

# Fabrication, properties and usage of single-crystalline YAG fibres

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## Abstract

Single crystalline YAG fibres are now produced by an internal crystallization method. Essentially, the method is crystallization of the oxide melt infiltrated into continuous channels made in an auxiliary matrix, normally molybdenum, and then extracting the fibre from the auxiliary matrix by chemical dissolution of it. The method is relatively simple and requires sufficiently small energy input into a real process of the fibre production. Because crystallising a batch of the fibres by using ICM is actually similar to making bulk crystals an expected cost of ICM fibres is of the same order of magnitudes as that of bulk crystals. It is shown that usage of ICM–YAG-fibres in a Ni-based matrix yields composites with high fibre/matrix interface strength and, hence, high creep resistance at very high temperatures for Ni-based materials. © 2002 Published by Elsevier Science Ltd.

*Keywords:* Fibres; Heat-resistant material; YAG;  $Y_3Al_5O_{12}$

## 1. Introduction

Single-crystalline and eutectic oxide fibres shall inevitably be a most important reinforcement for the family of heat-resistant materials composed of metal-, intermetallic- and ceramic–matrix composites. This is true despite experimental data on composites with sapphire fibres published up to now yield rather controversial conclusions especially while discussing metal- and intermetallic matrix composites. Asthana and Tewain<sup>1</sup> reviewed data on sapphire fibre strength and interfacial strength in sapphire-fibre/Ni-based-matrix composites. They concluded that (i) the interaction of alumina with Ni-based melt yielded a drastic decrease in strength characteristics of the fibres extracted from the matrix and (ii) the interface strength was influenced by a number of the factors. It has been also shown that doping a Ni-based matrix by Yb (and, perhaps, yttrium as well as other rare-earth elements) causes an increase in the interface strength. This is an important observation since to make use of high fibre strength characteristic for short fibre length; sufficiently high interface strength in brittle-fibre/ductile-matrix composites has to be provided.<sup>2</sup> Observing an influence of rare-earth metals in the matrix on the interface strength, which relates cer-

tainly to a rather old observation of better adherence of complex oxides containing rare-earth metals to the surface of Ni-based alloys, leads to the idea of using fibres of complex oxides as reinforcement for Ni-based matrices. YAG ( $Y_3Al_5O_{12}$ ) can be considered as an appropriate candidate for such a purpose. Another possible application of YAG fibres in structural materials is reinforcement in oxide-fibre/oxide-matrix composites. In this application, higher possible temperatures of the usage can be expected with YAG fibre than those with sapphire fibre due to higher creep resistance of the former.

Burrus and Stone<sup>3</sup> were certainly the first to report fabricating single crystalline YAG fibre by using the laser heated pedestal growth (LHPG) method. They grew Nd-doped fibre of a diameter of 50  $\mu\text{m}$  and length of about 200 mm for laser applications. However, the usage of such fibres in structural applications, similarly to other oxide fibres grown by the EFG method, launched certainly by LaBelle and Mlavsky,<sup>4</sup> will be prevented by high cost. A recent development of YAG fibres of a diameter between 0.5 and 2  $\mu\text{m}$  and a length up to 550 mm by using the micro-pulling-down method with Ir crucibles and RF heating by Chani et al.<sup>5</sup> can also hardly be used for producing fibres for structural applications for the same reason.

Under these circumstances, a way to produce single-crystalline and eutectic oxide fibres based on the internal crystallization method (ICM) looks a promising

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starting point for the development of a fibre technology aimed at structural applications. The ICM is crystallization of the oxide melt infiltrated into continuous channels made in a auxiliary matrix, i.e. molybdenum one,<sup>2,6</sup> and then extracting the fibre from the auxiliary matrix by chemical dissolution of it.<sup>7,8</sup> The method is relatively simple and requires sufficiently small energy input into a real process of the fibre production. The method allows us to obtain a variety of the oxide fibres, such as sapphire of a homogeneous crystallographic orientation<sup>8</sup> alumina-YAG eutectic,<sup>7</sup> mullite<sup>9</sup> and some others.

In the present paper, growth of single crystalline YAG fibres by using ICM, their strength characteristics and a possible usage in heat resistant composites are described.

## 2. Fibre fabrication

The method of internal crystallisation includes the following fabrication steps:

1. Formation of continuous cylindrical channels in an auxiliary matrix.
2. Infiltration of the channels with a melted fibre material.
3. Crystallisation of fibres in the channels.
4. Extraction of either a loose or bounded fibre bundle from the auxiliary matrix.

The auxiliary matrix, which is a molybdenum block with continuous channels, is obtained by diffusion bonding an assemblage of molybdenum wire and foil. A cross-section of the block is presented in Fig. 1. All details of the fabrication process to produce such a block of the auxiliary matrix are given in Refs. 6–8.

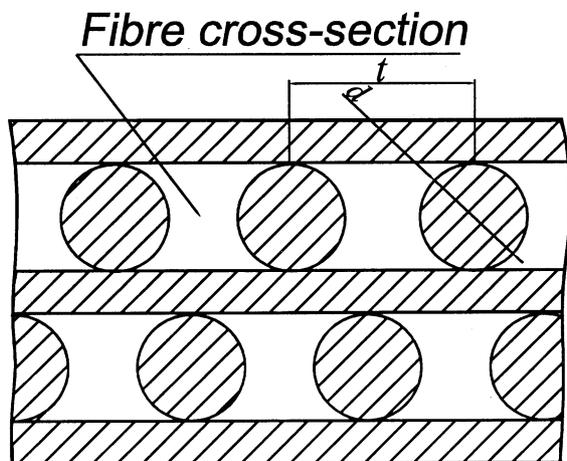


Fig. 1. A cross-section of the molybdenum carcass; the fibre cross-section of a future fibre is shown.

Infiltration of the channels with a melted fibre material and crystallisation of fibres in the channels are the most important steps of the whole procedure. The infiltration occurs due to capillary forces. A particular configuration of the *molybdenum block*—a *crucible*, containing raw material,—a *seed* of the determining crystallographic orientation of the fibres to be grown can vary. One such configuration was disclosed in a description of growth process of sapphire fibres.<sup>8</sup> Here we present a configuration without a crucible, in which case the seed contains enough material to crystallise a whole batch of the fibres, Fig. 2. YAG and YAG:Nd single crystals produced by the conventional Chochralsky technique were used as seed crystals. The seed is oriented in such a manner as to provide the  $\langle 111 \rangle$  orientation to the fibre axis.

Fig. 2 illustrates a sequence of processes at the fibre crystallisation. First, a susceptor/molybdenum-block/seed system (the first picture from the left) is heated up to the beginning of melting of the YAG seed at a zone of the seed/matrix-block contact. At a temperature just above the melting point, the melt infiltrates the channels of the molybdenum block (the second picture). After the whole block has been infiltrated (the third picture), the fibre growth starts with pulling up the matrix/seed system (the fourth picture). Crystallization rate can be from 15 to 25 mm/h. Crystallographic growth axes are determined by pre-oriented seed crystals. Moving the matrix/seed system to a cold zone of the furnace makes a solid/liquid interface in each channel move from the top to the bottom and the pulling-up is continued until the fibres along the whole channel length have been crystallised (the last picture). The molybdenum block with YAG fibres is then cooled for 4–6 h.

The infiltration and crystallization procedures are conducted in either a commercially available universal crystallising machine CRYSTAL or a special machine, CHIR, built for the particular aim of fabricating ICM-fibres. CRYSTAL has the process zone with an 8 kHz-induction-heated-graphite-susceptor/molybdenum-crucible set-up. The zone limits the length of the molybdenum carcass to about 100 mm. The CHIR's process zone with a radiant tungsten heater allows dealing with molybdenum blocks up to about 250 mm. An ambient atmosphere in CRYSTAL is argon gas under a pressure just above 1 atm. CHIR has a vacuum chamber. While dealing with 5 and 8 molybdenum blocks in one process, 120–150 g of fibres can be produced in the CRYSTAL machine. Productivity rate of CHIR is about one order of magnitude higher. Because crystallising a batch of the fibres by using ICM is actually similar to making bulk crystals an expected cost of ICM fibres is of the same order of magnitude as that of bulk crystals.

Extraction of the fibres after crystallization is achieved by dissolving the molybdenum matrix in acid

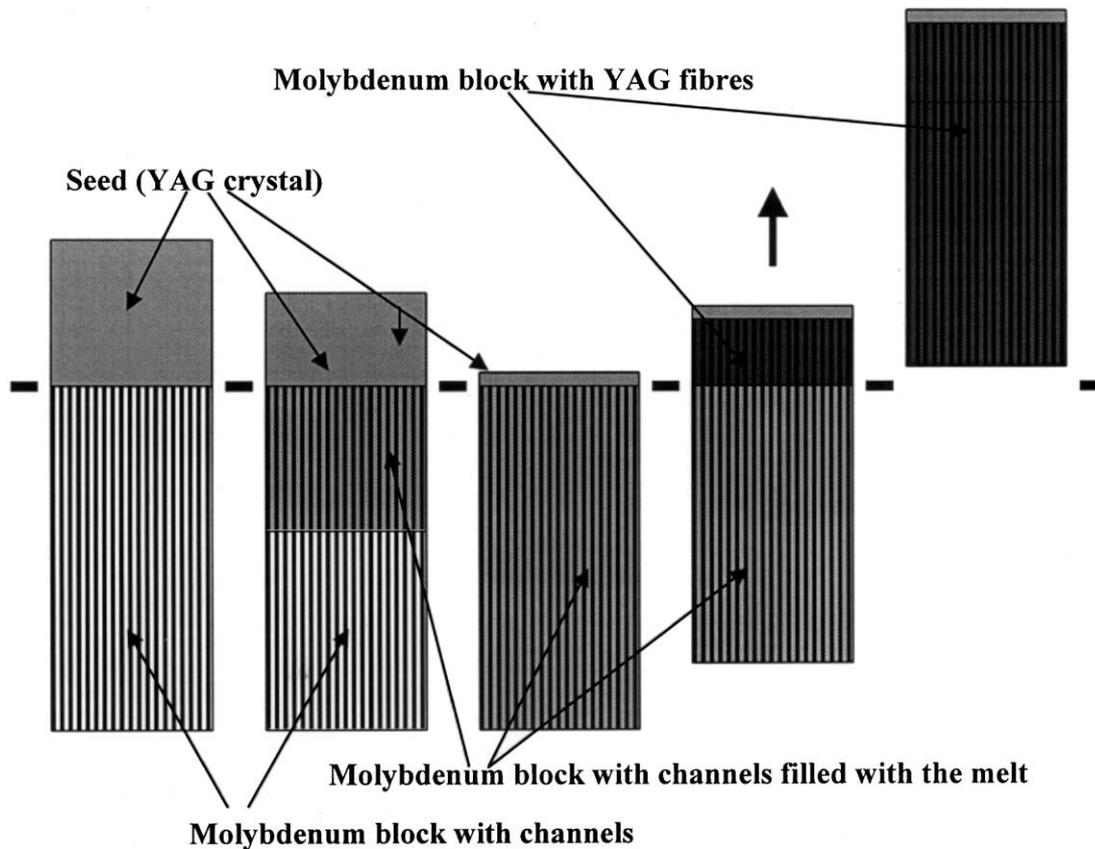


Fig. 2. Schematic of the experimental arrangement for crystallising homogeneously oriented YAG fibres without using a crucible.

mixtures. YAG fibres obtained by this method are shown in Fig. 3.

### 3. Mechanical characterisation

A special shape of the fibre cross-section, Fig. 1, makes measuring the tensile strength of the fibres in a straightforward way impossible. Hence, a kind of testing based on fibre bending seems to be appropriate. A procedure to measure bending strength of ICM-fibres is described in detail elsewhere.<sup>10</sup> A fibre of height  $d$  is looped over the rigid cylinder of a sufficiently large radius,  $R$ . Then the number of fibre breaks is counted. If the number of breaks is small enough, that means that the ratio of an average distance between neighbouring breaks to a characteristic size of the fibre cross-section is larger than 10, which ensures a possibility of neglecting the end effects, then the fibre is bent over the cylinder of a smaller radius. A new total number of the breaks are counted. The process is repeated until the average distance between the breaks becomes less than about  $10d$ .

The maximum fibre stress corresponding to the rigid cylinder radius is calculated according to

$$\sigma = E \frac{d}{2R} \quad (1)$$

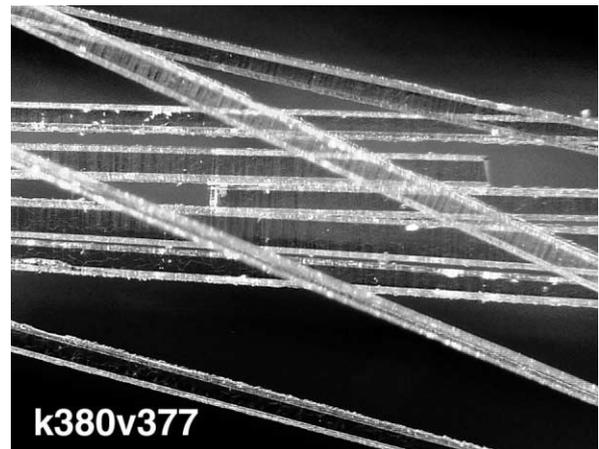


Fig. 3. YAG fibres.

where  $E$  is the Young's modulus of the fibre material. To a first approximation,  $\sigma$  can be assumed to be the fibre bending strength at a length equal to the average distance between the fibre breaks. Such a procedure obviously makes the fibre strength in a consequent analysis higher than the actual strength. However, using a series of the cylinders with a small increment of the radius makes the error small enough. Original experimental data obtained by testing fibres extracted from four YAG-fibre/molybdenum blocks are presented in Fig. 4.

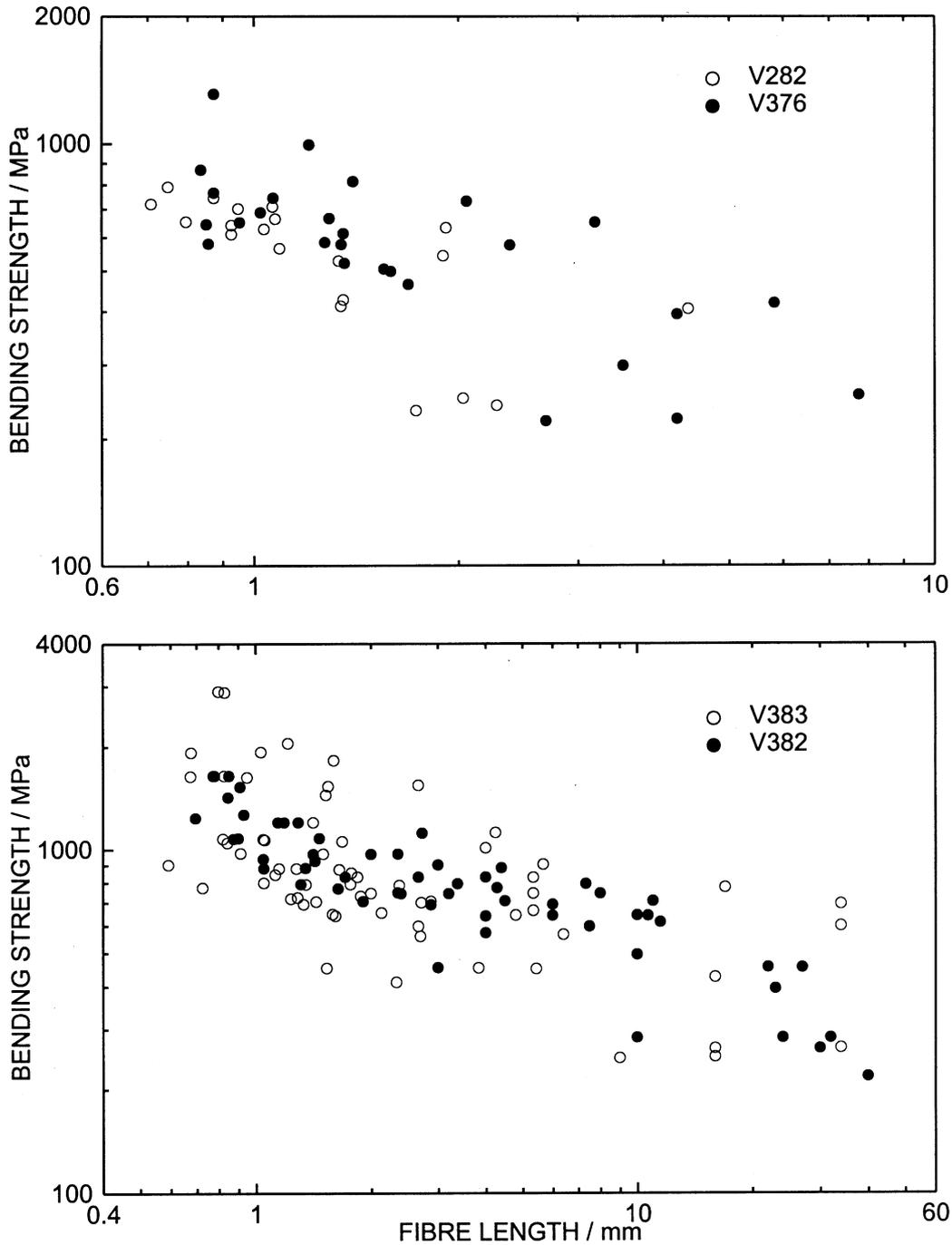


Fig. 4. Bending strength of the fibres of four batches versus distance between fibre breaks.

These data can be used to calculate the Weibull parameters to describe the fibre *tensile* strength via a cumulative distribution function of the form<sup>10</sup>

$$P(\sigma, g) = 1 - \exp\left(-\frac{g}{g_0} \left(\frac{\sigma}{\sigma_0}\right)^\beta\right) \quad (2)$$

where  $g$  is a geometrical parameter equal to either fibre length  $l$ , or fibre surface  $S$  or fibre volume  $V$  depending on a particular pattern of the characteristic defects location,  $g_0$  is a corresponding constant ( $l_0$ ,  $S_0$  or  $V_0$ ),  $\sigma_0$

and  $\beta$  are two other Weibull parameters. Obviously, one of the three parameters introduced can be chosen arbitrarily. Eq. (2) yields the average fibre strength of size  $g$  as

$$\langle\sigma(g)\rangle = \int_0^\infty \sigma dP(\sigma) = \sigma_0 \left(\frac{g}{g_0}\right)^\beta \Gamma\left(1 + \frac{1}{\beta}\right) \quad (3)$$

where  $\Gamma(\bullet)$  denotes the gamma-function.

It has been shown previously<sup>10</sup> that the *length* hypothesis does not fit experimental data; however, mechanical testing results do not allow us to decide

which hypothesis of the two (*surface* and *volume*) is true. Moreover, the presence of two populations of the defects that determine fracture of a fibre should not be excluded. Hence, one has to consider both cases.

In Ref.10, the strength scatter characterized by a set of fibres of a particular oxide-fibre/molybdenum-matrix block is used to calculate the Weibull exponent,  $\beta$ , which is assumed to be a characteristic value for the whole batch of the fibres. Then  $l^0$  is chosen as a constant characteristic length of the fibre. For a batch of the fibres,  $g$  will be either  $V_o = l^0 A_{av}$  or  $S_o = l^0 P_{av}$  where  $A_{av}$  and  $P_{av}$  are the average values of the cross-sectional area and perimeter for the fibre batch. The values of  $V_o$  and  $S_o$  are used in scale dependencies

$$\sigma^*(V) = \sigma_{oV}^b \left( \frac{V}{V_o} \right)^{-1/\beta}, \quad \sigma^*(S) = \sigma_{oS}^b \left( \frac{S}{S_o} \right)^{-1/\beta}$$

in which the Weibull parameters,  $\sigma_{oV}^b$  and  $\sigma_{oS}^b$ , can be obtained from experimental data. Also in Ref. 10, a procedure of calculation of the Weibull characteristics for tension based on either *volume* or *surface* hypothesis and an appropriate integration of Eq. (2) is outlined, which yields

$$\sigma'_{oV} = M_V(\beta)\sigma_{oV}^b, \quad \sigma'_{oS} = M_S(\beta)\sigma_{oS}^b$$

where  $M_V$  and  $M_S$  are given by

$$M_V = \left( \alpha - \frac{\pi}{4} \right)^{-1/\beta} \left[ \int_0^1 \xi^\beta \left( \alpha - \sqrt{1 - \xi^2} \right) d\xi \right]^{1/\beta},$$

$$M_S = \left( \frac{(\beta + 1)(\pi + 2\alpha)}{2} \right)^{-1/\beta}.$$

Here  $\alpha = t/d$  (see Fig. 1).

Applying this procedure to experimental data presented in Fig. 4. yields the Weibull parameters given in Table 1. The data obtained show that YAG fibres of the ICM type have room temperature strength which is strongly dependent on the length. This means that rather strict requirements to the fibre/matrix interface should be obeyed to make short fibre pieces to contribute

to the composite strength and creep resistance.<sup>2</sup> Experiments with ICM-fibres of sapphire<sup>8,11</sup> and alumina-YAG eutectic<sup>2,7,11</sup> have shown that coating a fibre with a thin layer of either metal, or carbide, or carbon yields an essential enhancement of their strength characteristics. This yields an important conclusion: an essential portion of defects determining the fibre strength is located on the fibre surface.

#### 4. Possible usage: reinforcing a Ni-based matrix

##### 4.1. Heat resistant metal matrix composites

Up to now, single crystalline YAG fibres under consideration have been used to reinforce Ni-based matrices. According to a previous experience described in Section 1, sufficiently good bonding of yttrium-containing fibres to a Ni-based matrix can be expected, which is necessary to exploit high strength of the fibres in the composites.

A series of composite specimens with two heat-resistant matrices described in Table 2 were obtained by using a pressure-casting technique. Details of the fabrication procedures are presented elsewhere.<sup>2,11</sup> The specimens are 65 mm in length and between 4 and 5 mm in diameter. The microstructure of a composite is illustrated in Fig. 5. A number of indications of rather intensive fibre/matrix interactions can be seen. In particular, the tips of the fibres, which had initially one straight side, become bowed (a fibre in the centre of the bottom); routes of the tips are thinned and, sometimes, the tip separates from the fibre (a fibre in the low left corner). Such interaction can yield both a degrading of a fibre and strengthening of the interface.<sup>12</sup>

Measuring the interface strength by using a modified push-out technique applicable to a fibre of non-circular cross-section<sup>13</sup> yields the results presented in Fig. 6.

It is interesting to emphasize two points:

1. The apparent interface strength depends on the disk thickness; this phenomenon is discussed in Ref. 13, the discussion yields a recommendation to use an average maximum value of the

Table 1  
Strength characteristics of single crystalline YAG fibres. The Young's modulus is taken as 392 GPa. Base fibre length is 1 mm

Block No.	Atmosphere in growth chamber	Pulling rate mm/h	Volume hypothesis				Surface hypothesis			
			$\beta$	$\sigma_{oV}^b$ MPa	$\sigma'_{oV}$ MPa	$V_o$ mm <sup>3</sup>	$\beta$	$\sigma_{oS}^b$ MPa	$\sigma'_{oS}$ MPa	$S_o$ mm <sup>2</sup>
V382	Vacuum	22	4.74	1013	728	.00911	4.74	681	352	3.930
V383	Argon	15	2.71	1225	796	.00889	2.74	693	260	2.810
V376	Argon	30	3.83	668	461	.00711	3.83	550	253	1.120
V282	Argon	15	4.57	575	407	.01041	4.63	532	265	0.894

Table 2  
Chemical composition of matrix alloys (Ni - bal.)

Trade mark of the alloy	Al	Cr	Mo	W	Ta	Co	Nb	Re	C	B	Ti
GS-32	6.0	5.0	1.0	8.3	4.0	9.0	1.5	4.0	0.015	0.015	
VKNA-4U	8.5	4.8	2.2	3.9		4.4		0.11			1.1

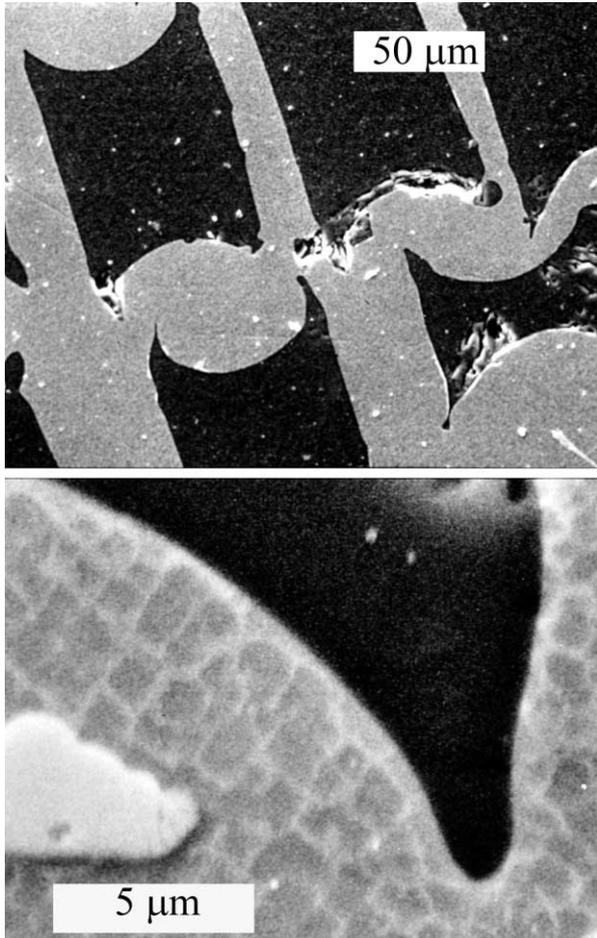


Fig. 5. Scanning electron micrographs of a YAG-fibre/GS-32-matrix composite specimen. Courtesy to Natalie Sarkissyan and M. Starostin.

apparent strength as an approximation to a real value of the strength. For the case under consideration, this value is  $\sim 200$  MPa.

2. A maximum value of the interface shear stress reaches 350 MPa, which has not been perhaps registered yet in oxide-fibre/Ni-based-matrix composites.<sup>1</sup>

Since ICM-fibres are characterised by a strong scale dependence (see Fig. 4), it is important to have high fibre/matrix interface strength in metal–matrix composites. This allows us to load strong short segments of the fibres, which yields high creep resistance. The quantitative evaluation of an effect of the interface strength

Table 3  
Estimated creep resistance (tensile stress to cause creep strain 1% for 100 h) of YAG-fibre/VKNA-4U-matrix composites at 1100 °C

Specimen No.	Fibre volume fraction	Creep resistance (MPa)
a1307	0.25	169
a13663	0.15	154

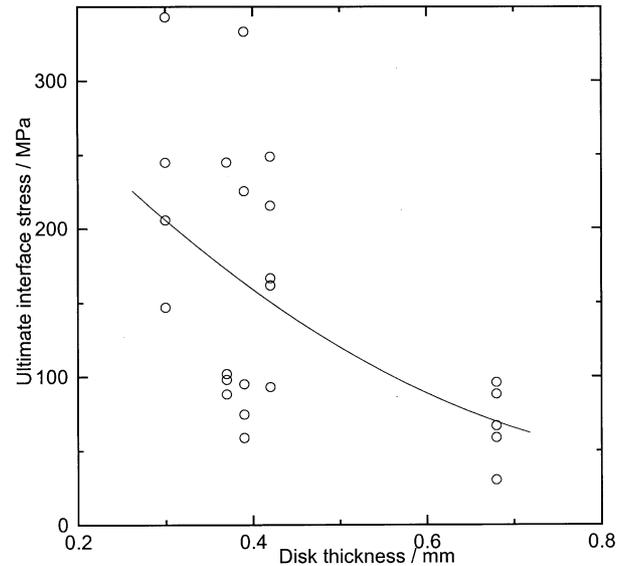


Fig. 6. Ultimate shear stress on the fibre/matrix interface in the YAG/VKNA-4U composite (Specimen a13662).

on creep resistance is given in Ref. 14 Preliminary experiments were conducted in bending in such a manner as to obtain creep characteristics of the composite by testing one specimen.<sup>14</sup> The creep strength of two composite specimens tested at 1100 °C is presented in Table 3. Obviously, a temperature of 1100 °C is a working temperature for this composite and the creep resistance of the specimens is higher than that of other tested composite systems.<sup>11</sup> Note that such values of the creep resistance are characteristic for modern super-alloys at temperatures around 1050 °C.<sup>15</sup> Prospective alloys now under development can hardly reach a working temperature of 1100 °C. Testing composites with a GS-32 matrix at higher temperature, 1150 °C, yields a result presented in Fig. 7. Since reinforcing a Ni-based matrix with oxide fibres gives a material with a density between 7 and 7.5 g/cm<sup>3</sup>, a temperature of 1150 °C seems to be a usage temperature for such composites.

#### 4.2. Other possibilities

Sapphire fibres produced by ICM were shown to be suitable for reinforcing polycrystalline alumina matrix.<sup>16</sup> This yields composites with enhanced fracture toughness

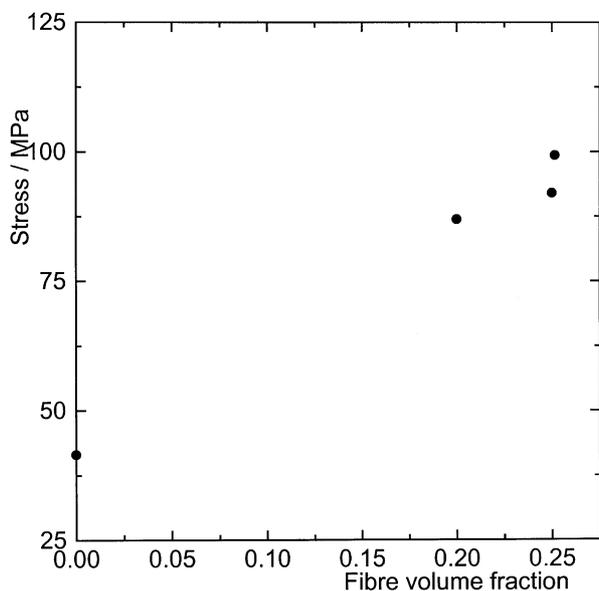


Fig. 7. Creep resistance (tensile stress to cause 1% creep strain for 100 h) at 1150 °C of YAG-fibre/GS-32-matrix composites obtained from bending tests versus fibre volume fraction.

value. There can be seen no obstacles to using YAG fibres in such applications.

The fibres are optically transparent (see Fig. 3) so they can also be used in applications where a combination of optical properties with high strength is essential.

## 5. Conclusions

Single crystalline YAG fibres are now produced by an internal crystallization method that can yield sufficiently non-expensive materials.

It is shown that use of ICM–YAG-fibres in a Ni-based matrix yields composites with high fibre/matrix interface strength and, hence, high creep resistance at very high temperatures for Ni-based materials.

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