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The toughness of free-standing CVD diamond

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A four-point bend test was used to determine the fracture toughness of mechanical grade and di-electric (optical) grade chemical vapour deposited (CVD) diamond. The validity of the test was first confirmed by measuring the toughness of alumina and confirming the results with literature values. The toughnesses of both types of CVD were similar; 8.5 ± 1.0 and 8.3 ± 0.4 MPa$\sqrt{\text{m}}$ respectively. This is higher than the value of 3.4 ± 0.5 MPa$\sqrt{\text{m}}$ measured for good quality natural diamond by Field and Freeman, [1] using an indentation technique. It is suggested that this is primarily due to differences in surface roughness. There were enough samples to make a preliminary study of the effect of temperature and these data are reported. *© 2004 Kluwer Academic Publishers*

1. Introduction

Chemically vapour deposited (CVD) diamond is finding increased application where the exceptional properties attained by diamond are required. For example, in applications where high wear or erosion resistances are important, or as optical, infrared and microwave "windows" in aggressive environments [2–5]. The stimulus for the present research was the use of CVD diamond as a window in a millimetre wave heating system for fusion research. Attractions are the low loss tangent ($\tan\delta = 2 \times 10^{-5}$ at the 100 GHz band), high thermal conductivity (1800–2000 W m^{-1} K^{-1}) and high mechanical strength [6–8]. Additionally, the window can also act as a barrier to tritium and radio-active dusts from the heating system [8, 9]. Clearly it is important for the window design to have data on the mechanical properties. This paper reports experiments on the toughness of both mechanical grade and di-electric (optical) grade CVD diamond.

Toughness is a measure of the energy required to propagate a crack. It is usually expressed in terms of the critical stress intensity factor, K_{IC},

$$K_{\text{IC}}^2 = 2\psi E\gamma, \qquad (1)$$

where ψ is a surface roughness factor.

K_{IC} can be obtained by monitoring the growth of fracture caused by indentation. Indenters of a variety of shapes and sizes have been used to measure the fracture toughness of diamond. There are two basic approaches, namely those which use a blunt indenter and those which use a sharp indenter. For reviews, see [10, 11].

Field and Freeman were the first to measure the K_{IC} of diamond [1, 11, 12]. They used a blunt indenter technique, first devised by Roesler [13] which produces a "cone" crack in isotropic materials such as glasses and a modified cone in diamond since the crack growth is influenced by the {111} cleavage planes in diamond [1]. A value of K_{IC} of 3.4 MPa$\sqrt{\text{m}}$ was found for diamond when good quality stones were indented on {100} planes. Field and Freeman noted that this was very close to the value predicted by Ramachandran [14] for {111} cleavage planes assuming simple bond breaking and no contribution from plasticity. For a surface roughness factor of $\Psi = 2$, then K_{IC} would increase to ~5.0 MPa$\sqrt{\text{m}}$.

Naletov *et al.* [15] propagated cracks from a Berkovich indenter and recorded K_{IC} data for synthetic and natural diamond in the range 7–11 MN $\text{m}^{-3/2}$. However, Novikov and Dub [16] have pointed out that the equation used by Naletov *et al.* for calculating K_{IC} overestimates the values obtained by a factor of 1.6; this comment was based on data from silicon where K_{IC} was measured by both the indentation technique and the more standard double-torsion method. Novikov and Dub [16] monitored the growth of cracks produced by a Vickers indenter in ⟨011⟩ directions along {011} planes and obtained values of about 5 MPa$\sqrt{\text{m}}$ for both synthetic and natural diamond crystals. Cleavage energies on (110) planes should be higher than on (111) planes by about 20% which would correspond to a higher K_{IC} value of about 10% (see Equation 1).

In 1996, Novikov and Dub [17] used a Vickers indentation to measure the fracture toughness of single crystal diamond obtaining values between 5 and 14 MPa$\sqrt{\text{m}}$. However, measurement of K_{IC} using the

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Vickers indentation technique often results in damage of the tip of the indenter with departure from the assumed stress field. Further, if the Vickers indenter is not oriented to produce cracks on the {111} cleavage planes the cracks are "stepped" as the crack seeks out the nearest cleavage plane. This gives a value for Ψ (see Equation 1) greater than unity. The spread in the data for the Novikov and Dub work is indicative of the problems.

Other researchers have used indentation techniques to measure K_{IC} and a range of values are quoted. For example, 6 to 7 MPa$\sqrt{\mathrm{m}}$ [18], 6.0 ± 1.5 MPa$\sqrt{\mathrm{m}}$ [2]. Drory and Gardinier [19] and Drory *et al.* [20] report a value of 5.3 ± 1.3 MPa$\sqrt{\mathrm{m}}$.

Jiang *et al.* [21] used three-point bending on free-standing CVD films and obtained a value of ~8 MPa$\sqrt{\mathrm{m}}$.

Preliminary work in this laboratory using the four-point bend test is described by Telling and Field [22] and Telling [23]. They found $K_{\mathrm{IC}} = 6.2 \pm 0.6\,\mathrm{MPa}\sqrt{\mathrm{m}}$ for a batch of di-electric (optical) grade CVD.

When indentation is used to determine strength, it has to be remembered that all indentation techniques are only representative of the small area tested and an average of many values is necessary for a reliable result [24, 25]. Furthermore, the strength value obtained depends on the indenter size [26, 27]. The work of Ruoff and Wanagel [28], Ikawa and Shimada [29] and Ikawa *et al.* [30] show much higher strengths are obtained when using small indenters. Interfacial frictional stresses can also be important [11, 31].

2. Experimental

2.1. The four-point bend test

A better method of ascertaining the fracture toughness involves initiating a crack in a thin plate by compressing the plate between four loading points. This is the four-point bend test, though the geometry employed is also referred to as the double torsion rig. Fig. 1 is a schematic of the test. Note the sample is grooved to maintain the crack propagation direction. A load cell records the force being applied to the lower load points with time, and the peak load at failure, P, can be used to calculate the fracture toughness, K_{IC}, using Equation 2 [32].

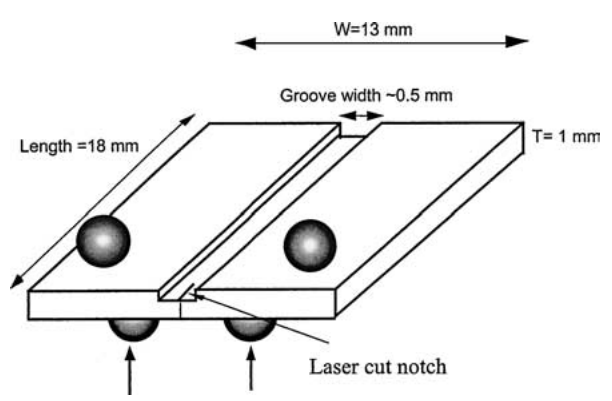

Figure 1 Schematic of the four-point-bend test apparatus. The groove depth is 0.21 mm. The distance between the outer and inner spherical contact points is 4 mm.

$$K_{\mathrm{IC}} = PD\sqrt{\frac{3}{Wt(1-\nu)\xi T^3}}, \qquad (2)$$

where D is the distance between inner and outer spherical contact points. W and T are the plate width and thickness, t is the plate thickness in the plane of the crack, ν is the Poisson ratio and ξ is a correlation factor determined from T and W.

$$\xi = 1 - 0.63\left(\frac{2T}{W}\right) + 1.2\left(\frac{2T}{W}\right)e^{\frac{\pi W}{2T}} \ldots \qquad (3)$$

The effective flaw size may be determined if both the strength, σ, and the toughness, K_{IC}, of a material are known, from Equation 4 [33] assuming the flaw has a half-penny geometry. Conversely, for materials of a given flaw size, C, the fracture toughness can be used to measure the strength, or so-called critical fracture stress.

$$C = \left(\frac{K_{\mathrm{IC}}}{1.24\sigma}\right)^2 \qquad (4)$$

2.2. Measurements of K_{IC} on CVD diamond

The fracture toughnesses of mechanical and di-electric (optical grade) CVD diamond were ascertained using the four-point-bend apparatus illustrated in Fig. 1. Both the mechanical grade CVD diamonds and the di-electric (optical quality) CVD diamonds were provided by De Beers Industrial Diamonds Ltd (UK), now Element Six Ltd.

Before inserting the plates into the test rig, a 3 mm long, ~30 μm wide starter flaw was laser cut all the way through one end of each specimen. The lower two loading points were then moved upwards, resulting in tension at the starter flaw. The lower load points were driven by a micrometer thread driven piston, as illustrated schematically in Fig. 2.

Since the loading geometry was symmetric about the central starter flaw, the crack tended to propagate through the middle of the sample. A deep groove was laser cut along the centre of each smaple, to ensure that the crack propagated the entire length. The depth of the

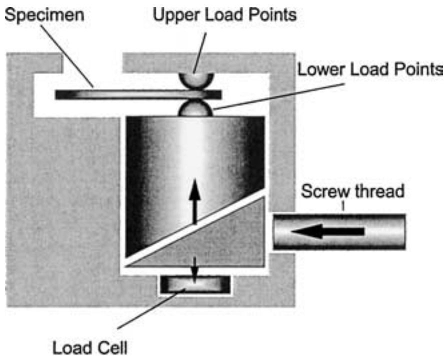

Figure 2 A cross-section schematic of the double torsion rig.

guiding groove was measured using digital calipers and confirmed using a calibrated optical microscope.

The increments in loads applied to the sample by the lower load points were measured using a Novotech F250 load-cell, located underneath the wedge that drove the piston. This was made possible by the geometry of the wedge, which applied an equal load to the piston and the load-cell. The load-cell, interfaced with a computer, was calibrated using an Instron Universal Testing Machine, model 4466.

3. Results

3.1. Calibration using alumina

The fracture toughness of 13 mm by 18 mm alumina plates was determined to ensure that the apparatus was well-calibrated. A 150 ± 50 μm groove was engraved along the centre of the 1 mm thick alumina. The load cell recorded the force being applied to the lower load points every second, and the peak load at failure was recorded, P. This critical failure load was then used to calculate the fracture toughness of alumina, K_{IC}, using Equation 2. The fracture toughness of alumina was measured to be 3.6 ± 0.4 MPa. This value compares well with those obtained by Telling [23] and McColm [34] who recorded values of 3.7 ± 0.2 MPa and 3.5–4.1 MPa, respectively. This gave confidence in the test method.

3.2. CVD diamond

The process was repeated for three mechanical and seven di-electric CVD diamond plates, each having a central groove that was 0.21 ± 0.1 mm deep; see Table I. However, one of the mechanical grade and two of the dielectric plates gave invalid results because the fracture failed to nucleate at the starter crack and the resulting catastrophic failure occurred at high load. A typical load to failure curve is shown in Fig. 3.

The average fracture toughness of mechanical grade CVD diamond was calculated to be 8.5 ± 1 MPa$\sqrt{\mathrm{m}}$. This value agrees reasonably with that given by Jiang *et al.* [21], who determined the fracture toughness of free-standing CVD films using three-point-bending to be $\sim$8 MPa$\sqrt{\mathrm{m}}$. The average fracture toughness of di-electric grade CVD diamond was 8.3 ± 0.4 MPa$\sqrt{\mathrm{m}}$. Earlier research in this laboratory on a different batch of di-electric grade CVD diamond plates found $K_{\mathrm{IC}} = 6.2 \pm 0.6$ MPa$\sqrt{\mathrm{m}}$ [22, 23].

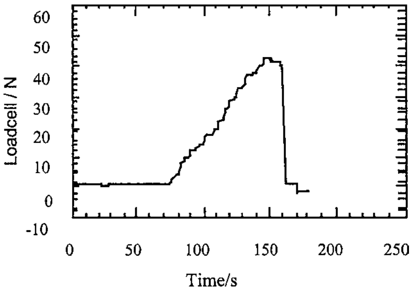

Figure 3 A typical load to failure curve. The sample in this case was mechanical grade CVD.

TABLE I Fracture toughness of CVD diamond

Grade of CVD diamond	Fracture toughness (MPa$\sqrt{\mathrm{m}}$)
Mechanical grade	10.6[a]
Mechanical grade	7.7
Mechanical grade	9.2
Average mechanical grade	8.5 ± 1.0
Di-electric grade	8.05
Di-electric grade	7.95
Di-electric grade	8.90
Di-electric grade	7.80
Di-electric grade	8.60
Average di-electric (optical) grade	8.3 ± 0.4

[a] Not used; see text.

TABLE II Fracture toughness of CVD diamond at 500 ± 50°C

	Fracture toughness(MPa$\sqrt{\mathrm{m}}$)
Di-electric grade	9.2
Di-electric grade	7.8
Average	8.5 ± 1

3.3. Fracture toughness of CVD diamond at 500 ± 50°C

The toughness of di-electric grade, CVD diamond was measured using the four-point bend test while the samples were exposed to the flame of a butane gas torch. Thermocouples on the sample and the load cell confirmed that the sample was at 500 ± 50°C while the load remained close to ambient.

The values obtained for the two available samples are shown in Table II.

The fracture toughness value of 8.5 ± 1 MPa$\sqrt{\mathrm{m}}$ at 500 ± 50°C had a higher error margin than that measured at room temperature because fewer samples were tested but was not significantly different to the room temperature value of 8.3 ± 0.4 MPa$\sqrt{\mathrm{m}}$.

3.4. Direct measurement of crack path lengths

It is clear when comparing fracture paths in CVD diamond that they are more circuitous (rougher) than a cleavage crack in gem-quality diamond. As noted by various workers, the fracture paths in CVD diamond are a mixture of intergranular and transgranular fracture [35] and with no suggestion that the grain boundaries are inherently weaker than the diamond grains. However, the boundaries and grain structure will cause cracks to divert producing rougher surfaces. We have quantified this by using optical microscopy to view cracks edge on. The ratio of actual path length to direct path length varies between samples of different grain sizes but is typically in the range 1.3 to 1.6. Cross-sections across fracture fronts gave similar values. Surface roughness may not be the only factor explaining CVD diamonds higher K_{IC} compared with gem-quality diamond but it is clearly a dominant factor.

4. Conclusions

The cost of diamond samples is high which explains why indentation tests with either "sharp" or "blunt" indenters have been traditionally used for strength and toughness measurements. However, as noted earlier a problem with using indentation for a strength test is that only small volumes are stressed. For K_{IC} measurement for diamond with a sharp indenter, the results can be affected by the quality of the tip of the indenter and its orientation. The present workers were fortunate to have plate samples which allowed use of the four point bending test (or double torsion) geometry.

The average fracture toughness of mechanical grade diamond was found to be $8.5 \pm 1.0\ \mathrm{MPa}\sqrt{\mathrm{m}}$ and $8.3 \pm 0.4\ \mathrm{MPa}\sqrt{\mathrm{m}}$ for di-electric (optical) grade CVD. This is consistent with the mechanical grade having more flaws and imperfections [5] which deflect the crack path and give increased surface roughness.

In general, all CVD material has a higher toughness than the value of $3.4\ \mathrm{MPa}\sqrt{\mathrm{m}}$ obtained by Field and Freeman [1] for good quality natural diamond. The grain boundaries in CVD diamond and different grain oritentations deflect the crack path causing increased surface roughness which increases K_{IC}. Attempts to quantify the increased surface roughness suggest that this must be a key factor in determining the toughness of CVD diamond. The role of residual stresses needs evaluation since these are also likely to increase fracture surface toughness. The fact that CVD diamond is 2 to 3 times tougher than natural diamond is important for various applications.

Only two samples were available for the experiments at 500 ± 50°C. However, they showed that the fracture toughness was not significantly different. This is of practical importance since various potential applications of CVD diamond involve temperatures of this order.

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