Optical-grade ceramics: Historical turning point for the design of optical elements

By Akio Ikesue

Though traditionally believed impossible, polycrystalline ceramics can achieve optical properties on par or superior to those of single crystals—and may lead to a historical turning point in the design of optical elements.

When observing the microstructure of ceramics, researchers frequently see residual pores and structural defects, such as segregation at grain boundaries. These defects serve as scattering sources of incoming light, which affect the optical properties of ceramics and make them inferior to single crystals in optical applications.

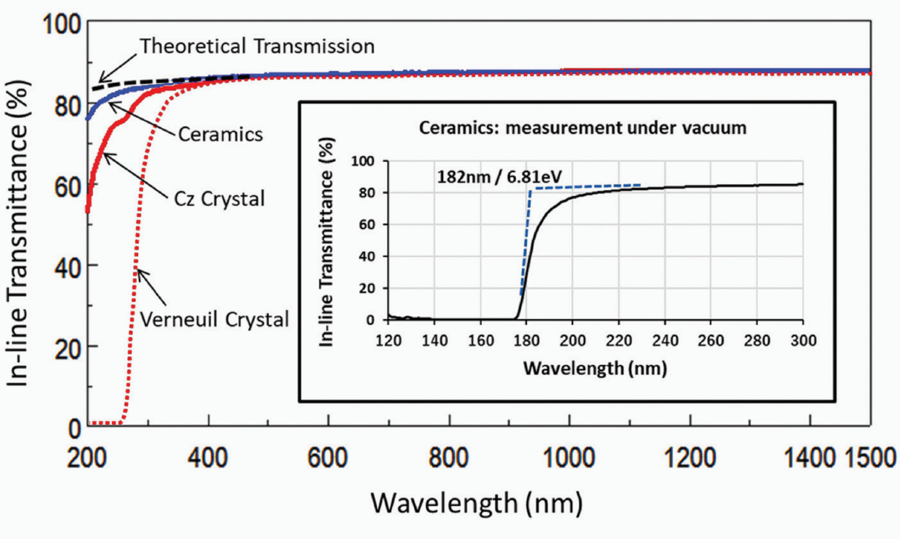
But what if these defects are removed? Could ceramics that contain pristine grain boundaries achieve optical properties similar to single crystals without grain boundaries?

Traditionally, the answer to this question is no. In the 19th century, Lord Rayleigh was awarded the Nobel Prize in Physics for constructing his own scattering theory based on computational science. According to his theory, even if these micrometer- to nanometer-scale defects are removed from the ceramic, scattering from grain boundaries (subnanometer scale) cannot be avoided.

Therefore, most materials scientists believed that the scattering of ceramics with grain boundaries would always be much larger than that of single crystals. For this reason, until the 1990s, there were very few studies on developing optoceramics for solid-state laser gain media, an application in which optical homogeneity is extremely important to ensure high beam quality of the generated laser light.

However, in 1995, the author and his colleagues successfully fabricated a polycrystalline yttrium aluminum garnet ($Y_3Al_5O_{12}$, or YAG) ceramic with optical properties on par with single crystals.[1] They demonstrated highly efficient laser oscillation using this ceramic as the laser gain media. Since then, they also reported on Faraday rotator ceramics with performance exceeding that of single crystals.[2,3] Furthermore, in 2020, they demonstrated spinel ceramics with performance that cannot be achieved with single crystals.[4]

How did they achieve these results? Low optical loss and high optical uniformity are requirements for optical



Credit: Ikesue and Aung, J. Eur. Ceram. Soc.

Figure 1. In-line transmittance curves of polycrystalline spinel ceramics and spinel single crystals synthesized by Verneuil or Czochralski methods, measured under air. Inset shows transmittance at ultraviolet region measured under vacuum atmosphere. Republished with permission from Reference 4.

materials. They discovered ceramics obtained from a solid-state reaction process can overcome obstacles faced by single crystals when pursuing these characteristics.

Single crystals are grown using the melt-growth method, which generally causes optical distortion and segregation at the solid–liquid interface where the single crystal is grown. On the other hand, the solid-state reaction process allows for the fabrication of ceramics without segregation and with defect-free (barring dislocations) grain boundaries. While the scattering from the grain boundaries has not completely disappeared, the scattering properties and optical uniformity is superior to that of single crystals synthesized by the conventional melt-growth method.

Figure 1 compares the transmission spectra of polycrystalline spinel ceramics to the transmission spectra of spinel single crystals with a thickness of 10 mm synthesized by Verneuil or Czochralski methods. Only one residual pore of about 2 μm was detected in the spinel ceramic, so the calculated porosity is as low as 10^{-13}.[4] Therefore, if there is scattering, it must be due to grain boundaries only.

In the visible to infrared wavelength region above 400 nm, the properties of each material look similar, but the optical loss of spinel ceramics was the smallest at 0.07%/cm. In contrast, optical losses of Czochralski and Verneuil single crystals are 0.12 and 0.28%/cm, respectively.

The major difference between ceramics and single crystals, based on the measurement results, is that the transmission characteristics of ceramics with grain boundaries in the short wavelength region (vacuum ultraviolet region) are close to the calculated theoretical transmission characteristics, and the optical bandgap is 6.81 eV. According to Rayleigh's scattering theory, grain boundaries are expected to cause large scattering at short wavelengths. But the measurement results overturned the conventional concept.

Figure 2 shows the optical quality of the polycrystalline ceramic and the spinel single crystals. As can be seen in the figure, the most uniform material is the ceramic. Even for the single crystal produced by the Czochralski method, which generally is used to produce high-quality single crystals, birefringence and a domain structure with mismatched refractive indices are observed.

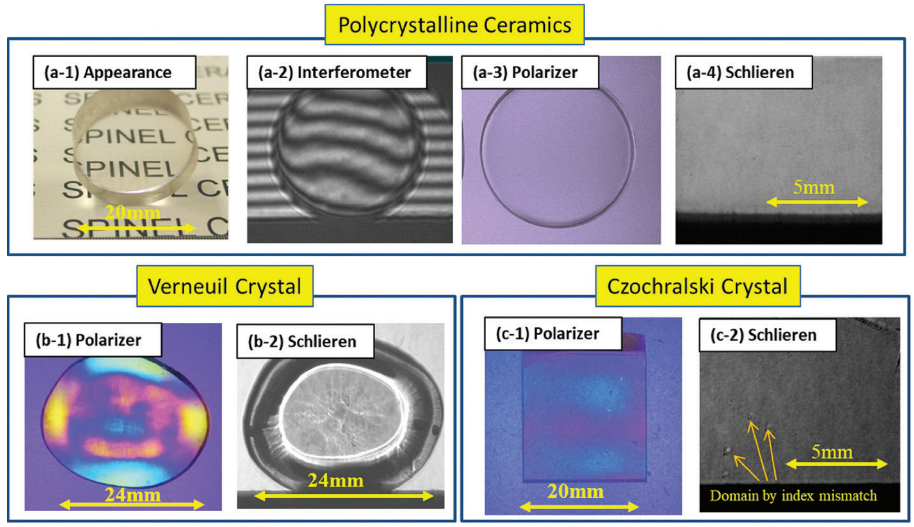


Figure 2. Optical inspection of polycrystalline spinel ceramics produced by sintering method and spinel crystals by Verneuil and Czochralski methods. Republished with permission from Reference 4.

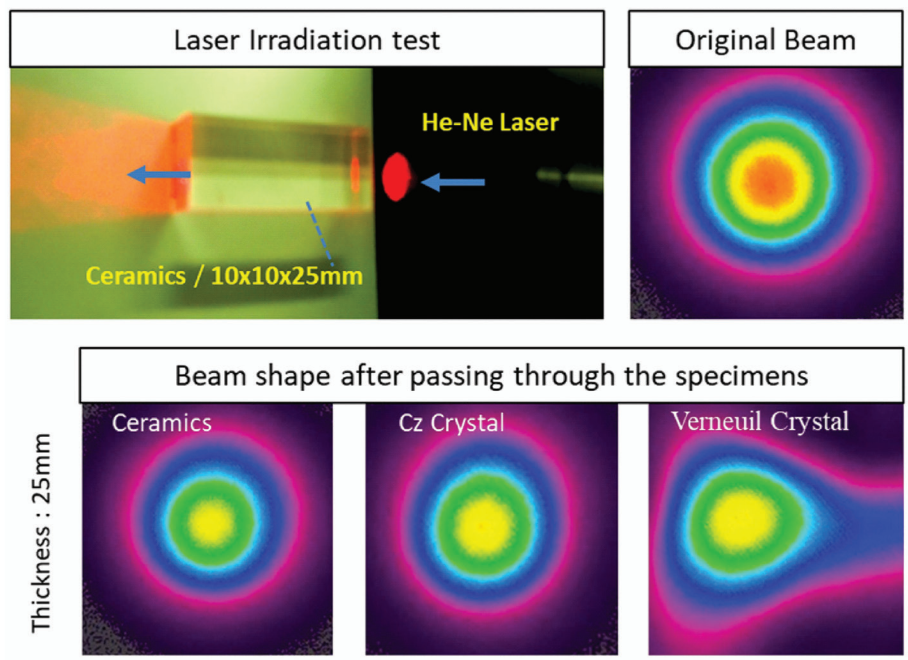


Figure 3. He-Ne laser irradiation test and change of beam pattern after passing through various specimens. Republished with permission from Reference 4.

It should be noted that spinel belongs to the cubic crystal structure, and in principle there is no birefringence in materials with the cubic crystal structure. However, when observed with a polarizing plate, the commercially available single crystals still have significant birefringence. In contrast, ceramics are free of birefringence and have an extremely high extinction ratio of more than 40 dB. Furthermore, grain boundary scattering, which was measured using laser tomography with a wavelength of 633 nm, was not detected in the ceramics.

Figure 3 shows the beam shape of a He-Ne laser (λ = 633 nm) that passed through each material with a thickness of 25 mm. The single crystal material distorted the beam significantly, while the beam transmitted through the ceramics with high optical homogeneity (i.e., it retained its original shape).

Because of the long-held belief in Rayleigh's scattering theory, investigation of ceramics as an optical material has been hampered to date. However, now can be a historical turning point when optical elements will shift from single crystals to polycrystalline ceramics.

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'Transparent' ceramics versus 'optical-grade' ceramics

Transparent ceramics do not necessarily have excellent optical quality. In general, transparent ceramics are of a quality level that allows the surrounding scenery to be clearly observed through the material, and most transparent ceramics have a scattering loss that is 10^2 to 10^6 times larger than that of single crystals.

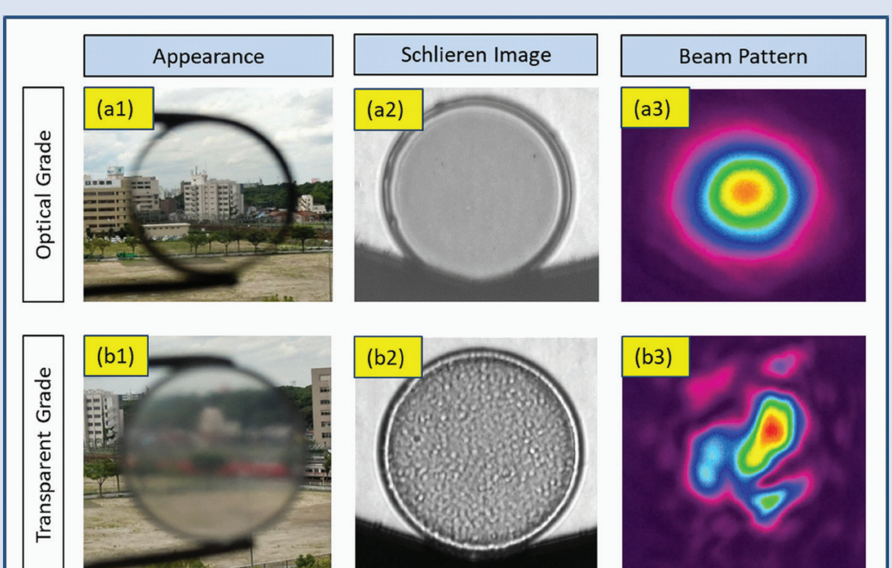
Optical-grade ceramics are ceramics with an optical loss of about 0.1%/cm, excellent uniformity, and optical performance equal to or greater than that of single crystals.

In the figure, images (a1) and (b1) demonstrate the difference between two Y_2O_3 ceramics with a thickness of 20 mm: an optical-grade sample and a transparent sample with low optical loss but poor optical homogeneity. (Y_2O_3 is a promising laser material, and research on laser oscillation from this transparent polycrystalline ceramic has been actively carried out.) The surrounding scenery can be clearly observed through the optical-grade ceramic sample (a1), while for the transparent ceramic with insufficient uniformity, the image of the surrounding scenery is blurry and distorted (b1). However, if the thickness of this transparent ceramic is reduced to less than 5 mm, the surrounding scenery can be observed clearly through the sample.

Many researchers may think that because Y_2O_3 ceramics have a single composition, they do not show nonuniformity after sintering. However, birefringence (optical distortion) occurs in the material due to insufficient grinding or ball milling conditions during the processing of the material. Therefore, although pristine Y_2O_3 has a cubic crystal structure in which birefringence does not exist, due to a slight mismatch in processing conditions, birefringence can generate in the material.

Images (a2) and (b2) show Schlieren images of optical grade and transparent grade Y_2O_3 ceramics, respectively. The Schlieren image allows accurate detection of the refractive index distribution inside the material by passing parallel light source with the same phase. The optical-grade sample has no detectable nonuniformity, whereas the transparent grade has significant nonuniformity.

Images (a3) and (b3) show the beam shapes of the 1,064 nm Nd:YAG laser transmitted through each material. The laser beam transmitted through optical-grade Y_2O_3 ceramics maintains high beam quality, similar to the high-quality spinel shown in Figure 3 (see main text). On the other hand, although the intensity of the laser transmitted through the transparent Y_2O_3 ceramics with insufficient uniformity and residual birefringence is high, most of the beam is diffused and the center part is remarkably distorted.[5] ■



Credit: Aung and Ikesue, *J. Eur. Ceram. Soc.*

Appearance (a1)(b1), Schlieren image (a2)(b2), and transmitted beam pattern (a3)(b3) for optical-grade and transparent Y_2O_3 ceramics. Republished with permission from Reference 5.