Published in final edited form as:

Opt Mater (Amst). 2013 March 1; 35(5): 827–832. doi:10.1016/j.optmat.2012.09.039.

Optical Properties of Anisotropic Polycrystalline Ce⁺³ activated LSO

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Abstract

Polycrystalline cerium activated lutetium oxyorthosilicate (LSO:Ce) is highly desirable technique to make cost effective and highly reproducible radiation detectors for medical imaging. In this article methods to improve transparency in polycrystalline LSO:Ce were explored. Two commercially available powders of different particulate sizes (average particle size 30 and 1500 nm) were evaluated for producing dense LSO:Ce by pressure assisted densification routes, such as hot pressing and hot isostatic pressing. Consolidation of the powders at optimum conditions produced three polycrystalline ceramics with average grain sizes of 500 nm, 700 and 2000 nm. Microstructural evolution studies showed that for grain sizes larger than 1 μm , anisotropy in thermal expansion coefficient and elastic constants of LSO, resulted in residual stress at grain boundaries and triple points that led to intragranular microcracking. However, reducing the grain size below 1 μm effectively avoids microcracking, leading to more favorable optical properties. The optical scattering profiles generated by a Stover scatterometer, measured by a He-Ne laser of wavelength 633 nm, showed that by reducing the grain size from 2 μm to 500 nm, the in-line transmission increased by a factor of 10^3 . Although these values were encouraging and showed that small changes in grain size could increase transmission by almost 3 orders of magnitude, even smaller grain sizes need to be achieved in order to get truly transparent material with high in-line transmission.

1. Introduction

Transparent polycrystalline ceramics are being widely studied for various applications such as infrared windows, optical lenses [1], solid state lasers [2], radiation detectors [3], military armor [4] and LCD displays [5]. The common premise in all cases is to achieve high optical transmission without compromising mechanical, chemical or electronic properties. Since transmission in polycrystalline materials is dependent on scattering sites that are in the path of the light beam, it is evident that a decrease in transparency is directly related to increase in the size and frequency of such sites, which can be porosity, grain boundaries and in some cases extrinsic secondary phases. It is evident that low porosity is required to minimize scattering, however, the role of grain boundaries is difficult to quantify. It is intuitive to infer that larger grains, requiring a light beam to traverse fewer grain boundaries, would minimize scattering and thus increase transmission. On the other hand, in polycrystalline alumina,

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reducing grain size down to 1–2 μm from 15–20 μm has been reported to increase its in-line transmission by orders of magnitude [6]. Therefore fundamentally the grain size itself is taken into consideration rather than grain boundary area, to act as a source of scattering. Moreover the grain size of a dense polycrystalline material also has an effect on the size of the residual pores, which act as scattering sites for the incident light. Therefore for smaller grain sizes the residual porosity at the grain boundaries is similarly smaller in size [7]. Hence, if the size of the scattering site could be reduced well below the incident wavelength there will be no deviation in the path of the incident light. This has involved many researchers to explore nano-sized grains to make optical ceramics with appreciable increase in transparency [8, 9, 10].

Cerium doped lutetium oxyorthosilicate (LSO:Ce) is a particularly attractive optical material for detection of γ -radiation used in PET and CT scanners [11]. Typically single crystals of LSO:Ce are used as detectors due to their good stopping power for x-rays and γ -rays (high density of 7.41 g/cm^3), high light output ($\sim 30,000$ photons/MeV) coupled with fast decay ($\sim 40 \text{ ns}$), and excellent optical transparency. LSO crystals, however, suffer from drawbacks such as cost and reproducibility of useable crystals. The price is driven not only by the inherent expense of the high-temperature Czochralski process by which they are grown, but also by the poor reproducibility of high quality crystals. Melcher *et al.* reported a statistical analysis where they found that the crystals have a reproducibility of mere 15% in terms of its light output which varies by more than a factor of ten [12, 13]. Thus a sustainable alternative involves manufacturing polycrystalline LSO:Ce ceramic by a powder metallurgy method [14]. Although the scintillation properties are close to that of single crystal, the progress towards making polycrystalline LSO:Ce has affected the overall achievable transparency of the ceramic [3]. This is particularly important because it needs to be transparent in the wavelength of cerium, Ce^{3+} , emission (420 nm) [11–13]. The anisotropy in optical properties of scintillator materials can be deduced using first principles calculation [15]. For many anisotropic halide based scintillators with a monoclinic symmetry, the optical isotropic properties are isotropic even though the crystal symmetry may be anisotropic. This however, remains to be evaluated for Lu based silicates with anisotropic symmetry like LSO. Thus the need for increasing the transparency in LSO:Ce has motivated us to examine its microstructure and correlate that to its optical properties.

This investigation focuses on the fabrication of polycrystalline LSO:Ce at different grain sizes. Two commercially available LSO:Ce powders were evaluated and their densification conditions optimized to make viable dense and transparent ceramics. These ceramics were characterized to correlate their microstructure to their optical properties and compare it to an optical scatter model.

2. Theory

Normally, transparent ceramics can be formed only from materials that are optically isotropic i.e., have a cubic structure. When light travels in a non-cubic material across a grain boundary from one randomly oriented grain into another, it encounters substantially different refractive indices, resulting in directional change by refraction. The collective effect of these multiple refractions across grain boundaries assures that some degree of deviation of light from its original path will prevent the material from obtaining full or complete transparency with high real in-line transmittance. LSO has a non-cubic (monoclinic, C_{2h}^6) crystal structure with anisotropic multiple refringence ($n_a=1.825$, $n_b=1.802$, $n_c=1.797$) [14, 16] which highly limits its transparency.

The transmission values for such anisotropic materials can be precisely measured by a Stover scatterometer which plots the scattering profile against the deviation in the path of

the incident beam. The plot basically features a broad low-level base, which represents light that has been diverted from its original path by scattering, and a sharp central peak many orders of magnitude higher, which corresponds to light that passes through without scattering (and is therefore a direct measure of *far-field* transparency) [17]. The ratio of the height of this peak, I (corrected for losses due to reflection at the surfaces), to that obtained from air alone (*i.e.*, with no specimen in the beam), I_0 , provides an experimental value for calculation of transmissivity $T=I/I_0$.

A theoretical model for calculation of the real in-line transmission can also be effectively used for explaining the grain boundary scattering in anisotropic materials [6]. It assumes that the grains are monosize spherical particles with radius r , which are randomly distributed spatially and their average separation is great enough to assure negligible probability that any given photon will experience more than one scattering event. On this basis a theoretical transmission T_{th} can then be calculated by means of the Mie scattering model and simplified by the Rayleigh-Debye-Gans approximation, is given by Equation 1 [5, 18]:

$$T_{th}=T_{max}\exp\left(-3\pi^2\frac{\Delta n^2 r t}{n^2 \lambda^2}\right) \quad (1)$$

where $T_{max}=16n^2/(n+1)^4$ is the maximum possible transmittance as limited by surface reflection, λ is the wavelength of the incident light, $2r$ is the average grain diameter, t is the thickness of the sample, n is the average refractive index and $n=n_{max}-n_{min}$ is the difference in the index of refraction.

3. Materials and Method

Nano-sized LSO powder doped with 0.2 mol% Ce was procured in bulk from Nanocerox Inc., Ann Arbor, MI. The coarser micron-grained LSO powder, also doped with 0.2 mol% Ce, was procured from Nichia Corporation, Japan. The specific surface areas of the powders were measured by the BET technique and used to calculate the average particle size. High-resolution scanning electron microscopy (SEM, Zeiss Supra55) was also used to characterize the powders and the final samples. Hot pressing was done in a graphite furnace (Centorr Associates Inc., Nashua, NH) equipped with high vacuum capabilities ($\sim 10^{-4}$ Torr). Hot Isostatic Pressing (AIP Inc., Columbus, OH) was performed under high pressure in a crucible sealed by LSO grog. Annealed was done in a box furnace (Carbolite RF1600) in air and ultrasonically cleaned and manually ground and polished to a mirror finish. The transparency of the final polished samples was quantified with a Stover scatterometer (Schmitt Industries, Portland, OR), which measures the angular distribution of light from a linearly polarized He-Ne laser source after it has passed through the specimen.

4. Results

4.1. Powder characterization

Commercially procured coarse and fine LSO:Ce powders of different average particulate sizes were evaluated for consolidation into dense ceramic discs. The coarser powder had an average particle size of $\sim 1.5 \mu\text{m}$, and the finer nano-sized powder had an average particle size of 60 nm. Figure 1 shows the particle size distribution and SEM micrographs of both powders alongside for comparison. While the coarser powder has an approximate Gaussian distribution (on a logarithmic horizontal scale), the size distribution of the finer powder is distinctly bimodal, indicating a substantial content of oversize agglomerates, presumably formed by the fusion of many smaller nanoparticles. Even these, however, are substantially smaller than any in the coarser material.

The size differential is also clearly evident in the micrographs, where the powder designated as nano does have much smaller average individual particle size, 30 nm compared to 1500 nm for the coarser material. Although these values are somewhat smaller than in the respective distributions, they clearly confirm the statistical size differential. The micrographs also indicate that some degree of agglomeration is present in both powders. In light of such agglomeration pressure assisted densification methods, such as hot pressing and hot isostatic pressing, were considered as suitable densification methods capable of breaking the agglomerates and consolidating the powder to near theoretical density without considerable grain growth.

4.2. Ceramic Fabrication

Hot pressing of the powders was done in a graphite die and graphite furnace under vacuum. Hot isostatic pressing was done in an alumina crucible filled with scrap LSO as grog material for the non-encapsulated material. The nanopowder turned out to be more sensitive to pressing temperature, but both powders reached maximum density (>99%) in the same temperature range, between 1400 – 1450°C. A hybrid processing method of hot pressing to obtain closed porosity (at 96% density) followed by hot isostatic pressing to obtain full density was explored here. The optimized conditions of temperature and pressing time for the different densification methods explored are summarized in Table 1.

The dense ceramics have a somewhat dark color, associated with loss of oxygen during densification (at such elevated temperatures, carbon is one of the strongest reducing agents known). Since the proximity of graphite (dies, heating elements) inevitably generated oxygen vacancies, the resultant discoloration had to be removed. This was accomplished by annealing the disks in air, allowing oxygen to diffuse in and fill the vacancies. Annealing was done at the lowest temperature that was found effective in removing the discoloration (1050 to 1200°C). Typical disks produced by the conditions listed in Table 1 are 15 mm in diameter and 0.8 mm in thickness. A visual inspection shows that the disks have comparable high contact transparency, a typical photograph of which has been shown in Figure 2. It is imperative to mention here that the contact transparencies of all the disks are similar and thus have not been shown here. The distance or far-field transparency is the most important factor in deciding for its application and remains to be quantified in each of the three cases. Thus the microstructures of the samples were explored and distance transparency evaluated by a Stover scatterometer. These are correlated and discussed below.

4.3. Characterization of Microstructure

When characterized by x-ray diffraction, all three ceramics showed the pure monoclinic LSO phase (PDF#041-0239) and were identical in all respects. Figure 3 shows their SEM micrographs after annealing. The average grain size, measured by the linear intercept method, for LSO I, LSO II and LSO III were $2\pm0.4\ \mu\text{m}$, $0.7\pm0.2\ \mu\text{m}$ and $0.5\pm0.2\ \mu\text{m}$, respectively. The coarser-grained ceramic contained some porosity within the grains as well as micro-cracks extending between grains. In most cases such pores are trapped within the grains themselves, isolated from the grain boundaries and with no kinetically viable path for annihilation. The finer nanocrystalline ceramics, on the other hand, did not contain any noticeable porosity or any secondary phases. The presence of more grain boundaries in the nano-sized ceramics provided sufficient paths for the movement and annihilation of the pores. Since the processing of the samples was done in an identical manner, any difference in their properties could be presumed to be linked to the difference in their microstructural features.

4.4. Optical Properties

Optical properties of the samples were quantified by a Stover scatterometer, which measures the amount of light transmitted by a specimen in the forward direction as a function of angle relative to the incident beam. This enables clear distinction between light that has been deviated from its original path by scattering and light that has passed through without being scattered at all. This provides a measure of the real in-line transmission for highly scattering materials which have low transmission values in the UV range. Figure 4 shows the transparency values measured by the Stover scatterometer for the LSO I to be 0.001% and that for nanocrystalline LSO II and III to be 0.2% and 1.2%, respectively. The relevance of these values with regard to scattering due to grain boundaries is discussed below.

5. Discussion

It is evident that the initial particle size of the powders together with the processing method greatly influences the final grain size of the hot pressed ceramic. Let us first consider the coarser of the two powders. The coarser particles had a somewhat tighter size distribution with a bell curve like distribution peaking at 1.5 μm . The average particle size was close to the final grain size, 2.4 μm in the hot pressed sample, and indicates an increase of 1.5 times [19]. The microstructure of the coarse grained LSO I ceramic (Figure 3a), even at the optimized parameters, showed a large number of pores. Such porosity is particularly deleterious for transparent ceramics since scattering is a strong function of the difference in refractive index (see Equation 1), which is of course much greater between pore and ceramic than between any two of the indices of the anisotropic grains. These trapped pores are apparently a consequence of the initial powder morphology, where voids were observed in the hard agglomerates (Figure 1b). This indicates that the uniaxial hot pressing process did not break down all the hard agglomerates before the particles around the voids densified fully, essentially locking them in position. To remove such porosity higher hot pressing temperatures and longer sintering times were evaluated. However, the density of the discs decreased sharply and the samples became opaque. It can be inferred that the formation of these pores probably occurs at an early stage in the densification process and at higher temperatures and/ or longer holding time the pores act as source points for crack formation, compromising the physical integrity of the ceramic. This is evident in Figure 5, which shows the respective microstructures obtained by hot pressing at 1450°C for 7 h and 1700°C for 2 h. The micrographs show large grains ($\sim 10 \mu\text{m}$) with intragranular fracture around the isolated pores caused by either anisotropy in thermal expansion and/or triggered by chemical considerations (*e.g.*, oxygen depletion at high temperatures) [20].

The LSO II and LSO III ceramics formed from the nano-sized particles, on the other hand, show tremendous grain growth, with an increase in the average size by 8–12 times, respectively, compared to the initial particle size of the powder. This is apparently related to the wide particle size distribution in the nano-sized powder which acts as a driving force for mass transport. The microstructure of the LSO II and III, were observed to be significantly better (basically pore- and crack-free) than the coarse-grained ceramic. However, closer examination did reveal some nano-sized porosity along the grain boundaries and inside the grains. These are residual pores left behind along the grain boundaries. Since the size of such pores is well-below 10 nm, their contribution to scattering is negligible and can be ignored for theoretical comparisons. Although the final densification temperature is similar in both LSO II and LSO III, the grain size in LSO III is at least 200 nm less than LSO II. This can be attributed to the porosity associated with the HIP sample that reduces the grain boundary migration and thus inhibits grain growth [21].

It is, however, interesting to note that the coarser grained ceramic contained intragranular cracking which is conspicuously absent in both the nanocrystalline ceramic. The origin of

these cracks in the coarser-grained ceramic could be due to (i) expansion of gases trapped in an isolated pore and/or (ii) residual stresses due to anisotropy in coefficient of thermal expansion, of LSO. The for LSO has a high anisotropy, with b and c almost 10 times higher than a where a, b, c are the axis directions of the C2/c space group. Therefore thermal contraction during cooling could generate stresses along grain boundaries and at triple point junctions that lead to spontaneous crack formation. It has been shown that formation of such spontaneous microcracks is related to both anisotropy (thermal expansion coefficient and elastic) and grain size [22]. When grain size increases beyond a critical value, the strain energy release rate from flaws such as pores becomes critical leading to micro-cracking [23]. Originally reported by Rice and Pohanka [24] and recently cited by Yousef *et al.*, this critical grain size value for different materials is directly dependent on its inherent anisotropy [23, 25, 26].

The optical quality of these ceramics is dependent on their microstructures (shown in Figure 3) which shows that the average grain size differs by a factor of 4. However, the transparency, as professed by the human eye, can be deceiving and does not register a clear difference. Therefore, although contact transparency, as perceived by text in contact with the ceramics, is similar for all, the far-field or distance transparency is significantly higher for both the nanocrystalline ceramics. This was confirmed by comparing the scatterometer measurements of the three ceramics in Figure 4. The distance transparency is several times higher for the nanocrystalline ceramic disks than for the coarse grained ceramic disk. This was compared to the theoretically calculated distance transparency expected for the three ceramics. The in-line transmission (T_{th}) was calculated by the Mie scattering model (given by Equation 1) and is shown in Figure 6. This also shows the experimentally value of transmission measured by the scatterometer, according to the Beer-Lambert law, as explained in **section 2**. The ceramics have about 50% of their theoretically predicted transmission values. This disparity is related to the microstructural flaws in the ceramics which reduces their transmission values. Also the Mie scatter model is simplistic in nature and assumes the grains to be spherical with enough separation to ensure minimum multiple scattering. These values of real in line transmission, at ~1%, show that the nanocrystalline LSO ceramic is of translucent quality. However, it is imperative to consider that the in-line transmission, as shown in Equation 1, is dependent mostly on the anisotropy in the refractive index of LSO. LSO has a non-cubic (monoclinic, C_{2h}^6) crystal structure with anisotropic multiple refringence ($n_g=1.825$, $n_m=1.802$, $n_c=1.797$). The difference in refractive index, n , is 0.03, which is an order of magnitude higher than other transparent oxide ceramics that are also anisotropic (such as quartz, $n=0.009$, and alumina, $n=0.008$). Therefore the theoretical limit for LSO, due to its anisotropy, is low to begin with [27]. This also decreases the attainable transparency of LSO with pressure-assisted densification methods as compared to alumina or quartz of the same grain size [18]. Thus a decrease in average grain size from 2000 nm to 500 nm increases the transmission, measured at the wavelength of 633 nm, by 1000 times. This shows a trend in significant increase of transmission values by decreasing the grain sizing to the nano-size regime. However, it is still not sufficient to propel the optical quality of this ceramic LSO into a truly transparent polycrystalline material. An estimation by the model shows that the transmission values become substantial when the grain size is decreased to ~ 100 nm. However, the present improvements in transmission values are significant enough to provide sufficient advancement in performance for the new layered detectors now being developed [28, 29], and for currently used CT detectors as well.

6. Conclusion

Two commercially procured LSO:Ce powders having average particulate sizes of 30 nm and 1500 nm were hot pressed at 1400 to 1450°C to get fully dense material. A post-

densification annealing in air was required to remove all the discoloration in the material due to oxygen deficiency. This was accomplished by heat treating in air between 1050 to 1200°C. The two ceramics were found to be equally translucent with good contact transparency. However, the far field or distance transparency, as measured by the Stover scatterometer, indicated that the nanocrystalline ceramic exhibits much better optical properties with transmission values 2 orders higher. This was related to the differences in the microstructure between the two materials. Apart from the average grain size difference of 700 nm and 2 μ m, for the nano-sized and micron-sized powders respectively, the coarse grained ceramic contained trapped porosity and intragranular cracks.

The Mie optical scatter model gave a good approximation for the contribution of grain size to optical scattering. The measured transmission values of the nano- and micron-sized material were close to the calculated theoretical limits. It is projected that by reducing the grains size to ~ 100–200 nm, the transmission values would increase to ~ 40%, which would increase the far field or distance transparency tremendously, making the ceramic material an attractive alternative to single crystal LSO.

Acknowledgments

The authors acknowledge the Department of Energy, USA, grant #DE-FG02-07ER84892 for funding and supporting this work. We also thank Gary Baldoni, Senior Scientist at Radiation Monitoring Devices (RMD), Watertown, MA for the help with coordinating the pressing at American Isostatic Presses, Columbus, OH and Stuart Miller, Scientist at RMD, Watertown, MA for his help with the scatterometer experiments.

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Highlights

1. Powders of Ce^{3+} doped LSO were used to fabricate transparent samples with average grain sizes, 2000 to 500 nm.
2. They produced microstructural variations due to the highly anisotropic properties of monoclinic LSO.
3. The in-line transmittance in the near-UV region changes by 3 orders of magnitude by simply changing the grain size.
4. A model using the Mie scatter theory compares these experimental findings to theoretical predictions optical behavior.
5. For truly transparent LSO:Ce with high in-line transmittance the model predicts material with grain size of about 100 nm.

Figure 1. Characteristics of (a) nano-sized and (b) coarse powders, showing particle size distribution graphs *top row*, and SEM micrographs (note difference in magnifications) *bottom row*.

Figure 2.
Typical photograph of dense LSO:Ce disk in contact with text (back light).

Figure 3. SEM micrographs of the (a) LSO I, (b) LSO II and (c) LSO III (note difference in magnifications). Samples were thermally etched at 800°C in air for 4 h.

Figure 4.

Scatterometer measurements on LSO ceramics. (Horizontal axis is scaled as the square root of the angle, to display a larger angular range without losing small-angle details; vertical axis is logarithmic.)

Figure 5.
Microstructures of disks hot pressed (a) 1450°C for 7 h (b) 1700°C for 2 h.

Figure 6. Transmission of polycrystalline LSO:Ce calculated from the scattering model and compared to that measured for the ceramics (shown by diamond shaped pointers).

Table 1

Conditions Used in Obtaining Dense LSO Ceramics

	Hot Pressing			Hot Isostatic Pressing		
	Temperature (°C)	Pressure (MPa)	Density (g/cm ³)	Temperature (°C)	Pressure (MPa)	Density (g/cm ³)
Micron sized Powder	1450	78	7.38	-	-	-
Nano-sized Powder	1400	78	7.39	-	-	-
	1275	78	7.10	1400	120	7.40
						7.40

LSO I

LSO II

LSO III