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Thermomechanical properties of rare-earth-doped AlN for laser gain media: The role of grain boundaries and grain size

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Abstract—The low thermal conductivity and poor fracture toughness of traditional laser gain media (rare-earth-doped single crystals) limits the overall power deliverable from a laser system. We present an investigation of the thermomechanical properties of a promising laser gain candidate, Tb-doped aluminum nitride (Tb:AlN). We pay special attention to the effect of the average grain size and the dopant segregation at the grain boundaries on the relevant properties: Vickers hardness, fracture toughness and thermal conductivity. We find that all properties are affected by grain boundaries and/or dopant segregation. However, the thermal conductivity is significantly more affected than the mechanical properties and therefore dominates the thermal shock figure of merit, R_s. We obtained a thermal shock figure of merit in Tb:AlN more than 60 times that of the state-of-the-art laser gain material Nd:YAG.

Keywords: Transparent ceramics; Spark plasma sintering (SPS); Current-activated pressure-assisted densification (CAPAD); Thermal conductivity; Indent fracture toughness

1. Introduction

Thermal management continues to be a significant problem in the design of high-performance, high-energy lasers. For instance, thermal gradients in laser gain materials can lead to performance issues such as thermal lensing and ultimately irreparable failure through thermal shock. A gain material with high thermal conductivity will be able to dissipate more heat, reducing thermal gradients and thus producing more laser power for a given pumping–cooling scheme. Similarly, materials with high fracture toughness can tolerate higher thermal stresses before failure. Thus, the important design criterion for maximizing laser energy is dependent on both mechanical and thermal properties of the gain material, with a thermal shock resistance figure of merit given by [1]:

$$R_s = \frac{k(1-\nu)}{\alpha E}\sigma_F \qquad (1)$$

where σ_F is the fracture stress, k is the thermal conductivity, E is Young's modulus, α is the coefficient of thermal expansion and ν is Poisson's ratio.

Traditionally, laser gain materials like Nd:YAG have been single crystals with relatively low thermal conductivity and low toughness. Polycrystalline ceramics are often tougher than single crystals, but can suffer from inferior optical properties (because of scattering at pores and grain boundaries). This optical property challenge has been alleviated by the advent of advanced processing procedures. For example, Ikesue and others [2,3] have shown that slope efficiencies of polycrystalline ceramics are on a par with single crystals. Recently, current-activated pressure-assisted densification (CAPAD) has been receiving considerable attention for processing optical ceramics [4–9]. A clear advantage of CAPAD is that fast processing kinetics can create metastable phases that are not possible with single-crystal growing techniques or pressureless sintering. We recently demonstrated over-equilibrium doping of active rare-earth (RE) elements, leading to luminescence in the AlN [10] and Al_2O_3 [11] systems. Since the thermal conductivity and fracture toughness of these ceramics are inherently higher than the state-of-the-art laser gain material Nd:YAG, metastable RE:Al_2O_3 and RE:AlN hold significant promise for next-generation high-energy lasers.

As alluded to above, extended defects like grain boundaries can dominate the optical, thermal and mechanical properties of ceramics. The effects of grain boundary phases in AlN with RE additions have been reported [12,13], but in these cases the goal was to make tough, highly thermally conductive AlN ceramics, not photoluminescent, light-transmitting ceramics. Thus high concentrations (~5 wt.%) of RE oxides were added. Very little

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attention has been given to microstructural features that influence the thermomechanical performance of polycrystalline ceramics for optical applications.

Here we present a study on the influence of grain boundaries on the mechanical and thermal properties of translucent, photoluminescent rare-earth-doped aluminum nitride (RE:AlN). The ceramics were CAPAD processed under various conditions and with varying Tb dopant concentrations such that they had different grain sizes (and thus different concentration of grain boundaries) and different degrees of dopant segregation at the grain boundaries. The relevant properties examined are hardness, fracture toughness, thermal conductivity and the thermal shock resistance figure of merit. We find that all properties are affected by grain boundaries and/or dopant segregation. However, the thermal conductivity is significantly more affected than the mechanical properties, and therefore dominates the thermal shock performance. The thermal conductivity is strongly impacted by grain boundary scattering (made stronger by dopant segregation) at all temperatures, with Umklapp scattering also important at higher temperatures.

2. Experimental procedure

2.1. Powder processing

All samples discussed in this paper were processed from commercially available aluminum nitride (97% purity as AlN, 1–2 μm particle size, Tokuyama Co., Japan) and rare earth powders (99.9% purity (REO) 40 mesh Tb powder, Alfa Aesar, USA) using CAPAD. The samples discussed in this paper were made from powders in three different conditions: as-received powders, low-energy ball-milled (LEBM) powders and powders milled with high-energy planetary ball milling (HEBM). Tb dopants were added either as pure Tb metal or as TbN (nitride synthesis described below). The dopant powders were mixed with AlN using either LEBM or HEBM. For HEBM, powders were planetary ball milled under argon using a Fritsch Pulverisette 7 premium line. The high-energy milling parameters were a ball:powder mass ratio of 10:1, at 450 rpm for 3 h. Either pure Tb or TbN was used as the dopant source. In order to synthesize TbN, Tb powder was nitrided in a tube furnace at 600 °C for 6 h under flowing nitrogen. The Tb was loaded into the furnace tube under an argon atmosphere and sealed before transfer to the furnace, initiating nitrogen flow and subsequent heating. The dopant concentrations, dopant source and ball milling method are available in Table 1.

2.2. CAPAD processing

For consolidation using CAPAD, powders were loaded into a graphite die and plunger set under an argon atmosphere in an inert atmosphere glovebox. The die and plunger assembly was then loaded into our custom-built CAPAD apparatus. Once the sample was in the CAPAD, a mechanical vacuum was applied before processing took place. Densification was conducted using 105 MPa of applied pressure and a heating rate of approximately 500 °C $\min^{-1}$. The maximum hold temperature was 1700 °C, and the processing times were less than 15 min.

3. Characterization and property measurement

3.1. Microstructural characterization

The relative density of the samples was measured using the Archimedes method. The microstructure of the samples was characterized using a Philips FEG30 scanning electron microscope (SEM) with both secondary (SE) and backscatter electron (BSE) detectors, for topography and phase distribution, respectively. Samples were also characterized for phase using a BrukerD8 Advance X-ray diffractometer with Cu K_α radiation. Samples were characterized for grain size by measuring individual grains on SEM micrographs using ImageJ. The results reported are averages of at least 100 grain measurements.

3.2. Measurement of hardness and fracture toughness via Vickers indentation

AlN and Tb:AlN samples to be tested by Vickers indentation were wet polished with successively finer grits of diamond abrasive wheels followed by alumina paste on felt wheels to 0.5 μm abrasive particle size. Vickers indentations were made on a Wilson Tukon 2100 using an $F = 10$ kgF indentation force. Under these indentation conditions, uniform easily measureable indents and cracks were produced. In addition, we measured two commercially available materials, laser-quality single-crystal Nd:YAG (1.1% Nd) and undoped, polycrystalline AlN (Coorstek AlN 170). The indents used for hardness and fracture toughness measurements on Nd:YAG were made using $F = 0.25$ kgF indentation force since they shatter using an $F = 10$ kgF indentation force. An example of the indents produced from Vickers indentation of our Tb:AlN samples can be seen in Fig. 1. A micrograph of an indent on Nd:YAG is included for comparison. The indent diagonals and crack lengths were measured using an optical microscope, and the Vickers hardness was then calculated using the relation:

$$HV = \frac{(F)\cdot 2\sin(136^{\circ}/2)}{(2a)^2} \approx \frac{1.8544F}{(2a)^2} \quad (2)$$

where F is the applied load in kgf and $2a$ is the average length of the diagonal in mm.

The mode 1 fracture toughness, K_{IC}, was calculated using the indentation procedure described above and the Niihara relation as described in Ref. [14], with data measured using the indentation procedure described as follows:

$$K_{IC} = 0.035a^{\frac{1}{2}}E^{\frac{2}{5}}HV^{\frac{3}{5}}\left(\frac{l}{a}\right)^{\frac{-1}{2}}\phi^{\frac{-3}{5}} \quad (3)$$

where a is the half diagonal, l is the crack length and ϕ is the constraint factor. $\phi = 3$ [14] and $E = 308$ GPa [19] were used. The values of K_{IC} and HV listed in Table 1 were calculated from averages of at least 10 Vickers indents.

3.3. Thermal conductivity measurement

Due to the significant influence of thermal conductivity on the power limits of laser host materials (Eq. (1)), it is important to understand the influence of microstructure (both grain size and grain boundary cleanliness) and dopant concentration on the thermal conductivity of RE-doped AlN. A standard 3ω method [15] was used to measure the