O U R N A L O F

Ceramic Processing Research

# Polycrystalline YAG; structural or functional?

#### Tai-II Mah\*, T.A. Parthasarathy and Hee Dong Lee<sup>a</sup>

Air Force Research Laboratory, Materials and Manufacturing Directorate, AFRL/MLLN Wright-Patterson AFB, OH 45433-7817 <sup>a</sup>UES Inc., 4401 Dayton-Xenia Rd. Dayton, OH 45432-1894

The current status and advances of polycrystalline YAG in both structural (high temperature) and functional applications are reviewed. A historical background of YAG and its progressive development to its current stage in functional and structural applications is reviewed. This review emphasizes the important aspects of polycrystalline YAG processing-microstructure-property relationships. For high temperature structural applications, the published mechanical properties (both single and polycrystalline) of YAG are described. Selected structural applications of polycrystalline YAG (e.g., monolith, fiber and CMC matrix) are discussed along with various aspects of processing. For functional applications, the current status of polycrystalline YAG, both doped and undoped, is reviewed from the processing point-of-view. Certain functional applications, including IR/ laser window materials, laser hosting materials, and high intensity discharge lamp materials, are reviewed. Recent research activities carried out in our lab for processing transparent polycrystalline YAG are also described.

Key words: YAG, high temperature structural material, laser hosting material.

#### Introduction

Single crystal yttrium aluminum garnet (YAG or  $Y_3Al_5O_{12}$ ) has been recognized and utilized widely as a laser gain host material for the last four decades. In the mean time, research work on polycrystalline YAG has been reported over only the past 20 years. Over these years, polycrystalline YAG has evolved to promising levels in two different classes of applications, one as a structural material for high temperature applications and the other as a functional material for uses such as laser windows and laser-gain materials. The purpose of this article is to review the current status of these two advances.

For high temperature structural applications, a background on YAG and its relevant published mechanical properties (both single and polycrystals) will be described. The structural application of polycrystalline YAG will be discussed along with various aspects of YAG processing for particular applications (e.g., monolith, fiber and CMC matrix).

For functional applications, we will limit our discussions only to polycrystals. The current status of polycrystalline YAG, both doped and undoped, will be reviewed from the processing point-of-view. Toward the end, recent research activities carried out in our laboratory will be described. The important aspects of polycrystalline YAG processing-microstructure-property relationships will be discussed in detail.

#### Background

There are many proposed phase diagrams for the system  $Al_2O_3$ - $Y_2O_3$ , and many discrepancies exist among them [1-5]. Cockayne [5] reviewed the chronological development of the  $Al_2O_3$ - $Y_2O_3$  phase diagram. The main differences in the proposed phase diagrams are the solubility of  $Y_2O_3$  in  $Al_2O_3$  and vice versa, the melting temperatures of the binary compounds, the stability and melting behavior of YAIO<sub>3</sub>, the eutectic compositions, and the eutectic temperatures. Some of these discrepancies were resolved on the pseudobinary between  $Al_2O_3$  and YAIO<sub>3</sub> [4]. Mah and Petry [6] reported the eutectic composition between  $Y_4Al_2O_9$  and  $Y_2O_3$  and  $Y_2O_3$  as shown in Fig. 1.

The pseudobinary compound Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> (Yttrium Aluminum Garnet, YAG) has attracted attention for many decades as a laser gain host material owing to its capability to accept trivalent laser activator ions ranging from transition metals to rare earth elements. Since the initial reports [7, 8] of Nd<sup>+3</sup> doping and co-doping with Cr<sup>+3</sup>, the utilization of Nd : YAG proliferated in the military, industry, and medical communities. Starting from about 20 years ago, attempts to fabricate optical quality, transparent polycrystalline YAG were reported [9-22]. The fabrication of transparent polycrystalline YAG, which has a comparable optical quality to the single crystal, was reported through vacuum sintering processes with highly reactive solid precursors or YAG powders [12-16]. Recently, we reported the fabrication of transparent polycrystalline YAG with a small grain size utilizing a sinter/HIP process [23]. The advancement of doped and

<sup>\*</sup>Corresponding author: Tel : +1-937-255-9829 Fax: +1-937-656-7292

E-mail: Taiil.Mah@wpafb.af.mil



Fig. 1. Pseudo-binary phase diagram of Al<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub>.

un-doped transparent polycrystalline YAG fabrication opened new opportunities, since the utilization of conventional ceramic processing provides fast, low-cost, and large size production. It also provides the possibility to incorporate a high concentration of dopants.

Interest in polycrystalline YAG as a high temperature structural material started from the current authors group in the late 1980's. As part of a U.S. Air Force initiative to develop high-temperature materials for use in future turbine engines, YAG was identified as a potential candidate for an oxide-oxide composite matrix as well as a reinforcement fiber. Research efforts ranged from the synthesis of stoichiometric YAG powder to the measurement of high temperature strength, toughness, and creep properties of single- and poly-crystals. The most outstanding outcome of the mechanical property measurements on single crystal YAG is that YAG is the most creep-resistant high temperature oxide [17, 18].

In the following sections, the structural applications of YAG will be presented first. It will be followed by the functional applications of YAG and will finish with our most current work on transparent polycrystalline YAG production.

#### **Structural Applications**

The use of YAG for high temperature structural applications was not considered by the materials science community until a concerted effort, led by the ceramics group in the U.S. Air Force, began in this area. A series of research programs were funded through various organizations to study the potential applications of YAG for high temperature structural components, especially in gas turbine engines. In the following subsections, a few examples of results, generated based on those efforts and the related applications of YAG, will be presented.

# Characterization of YAG Properties – Creep Properties of YAG

Corman [24, 25] studied the compression creep behavior of undoped YAG in helium in the temperature and stress ranges of 1650° to 1850 °C and 50 to 280 MPa, respectively. The creep experiments were carried out along the [100], [110], and [111] axes. The stress and temperature dependence of the steady state creep rate were calculated based on the equation by Cannon and Langdon [26, 27]. Corman found unusual creep behavior for the [100] and [110] directions, where the stress exponent and the activation energy appear to increase abruptly with an increase in either temperature or stress. No specific mechanism was given to explain this anomaly. Blumenthal and Phillips [28], Karato et al. [29], and Wolfenstine [30] also studied the creep behavior of single crystal YAG in various orientations. Based on their creep experiment along the [135] direction and subsequent slip trace analysis, Blumenthal and Phillips confirmed that  $\{110\} < 1-11 >$  was the primary slip system and the rate-controlling deformation mechanism above 1600 °C and below 300 MPa is dislocation glide. Another important aspect of the creep experiments on single crystal YAG by various researchers is that the steady state creep rate of YAG is almost independent of orientation and the stress exponent is about 3.3. Figure 2 is a collection of reported data plotted on one graph, and all of the data points (not shown) lie close to the drawn line. The conclusion of these studies is that single crystal YAG is the most creep resistant oxide known at present. The creep behavior of YAG is compared with several other refractory oxide single crystals in Fig. 3. It clearly shows that the creep rate of YAG is almost an order of magnitude lower than the creep rate of c-axis sapphire, which is one of the best creep resistant oxides.



**Fig. 2.** Steady state creep rate vs. stress of single crystal YAG from a collection of publications (data points not shown). The creep rate is almost independent of orientation and the stress exponent is about 3.3.



Fig. 3. Creep behavior of several oxide single-crystals at 100 MPa.



#### - Fracture Toughness of Single Crystal YAG

Since fracture resistance is a primary concern in structural ceramics, it is necessary to investigate the fracture toughness of YAG [31, 32]. With this rationale, the present authors studied the fracture toughness of single crystal YAG as a function of temperature up to 1600 °C in air and vacuum. The fracture toughness values measured at various temperatures in air are shown in Fig. 4 and are plotted as a function of crystallographic orientation of the specimen. The measured fracture toughness, using the single-edge-notched-beam (SENB) technique, was 2.2 MPa  $\sqrt{m}$  at room temperature and it increased with temperature above 1200 °C, reaching a value of 4.5 MPa  $\sqrt{m}$  at 1600 °C in air. The toughness increased further to 5.5 MPa  $\sqrt{m}$  when tested under



**Fig. 5.** Flexural strength and fracture toughness of polycrystalline YAG as a function of temperature.

vacuum ( $10^{-5}$  torr) at 1600 °C. The effects of crystallographic orientation and oxygen partial pressure on toughness were studied at 1600 °C and both temperature and testing environments were found to influence the fracture toughness. However, as in the case of creep tests, there was no significant effect of crystallographic orientation.

#### - Mechanical properties of Polycrystalline YAG

The present authors fabricated polycrystalline YAG without any sintering aid (e.g., SiO<sub>2</sub>) using two different processing routes, sintering and hot-pressing. The starting YAG powder was synthesized using a mixed alkoxide method. The strength and fracture toughness of the polycrystalline YAG as a function of temperature were measured using 4-point bending and SENB configurations, respectively (Fig. 5) [33]. The hot-pressed YAG had a strength of 234 MPa at room temperature and maintained its strength up to 1400 °C. The sintered YAG showed a much lower strength value (102 MPa) due to the high porosity (>4%) and large grain size (8 µm compared to 3 µm for hot-pressed YAG). The fracture toughness of the hot-pressed YAG was approximately 1.5 MPa  $\sqrt{m}$  at room temperature with a gradual drop to 1.3 MPa  $\sqrt{m}$  at 1400 °C. The sintered YAG showed similar behavior. The creep behavior of the hot-pressed YAG was also studied in the temperature range of 1400 °C to 1610 °C using constant strain rate compression tests under strain rates ranging from  $10^{-5}$ /s to  $10^{-3}$ /s [34]. The temperature dependence of creep of single crystal and polycrystalline YAG are compared in Fig. 6. The observed similar activation energies in both single crystal and polycrystals suggest that the creep of YAG is lattice diffusion controlled. The stress exponent of the creep rate, the activation energy in comparison with that for single crystal YAG, and the grain size dependence suggest that the Nabarro-Herring creep rate, limited by the bulk diffusion of one of the cations (Y or Al), is the operative mechanism.



**Fig. 6.** Temperature dependences of the single crystal and poly crystalline YAG show that the apparent activation energies are similar.

#### **YAG Fibers**

#### - Single Crystal Fibers

The aforementioned superior creep resistance of single crystal YAG indicates that YAG is a perfect candidate for a reinforcement fiber for high temperature ceramic matrix composites. The absence of crystallographic orientation dependence and isotropic cubic structure gives YAG a significant advantage over sapphire fibers. Under this rationale, Saphikon, Inc., carried out a research effort (under U.S. Air Force contract, '94) [35] designed to produce a large quantity, reasonably small diameter YAG fiber through the Edge-defined-Film-fed-Growth (EFG) technique. As expected, the EFG YAG fiber showed excellent creep resistance  $(1.48 \times 10^{-8})$ /s at 1600 °C and 200 MPa) and 2.36 GPa room temperature strength. However, due to the intrinsic difficulties (phase instability, difficult growth parameter control, etc.) of producing YAG fibers via EFG and the high degree of variability (fiber orientation and strength), Saphikon concluded that single crystal YAG fiber has poor manufacturability potential. No other effort has been pursued for single crystal YAG fiber growth.

# - Polycrystalline Fibers

The present authors also compared the creep properties of polycrystalline YAG to that of alumina with similar grain sizes and found that polycrystalline YAG has a lower creep rate than alumina [34]. This comparison clearly indicates the potential of polycrystalline YAG as refractory fibers. Under a U.S. Air Force funded program, two organizations, General Atomics [36] and the University of Michigan [37], investigated the processing of small diameter polycrystalline YAG fibers. Chen *et al.* [36], identified a reproducible method for the synthesis and processing of a sol-gel derived precursor YAG viscous solution through organic acid modifications of metal alkoxides. Using the viscous solution, continuous long length, flexible precursor, 30-50 mm diameter monofilaments were spun at ~50 m/minute. These pure YAG fibers (sintered at 1600-1650 °C) had low room temperature strength because of a large grain size, weak grain boundaries, and large residual pores in the fibers that could not be eliminated.

Researchers at the University of Michigan studied the feasibility of producing YAG fibers from colloidal sols and metal-organic precursors [37, 38]. King and Holloran [37] used commercially available colloidal sols of Y<sub>2</sub>O<sub>3</sub> and AlOOH, while Lin et al. [38], used mixtures of either yttrium acetate hydrate and aluminum formate hydrate or yttrium isobutyrate and aluminum isobutyrate. King and Holloran [37] added polyethylene oxide to their mixed sols to aid stabilization as well as a spinning aid at various stages of aging. After the addition of water to the mixture, the green fibers were easily spun into a diameter of 150 µm. Lin et al. [38], also drew fibers either by hand or by a commercial spinneret after purification and evaporation of the precursor mixtures into viscous solutions. The thinnest diameter of the drawn fiber was ~20 µm. In both cases (colloidal sol mixture and metalorganic precursors), the green fibers produced were dense and mostly defect free. As in the case of commercial ceramic fibers (e.g., 3M's Nextel<sup>TM</sup> series), the green fibers have to maintain their structural integrity during drying and crystallization/sintering processes. These two steps involve large amounts of volume shrinkage. The primary difference between the YAG fiber and the Nextel<sup>TM</sup> series fibers is the crystallization and sintering behaviors of YAG. Nextel<sup>TM</sup> series fibers (alumina or aluminosilicate) are rather easy to crystallize and sinter, due to the high sinterability of alumina and the viscous nature of silicates (with a small addition of a sintering aid, like boria). As will be briefly discussed in the YAG powder production section, the crystallization of YAG from chemical precursors follows very complicated stages with increasing temperature before the final YAG crystallization. Not only the complicated crystallization, but also the high creep resistance of YAG plays an opposite role in terms of sintering (low lattice diffusion rate). The evidence of this low diffusion rate is that the grain growth rate of polycrystalline YAG is extremely slow until the temperature reaches near  $0.85T_m$  [34]. These factors (crystallization and sintering) are the current impediments to the full scale YAG fiber production. Other researchers [39-41] have also experienced similar drawbacks, as described above.

#### **Other Applications of YAG**

#### – YST

The authors studied another application of YAG as a matrix material for a ceramic composite (YST:YAG, SiC<sub>w</sub>, and TiC) cutting tool material for the improved machining of titanium alloys [42]. The rationale of using YAG as a matrix was based on the thermodynamic and reaction couple studies of the cutting tool material,



**Fig. 7.** SEM photomicrograph of YST (light = YAG, grey = TiC, and dark =  $SiC_w$ ).

as well as YAG single crystals, with commercial grade titanium. The reaction couple study showed the formation of a thin passivating layer of  $Y_2O_3$  next to the composite, which is exceptionally resistant to attack by titanium. This cutting tool is electrically conductive and was easily machined using electro-discharge machining. A typical microstructure of the YST is shown in Fig. 7. Further details (processing, microstructure, and properties) of this material can be found in this issue of the Journal [43].

#### - Fiber Coating and Matrix for CMCs

Ceramic matrix composites have been studied for almost three decades based on the fact that high damage tolerance can be achieved through distributed damage mechanisms. These mechanisms are dependent on matrix cracks deflecting into matrix/fiber interfacial debonding cracks. One way of achieving the interfacial debonding crack is the introduction of a weak coating on the fiber. YAG was utilized for this purpose in the form of porous coatings on Nextel<sup>™</sup> 610 fibers [44]. A polymeric solution-derived YAG with a fugitive carbon phase was used to develop the porous fiber coating. The porous YAG fiber coating did not reduce the strength of the tows when heated in argon, and they degraded the tow strength by only ~20% after heating in air at 1200 °C for 100 hrs. Minicomposites containing porous YAG coated fibers were nearly twice as strong as those containing uncoated fibers. Long-term exposure of the composite induced the densification of the porous YAG coating, which made the coating ineffective. However, below the threshold of porous YAG coating densification, the porous YAG coating is effective for crack deflection.

YAG was also used as a filler material for Nextel<sup>TM</sup> 610 fiber reinforced porous alumina matrix composites [45, 46]. Various ratios of alumina and YAG were used as matrices to determine the effect of YAG on the retained tensile strength of the composites after heating to 1100 and 1200 °C. A plot of average tensile strengths of various composites containing either YAG or a yttrium-



**Fig. 8.** Average tensile strengths of Nextel<sup>TM</sup> 610 fiber reinforced Alumina based composite with YAG and/or yttrium addition in the matrix.

doped matrix versus heat-treatment conditions is shown in Fig. 8. It was found that in all cases, the YAG containing composites gave higher ultimate stresses and greater strains to failure than the all-alumina composites, when processed at 1200 °C for times up to 100 hrs. No correlation was found between strength and the amount of YAG in the matrix; however, composites with matrices containing YAG vs. alumina as the binder phase had higher strengths. YAG was found to greatly hinder densification of alumina at 1200 °C, similar to the case of mullite in Nextel<sup>TM</sup> 720 fiber reinforced alumina-mullite matrix composites [47]. The observed high strength retention of YAGcontaining composites may be attributed to the matrix resistance to densification during high temperature exposure.

# **Functional Applications**

Since the first report of the successful production of optical quality polycrystalline YAG, a series of concentrated research efforts were carried out throughout the world. It has reached the stage that some of the doped YAG materials are available commercially. In the following subsection, we will start with the various processes used to produce stoichiometric YAG powder. It has been our experience that the correct stoichiometry is the most important pre-requisite to achieving high optical quality, transparent polycrystalline YAG. This will be followed by discussions on some of the functional applications of transparent polycrystalline YAG, including the densification processes.

#### YAG powder Synthesis

YAG powders are commercially available through Ceralox (Condea Vista Company, Tucson, AZ 85706). This commercial powder is produced through the high temperature reaction of  $Al_2O_3$  and  $Y_2O_3$ , followed by extensive particle size reduction steps (e.g., crushing, milling, etc). Consequently, the average particle size is large and has a wide particle size distribution. Due to the high temperature solid-state reaction process, it often contains un-reacted starting constituent phases. In order to overcome the problems (i.e., particle size, stoichiometry, phase purity, etc.), the production of YAG was pursued via various chemical processes. The reported YAG powder syntheses are by a mixed alkoxide process [34, 48, 49], by metalloorganic precursors [50], by co-precipitating water-based precursors (nitrates, sulfates, chlorides, citrates) with different precipitants (urea, ammonium hydroxide, ammonium hydrogen carbonate, etc.), or some combination of precursors and variations of processes (combustion, flame pyrolysis, etc.) [51-75]. The difficulties involved in YAG powder production are the direct crystallization of YAG from precursor materials and the control of the final particle size. Typical sol-gel processing of YAG powder invariably results in the formation of intermediate yttrium aluminates (Y<sub>4</sub>Al<sub>2</sub>O<sub>9</sub> and YAlO<sub>3</sub>) with temperature before the final transformation to YAG, even though the overall starting composition of  $Y_2O_3$  and  $Al_2O_3$  is 3:5. The temperature where the intermediate yttrium aluminate phase forms and the ultimate YAG transformation occur varies a little with the precursors and processing methods. However, the most important factor, which governs the direct crystallization of YAG from the precursors, is the atomistic mixing/bonding of yttrium and aluminum and their close proximity throughout the whole duration of the processing stages.

Recently, the present authors synthesized YAG powder through two different combustion processes.<sup>23</sup>



Fig. 9. Processing flow chart for YAG powder synthesis and densification.

The schematic processing flow chart is shown in Fig. 9. First, a partial combustion (PC) route was attempted by mixing commercial  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (AKP53,  $d_{50}$ :0.2 µm, Sumitomo Chemical) with an aqueous yttrium nitrate solution at a mole ratio of 5:3 of Al:Y, and to this, the appropriate amounts of reductants (amino-acids) were added. Second, a full combustion (FC) process was evaluated by dissolving yttrium nitrate and aluminum nitrate in water with a mole ratio of 5:3 of aluminum and yttrium, and the proper amounts of reductant (amino-acids) and oxidant (ammonium nitrate) were also added. After drying water from both precursors, they were combusted at temperatures around 200°-240 °C. The as-combusted PC-YAG and FC-YAG were further calcined at 1000° and 1100 °C with holding for 2 hours. The as-calcined powders were mildly ball milled for 24 hours with high purity alumina grinding media (Tosoh) using ethyl alcohol as a liquid medium. The alumina balls were weighed before and after ball milling to monitor potential contamination and a negligible amount of contamination was observed. Both of the as-combusted powders were snow-like flaky and porous large agglomerates. The agglomerates were very soft and could be readily de-agglomerated during ball milling. The agglomerates also were well dispersed during ultra-sonification. The morphology and size of the FC-YAG powder, calcined at 1000 °C, are shown in Fig. 10. The primary particle size of the 1000 °C calcined FC-YAG was estimated to be ~30 nm, whereas the PC-YAG consisted of 50-100 nm sized particles. The results of x-ray analyses of FC-YAG are shown in Fig. 11. The as-combusted FC-YAG consists of a small amount of hexagonal YAP (YAlO<sub>3</sub> : ICDD file No. 16-0219) with an amorphous phase. After a 2 hour calcination at 1000 °C, only the single phase cubic YAG was detected. Further heating at 1600 °C confirmed the single phase cubic YAG with an increase in peak intensity.



Fig. 10. SEM photomicrograph of FC-YAG powder after calcination at 1000 °C.



Fig. 11. XRD patterns of FC-YAG powder after various heat-treatment temperatures.

#### **IR/Laser Window Materials**

As mentioned in the introduction, YAG possesses a high efficiency of energy transfer and radiation damage resistance, which has made it a popular laser host material. Another attractive application of polycrystalline YAG is as IR and laser window materials. Since the transmittance of YAG extends from the UV  $(0.2 \,\mu m)$ range to the mid-IR (5.5 µm) range, YAG can be potentially used as IR and laser windows, along with missile domes. It is reported that high power laser window materials should possess a very low distortion during use and a high hardness (for scratch and erosion resistance) while retaining its structure and optical properties for the lifetime of the component, which for airborne missile applications is about 20 years [76-81]. Optical flatness is the desired property and YAG is cubic in crystal structure, which imparts optical isotropy (unlike sapphire), and it exhibits no birefringence. Although YAG does not have a negative thermo-optic coefficient, it has very low values for both the thermooptic coefficient and the thermal expansion coefficient in the temperature range of 100 to 300 K [82], as well as a much better thermal conductivity than FS (fused silica) or low-OPD (optical path difference) glasses, which are the currently used materials. Further, it has a very low absorption coefficient, making its optical figure of merit comparable to FS and low-OPD glasses [83]. These factors, along with its far superior wear/ abrasion resistance, higher fracture toughness/strength, and ultra-high environmental stability, make polycrystalline YAG an ideal choice for bulkhead laser window applications in airborne and space vehicles.

# - Transparent Polycrystalline YAG Processing

For IR/laser applications, high optical transparency is demanded, and single crystal YAG can be preferred. As mentioned earlier, transparent polycrystalline YAG, which possesses similar optical qualities as single crystal YAG, can successfully be fabricated through vacuum sintering processes with highly reactive solid precursors or YAG powders [12-22]. The production of high quality transparent YAG hinges on the complete elimination of pores, since significant optical scattering can occur from the residual pores. The total volume of the residual pores must be restricted below a few ppm in order to achieve the desirable optical quality. Because of such an extreme requirement, the current processes that produce transparent polycrystalline YAG have mostly been conducted under high vacuum (<10<sup>-6</sup> torr) and high temperatures (> 1750 °C) with lengthy soaking periods (> tens of hours).

Recently, the present authors studied the fabrication of transparent polycrystalline YAG through a sinter/ HIP process using the YAG powder produced in the authors lab via combustion process [23]. By combining these two technologies (powder synthesis and a two step consolidation process), we were able to produce transparent polycrystalline YAG that exhibited an excellent optical quality comparable to single crystal YAG. As shown in the processing flow chart (Fig. 9), the process can be carried out at much lower temperatures with shorter processing times and can be easily scaled up. Thus, it is believed to be highly economic. A brief description of the consolidation process will be presented. Due to the limited space for the current article, we will limit our discussion of polycrystalline YAG processed using the FC-YAG powder only.

The calcined powders were mildly milled and further processed with binder (PEG), and finally granulated by passing through a 200-mesh nylon sieve. The granulated powders were cold pressed into a disc shape with dimensions of 12.7 mm (1/2") diameter and 3 mm thickness. The cold pressed discs were further cold



Fig. 12. SEM photomicrographs 2 atom. % Nd-doped YAG, calcined at 1000° and 1100°C, sintered at 1650 °C, and HIPed at 1550 °C.



Fig. 13. Transmittance of HIPed (a) FC-YAG at 1500°C and (b) PC-YAG at 1550 °C, and (c) single crystal YAG. (inset picture corresponds to HIPed FC-YAG).

isostatically pressed at 200 MPa and the measured green density was 45°-50% of the theoretical density  $(\rho_{YAG} = 4.55 \text{ g/cm}^3)$ . The cold-isostatically-pressed discs were heated at 700 °C in air for binder removal. The calcined discs were sintered in air at temperatures between 1550° and 1650 °C for 5 hours in an alumina tube muffle furnace. The translucent as-sintered bodies were single phase YAG, identified with x-ray diffraction analysis, and the densities were 95.0-99.0% theoretical density (measured by Archimedes method). Finally, the as-sintered YAG was hot isostatically pressed (HIPed) at temperatures of 1450°-1550 °C for 5 hours under 200 MPa argon pressure. Fig. 12 shows the microstructure of HIPed YAG specimens. The pores were apparently completely eliminated during HIPing, and the grains are uniformly distributed with sizes ranging from 1-2 µm. Both HIPed YAG showed a similar grain size distribution even at slightly different HIPing temperatures. The average grain size of the HIPed YAG (1-2 µm) is considerably smaller (about an order of magnitude) than the published vacuum sintered polycrystalline YAG (10-20  $\mu$ m). It is therefore anticipated that the HIPed YAG would posses superior mechanical properties than the vacuum sintered YAG.

The HIPed samples were surface ground and polished with a 1 µm diamond slurry for optical transmittance measurements. The transmittance in the wavelength from the visible (400 nm) to near I.R. (2500 nm) was measured by a Cary 5E spectrophotometer. The sample dimension for transmittance measurements was 12 mm diameter with a 2 mm thickness. The transmittance results are plotted in Fig. 13 along with a single crystal Nd doped YAG with thickness of 1.2 mm. The transmittance of the HIPed FC-YAG looked nearly identical as the single crystal. The small deviation can be attributed to the slightly different polishing quality. The great advantage of PC-YAG can be found in its simple powder production and highly economic aspect; however, further processing optimization is needed.

### High Intensity Discharge Lamp Material

Another interesting potential application of polycrystalline YAG would be as arc tubes in high-pressure sodium or metal halide (e.g., NaI, CsI or  $HgI_2$ ) discharge lamps [84, 85] Lucalox<sup>TM</sup> has been used successfully for many decades. Lucalox<sup>TM</sup> is a patented material from GE and is a translucent alumina, which was based on the pioneering work by Coble [86] MgO was used as an additive and its role is still somewhat controversial; however, it does reduce grain boundary mobility and thus prevents discontinuous grain growth [87]. The suppression of grain growth promotes the reduction of pore entrapment in the grain and effectively eliminates pores during sintering. An almost theoretical density is needed to achieve the desired translucency for lighting applications. The corrosionresistant Lucalox<sup>TM</sup> made it possible to contain sodium vapor at high temperature, which produces goldenwhite light at high efficiency. Recently, Krell et al. [88, 89] reported highly translucent Al<sub>2</sub>O<sub>3</sub> having grain sizes of 0.4-0.6 µm and high hardness and strength (20-21 GPa and 600-700 MPa). The intention of their work (improved mechanical properties) was to develop metal halide lamps with transparent and strong Al<sub>2</sub>O<sub>3</sub> lamp envelopes, which resist the high internal pressure of the discharge plasma. They were successful in achieving 55-65% real in-line transmission through the polished 0.8 mm thick plates. Wei et al. [90], studied the sodium attack on translucent polycrystalline alumina. They found the degradation (reaction of sodium and alumina to form sodium aluminates and aluminum) of the alumina depends on the grain boundary diffusion of aluminum (reaction product) through the alumina wall. They claimed a significant improvement against sodium attack was achieved through the reduction of the MgO addition, doping with tetravalent cations (e.g.,  $Zr^{+4}$ ) to chargecompensate the Mg<sup>+2</sup> at aluminum lattice sites, and form a second phase to absorb MgO in-situ. YAG has an excellent corrosion resistance to sodium or metal halide vapors compared to alumina. The light transmittance of transparent YAG is far superior to polycrystalline alumina (Fig. 13), simply owing to the isotropic cubic structure of YAG. Recently, Konoshima [91] (Konoshima Chemical Company) and Toshiba [92] (Toshiba Lighting & Technology Corporation) obtained patents using YAG as high-voltage discharge lamp material. Due to the reaction of rare-earth halides, which provides higher radiation efficiency than metal halides (e.g., NaI, CsI or HgI<sub>2</sub>) with YAG, the patents involve corrosion-resistant coatings inside of YAG tubes or a YAG lamp designed to circumvent the reaction. The current research activities of using YAG as high intensity discharge lamp envelope material clearly exhibits another versatile utilization of transparent polycrystalline YAG.

# **Concluding Remarks**

The current status and the advances of two different classes of applications, high temperature structural and functional, of polycrystalline YAG have been reviewed in this paper.

As is evident from the discussions in the preceding sections, polycrystalline YAG provides many options to the materials scientists to select YAG as materials for their specific needs and applications. One particular area, which the current authors believe needs to be further explored, is the development of small diameter polycrystalline YAG fibers for CMC reinforcements. The current limitation of the full utilization of oxide CMCs in high temperature gas turbine engine applications is the non-availability of suitable creepresistant reinforcement oxide fibers. Since most of the components are creep limited, availability of highly creep resistant refractory YAG fibers will provide numerous options to the materials scientists and engineers for their particular applications.

For functional applications, the importance of YAG powders in terms of purity, stoichiometry, and size for the final transparency were discussed. Limited discussions of polycrystalline YAG for IR and laser windows clearly show the potential future applications of YAG in this unexplored field. The advantages of doped polycrystalline YAG, in the field of laser applications, over the current single crystals are numerous and the commercial application of this cost-effective polycrystalline YAG seems unlimited. Another potential application of the polycrystalline YAG as a high intensity discharge lamp envelope material needs to be explored further for the development of the next generation lighting industry.

#### References

- S.J. Schneider, R.S. Roth, and J.S. Waring, J. Res. Natl. Bur. Stand. A (U.S.) 65 (1961) 345.
- L.E. Olds and H.E. Otto, Phase Diagrams for Ceramists, 1969. Edited by E.M. Levin, C.R. Robbins, and H.F. McMurdie, American Ceramic Society, Columbus, OH; Fig. 311.
- 3. D. Viechnicki and F. Schmid, Mater. Res. Bull. 4 (1969) 84-88.
- 4. J.L. Caslavsky and D.J. Viechnicki, J. Mater. Sci. 15 (1980) 1709-1718.
- 5. B. Cockayne, J. Less-Common Met. 114 (1985) 199-206.
- T. Mah and M.D. Petry, J. Am. Ceram. Soc. 75[7] (1992) 2006-2009.
- J.E. Geusic, H.M. Marcos, and L.G.Van Uitert, Appl. Phys. Lett. 4 (1964) 182-184.
- Z.J. Kiss and R.C. Duncan, Appl. Phys. Lett. 5 (1964) 200-202.
- 9. G. de With and H.J.A. van Dijk, Mater. Res. Bull. 19 (1984) 1669-1674.
- 10. G. deWith, Phillips J. Res. 42 (1987) 119-130.

- 11. M. Sekita, H. Haneda, T. Yanagitani, and S. Shirasaki, J. Appl. Phys. 67[1] (1990) 453-458.
- 12. M. Sekita, H. Haneda, S.i Shiransaki, and T. Yanagitani, J. Appl. Phys. 6[6] (1991) 3709-3718.
- A. Ikesue, I. Furusato, and K. Kamata, J. Am. Ceram. Soc. 78[1] (1995) 225-228.
- A. Ikesue, K. Kamata, and K. Yoshida, J. Am. Ceram. Soc. 78[9] (1995) 2545-2547.
- A. Ikesue, T. Kinoshita, K. Kamata, and K. Yoshida, J. Am. Cearm. Soc. 78[4] (1995) 1033-1040.
- J. Li, T. Ikegami, J-H. Lee, and T. Mori, J. Am. Ceram. Soc. 83[4] (2000) 961-963.
- I. Shoji, S. Kurimura, Y. Sato, T. Taira, A. Ikesue, and K. Yoshida, App. Phy. Lett. 77[7] (2000) 939-941.
- S. Nakayama, A. Ikesue, and M. Sakamoto, Nippon Kagaku Kaishi (6) (2000) 437-440.
- T. Tachiwaki, M. Yoshinaka, K. Hirota, T. Ikegami, and O. Yamaguchi, Solid State Comm. 119 (2001) 603-606.
- 20. Akio Ikesue, Optical Materials 19 (2002) 183-187.
- J. Lu, K. Ueda, H. Yagi, T. Yanagitani, Y. Akiyama, and A. A. Kaminski, J. Alloys and Compounds 342 (2002) 220-225.
- 22. Y. Rabinovitch, D. Tetard, M.D. Faucher, and M. Pham-Thi, Optical Materials 24 (2003) 345-351.
- 23. H.D. Lee, T. Mah, and T.A. Parthasarathy, Ceram. Eng. Sci. Proc. 25 (2004) in press.
- 24. G.S. Corman, Ceram. Eng. Sci. Proc. 12[9-10] (1991) 1745-1766.
- 25. G.S. Corman, J. Mater. Sci. Lett. 12 (1993) 379-82.
- W.R. Cannon and T.G. Langdon, J. Mater. Sci. 18 (1983) 1-50.
- 27. Idem, ibid. 23 (1988) 1-20.
- 28. W.R. Blumenthal and D.S. Phillips, J. Am. Ceram. Soc. 79[4] (1996) 1047-1052.
- S. Karato, Z. Wang, and K. Fujino, J. Mater. Sci. 29 (1994) 6458-6462.
- 30. J. Wolfenstine, Scripta Materialia 38[6] (1998) 965-968.
- 31. T. Mah and T.A. Parthasarathy, Scripta Metall. et Mater. 28[11] (1993) 1383-1385.
- T. Mah and T.A. Parthasarathy, J. Am. Ceram. Soc. 80[10] (1997) 2730-2734.
- K. Keller, T. Mah, and T.A. Parthasarathy, Ceram. Eng. Sci. Proc. 11 (1990) 1122-1133.
- 34. T.A. Parthasarathy, T. Mah, and K. Keller, J. Am. Ceram. Soc. 75[7] (1992) 1756-5179.
- J.M. Collins, H.E. Bates, and J.J. Fitzgibbon, WL-TR-94-4085 (1994).
- K.C. Chen, K.S. Mazdiyasni, and H.H. Streckert, WL-TR-94-4022 (1993).
- B.H. King and J.W. Holloran, J. Am. Ceram. Soc. 78[8] (1995) 2141-2148.
- Y. Lin, Z.F. Zhang, J.W. Holloran, and R.M. Laine, J. Am. Ceram. Soc. 81[3] (1998) 629-45.
- R.C. Pullar, M.D. Taylor, and A.K. Bhattacharya, J. Eur. Ceram. Soc. 18 (1998) 1758-1764.
- 40. R.C. Pullar, M.D. Taylor, and A.K. Bhattacharya, J. Eur. Ceram. Soc. 19 (1999) 1747-1758.
- R. Kruger, W. Glaubitt, and P. Lobmann, J. Am. Ceram. Soc. 85[11] (2002) 2827-2833.
- T. Mah, T.A. Parthasarathy, and M.K. Cinibulk, U.S. Patent 6,620,756,B2, Sept. 16, (2003).
- 43. T. Mah, H.D. Lee, T.A. Parthasarathy, and M.K. Cinibulk, this issue of J. Ceram. Proc. Res.
- 44. M.K. Cinibulk, T.A. Parthasarathy, K.A. Keller, and T. Mah, J. Am. Ceram. Soc. 85[11] (2002) 2703-2710.

- M.K. Cinibulk, K.A. Keller, T. Mah, and T.A. Parthasarathy, Ceram. Eng. Sci. Proc. 23 (2002) 629-636.
- 46. M.K. Cinibulk, K.A. Keller, and T. Mah, J. Am. Ceram. Soc., in press
- 47. C.G. Levi, J.Y. Yang, B.D. Dalgleish, F.W. Zok, and A.G. Evans, J. Am.Ceram. Soc. 81[8] (1998) 2077-2086.
- M. Steinmann and G. de With, Euro-Ceramics, Vol. 1. Processing of Ceramics; Maastricht; The Netherlands; 18-23 June 1989 (1989) p.1109-1113.
- O. Yamaguchi, K. Takeoka, and A. Hayashida, J. Mater. Sci. Lett. 10 (1990) 101-103.
- 50. Y. Lin, Z.F. Zhang, B. King, J.W. Holloran, and R.M. Laine, J. Am. Ceram. Soc. 79[2] (1996) 385-943.
- 51. D.R. Messier and G.E. Gazza, Am. Ceram. Soc. Bull. 51 (1972) 692-95.
- 52. Q. Zhang and F. Saito, Powder Technol. 129 (2003) 86-91.
- T. Takamori and L.D. David, Am. Ceram. Soc. Bull. 65[9] (1986) 1282-1286.
- 54. J.W.G.A. Vrolijk, et al., J. Eur. Ceram. Soc. 6 (1990) 47-51.
- M. Inoue, H. Otsu, H. Kominami, and T. Inui, J. Am. Ceram. Soc. 74[6] (1991) 1452-54.
- 56. P. Apte, H. Burke, and H. Pickup, J. Mater. Res. 7[3] (1992) 706-711.
- 57. M. Gomi and T. Kanie, Jpn. J. Appl. Phys. 135[3] (1996) 1798-1801.
- Y. Hakuta, K. Seino, H. Ura, T. Adschiri, H. Takizawa, and K. Arai, J. Mater. Chem. 9 (1999) 2671-2674.
- 59. S.M. Sim, K.A. Keller, and T. Mah, J. Mater. Sci. 35[3] (2000) 713-717.
- 60. S.H. Yu, J. Ceram. Soc. Japan 109[5] (2001) S65-S75.
- K. Kinsman, J. McKittrick, E. Sluzky, and K. Hesse, J. Am Ceram. Soc. 77[11] (1994) 2866-2872.
- K. Han, H. Koo, and C. Lim J. Am. Ceram. Soc. 82[6] (1999) 1598-1600.
- N. Matsushita, N. Tsuchiya, K. Nakatsuka, and T. Yanagitani, J. Am. Ceram. Soc. 82[8] (1999) 1977-1984.
- 64. T. Tachiwaki, M. Yoshinaka, K. Hirota, T. Ikegami, and O. Yamaguchi, Solid State Comm. 119 (2001) 603-606.
- J.G. Li, T. Ikegami, J.H. Lee, and T. Mori, J. Am. Cearm. Soc. 83[4] (2000) 961-963.
- J. Lu, K. Ueda, H. Yagi, T. Yanagitani, Y. Akiyama, and A. A. Kaminski, J. Alloys and Compounds 342 (2002) 220-225.
- 67. Y. Rabinovitch, D. Tetard, M.D. Faucher, and M. Pham-Thi, Optical Materials 24 (2003) 345-351.
- N. Matsushita, N. Tsuchiya, K. Nakatsuka, and T. Yanagitani, J. Am Ceram Soc. 83[4] (2000) 961-963.
- J.G. Li, T. Ikegami, J.H. Lee, and T. Mori, J. Mater. Res. 15[11] (2000) 2375-2386.
- J.G. Li, T. Ikegami, J.H. Lee, and T. Mori, J. Mater. Res. 15[7] (2000) 1514-1523.
- 71. J. McKittrick, L.E. Shea, C.F. Bacalski, and E.J. Bosze, Displays 19 (1999) 169172.
- 72. Y.C. Kang, Y.S. Chung, and S.B. Park, J. Am Ceram. Soc. 82[8] (1999) 2056-2060.
- 73. G. Gowda, J. Mater. Sci. Lett. 5 (1986) 1029-1032.
- 74. M.K. Cinibulk, J. Am. Ceram. Soc. 83[5] (2000) 1276-1278.
- 75. G. Gowda, J. Mater. Sci. Lett. 5 (1986) 1029-32.
- Handbook of Laser Science and Technology, M.J. Weber, ed., CRC Press (1986).
- P.S. Carlin, Proceedings of IEEE Aerospace Conference, 4, March 18 (2000) 169-181.
- 78. K.W. Billman, D.C. Tran, L.F. Johnson, M.B. Moran, S.F.

Nee, J.A. Detrio, S.M. Daigneault, and A.P. Bukley, W.E. Thompson, P.H. Merritt, Eds., Laser Weapons Technology II, Proc. SPIE (2001).

- S.F. Nee, L.F. Johnson, M.B. Moran, J.M. Pentony, S.M. Daigneault, D. Tran, K.W. Billman, and S. Siahatgar, A.J.M. III, E.G. Arthurs, Eds., Inorganic Optical Materials II, Proc. SPIE (2000).
- 80. C.A. Klein, Opt. Eng. 36[6] (1997) 1586-1595.
- 81. Transparent Armor, The AMPTIAC Newsletter 4[3] (2000) 1-13.
- R. Wynne, J.L. Daneu, and T.Y. Fan, Applied Optics 38[15] (1999) 3282-3284.
- S. Musikant. in Optical Materials: An Introduction to Selection and Applications, Marcell Decker, NY, 1985.
- 84. V. Rohe and C.M. Schoene, Ceram. Eng. Sci. Proc. 5[7-8]

(1984) 752-764.

- G. De With and H.J.A. Van Dijk, U.S. Pat. No. 4841195, June 20 (1989).
- 86. R.L. Coble, U.S. Pat. No. 3026210 (1960).
- 87. J.E. Burke, MRS Bulletin, June (1996) 61-68.
- A. Krell, P. Blank, H. Ma, T. Hutzler, M.P.B. van Bruggen, and R. Apetz, J. Am. Ceram. Soc. 86[1] (2003) 12-18.
- Rolf Apetz and Michel P.B. van Bruggen, J. Am. Ceram. Soc. 86[3] (2003) 480-486.
- George C. Wei, Arlene Hecker, and David A. Goodman, J. Am. Ceram. Soc. 84[12] (2001) 2853-2862.
- T. Yanagitani, H. Yagi, S. Imagawa, and H. Kubo, U.S. Pat. No. 6200918 B1, March 13 (2001).
- H. Honda, S. Ashida, K. Saita, T. Otabe, M. Shibuya, and N. Watanabe, U.S. Pat. No. 6215254 B1, April 10 (2001).