation environment the electrical conductivity is greatly increased.[6-9] The effect of this increased conductivity on the thermal conductivity due to an increase in phonon-electron scattering must be determined in order to predict any significant decrease in the thermal conductivity that may occur.

The lattice thermal conductivity is given by the integral

$$K=\frac{1}{3}\int_0^{\omega_D} C(\omega)v^2\tau(\omega)d\omega, \tag{1}$$

where $C(\omega)$ is the contribution to the specific heat per frequency interval for phonons of frequency ω, v is the phonon velocity, $\tau(\omega)$ is the phonon relaxation time (the average time a phonon of frequency ω exists before being scattered), and ω_D is the Debye frequency. The phonon relaxation time is additively composed of the reciprocal relaxation time for each interaction,

$$\frac{1}{\tau(\omega)}=\sum_\alpha \frac{1}{\tau_\alpha(\omega)}, \tag{2}$$

where the sum is over phonon scattering mechanisms.

Following Klemens,[12] the fractional reduction in the thermal conductivity due to phonon-electron scattering at high temperatures ($T\gg\theta/3$, where θ is the Debye temperature) is found as follows. The reciprocal relaxation time due to intrinsic three-phonon scattering is of the form[13,14]

$$1/\tau_i(\omega,T)=B\omega^2, \tag{3}$$

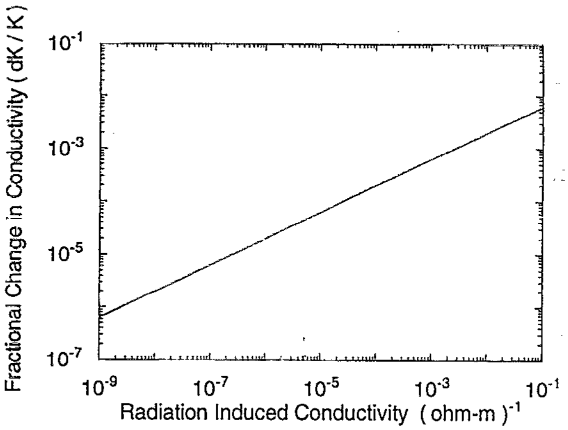

FIG. 1. Fractional change in the thermal conductivity of alumina vs the radiation-induced electrical conductivity at 400 K.

where T is the temperature and B is a material-dependent parameter proportional to T. Assuming a constant phonon-electron relaxation time for the phonon-electron interaction, then

$$1/\tau_{p-e}=C. \tag{4}$$

An expression for C is given in Eq. (16). Energy and wave-vector conservation in the phonon-electron interaction dictates a maximum wave vector for those phonons allowed to interact with the electrons, and an expression for the maximum interacting phonon frequency is given in Eq. (14). Using the high-temperature limit for the specific heat,

$$C(\omega)=(3K_B/2\pi^2v^3)\omega^2, \tag{5}$$

where K_B is the Boltzmann constant, the thermal conductivity with phonon-electron scattering included is found by substituting in the combined relaxation time for intrinsic and phonon-electron scattering along with this expression for the specific heat into Eq. (1). Denoting the phonon-electron interaction cut off frequency by ω_c the thermal conductivity is

$$K_{p-e}=\frac{K_B}{2\pi^2v}\left(\int_0^{\omega_c}\omega^2\frac{1}{B\omega^2+C}\,d\omega+\int_{\omega_c}^{\omega_D}\omega^2\frac{1}{B\omega^2}\,d\omega\right). \tag{6}$$

Evaluating the expression in Eq. (6) and noting that the intrinsic conductivity is given by

$$K_i=\frac{K_B}{2\pi^2v}\int_0^{\omega_D}\omega^2\frac{1}{B\omega^2}\,d\omega=\frac{K_B\omega_D}{2\pi^2vB}, \tag{7}$$

then

$$K_{p-e}=K_i-\frac{K_i}{\omega_D}\left(\frac{C}{B}\right)^{1/2}\tan^{-1}\left[\omega_c\left(\frac{B}{C}\right)^{1/2}\right]. \tag{8}$$

The fractional change in the thermal conductivity is then given by

$$\frac{\delta K_{p-e}}{K_i}=\frac{1}{\omega_D}\left(\frac{C}{B}\right)^{1/2}\tan^{-1}\left[\omega_c\left(\frac{B}{C}\right)^{1/2}\right]=\frac{\omega_0}{\omega_D}\tan^{-1}\left(\frac{\omega_c}{\omega_0}\right), \tag{9}$$

where $\omega_0=(C/B)^{1/2}$, which is the frequency at which the intrinsic scattering relaxation time is equal to the phonon-electron scattering relaxation time.

B. Vacancy scattering

The fractional change in the thermal conductivity due to point defects can be determined in a similar manner. It has been shown[5,12,15] that at high temperatures the fractional decrease in the thermal conductivity due to point defects is

$$\delta K_p/K_i=1-(\omega_p/\omega_D)\tan^{-1}(\omega_D/\omega_p), \tag{10}$$

where $\omega_p=(B/A)^{1/2}$. In this expression A is a parameter which appears in the reciprocal phonon relaxation time due to point defects, which is of the form[16]

$$1/\tau_p=A\omega^4, \tag{11}$$

and is proportional to the defect concentration and depends on the nature of the defect. The parameter B is defined in Eq. (3), and thus ω_p is the frequency at which the intrinsic scattering relaxation time is equal to the point defect scattering relaxation time.

C. Extended defect scattering

The reciprocal relaxation time for extended three-dimensional defects has the form[15,17,18]

$$1/\tau_x=NAv=(3v/4)(x/r)=D, \tag{12}$$

where N is the number of defects per unit volume, A is their cross-sectional area, x is the volume fraction of defects, and r is their radius. Using this relaxation time the fractional change in the thermal conductivity can be shown to be[15,19,20]

$$\delta K_x/K_i=(\omega_x/\omega_D)\tan^{-1}(\omega_D/\omega_x), \tag{13}$$

where $\omega_x=(D/B)^{1/2}$. Once again, B is the parameter from the intrinsic relaxation time expressed in Eq. (3), and in this case ω_x is the frequency at which the intrinsic scattering relaxation time is equal to the extended defect scattering relaxation time.

III. EVALUATION OF THE FRACTIONAL CHANGE IN CONDUCTIVITY

The fractional change in the thermal conductivity due to phonon-electron scattering is given in Eq. (9). In order to evaluate this expression one must know ω_D the Debye frequency, ω_c the phonon-electron cutoff frequency, and $\omega_0\equiv(C/B)^{1/2}$. The Debye temperature of alumina is 1000 K (Ref. 21) and thus $\omega_D=\theta_D K_B/\hbar=1.31\times10^{14}\ \mathrm{s}^{-1}$. If the maximum electron energy is chosen to be K_BT then wave-vector conservation dictates that the maximum frequency of interacting phonons is[17,18]

$$\omega_c=(4\pi v/h)(2mK_BT)^{1/2}, \tag{14}$$

TABLE I. Material constants.

Constant	Alumina
$\theta_D^{\rm a}$ (10^3 K)	1.0
$\Omega^{\rm b}$ (10^{-30} m^3)	8.5
v (10^4 m/s)	1.0
$\mu^{\rm c}$ (10^{-4} m^2/V s)	3.0

[a]See Ref. 21.
[b]See Ref. 22.
[c]See Ref. 23.

where m is the electron mass. Expressing the parameter B in terms of the intrinsic thermal conductivity, it can be seen from Eq. (7) that

$$B=K_B\omega_D/2\pi^2 vK_i. \tag{15}$$

The phonon-electron reciprocal relaxation time has been calculated using a momentum balance argument[17,18] and is

$$\frac{1}{\tau_{p-e}}=C=\frac{n^2e^2v^2}{\mathcal{N}K_BT\sigma}\frac{1}{\alpha}\left(\frac{\theta}{T}\right)^{3/2}, \tag{16}$$

where

$$\alpha=4^3(\pi\Omega/6)^{1/2}(vm/h)^{3/2}.$$

Here Ω is the volume per atom, e is the electron charge, n is the conduction-band electron concentration, $\mathcal{N}$ is the number of atoms per unit volume, and σ is the electrical conductivity. Using the expression $n=\sigma/e\mu$ where μ is the electron mobility, Eq. (16) becomes

$$\frac{1}{\tau_{p-e}}=C=\frac{\sigma v^2}{\mu^2\mathcal{N}K_BT}\frac{1}{\alpha}\left(\frac{\theta}{T}\right)^{3/2}. \tag{17}$$

Table I gives the material constants for alumina. Substituting these values into the expressions for ω_0 and ω_c we obtain $\omega_0=5.86\times10^{14}(K_i^{1/2}\sigma^{1/2}/T^{5/4})$ s^{-1} and $\omega_c=9.51\times10^{11}T^{1/2}$ s^{-1}. Using these values in Eq. (9) gives the fractional change in the thermal conductivity in terms of the electrical conductivity, intrinsic thermal conductivity, and the temperature,

$$\frac{\delta K_{p-e}}{K_i}=4.47\frac{\sigma^{1/2}K_i^{1/2}}{T^{5/4}}\tan^{-1}\left(\frac{1.62\times10^{-3}T^{7/4}}{\sigma^{1/2}K_i^{1/2}}\right). \tag{18}$$

In order to evaluate the expression for the fractional decrease in the thermal conductivity due to vacancy scattering given in Eq. (10) we must evaluate $\omega_p=(B/A)^{1/2}$. For vacancies[14,24]

$$A=C_v\Omega(9/4\pi v^3), \tag{19}$$

where C_v is the vacancy concentration per atom. Thus $\omega_p=3.89\times10^{13}/(K_iC_v)^{1/2}$ and Eq. (10) becomes

$$\frac{\delta K_v}{K_i}=1-\frac{2.96\times10^{-1}}{K_i^{1/2}C_v^{1/2}}\tan^{-1}\left(\frac{K_i^{1/2}C_v^{1/2}}{2.96\times10^{-1}}\right). \tag{20}$$

Finally for the fractional decrease in the conductivity due to extended defects, $\omega_x=9.05\times10^8(K_ix/r)^{1/2}$ s^{-1} and Eq. (13) becomes

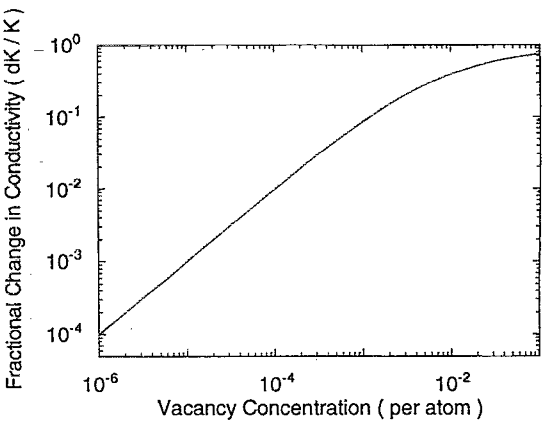

FIG. 2. Fractional change in the thermal conductivity of alumina vs vacancy concentration at 400 K.

$$\frac{\delta K_x}{K_i}=6.91\times10^{-6}\left(\frac{K_ix}{r}\right)^{1/2}\tan^{-1}\left[1.45\times10^5\left(\frac{r}{K_ix}\right)^{1/2}\right]. \tag{21}$$

IV. RESULTS

The fractional change in the conductivity versus the electrical conductivity is plotted in Fig. 1 at 400 K. The intrinsic thermal conductivity was taken to be[25] 26.4 W/m K. From this plot it is seen that a 0.1% change in the thermal conductivity corresponds to a radiation-induced electrical conductivity (RIC) of 3×10^{-3} $(\Omega\,\mathrm{m})^{-1}$. As seen in the following section the upper limit on the RIC expected in the fusion environment is on the order of 10^{-5} $(\Omega\,\mathrm{m})^{-1}$ and thus the effect of phonon-electron scattering on the thermal conductivity of alumina is not expected to be of any consequence in the fusion environment.

Figure 2 is a plot of the fractional change in the thermal conductivity versus vacancy concentration, also at 400 K. From this plot a vacancy concentration of 1×10^{-4} corresponds to a fractional change in the thermal conductivity of 1%. As shown previously[15] vacancy concentrations on the order of 1% have been observed in alumina neutron irradiated to a fluence of one displacement per atom (dpa) or more, and large decreases in the thermal conductivity are expected as a result of phonon scattering by vacancies in neutron-irradiated ceramics.

The fractional change in the thermal conductivity versus volume concentration of 10-nm-diam extended defects is plotted in Fig. 3 at 400 K. Extended defects produced by neutron irradiation[26] and by proton irradiation[27] in the form of voids and alumina precipitates have been shown to produce significant decreases in the thermal conductivity.[15,19,20] From this plot it can be seen that a fractional change in the conductivity of 1% corresponds to a volume fraction of only 2×10^{-4} or a number density of 3.8×10^{20} m^{-3} 10-nm-diam defects.

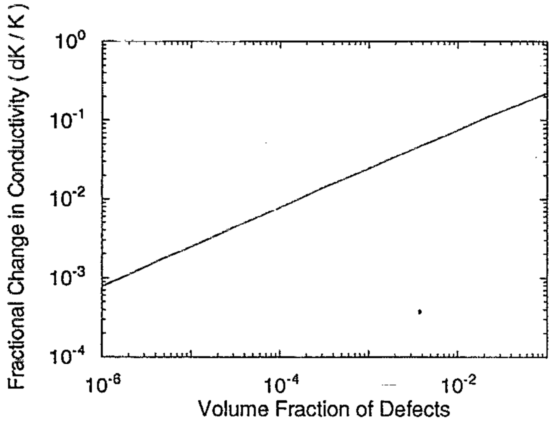

FIG. 3. Fractional change in the thermal conductivity of alumina vs volume concentration of 10-nm-diam spherical defects at 400 K.

V. DISCUSSION

A. Prompt effects

As an example of the prompt fractional change in the thermal conductivity expected under radiation conditions where the dielectric loss was observed to increase, the conditions for the dielectric loss measurements in alumina of Stoller and co-workers[10] will be used. In these experiments the alumina samples were irradiated at 306 K in a pulsed fission reactor for approximately 1.7×10^{-2} s. The ionizing rate was 5.83×10^{4} Gy/s and the dielectric loss tangent at 100 MHz was observed to increase by 1.6×10^{-4} in a 99.8% purity alumina. The loss tangent has been observed to increase by 3.5×10^{-4} in 97% purity alumina.[28] The change in the loss tangent of a dielectric associated with the RIC is

$$\Delta \tan \delta(\omega) = \sigma_{dc}/\omega\epsilon_0\epsilon_r, \tag{22}$$

where σ_{dc} is the dc electrical conductivity, ω is the angular frequency, ϵ_0 is the permittivity of free space, and ϵ_r is the relative permittivity of alumina. Using this expression the RIC corresponding to a change in the loss tangent can be determined. Using $\epsilon_r=9$ these changes in the dielectric loss tangent, $\Delta \tan \delta$, correspond to conductivities of 8.0×10^{-6} $(\Omega\,\mathrm{m})^{-1}$ and 1.7×10^{-5} $(\Omega\,\mathrm{m})^{-1}$, respectively, in the two different grades of alumina. Pells *et al.*[9] determined the RIC versus the ionizing dose rate for polycrystalline alumina irradiated with 20 MeV protons. Extrapolating this data to an ionizing dose rate of 5.8×10^{4} Gy/s, it is estimated that the RIC in these experiments would be $\sigma=1.8\times10^{-6}$ $(\Omega\,\mathrm{m})^{-1}$.

The expected ionizing dose rate in a fusion reactor ranges from 10^3 to 10^5 Gy s^{-1},[29] which according to the data of Pells corresponds to a range of conductivities of from 1.0×10^{-8} to 3.7×10^{-6} $(\Omega\,\mathrm{m})^{-1}$. Substituting these values of the RIC into Eq. (18), along with $K_l(306\text{ K})=35.4$ (Ref. 25), the fractional change in the thermal conductivity may be determined. Table II gives the fractional change expected in the thermal conductivity for the range of RIC values expected in a fusion environment. From this table it can be seen that a negligibly small change in the lattice thermal conductivity is expected due to phonon-electron scattering under these conditions.

TABLE II. Range of RIC values expected in the fusion environment and the corresponding fractional reduction in the lattice thermal conductivity at 306 K.

σ $(\Omega\,\mathrm{m})^{-1}$	$\delta K/K$
1.0×10^{-8}	3.3×10^{-6}
1.7×10^{-5}	1.3×10^{-4}

It is also not expected that the increase in the electronic thermal conductivity due to RIC will be significant. This can be seen by using the Lorenz ratio for metals, 2.5×10^{-8} W Ω K^{-2} (this value of the Lorenz ratio will lead to an overestimate of the thermal conductivity in these materials), to determine the corresponding thermal conductivity. A RIC of 1×10^{-5} $(\Omega\,\mathrm{m})^{-1}$ corresponds to a thermal conductivity of only 7.5×10^{-11} W/m K at 300 K. Therefore, the increase in the thermal conductivity due to the RIC is negligible.

The vacancies produced during neutron irradiation will also scatter phonons and lead to a reduction in the thermal conductivity. During displacive irradiation the point defect concentration initially increases at a linear rate.[30] The time at which the defect concentrations deviate from this linearity is a function of temperature and the sink strength. For room-temperature irradiation of ceramics it is reasonable to assume this relationship holds over the short time of the reactor pulse used in the experiments of Stoller and co-workers,[10] 1.7×10^{-2} s^{-1} (this will give an upper bound to the fraction change in any case). Then

$$C_v = Gt, \tag{23}$$

where G is the defect generation rate for mobile monodefects per second and t is the time of the irradiation. The use of Eq. (23) neglects any influence of the displacement cascades themselves. In this case Eq. (23) gives $C_v=3.06\times10^{-9}$. Substituting this value into Eq. (20) at 306 K we obtain $\delta K_v/K_l=4.12\times10^{-7}$. This is also a negligibly small prompt change in the thermal conductivity. Thus, for experimental conditions where the prompt effects on the electrical conductivity and dielectric loss tangent in a radiation field are great, there is no significant prompt effect on the lattice thermal conductivity.

B. Accumulated dose effects

With accumulated dose the effects of aluminum precipitates, voids, and the accumulated vacancy concentration on the lattice thermal conductivity of alumina must be taken into account.[15,19,20] Pells and Shikama[27] studied void and aluminum precipitate formation in 1 MeV electron-irradiated alumina. The results of this work can be applied to determine the resulting reduction in the thermal conductivity. For example, Table III gives the typical defect size and concentrations found in a sample irradiated at 1130 K to a fluence of 124 MC/m^2 which corresponds to 6.2 dpa.

TABLE III. Defect type, size, and concentration.

Defect type	Most probable diameter ($\times10^{-9}$ m)	Vol %
Void	7	5.6
Al Precipitate	17	2.9

For these defects,

$$\omega_x=9.05\times10^8\left[K_i\left(\frac{x_1}{r_1}+\frac{x_2}{r_2}\right)\right]^{1/2}=1.05\times10^{13}\ \mathrm{s}^{-1}, \qquad (24)$$

where the subscripts 1 and 2 denote voids and precipitates, respectively. Using $K_i(1130\ \mathrm{K})=6.92$ W/m K (Ref. 23), the reduction in the conductivity given by Eq. (21) is $\delta K_x/K_i=11.9\%$ at 1130 K. This is a substantial reduction in the conductivity which must be taken into account when discussing applications of alumina in radiation environments where heat flows.

At high doses the accumulated vacancy point defect concentration also becomes significant. By referring to Fig. 2 one can see that at 400 K large reductions in the thermal conductivity are expected even for vacancy concentrations of less than 1%. A reduction is also expected at higher temperatures although due to the increase in intrinsic scattering the reduction will not be as great as in Fig. 2. As an example, for vacancies we have seen that $\omega_p=3.89\times10^{13}/(C_v K_i)^{1/2}\ \mathrm{s}^{-1}$, so that at 1130 K for a 0.1% isolated vacancy concentration $\omega_p=4.66\times10^{14}$ and from Eq. (20) $\delta K_v/K_i=2.5\%$. For a 1% vacancy concentration $\omega_p=1.47\times10^{14}$ and $\delta K_v/K_i=18.2\%$. Thus, for the range of vacancy concentrations expected in these materials a significant reduction in the thermal conductivity is expected. If $\omega_x<\omega_p$, which is true for the examples above, then the point defects, which strongly scatter phonons above ω_p, and extended defects, which strongly scatter phonons below ω_x, scatter phonons independently of one another and the reductions are additive. Thus a total reduction of 14.4% is expected in the case of a 0.1% vacancy concentration and a reduction of 30.1% is expected in the case of a 1% vacancy concentration. It is reasonable to expect that the total reduction in thermal conductivity for the sample in this example would be within this range.

VI. CONCLUSIONS

It has been shown that the phenomenon of RIC, which gives rise to a large prompt increase in the electrical conductivity and dielectric loss tangent, does not give rise to a corresponding prompt decrease of the lattice thermal conductivity. It has also been shown that vacancies produced during short irradiations do not cause a prompt decrease of the thermal conductivity. Thus, although the prompt increase in the dielectric loss tangent during irradiation must be considered in the design of microwave windows, it is not expected that there will be a corresponding prompt decrease in the thermal conductivity of any consequence.

However, it has been demonstrated that with an *increasing* dose when vacancies have accumulated and voids and precipitates have been produced a significant reduction in the thermal conductivity will occur. At 400 K the concentration of vacancies necessary to produce a 5% reduction in the thermal conductivity is $C_v=6\times10^{-4}$. The volume fraction of 10-nm-diam defects needed to produce a 5% reduction in the thermal conductivity at 400 K is $x=4\times10^{-3}$, which corresponds to a number density of $7.6\times10^{21}\ \mathrm{m}^{-3}$. These accumulated dose effects must be taken into account when designing microwave windows and studying their mechanical reliability in a radiation field.

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