THERMAL CONDUCTIVITY OF NATURAL AND ISOTOPICALLY ENRICHED DIAMOND - EFFECT OF NEUTRON IRRADIATION

D.P. White, Department of Physics, Merrimack College, N. Andover, MA 01845

OBJECTIVE

The objective of this work is to study the influence of defects produced by neutron irradiation on the thermal conductivity of diamond.

SUMMARY

Studies on the temperature dependence of the thermal conductivity of isotopically enriched (0.1% ^{13}C) diamond [1-5] have determined the form for the intrinsic scattering phonon relaxation times in diamond. In addition, low temperature thermal conductivity measurements and infrared spectra of lightly neutron-irradiated type IIa natural diamond [6] have determined the size and concentration of extended regions of disordered carbon responsible for phonon scattering. These results have been used to model the thermal conductivity changes expected in neutron irradiated diamond at higher temperatures, from 100 to 1000K. It was found that upon irradiation to a fluence of 4.5 X 10^{22} neutrons m^{-2} the thermal conductivity of natural diamond went from 2200 to 370 W/mK at 300K while the thermal conductivity of diamond with a 0.1% concentration of ^{13}C went from 3000 to 370 W/mK at 300K.

PROGRESS AND STATUS

Introduction

Electron cyclotron resonance heating (ECH) is a major candidate for plasma heating in ITER [7-11]. One of the milestones is the development of a 170Ghz, 1MW, continuous wave (CW) gyrotron. The development of a ceramic window capable of power transmission at these levels is critical. Sapphire and silicon nitride were not able to sustain CW operation at these levels [7,8] and thermal runaway occurred in these materials above ~170kW. Recent work has concentrated on diamond as it has a small dielectric loss tangent [7-11] and high thermal conductivity [1-5,12]. It is important to characterize the effect of neutron irradiation on the thermal conductivity of diamond, as this will have a direct influence on the final temperature of the window during CW operation of the gyrotron. In tests of diamond windows on 170Ghz gyrotrons [8,9] it was found that these windows could fulfill the requirements for CW transmission in the megawatt range. The thermal conductivity of the windows used in these tests was estimated to be about 1800 W/mK by comparing experimental results with numerical simulations. It is expected that upon neutron irradiation the thermal conductivity will be degraded [6,13-14] and thus the performance of these windows will be affected.

Theory

The thermal conductivity was calculated using the Callaway method [15]. This method takes into account the effects of intrinsic three-phonon normal (N) processes. These processes do not create thermal resistance directly but affect the thermal resistance by transferring phonons from frequencies where they are not scattered to frequencies where scattering is more efficient. The Callaway expression for the temperature dependent thermal conductivity, extended to take into account the differences in the transverse and longitudinal modes [1,3] is

$$K(T) = K_1(T) + K_2(T) \tag{1}$$

where,

$$K_1(T) = \frac{k^4 T^3}{6\pi^2 \hbar^3} \sum_{j=1}^{3} \frac{1}{v_j} \left(\int_0^{\theta_j / T} \tau_{cj} J_4(x) dx \right) \tag{2}$$

and

$$K_2(T) = \frac{k^4 T^3}{6\pi^2 \hbar^3} \sum_{j=1}^{3} \frac{1}{v_j} \frac{\left(\int_0^{\theta_j / T} \frac{\tau_{cj}}{\tau_{nj}} J_4(x) dx \right)^2}{\int_0^{\theta_j / T} \frac{\tau_{cj}}{\tau_{nj} \tau_{rj}} J_4(x) dx}. \tag{3}$$

The sum is over transverse and longitudinal modes, k is the Boltzmann constant, T is the absolute temperature, $\hbar$ is Plank's constant divided by 2π, v is the phonon velocity, $x = \hbar\omega / kT$, ω is the phonon angular frequency, and $J_4 = x^4 e^x (e^x - 1)^{-2}$. The relaxation times are defined as follows, the resistive relaxation time τ_r is given by, $\tau_r^{-1} = \sum_i \tau_i^{-1}$, where the sum is over all resistive scattering processes including intrinsic umklapp (U) processes, isotope scattering, extended defect scattering, etc.. The N process relaxation time is τ_n and τ_c is the combined relaxation time given by, $\tau_c^{-1} = \tau_r^{-1} + \tau_n^{-1}$.

Natural diamond has a Carbon-13 concentration of 1.1% and it is known that the thermal conductivity of isotopically enriched diamond is greatly increased over a wide temperature range compared to natural diamond particularly at the peak of the thermal conductivity curve. In order to explain the large increase (originally much larger than expected [16]) in terms of isotope scattering it was necessary to include the effects of N processes [5]. Subsequent analyses of the thermal conductivity of enriched diamond have included N processes. Wei et. al. [1] used the Callaway expression to fit experimental thermal conductivity data on diamond with the natural isotope concentration and isotopically enriched (0.1% ^{13}C) diamond over a wide temperature range (100-1000K). From this analysis they determined forms for the intrinsic phonon relaxation times. They found the U-process relaxation time to be,

$$\tau_u^{-1} = 4.7 \times 10^2 x^2 T^3 e^{\frac{-670}{T}} \tag{4}$$

and the N-process relaxation time to be,

$$\tau_n^{-1} = 1.49 x T^4 . \tag{5}$$

Olson et. al. [2] and Morelli et. al. [6] find similar expressions for the U-process relaxation time.

Morelli et. al. [6] measured the thermal conductivity of neutron irradiated diamond at low temperatures from 5-300 K. They determined that neutron irradiation caused phonon scattering by extended regions of disordered carbon. The size and concentration of these defects were

determined from curve fitting to their low temperature experimental data. They also found that the size and concentration thus determined correlated with those determined from infrared spectra. In addition they also saw an increase in the amount of simple point defect scattering (vacancy scattering) after neutron irradiation.

This information on intrinsic relaxation times and on neutron irradiation induced have been used here to determine the effect of neutron irradiation on the thermal conductivity of both isotopically enriched diamond and of diamond with the natural isotope concentration. To do this the phonon relaxation times were substituted into the Callaway expression, Eqs. 1-3.

The relaxation time due to isotope point defect scattering is given by [5,17],

$$\tau_{isotope}^{-1} = \frac{c(1-c)}{(12+c)^2}\Omega\frac{1}{4\pi v^3}\left(\frac{k}{\hbar}\right)^4 T^4 x^4 \qquad (6)$$

where c is the isotope concentration and Ω is the atomic volume. The relaxation time due to point defect vacancy scattering is given by [17-19],

$$\tau_{vac}^{-1} = 9c_v\Omega\frac{1}{4\pi v^3}\left(\frac{k}{\hbar}\right)^4 T^4 x^4 \qquad (7)$$

where c_v is the vacancy concentration. The relaxation time for extended defects is given by [3,6,20],

$$\tau_{ext}^{-1} = \frac{\rho v \pi a^2}{4} \text{ if } \frac{akTx}{v\hbar} > 1 \qquad (8)$$

$$\tau_{ext}^{-1} = \frac{\rho \pi a^6}{4v^3}\left(\frac{k}{\hbar}\right)^4 T^4 x^4 \text{ if } \frac{akTx}{v\hbar} < 1$$

where ρ is the extended defect volume density and a is the radius of the extended defect, assumed spherical. Extended defects scatter long wavelength phonons as point defects (Rayleigh scattering) and geometrically (boundary scattering) for short wavelength phonons.

Morelli et. al. [6] performed four irradiations with the following results.

Table I. Size and concentration of extended defects for various neutron fluences along with vacancy concentration necessary to produce point defect scattering in excess of that seen in unirradiated sample. F is the neutron fluence. Data from Ref. 6.

F (neutrons m^{-2})	ρ (m^{-3})	a (m)	c_v (per atom)
0	0	0	0
$3.0X10^{20}$	$4.5X10^{22}$	$8X10^{-10}$	$4.7X10^{-5}$
$1.2X10^{21}$	$1.9X10^{23}$	$8X10^{-10}$	$8.8X10^{-5}$
$6.0X10^{21}$	$1.0X10^{24}$	$8.5X10^{-10}$	$1.15X10^{-4}$
$4.5X10^{22}$	$5.6X10^{25}$	$5.5X10^{-10}$	$1.7X10^{-4}$

These values were used in the relaxation times above and used in the Callaway expression to calculate the thermal conductivity of diamond.

Results

The results of the calculations described above are displayed in the plot in Figure 1. The results for the irradiated isotopically enriched diamond are not shown as they fall on top of the natural diamond curves.

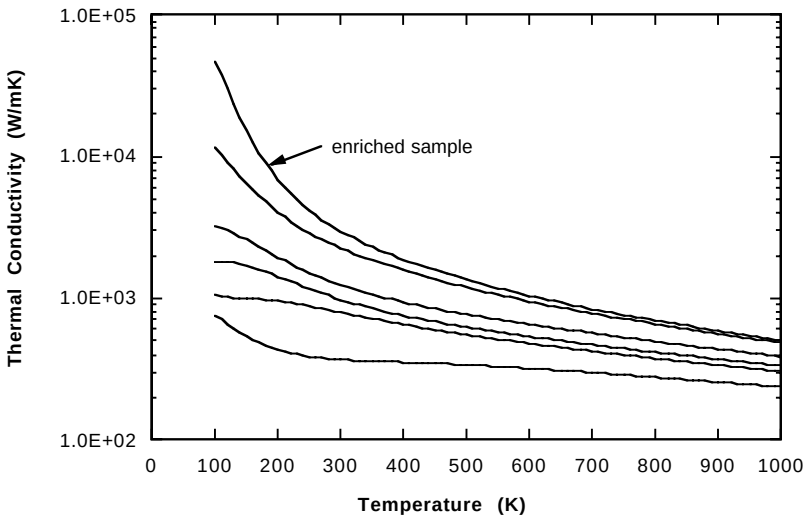

Figure 1. The thermal conductivity of diamond as a function of temperature. The upper curve is for unirradiated isotopically enriched diamond (0.1% ^{13}C). The other curves are for diamond with the natural isotope concentration of 1.1% in the same order as in Table 1.

CONCLUSIONS

From the data presented in Fig. 1 it is shown that the thermal conductivity of diamond is substantially degraded upon neuron irradiation. The decrease in the thermal conductivity of the isotopically enriched diamond is particularly pronounced and any advantage gained in terms of higher thermal conductivity is quickly eliminated upon neutron irradiation. An example of the decrease in thermal conductivity which occurs upon neutron irradiation, it was found that upon irradiation to a fluence of 4.5 X 10^{22} neutrons m^{-2} the thermal conductivity of natural diamond went from 2200 to 370 W/mK at 300K while the thermal conductivity of diamond with a 0.1% concentration of ^{13}C went from 3000 to 370 W/mK at 300K. This factor of 6 reduction in the case of natural diamond and the factor of 8 reduction in the case of the enriched diamond have implications in the design of ECH gyrotron windows.

FUTURE WORK

Future work will involve including other defect types into the calculations. With the model developed here it will be possible to determine the effect of each new type of defect on the thermal conductivity.

ACKNOWLEDGMENTS

The author is happy to acknowledge that this research was partially supported through a grant from the U.S. Department of Energy, Grant number DE-FG02-01ER54627 and through a course load reduction grant from the Faculty Development Grant Committee of Merrimack College.

REFERENCES

1. Lanhua Wei, P.K. Kuo, R.L. Thomas, T.R. Anthony, and W.F. Banholzer, Phys. Rev. Lett. 70, no. 24 (1993) 3764.
2. J.R. Olson, R.O. Pohl, J.W. Vandersande, A. Zoltan, T.R. Anthony, and W.F. Banholzer, Phys. Rev. B 47, no. 22 (1993) 14850.
3. J.E. Graebner, S. Jin, J.A. Herb, and C.F. Gardinier, J. Appl. Phys. 76, no. 3 (1994) 1552.
4. R. Berman, J. Phys. Chem. Solids 59, no. 8 (1998) 1229.
5. R. Berman, Phys. Rev. B 45, no. 10 (1992) 5726.
6. Donald T. Morelli, Thomas A. Perry, and John W. Farmer, Phys. Rev. B 47 (1993) 131.
7. K. Sakamoto, ITER EDA Newsletter vol. 8 (1999) 3.
8. K. Sakamoto, A. Kasugai, M. Tsuneoka, K. Takahashi, T. Imai, T. Kariya, and Y Mitsunaka, Rev. of Sci. Instru. 70, no. 1 (1999) 1.
9. O. Braz, A. Kasugai, K. Sakamoto, K. Takahashi, M. Tsuneoka, T. Imai, and M. Thumm, Int. J. of Infrared and Millimeter Waves 18, no. 8 (1997) 1495.
10. R. Heidinger, A. Ibarra, and J. Molla, J. Nucl. Mater. 258-263 (1998) 1822.
11. R. Heidinger, R. Spörl, M. Thumm, J.R. Brandon, R.S. Sussmann, C.N. Dodge, in Conf. Digest, 23rd International Conference on Infrared and Millimeter Waves, edited by T.J. Parker and S.R.P. Smith, Colchester, Essex UK, Sept. 7-11, (1998) 223.
12. J.E. Graebner, M.E. Reiss, L. Seibles, T.M. Hartnett, R.P. Miller, and C.J. Robinson, Phys. Rev. B 50, no. 6 (1994) 3702.
13. R. Berman, E.L. Foster, and H.M. Rosenburg, in Defects in Crystalline Solids –Report'54 Bristol Conference, edited by A.C. Strickland, The Physical Society, London (1954) 321.
14. D.T. Morelli, T.A. Perry, J.W. Vandersande, and C. Uher, Phys. Rev. B 48, no. 5 (1993) 3037.
15. J. Callaway, Phys. Rev. 113 (1959) 1046.
16. T.R. Anthony, W.F. Banholzer, J.F. Fleischer, L. Wei, P.K. Kuo, R.L. Thomas, and R.W. Pryor, Phys. Rev. B 42 (1990) 1104.
17. P.G. Klemens, in Thermal Conductivity, edited by R.P. Tye (Academic Press, NY) (1969).
18. P.G. Klemens, Nucl. Instr. and Meth. 229 (1984) 204.
19. D.P. White, J. Appl. Phys. 73 (1993) 2254.
20. J.W. Vandersande, Phys. Rev. B 15 (1977) 2355.