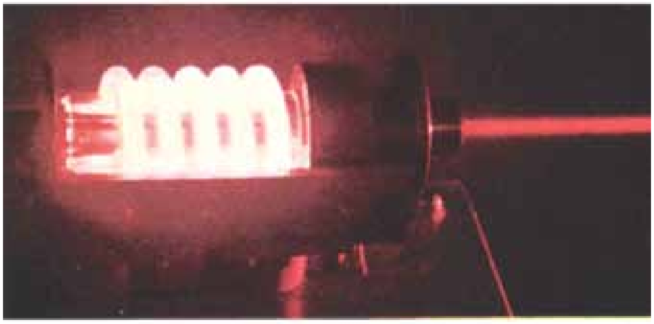
Transparent ceramics for lasers — A game-changer

Martin Richardson and Romain Gaume

Since the birth of the laser age, most high-power solid-state lasers have been based on crystalline or glass material. As the laser field has matured — especially with the introduction of efficient monochromatic diode-based optical excitation — laser architectures, power capabilities and configurations have outstripped the capabilities of conventional crystal fabrication. But, new methods of fabricating doped transparent ceramic laser materials promise to overcome these limitations and to lead to a transformation in advanced laser technology.


The first laser was demonstrated in 1960.

Introduction

The development of lasers always has been governed by the availability of suitable materials. Lasers are based on the efficient manipulation of the populations of excited electronic or vibrational states of specific ions, atoms or molecules in various host media by radiant or electronic energy from subsidiary sources. The first lasers were based on either solid-state single crystals or on gaseous media.

In the early days, researchers favored single crystals doped with rare-earth ions, particularly oxide crystals, as solid-state media for several reasons. First and most importantly, the energy-level structure of specific rare-earth ions (such as Nd^{3+} in yttrium aluminum garnet, or Cr^{3+} in Al_2O_3) offered the highest coefficients for stimulated emission on transitions in the near-infrared spectrum when irradiated with bright, pulsed pump light from the then-available white-light flashlamp sources.

Second, investigators selected materials based on the availability of single crystals with lengths sufficient for optical gain and their optical quality—namely reduced losses from microscopic optical defects, stress-induced refractive index changes and particles inclusions originating from crystal growth processes. Ruby (Cr^{3+}:Al_2O_3) was a preferred material because of its broad absorption bands in the green and blue regions of the spectrum. Because engineers for the watch industry had perfected the synthetic growth of large boules by the Verneuil method (to provide crystal bearings), cylindrical rods suitable for helical flashlamp pumping were available.

With the explosive development of lasers in the following decade, particularly incorporating the technique of Q-switching, which overnight increased the instantaneous power output of lasers by several orders of magnitude, it was not long before optical damage caused by the laser beam fluence (joules per square centimeter), itself, became a principal limitation. This forced high-energy laser developers to adopt glass rather than crystal host materials, despite the smaller optical gain because rods and slabs of this material were not limited in aperture size.

Another paradigm shift occurred in the 1980s and 1990s with the development of high-power, monochromatic continuous wave (cw) laser diode-pumping techniques. High-power continuously operating solid-state lasers necessitated a return to the use of crystalline media because of its vastly superior thermal conductivity. The combination led to the adoption of new diode-pumping architectures, including thin-disk, zig-zag and waveguide designs. This development path toward high-power cw solid-state lasers is approaching a similar power density limit, now defined broadly as thermal damage.

Enter the development of transparent polycrystalline ceramics. The transformative breakthroughs with transparent polycrystalline ceramics have occurred only within the past decade, principally in Japan by Akio Ikesue at World-Lab Co., Nagoya and at Konoshima Chemical Co.

Careful powder synthesis and sintering can dramatically reduce the presence of scattering centers, such as porosity and grain-boundary defects to levels, at least for Nd:YAG, less than or comparable with single-crystal materials.

This approach quickly led to the fabrication of ceramic laser blanks, 10-centimeters-square, with optical quality sufficient to sustain the highest laser powers then available (about 60 kilowatts).

Since then, the Department of Defense's Joint High-Power Solid-State Laser laser program has succeeded in demonstrating two 100-kilowatt diode-pumped Yb:YAG solid-state laser systems in the laboratories of Northrop Grumman Corp. and Textron Corp. This system has since been re-engineered for field tests at the High Energy Laser System Test Facility laser range at the White Sands Missile Range.

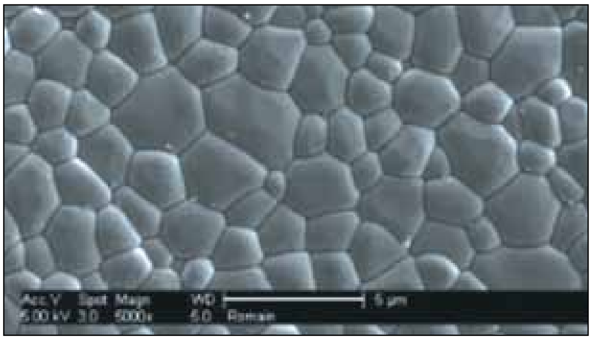
These developments alone signify that ceramic laser materials have come into their own. This is just the beginning; we can foresee this technology being more transformative to solid-state lasers than was the introduction of high-power laser diode pumping almost two decades ago. Ultimately these new materials will lead to completely new generations of solid-state lasers.

Fabrication of transparent laser ceramics

At the turn of the

(Credit: Nd:YAG photo courtesy VLOC)

Single laser crystals grown by conventional Czochralski growth are limited by boule size and corable rod diameters, typically a few centimeters.



(Credit: R. Gaume – Stanford University)

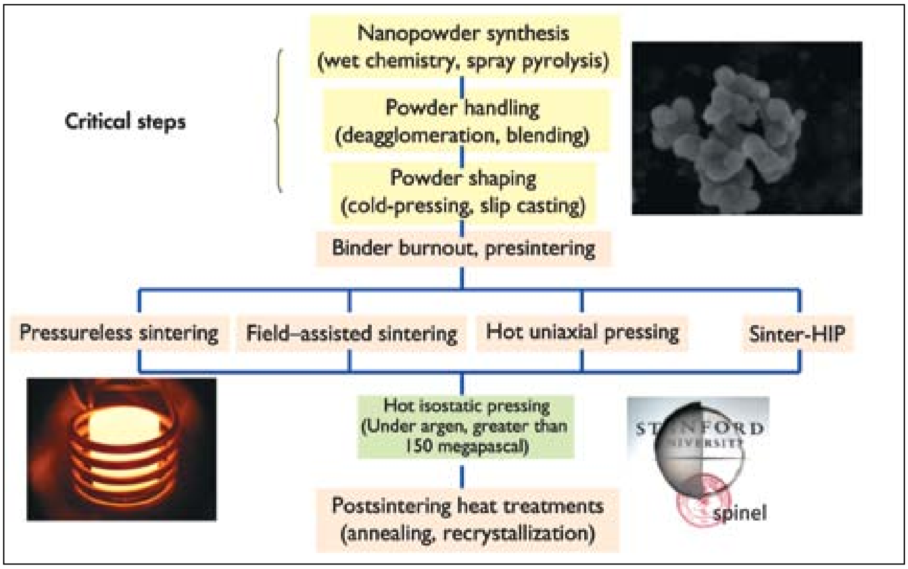
Reduced grain-boundary defects of transparent optical ceramics.

(Credit: R. Konoshima)

10 cm x 10 cm Nd:YAG ceramic laser slabs.

(Credit: Northrop Grumman Corp.)

Northrop Grumman 100-kilowatt JHPSSL laser system.



Critical steps In the fabrication of transparent ceramics

1960s, translucent ceramics attracted attention because of their superior strength, refractory character and possibility of near-net-shape and scalable fabrication.

Fully densified and nontextured ceramics composed of birefringent materials (such as Al_2O_3), however, present random Fresnel reflections at grain-boundaries. Thus, the development of laser ceramics naturally oriented itself toward optically isotropic cubic materials, such as YAG, Y_2O_3 and CaF_2.

There are stringent requirements at every step of the ceramic fabrication process. Longstanding efforts have been made to refine the various fabrication steps and reduce scattering losses. In particular, the use of highly sinterable, high-purity powders; the homogeneous shaping of flawless powder compacts; and the control of fine microstructure by adequate sintering are crucial.

The preferred techniques for producing laser-destined ceramic nanopowders rely on “bottom-up” syntheses, such as wet-chemistry, sol–gel methods or flame spray pyrolysis of organometallic compounds. This allows the production of fine (less than 100 nanometers) and nonagglomerated powders with relatively narrow size distributions. In the case of softer materials, such as fluorides, investigators found that “bottom-down” approaches — using grinding and milling of high-purity single crystals in dry and inert atmosphere — are effective.

Then, it is essential that no impurity be introduced and that the powder be phase-pure during fabrication. This last requirement can be particularly challenging where minuscule departure from stoichiometry can result in significant scattering losses in the final ceramic through the nucleation and growth of secondary phases (e.g., alumina or yttrium aluminum perovskite $YAlO_3$ in nonstoichiometric YAG).

Shaping can be achieved using standard cold pressing techniques, however, slip-casting, tape-casting or gel-casting are usually preferred because they minimize compaction gradients and allow the preparer to scale the size of the cast. Deflocculated aqueous slurries of ceramic powders can be cast into centimeter-thick parts and then carefully calcined to burn out any additive.

The removal of porosity is achieved by pressureless sintering (in a controlled atmosphere or vacuum) or by vacuum hot-pressing followed by hot isostatic pressing. In the case of pressureless sintering, firing can be pursued until complete densification when it is conducted under vacuum (10^{-6} torr (0.133 millipascal)) or until the removal of the open porosity (about 92 percent of theoretical density) when conducted on oxides in oxygen (typically 0.8 T_{mp} (in kelvin) for 10 hours). Good outgassing is critical during the temperature ramp-up to reduce gas entrapment in the collapsing porosity.

Techniques have been developed to favor the kinetics of densification over that of grain coarsening. In particular, several things allow for the complete densification while inducing minimal grain-growth: the addition of lasting sintering additives (e.g., SiO_2 in YAG); transient solid phases (e.g., La_2O_3 in Y_2O_3); transient liquid phases (LiF in $MgAl_2O_4$); or the use of a two-step sintering technique (e.g., Y_2O_3).

If the amount of lasting additive is kept below a few hundreds of parts per million of SiO_2 in YAG, no phase segregation occurs at the grain boundaries and the ceramic is scatter-free. However, at these concentrations, the color centers (likely created as a result of the solid solution) are thought to impact absorption levels in the final ceramic and perhaps degrade laser performance at high power. Such effects are still under investigation.

Hot uniaxial pressing, which can increase the driving force for densification, enables the complete densification of a powder compact at lower temperatures and shorter soaking duration (typically 0.4 T_{mp} (in kelvin) for 5 hours). This leads to ceramics with finer microstructures and superior strengths. The remaining closed porosity can be collapsed via a hot isostatic press.

The future

The unique flexibilities that this new type of laser medium offers are still largely unexplored, and many new pathways of investigation for laser development are available

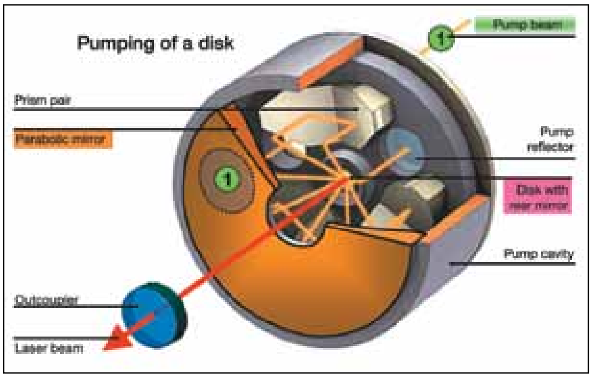
One of the first advantages of transparent ceramics is a greater range of rare-earth-ion-doping concentration. For instance, whereas single-crystal YAG cannot be grown with a Nd^{3+} rare-earth-ion concentration of much greater than 1 percent, with ceramics, concentrations above 5 percent already have been achieved.

This has major importance for several laser architectures, such as high-power, thin-disk lasers, or for compact, end-pumped devices where there are benefits to using thinner laser media with higher small-signal gain. In another example, ytterbium-doped

YAG is important for very-high-power systems where thermal losses must be minimized. With 970 nanometers pumping and an output wavelength of 1,060 nanometers, it possess the smallest quantum defect, the minimum fractional energy deposited directly into vibrational heating. Yet, this can now be made as a ceramic with a wide range of dopant concentrations.

The ceramics approach also extends to the host materials. The power scaling of solid-state lasers requires gain media with high thermal conductivity, low thermal expansion and high fracture strength, so that under extreme thermal loading they will not fracture. For those reasons, ytterbium-doped rare-earth sesquioxides Y_2O_3, Lu_2O_3 are good candidate materials in the development of high-power lasers that require high dopant concentrations, such as thin-disk or microchip lasers. Ceramic processing emerges as a promising approach because the high melting point of these materials (about 2,400°C) precludes the fabrication of large single crystals. Indeed, several groups have obtained encouraging results in their work on the development of laser-grade ceramics of ytterbium-doped sesquioxides, particularly Jasbinder Sanghera's laboratory at Naval Research Lab

Other hosts of interest include cubic fluorides, such as CaF_2, for high-power amplifiers or UV lasers. Although undoped single crystals or polycrystalline fusion-cast CaF_2 can be produced with diameters exceeding 35 centimeters, the fabrication of large-aperture doped fluorides with high optical homogeneity still remains challenging.



Thin-disk laser architecture.

(Credit: courtesy Morris et al, Trumpf, DEPS 2009)

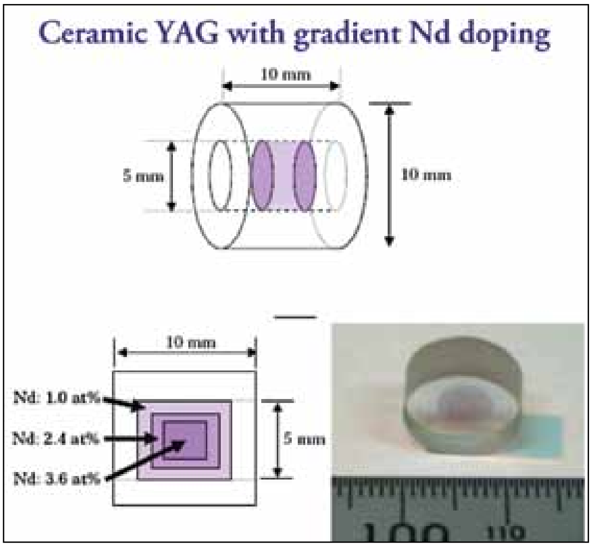
Fabrication is hampered by the high value of the dopant distribution coefficient between the melt and the crystallized solid, which destabilizes the crystallization interface by concentration supercooling. On the other hand, materials of better dopant uniformity and overall optical quality are expected via ceramic processing because it is size-scalable, does not involve such solid–liquid equilibria and allows for better microstructure control.

Transparent ceramics also permit investigators to structure the dopant three dimensionally, known as "dopant engineering." Various groups already have made initial demonstrations of thisprocess. This capability will ultimately allow the design of new laser architectures that will be even more efficient and produce better beam qualities.

The advantages of transparent ceramics are not limited to these transformational options for future lasers. Codoping of multiple-laser, rare-earth ions has already been achieved by researchers at Konoshima Chemical. This opens the possibility of novel lasers operating on new wavelengths and of multiple lasers operating in the same medium.

As explained earlier, the birefringent nature of some optically relevant gain media has long precluded the fabrication of laser-grade transparent ceramics composed of those materials. Recently, however, Takunori Taira's group at the Institute of Molecular Science (Japan) succeeded in producing highly textured and laser-grade ceramics of fluoro-apatites by magnetic field alignment of powders during slip casting.

There are strong economic forces building that will accelerate their development. The market for crystalline laser materials is fairly small (approximately $4 million), and the market for ceramic laser materials is even smaller, at present. But, as high-power and new novel lasers appear, these numbers will grow.



Demonstration of 3D dopant gradients in Nd;YAG ceramic.

(Credit: courtesy A. Ikusu)

The market for transparent ceramics will grow faster because their development is beginning to be driven by their applications in the markets for nuclear and radiological detection (scintillators) and for transparent armors. Both these markets are significantly larger than the market for laser media.

Thus, in summary, to modify an old pun in the laser field, "there's a bright future in store for ceramic laser materials."

About the authors

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Further reading

A. Ikesue, Y.L. Aung, T. Taira, T. Kamimura, K. Yoshida and G.L. Messing, "Progress in Ceramic Lasers," *Ann. Rev. Mater. Res.*, **36** [August] 397–429 (2006). ■