Influence of neutron irradiation on the thermal conductivity of vapor-deposited diamond

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(Received 2 February 1994; accepted for publication 18 April 1994)

The influence of neutron irradiation on the thermal conductivity κ of diamond films fabricated by hot filament (HF) and microwave plasma assisted (MPA) deposition has been studied. The additional thermal resistivity induced by irradiation is similar to that found in single crystal diamond and is due mainly to the formation of clusters of disordered carbon material. Despite a significant difference in κ prior to irradiation, the thermal conductivity of the HF and MPA films is almost the same after a cumulative dose of 2.7×10^{17} neutrons cm^{-2}.

INTRODUCTION

Great advances in the growth of chemically vapor deposited (CVD) diamond has paved the way for application of this material in a wide variety of technologies. One of the most promising uses of CVD diamond is as a heat transfer medium.[1] This application relies on the high thermal conductivity κ, in excess of three times that of copper, which can be achieved with high quality CVD material today. As a result, CVD diamond is quickly becoming the material of choice as a substrate for high power electronics, light emitting diodes, and lasers. In many of these applications, as well as in CVD diamond's proposed[2] use as a radiation detector, of critical importance is the behavior of this material's physical properties in a harsh radiation environment. Since the thermal conductivity is one of the most attractive physical parameters of CVD diamond and many of these uses rely on a high κ, we have undertaken an investigation of the influence of neutron irradiation on the heat-carrying ability of these materials. Neutron doses on the level of those which can cause significant degradation of the thermal conductivity can occur in nuclear reactor applications in medicine and industry in, relatively speaking, a fairly short time scale (months or a few years). In addition, disordered regions can also be formed by ion bombardment and electron irradiation, and neutron impingement offers a convenient and well calibrated method of producing such structural disorder for investigation in the laboratory.

EXPERIMENTAL PROCEDURE AND RESULTS

The irradiation procedure and thermal conductivity measurement technique have been described earlier.[3] The samples used in this study were from the same batch as those used in an earlier study.[4] One sample was grown by microwave plasma assisted (MPA) CVD and the second by hot filament (HF) CVD. Both of these samples possess a microstructure characteristic of CVD diamond,[5] namely, columnar grains about 1 μm in extent at the substrate surface and tens of μm in extent on the top (growth) surface. The grain size influences the thermal conductivity,[4,6,7] and, due to the columnar structure, produces an anisotropic[8] and locally varying[9] κ. In addition to these grain boundary effects, it has also been established that intragranular defects produce a depression in the temperature dependent thermal conductivity $\kappa(T)$ in the region of 30 K. The thermal conductivity of our samples prior to irradiation is shown in Figs. 1 and 2 and is consistent with these observations. Namely, at room temperature we find $\kappa(\text{MPA})=17$ W cm^{-1} K^{-1} and $\kappa(\text{HF})=12$ W cm^{-1} K^{-1}. Because the grain size and distribution of these two samples are very similar, most of this difference is due to the additional disorder-induced scattering in the HF sample which is clearly visible as a bending in the curve near 30 K. Measurements to lower temperature[4,10] indicate that this bending is actually the commencement of a dip in the thermal conductivity curve. This type of behavior has been shown to be consistent[3] with scattering by a cluster, or precipitate, approximately 15 Å in extent, and is presumably[4] associated with the presence of nondiamond carbon located at the grain interfaces.

After irradiation to cumulative levels of 3×10^{16} and 2.7×10^{17} neutrons cm^{-2}, $\kappa(T)$ of both samples is depressed throughout the entire temperature range. The effect of the irradiation is to enhance the magnitude of the dip near 30 K in the HF film and produce such a dip in the MPA film, which possessed no such anomaly prior to irradiation. In essence, neutron irradiation produces a κ very similar in magnitude and temperature dependence to that of more highly disordered films[6] containing appreciable amounts of sp^2-bonded carbon. The behavior of irradiated films is essentially identical to that in single crystal diamond,[3,11] for which detailed optical and thermal studies indicate that neutron irradiation produces clusters of disordered carbon approximately 15 Å in extent. A more direct comparison of the influence of neutron irradiation on the thermal conductivity of CVD and single crystal diamond can be made by plotting the additional thermal resistivity ΔW over that of the unirradiated sample as a function of temperature. This procedure, shown in Fig. 3, separates out the effect of defects induced by irradiation from that of grain boundaries and other native defects. We indeed find that the additional thermal resistivity due to neutron irradiation is essentially the same in CVD diamond as in the single crystal. It should be noted that at the

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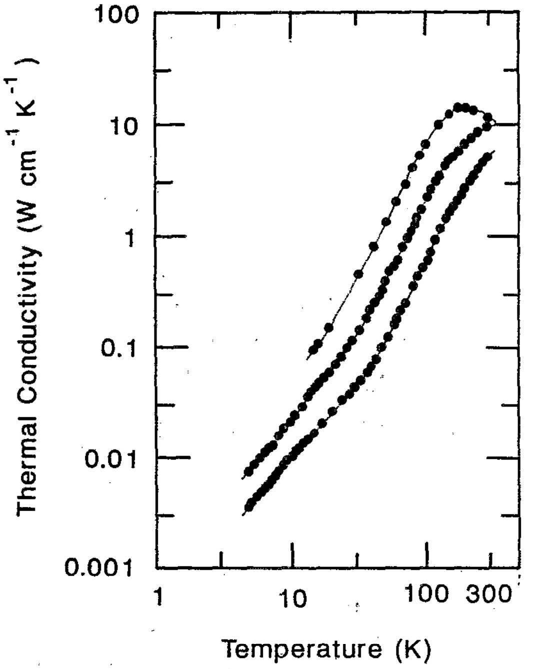

FIG. 1. Thermal conductivity of a synthetic diamond film grown by hot filament deposition before (top) and after irradiation with fast neutrons to a level of 3×10^{16} (middle) and 2.7×10^{17} (bottom) neutrons cm^{-2}.

level of 2.7×10^{17} neutrons cm^{-2}, most of the total thermal resistivity is due to the radiation-induced damage. This indicates that any advantage that might be gained in using higher thermal conductivity samples prior to irradiation will be lost when the cumulative fluence of neutrons reaches this level. It should be noted, however, that the room temperature values of 6–8 W cm^{-1} K^{-1} after the final irradiation still exceed those of copper and all other high thermal conductivity nonmetals.[12] Thus, in applications where fairly heavy doses of neutron irradiation can be expected over short periods of time, it may prove more cost-effective to use less expensive HF material since any advantage gained by going to more highly conducting MPA films will be quickly lost upon irradiation.

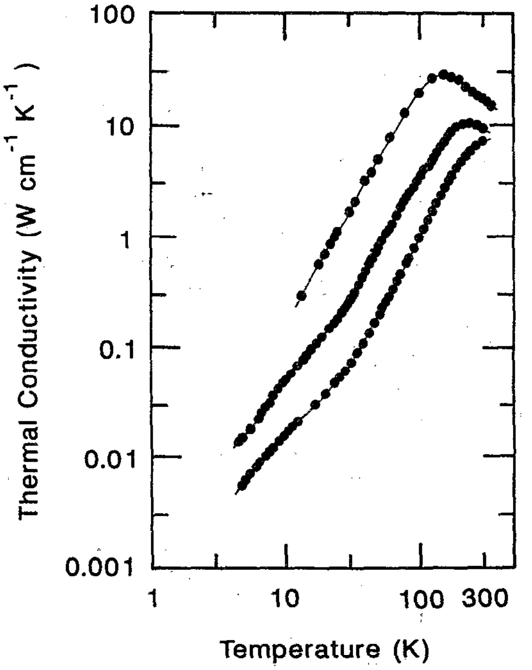

FIG. 2. Thermal conductivity of a synthetic diamond film grown by microwave plasma assisted deposition before (top) and after irradiation with fast neutrons to a level of 3×10^{16} (middle) and 2.7×10^{17} (bottom) neutrons cm^{-2}.

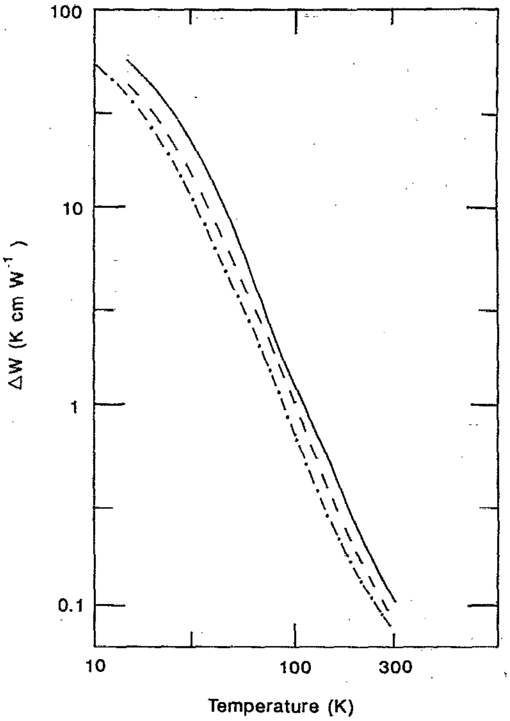

FIG. 3. Radiation-induced thermal resistivity of HF, MPA, and single crystal diamond after a fluence of 2.7×10^{17} neutrons cm^{-2}. Solid curve: HF sample; dashed curve: MPA sample; chain curve: single crystal.

ACKNOWLEDGMENTS

One of us (C. U.) acknowledges partial support from the University of Michigan Phoenix Memorial Project and the Office of Naval Research under Grant No. N00014-92-J-1335.

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[1] An often misused terminology in this context is the phrase "heat sink." A material will be a good heat sink if it has a high specific heat, which is not true of diamond.

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