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Filamentary Sapphire

Part 2 *Fracture Strength in Tension*

J. T. A. POLLOCK*
Tyco Laboratories Inc, Bear Hill, Waltham, Massachusetts, 02154, USA

The fracture strength in tension of nominally 0.026 cm diameter *c*-axis sapphire filament grown from the melt using the "edge-defined, Film-fed Growth" technique is presented. The strength is found to be a function of filament diameter, decreasing with decreasing diameter. This relationship is explained on the basis of the particular characteristics of the melt growth preparation method. A melt temperature controlling device, dependent on the infra-red emitted from the melt area at which the filament is withdrawn, was developed and used to grow filament of constant diameter in the growth rate range 2.5 to 7.5 cm/min. With this device strength is not found to vary significantly with growth rate. Average strengths in tension of 2.75×10^9 N/m^2 (400×10^3 lb f/in.^{2}) are reported using this temperature controlling method compared with 2.5×10^9 N/m^2 (360×10^3 lb f/in.^{2}) for similar diameter material grown using a constant power mode system.

1. Introduction

Sapphire is a high melting point oxide with substantial strength and hardness characteristics and, because of its thermodynamic compatibility with many metals and alloys, is of importance as a future structural reinforcement in composite materials. Using the "Edge-defined, Film-fed Growth" process (EFG) [1-5] an experimental characterisation programme, aimed at determining the optimum growth conditions for high tensile strength filamentary sapphire, has been completed. Part 1 of this programme [6] involved a visual study of the growth characteristics of *c*-axis sapphire filament in the growth rate range 2.5 to 7.5 cm/min using still and 16 mm motion photography. Characterisation of the microstructure using transmission metallographic techniques was also carried out. The present paper is concerned with tensile strength characterisation directly related to part 1. More recent data describing the tensile strength of material grown after incorporating some experimental changes are also presented.

2. Experimental

The experimental procedure involved the continuous growth of *c*-axis filamentary sapphire, in the growth rate range 2.5 to 7.5 cm/min, using the apparatus described in part 1. Melt temperature and growth rate changes were made without interrupting the continuity of growth during a given experiment. This continuity is of importance since minor orientation changes, and associated variations in Young's modulus [7] might be introduced after reseeding. Four feet of filament were grown for each set of growth rate and melt temperature conditions. Melt temperature increases at constant growth rate were limited such that minimum filament diameters were ~ 0.02 cm.

Two series of growth runs were completed using these growth procedures. These series differed only in the method used automatically to regulate the power supplied to the rf heating coil after a power level had been manually selected. The first series made use of an rf pickup coil to ensure constant current supply to the rf heating coil. The sensing coil operated by detecting changes in the rf heating coil field and inducing compensating adjustments in the 20 kW power generator. On the basis of the experimental observations described in part 1 and the fracture strength data obtained with filament grown using the latter constant power mode

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*Present address: AAEC Research Establishment, Materials Division, Lucas Heights, Sutherland 2232, New South Wales, Australia.

regulating system, another power control system based on detection of the infra-red spectrum emitted from the growth area was introduced. Details of this latter system and the reasons for its introduction will be presented in section 3.2, as well as fracture strength data for filament grown using it.

The lengths of filament were carefully broken off as they cleared the pulling belt and, although usually coated with paraffin, placed on foam rubber pads as a precaution against surface degradation. At least three samples were taken from the centre of these labelled lengths and after beading in an oxygen/hydrogen flame, fixed in cardboard tabs using epoxy in a form suitable for tensile testing. A 1 in. gauge length and strain rate of 5×10^{-3} min^{-1} was used to determine the tensile fracture stress of the samples.

3. Results and Discussion

3.1. Sapphire Filament Grown Using Constant Power Mode Temperature Control

Four growth runs of *c*-axis filament were completed using growth rates in the range 3.0 to 5.1 cm/min. Table I presents data relating the tensile fracture stress, temperature measured at the orifice and filament diameter for one of these growth runs. The data for other runs follow a similar trend, and indicate that for constant growth rate, increasing filament strength is measured with increasing filament radius, i.e. decreasing orifice tip (i.e. melt) temperature. Typically, the strength variation during a growth run would fall within the range 2.1 to 2.6×10^9 N/m^2 (300 to 375×10^3 lb f/in.^{2}).

This relationship of fracture stress with filament diameter is opposite from the relationship normally reported for brittle materials like sapphire, where surface and volume flaws play controlling roles in determining the fracture strength. However, two important parameters are changing during the growth of decreasing diameter sapphire filament from a constant diameter molybdenum orifice using EFG. The first is the possibility of increasing dissolution of Mo in the Al_2O_3 melt due to the higher melt operating temperature, the latter being reflected in the increasing orifice tip temperature. In extreme cases of overheating, 10 to 15 ppm Mo have been detected in filament which was up to 7×10^7 N/m^2 (100×10^3 lb f/in.^{2}) weaker in tension testing to fracture, than similar diameter material grown under conditions where the Mo content was less than 5 ppm. Metallographic investigations have indicated that this excess Mo tends to aggregate in small clusters near the surface of the filament. However, quantitative analysis of the sapphire grown and discussed in the present work indicated that the Mo content was less than 5 ppm. A second changing parameter of importance is the increasing meniscus film height which accompanies decreasing filament diameter. Details of this relationship at many growth rates are presented in part 1. Two photographs of the growth region (figs. 1 and 2) serve to illustrate the extreme cases. The controlling effect of the orifice edge on the geometric integrity of the filament decreases with

TABLE I Typical tensile fracture stress data for *c*-axis filamentary sapphire grown continuously at 4.4, 3.4 and 3.8 cm/min, using constant power mode temperature control.

Growth speed, cm/min	Growth temperature measured at orifice tip (°C)	Filament diameter cm	Average tensile fracture stress N/m^2	(lb f/in.^{2})
4.4	2060	0.024	2.62×10^9	(380×10^3)
4.4	2080	0.023	2.26×10^9	(327×10^3)
4.4	2120	0.023	2.35×10^9	(339×10^3)
4.4	2140	0.021	2.32×10^9	(336×10^3)
4.4.	2160	0.024	2.31×10^9	(335×10^3)
4.4	2170	0.019	2.28×10^9	(331×10^3)
3.4	2120	0.024	2.32×10^9	(336×10^3)
3.4	2180	0.021	2.15×10^9	(311×10^3)
3.8	2150	0.024	2.24×10^9	(325×10^3)
3.8	2120	0.026	2.37×10^9	(343×10^3)
3.8	2090	0.026	2.66×10^9	(385×10^3)

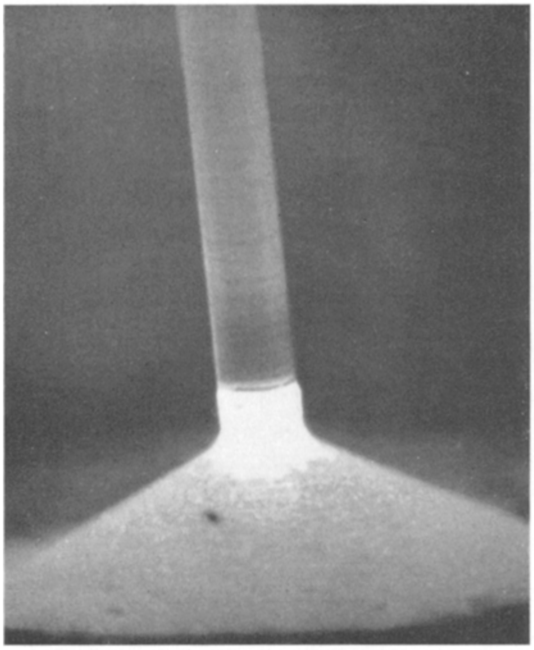

Figure 1 Photograph of growth region showing sapphire filament growth from a thin liquid meniscus film. Growth rate: 5.0 cm/min. Growth temperature: 2040°C. Film thickness: ~0.0006 cm.

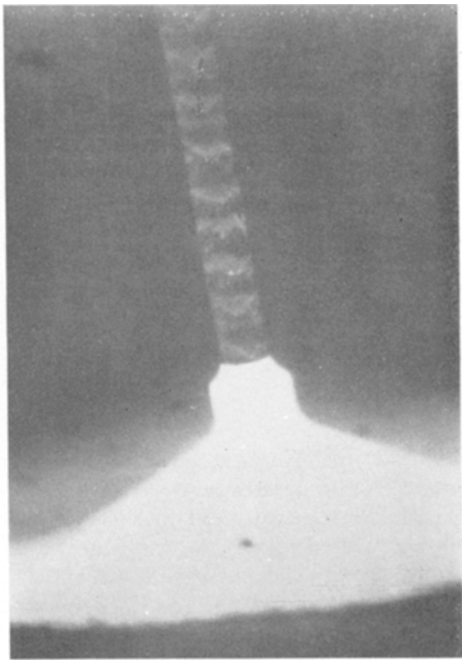

Figure 2 Photograph of growth region showing sapphire growth from a thick liquid meniscus film. Growth rate: 3.3 cm/min. Growth temperature: 2135°C. Film thickness: ~0.007 cm.

increasing meniscus height. The liquid sapphire in the meniscus becomes less viscous with increasing temperature [8] and filament movement in the furnace due to thermal convective currents can result in surface ripples. Surface striations produced by physically vibrating the filament while it is being withdrawn are easily observed. Also, it has been noted that with decreasing filament diameter, the variation in filament diameter increases. For example, 0.025 cm diameter filament will typically show diameter variation of less than 1% compared with greater than 5% for filament of 0.020 cm diameter. Thus, it is felt that the observed decrease in strength with decreasing filament radius is caused by a less smooth filament surface resulting from the loss of edge definition by the outside of the orifice.

Employing the data obtained over all four growth runs, tensile fracture stress versus growth speed is shown plotted in fig. 3, curve A. The bar range represents the standard deviation obtained over all the data for a given speed (12 to 16 samples). A small improvement in strength occurs with increasing growth speed and appears to be maximum in the region 3.8 to 4.4 cm/min. If filament of constant diameter is considered, an essentially similar relationship is observed. For example, when data for filament of ~ 0.026 cm diameter is treated, a maximum in tensile strength of 2.5×10^9 N/m^2 (360×10^3 lb f/in.^{2}) is obtained for filament grown at 4.4 cm/min. This is a more correct method of determining the relationship between growth rate and strength,

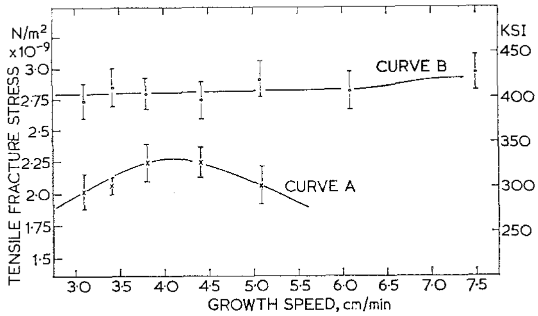

Figure 3 Tensile fracture stress versus growth rate. Curve A. Sapphire filament grown using constant power mode temperature regulation. Curve B. Sapphire filament grown using infra-red sensor temperature regulation.

since the surface area and volume of sapphire is constant for each fracture stress determination. Thus, variations in strength as a result of growth speed related fracture sources should be more readily detected.

The importance of comparing filament of constant diameter grown at different growth rates and the strength versus diameter relationship reported above, were directly responsible for the series of experiments to be described in section 3.2. Additional information was also provided by part 1 of this investigation, which made use of still and motion photography to study the growth process at various growth rates and temperatures. Examination of results of this study showed that the meniscus becomes increasingly turbulent with increasing growth temperature. This turbulence is reflected in the microscopic void distribution within the filament which, in the absence of surface defects, undoubtedly plays a role in determining the filament fracture strength. Details of fractographic investigations, supporting an internal fracture origin hypothesis in high strength filament, are presented elsewhere [5, 9].

3.2. Sapphire Filament Grown Using Infra-red Sensor Temperature Control

The desirability of maintaining the filament diameter constant during growth rate variations, coupled with operating evidence that the rf sensor control coil was not sufficiently sensitive, led to the use of a temperature measuring device utilising the infra-red portion of the spectrum. This device can be operated in various temperature ranges up to 2800° C and may be focused on an area as small as 0.10 cm diameter. The device was used by focusing it through a windowless port onto the area at and surrounding the molten meniscus from which the filament is withdrawn. As can be observed from figs. 1 and 2, the emissivity of liquid sapphire in the visible range is considerably higher than that of either solid sapphire or the Mo orifice ($\epsilon_{2000°C} \simeq 0.4$). The same relationship is generally true for the infra-red range. Also, as shown in part 1 [6] the emitting surface area of the meniscus region typically increases from 1.8×10^{-4} cm^2 to 6.2×10^{-4} cm^2 as the film thickness increases from 1.6×10^{-3} to 7.1×10^{-3} cm, respectively. Thus, the increasing heat of solidification per minute accompanying increasing growth rate, coupled with a constant power input to the rf coil, results in a thicker sapphire meniscus and small diameter filament, i.e. the level of infra-red emitted increases.

The signal generated by detecting the infra-red emitted from the growth area was used automatically to control the power input to the rf heating coil in a closed loop operation. Fig. 4 shows chart recordings for trial experiments during which three increases in growth speed, with their associated heats of solidification per minute, were made. In the first test (curve A) the detector was used to control the power level to the rf coil. The linearity of curve A (excepting background noise) indicates that by reducing the power input to the rf coil, and thus reducing the melt temperature, a constant infra-red detection level, i.e. constant meniscus height and filament diameter, was maintained during the growth rate changes. The sensitivity of the device was completely demonstrated in a second test (curve B) by disconnecting it from the controlling circuit, whereupon growth rate variations and concommitant meniscus height changes were observed to produce large changes in the level of infra-red detected.

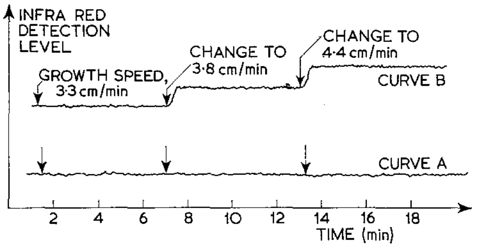

Figure 4 Chart recordings of infra-red sensor output as a function of growth rate. Curve A. Sensor used in closed loop control circuit to maintain meniscus height and filament diameter constant. Curve B. Sensor used simply to measure changes in infra-red level associated with changes in growth speed.

Three growth runs employing growth rates in the range 2.5 to 7.5 cm/min were completed using the infra-red device to control the melt temperature such that filament of constant diameter was grown. Data for one of these runs, typical of all three, are given in table II. It is apparent that the filament diameter (cross-sectional area) increased during growth at the first growth speed selected (5.1 cm/min). This is the result of a stabilising time, during which some Al_2O_3 spills over from the top of the orifice and increases the emissivity of the orifice/meniscus

TABLE II Typical tensile fracture strength data for *c*-axis sapphire grown continuously at 5.1, 4.4, 3.8, 3.4, and 3.0 cm/min, using an infra-red sensor as temperature controller.

Growth speed, cm/min	Filament diameter, cm	Average tensile fracture stress	
		N/m^2	(lb f/in.^{2})
5.1	0.0251 to 0.0256	3.1×10^9	(451×10^3)
4.4	0.026	2.77×10^9	(403×10^3)
3.8	0.026	2.71×10^9	(394×10^3)
3.4	0.026	2.75×10^9	(440×10^3)
3.0	0.026	2.74×10^9	(398×10^3)

region viewed by the infra-red detector. Thus, the controller causes the heat input to the crucible via the rf coil to decrease, and for a given growth rate a slightly large filament diameter is grown. Except for the filament grown during this settling period, the diameter was close to 0.026 cm during each of the three growth runs.

The average fracture stress over all the data (50 samples) was 2.82×10^9 N/m^2 (410×10^3 lb f/in.^{2}) with a standard deviation of 1.17×10^7 N/m^2 (17×10^3 lb f/in.^{2}). Fracture strength versus growth rate, averaged over all the data, is plotted in fig. 1, curve B. No continuing trend with growth speed is clear, an average fracture strength in tension of ~ 2.75 N/m^2 (400×10^3 lb f/in.^{2}) being measured irrespective of growth rate. Comparison of filament strength with that grown using the rf sensor coil as melt temperature controller indicates that a significant strength enhancement is obtained. For filament of 0.026 cm diameter, this strength increase ranges from $\sim 2.7 \times 10^7$ N/m^2 (40×10^3 lb f/in.^{2}) at 4.4 cm/min to 10.3×10^8 N/m^2 (150×10^3 lb f/in.^{2}) at 5.1 cm/min .

4. Conclusions

Using a constant power mode temperature control method, it is found that at constant growth rate, the tensile fracture strength of *c*-axis sapphire filament decreases with decreasing filament diameter in the range 0.026 to 0.020 cm. The latter dimension is controlled by the height of the molten meniscus film from which the filament is withdrawn, an increasing meniscus thickness and decreasing filament diameter accompanying melt temperature increases. The decrease in strength is considered to be related to a less perfect microstructure resulting from turbulence at the growth interface and decreasing edge definition from the orifice which attend increasing meniscus thickness. Average fracture strengths in the range 2.1 to 2.5×10^9 N/m^2 (300 to 360×10^3 lb f/in.^{2}) were measured, the maximum value being obtained at a growth rate of $\sim$ 4.4 cm/min.

Filament of constant 0.026 cm diameter was grown employing a temperature controlling device dependent on the infra-red emitted from the growth region. The fracture strength of this filament was not found to vary significantly with growth rate. Average strengths in tension of $\sim 2.75 \times 10^9$ N/m^2 (400×10^3 lb f/in.^{2}) were measured, representing a 2.7×10^7 N/m^2 (40×10^3 lb f/in.^{2}) enhancement over the maximum strength of similar diameter material grown under constant power mode temperature control.

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References

1. H. E. LABELLE, JUN and A. I. MLAVSKY, *Mater. Res. Bull.* 6 (1971) 571.
2. H. E. LABELLE, JUN, *ibid* 6 (1971) 581.
3. B. CHALMERS, H. E. LABELLE, JUN, and A. I. MLAVSKY, *ibid* 6 (1971) 681.
4. H. E. LABELLE, JUN and J. T. A. POLLOCK, *ibid* to be published,
5. H. E. LABELLE, JUN and G. F. HURLEY, *SAMPE* **6** (1970) 17.
6. J. T. A. POLLOCK, *J. Mater. Sci.* 7 (1972) 631.
7. J. B. WACHTMAN, W. E. TEFFT, D. G. LAM, and R. P. STINCHFIELD, *J. Res. Nat. Bur. Stand.* **64A** (1960) 213.
8. R. P. NELSON, Battelle North-West, private communication.
   $\mu = 0.7$ poise at the melting point, 2057°C.
   $\mu = 0.4$ poise at 2650°C
9. J. T. A. POLLOCK and G. F. HURLEY, submitted for publication.

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