Ceramic laser materials

The word 'ceramics' is derived from the Greek *keramos*, meaning pottery and porcelain. The opaque and translucent cement and clay often used in tableware are not appropriate for optical applications because of the high content of optical scattering sources, that is, defects. Recently, scientists have shown that by eliminating the defects, a new, refined ceramic material — polycrystalline ceramic — can be produced. This advanced ceramic material offers practical laser generation and is anticipated to be a highly attractive alternative to conventional glass and single-crystal laser technologies in the future. Here we review the history of the development of ceramic lasers, the principle of laser generation based on this material, some typical results achieved with ceramic lasers so far, and discuss the potential future outlook for the field.

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Solid-state lasers are widely used in metal processing[1–6], medical applications, such as eye surgery[7–15], red–green–blue (RGB) light sources in laser printers and projectors[16,17], environmental instrumentation measurements[18,19] and optical transmission systems, and have demonstrated potential for future nuclear-fusion applications[20–22]. Conventionally, single crystal or glass is used as the gain medium in solid-state lasers, the first example of which was the ruby laser devised by Maiman in 1960 (refs 23,24). Since the success in generating continuous-wave (c.w.) laser oscillation using an Nd:YAG single crystal at room temperature in 1964 (ref. 25), designs of solid-state lasers using single crystals[26–27] have continually progressed. However, there remains the challenge of finding an approach that can overcome the technological and economical issues of conventional single-crystal laser gain media[28,29].

Recently, ceramic laser technology has emerged as a promising candidate because of its numerous advantages over single-crystal lasers. First, ceramics can be produced in large volumes, which makes them attractive for high-power laser generation. Second, they can provide a gain medium for fibre lasers with high beam quality and can also be made into composite laser media with complicated structures that would otherwise be difficult to fabricate. Besides, ceramics can be heavily and homogeneously doped with laser-active ions. They can also be used to fabricate novel laser materials, such as sesquioxides, which cannot be produced by the conventional melt–growth process. In addition, single crystals with new structures can be fabricated from ceramics through sintering. This type of ceramic-derived single crystal has proved to have high resistance to laser damage and a long lifetime, and is very promising for high-power-density lasers. This novel laser gain medium cannot be produced by conventional single-crystal-growth technology and may offer new laser performance.

An effort to use ceramics as a laser gain medium began in 1964 with Dy:CaF_2 in cryogenic conditions[30,31]. In the 1970s, Nd:Y_2O_3–ThO_2 was successfully used for pulsed laser oscillation[32–34]. Although the achievement marked the onset of ceramic-laser technology, the considerably low oscillation efficiency of the laser disappointed material and laser scientists. In the 1980s, translucent YAG ceramics were developed[35–37], but poor laser oscillation was obtained because it was difficult to achieve high-optical-grade properties with these materials.

In the early 1990s, successful laser oscillation was first demonstrated in Japan using Nd:YAG ceramics[38]. However, the achievement was not widely recognized until a report of the finding was published in 1995 (ref. 39).

Since then, the development of this technology has led to the achievement of highly efficient laser oscillation in compact devices, ease of control of the laser mode, and the generation of highly focused coherent beams. Moreover, the realization of high-power lasers based on ceramic technology has brought the realization of ultrahigh-speed machining technology and nuclear fusion one step closer. In terms of cost and performance, the use of ceramics enables mass production, which may lead to low-cost commercial production of RGB lasers or the production of lasers with customized special functions.

At present, research and development of ceramic lasers are actively performed not only in Japan but also throughout Asia[40–42], America[43–45] and Europe[46–48]. Continuous attention has been paid to ceramic laser technology, and it is not an overstatement to say that a new paradigm based on this technology has gradually formed for the development of future solid-state lasers.

HIGH-EFFICIENCY LASER OSCILLATION

Ceramics are composed of randomly oriented microcrystallites, as shown in Fig. 1a. Fabrication of translucent ceramics began in the 1950s (refs 49,50), but laser-grade transparent ceramics were not fabricated until the achievement of highly efficient ceramic laser oscillation in the 1990s (ref. 39). The reason is simple: translucent ceramics contain many scattering sources, such as grain boundary phases, residual pores and secondary phases, which cause significant scattering losses that prohibit laser oscillation in the translucent ceramic laser gain medium.

The first laser oscillation using transparent Nd:YAG ceramics was demonstrated in 1995 (ref. 39), with a performance comparable to single-crystal laser oscillation. The microstructure of the reported

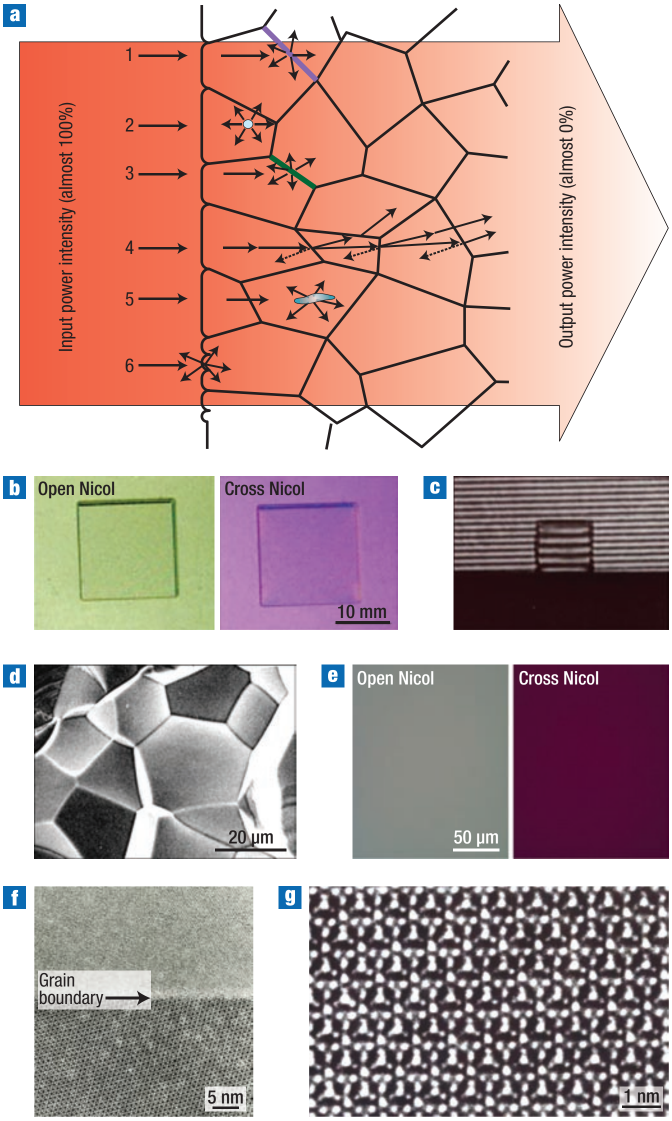

Figure 1 The microstructure of conventional and advanced transparent ceramics. **a**, A schematic showing the microstructure of conventional transparent ceramics, light scattering and the attenuation of input power through the ceramic body. Strong scattering owing to (1) a grain boundary, (2) residual pores, (3) secondary phase, (4) double refraction, (5) inclusions and (6) surface roughness in ceramics prohibits applications in optics. **b**, Polarized images of optical-grade 1% Nd:YAG ceramics on the macroscopic scale viewed through a Nicol (left) and a crossed Nicol polarizing prism (right), showing no double refraction or refractive-index fluctuation. **c**, An interferometry image of optical-grade 1% Nd:YAG ceramics on the macroscopic scale. Stress-free and straight fringes can be seen throughout the whole position of the specimen. **d**, Fracture surface and, **e**, polarized Nicol (left) and crossed Nicol (right) images of optical-grade 1% Nd:YAG ceramics on the microscopic scale. Perfect microstructure without pores and birefringence can be observed. **f**, Transmission electron microscope image of a grain boundary and, **g**, the lattice structure of optical grade 1% Nd:YAG ceramics. Clean grain boundaries with no atomic defects can be seen in optical-grade ceramics.

transparent ceramics is markedly distinct from conventional translucent ceramics. On the macroscopic scale, as shown in Fig. 1b,c, these materials show no double refraction or refractive-index fluctuation, indicating that the optical quality of transparent ceramics is very high. On the microscopic scale, as shown in Fig. 1d,e, no residual pores, secondary phases or optically inhomogeneous parts are observed. Only clean grain boundaries on the atomic scale were observed, as shown in Fig. 1f,g. The numbers of point and line defects and dislocations were also at a minimum level in the grains.

These observations suggest that the formation technology is very important to achieve a perfect microstructure. It is also clear that once the fabrication technology of transparent ceramics that have almost perfect optical homogeneity and microstructure becomes available, it could open the way for the development of ultralow-scattering-loss ceramics. As a result, highly efficient ceramic laser gain media with a performance comparable to single crystal could be developed. However, it has been pointed out that, from the physical aspect of polycrystalline ceramics, transparent ceramics with ideal microstructures will inevitably fail to achieve a high oscillation efficiency and beam quality owing to the grain boundaries.

Figure 2a shows the experimental set-up for c.w. and quasi-c.w. laser oscillations as demonstrated in 1991 for the first time using optical-grade Nd:YAG ceramics. An Ar-ion laser and a laser diode with an operation wavelength of 808 nm were used for side- and end-pumping, respectively. To compare the laser performance of a single crystal with that of ceramics, end-pumping with a laser diode was applied for c.w. laser operation with both gain media, and their lasing performance was reported in 1995 (ref. 39), as shown in Fig. 2b. It was proven that the laser performance of ceramics was almost equivalent to that of the single crystal. (Note that at the time of publication laser-diode excitation was not yet a mature technology, and an antireflection coating was not applied to the sample, resulting in a low oscillation efficiency.)

Now we have realized a slope efficiency of more than 60% using Nd:YAG ceramics. It has been confirmed that the transverse mode of ceramic gain media is single mode with a Gaussian distribution, and the longitudinal mode can also be generated as single mode. In ceramics there are dislocations at the grain boundaries where the crystal orientations are different, and Rayleigh scattering (scattering at grain boundaries) had been considered a fatal problem for ceramic laser gain medium. The demonstration of single-mode lasing using ceramic gain media proved that the grain boundaries do not affect the generation and amplification of a coherent beam.

Other research teams also reported successful laser oscillation from ceramic gain media with excellent results[40–48]. Figure 2c shows a blue and a green ceramic laser using secondary harmonic generation of nonlinear crystals $KTiOPO_4$ (that is, KTP) and $KNbO_3$ (that is, KN). These results also proved that scattering at grain boundaries in ceramics does not affect the laser oscillation mode or the short-wavelength laser oscillation.

THE CURRENT STATUS OF CERAMIC TECHNOLOGY

As single-crystal-growth technology is a process whereby the raw materials are melted and solidified again[51–53], the types of materials that can be produced are fairly limited. Technological issues due to heating of the raw materials above the melting point, such as heat fluctuation during melting and segregation of laser active ions at the interface of the solid–liquid phase during crystal growth, are yet to be resolved. Core, striation, facet and optical stress are often present in the melt–growth single crystals, thus making them optically inhomogeneous[52]. Improvement of the optical quality of the melt–growth single crystal seems almost impossible. In addition, the melting process requires a backup system against electricity breakdown and earthquakes, leading to issues regarding the high energy consumption and initial cost, and the limited productivity.

The development of a practical ceramic laser started in 1995 when c.w. laser oscillation was achieved using polycrystalline Nd:YAG ceramic laser gain medium[39], proving that ceramic materials could overcome both the technical and economical problems of melt–growth single crystals. In 1997, a microchip laser[54] and single-mode oscillation[55] were demonstrated by using heavily doped Nd:YAG ceramics, which could not be easily produced in melt–growth technology. Ultrashort-pulsed laser (pico- to femtosecond) oscillation using Nd:YSAG (yttrium scandium aluminium garnet) (ref. 56) and Yb:YSAG ceramics[57] were also subsequently demonstrated.

In recent years, an interesting result regarding power scaling has been reported. Lasing at 1.47 kW was successfully generated using an Nd:YAG ceramic rod in 2001 (ref. 58), even though its oscillation efficiency was 15% lower than that of single crystal. A 67-kW, quasi-c.w. laser was demonstrated using large-scale laser gain medium (100 mm × 100 mm × 20 mm, see Fig. 3)[59].

It is necessary to understand the damage caused to the grain boundaries of ceramics during high-power laser operation. There are no reports concerning the lifetime of laser gain media, but, in principle, ceramic technology does not limit the size of the laser gain medium. Hence high-power scaling with respect to the medium volume is an important merit for future developments. Although grain boundaries in ceramics are considered a kind of structural defect, it can, however, drastically improve the fracture toughness and thermal-shock resilience.

NOVEL LASER GAIN MEDIA AND FUNCTIONALITIES

By adapting the sintering of solid particles in ceramic processing, it is possible to produce high-melting-point materials at lower temperatures with very short delivery times. Sesquioxide materials (such as Y_2O_3, Sc_2O_3 and Lu_2O_3), for example, are promising laser materials, but their melting point is very high (above 2,400 °C) and the phase transition point is below the melting point. Ceramic technology enables fabrication of lasing devices using such materials. As a result of their high thermal conductivity and the feasibility of large-scale laser gain media, sesquioxide lasers have attracted much attention for the development of high-output-power and ultrashort-pulsed lasers, as an alternative to Ti:sapphire lasers.

Some excellent reports have been published. For example, transparent Nd-doped HfO_2–Y_2O_3 ceramics have been synthesized using hot isostatic pressing[60]; a cryostat laser was fabricated with 77% slope efficiency using high-performance Er:Sc_2O_3 ceramics[61]; and a broadband laser was developed with a 5-nm bandwidth (the bandwidth of normal Nd:Y_2O_3 is approximately 1 nm) by controlling the symmetry property of Nd:Y_2O_3 crystal. These reports demonstrate the potential for the development of tunable and ultrashort-pulsed lasers based on a new principle[62]. Short-pulsed laser oscillation was achieved through Kerr-lens mode-locking (KLM) using Yb:Lu_2O_3 and Yb:Sc_2O_3 ceramics[63,64], and a pulse width of 92 fs and average output power of 850 mW were obtained from Yb:Sc_2O_3 ceramics.

Regarding the shape and configuration, ceramic technology can provide gain media with complex structures that would be difficult to fabricate in single crystal. Figure 4A shows the ceramic forms used at present[65]. Round-type cladding–core element and fibre (simple and end-cap types) are typical examples (see Table 1 for properties. Reprinted with permission from ref. 65). In recent years, fabrication of several tens of micrometre-sized Nd:YAG microsphere lasers that have a truly spherical shape has been challenging[66]. Figure 4B shows the appearance of a composite ceramic that has a cladding–core configuration with neodymium concentrated in the core area. A ceramic composite laser is analogous to the single-crystal type in configuration, but they are different from each other with respect to technical advantages. In the case of a single-crystal composite, the neodymium doping profile is monotonic, whereas the ceramic composite has an almost ideal Gaussian distribution, which facilitates the generation of Gaussian-mode lasing. By controlling the distribution of laser-active ions in a ceramic laser element, the desired mode of laser generation will be readily available in the future.

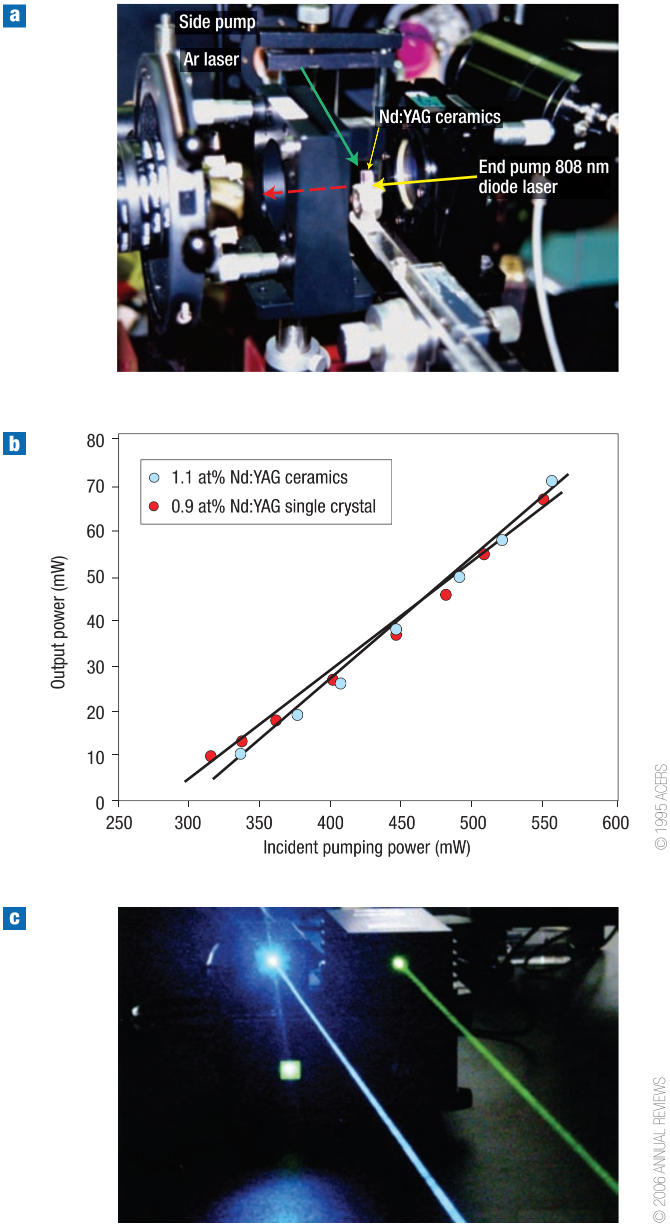

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Figure 2 Laser oscillation using optical-grade Nd:YAG ceramics. **a**, Experimental set-up for c.w. and quasi-c.w. laser oscillation performed in 1991 using optical-grade Nd:YAG ceramics. An Ar-ion laser was used for side-pumping and a laser diode with an operation wavelength of 808 nm was used for end-pumping. **b**, Continuous-wave laser performance achieved in 1995 (ref. 39). Comparison between polycrystalline ceramics (pale blue) and commercial single crystal (red) shows that the laser performance of polycrystalline ceramics is equivalent to that of single crystals. Lines show best fit to the experimental results. Reprinted with permission from ref. 39. **c**, Demonstration of blue and green laser oscillation using Nd:YAG ceramics. The grain boundaries in the ceramics do not affect the laser oscillation mode or short-wavelength laser oscillation. Reprinted with permission from ref. 71.

Figure 5a shows the distribution of neodymium ions in the length direction of the laser gain medium; the desired smooth distribution of neodymium ions was confirmed in the sintered body.

It is very important to control the temperature in laser gain media during laser operation because thermal lensing effects greatly

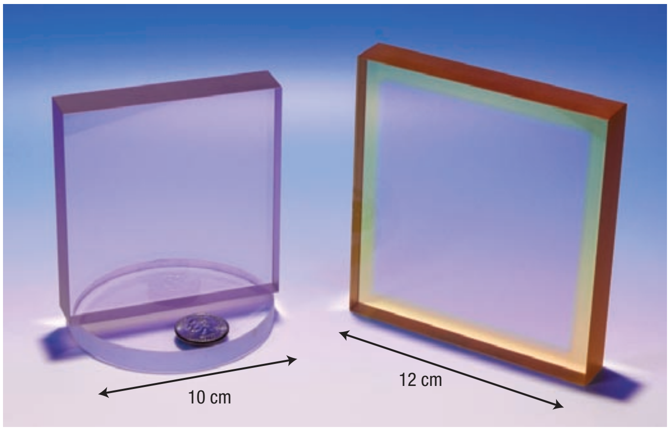

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Figure 3 Large-scale Nd:YAG ceramics developed by Konoshima Chemical. Ceramic with an active aperture, that is, doped with laser-active ions (left) and with samarium cladding (right). Laser generation at 67 kW was achieved using a laser amplifier with five large slabs of this Nd:YAG ceramic. Reprinted with permission from ref. 59.

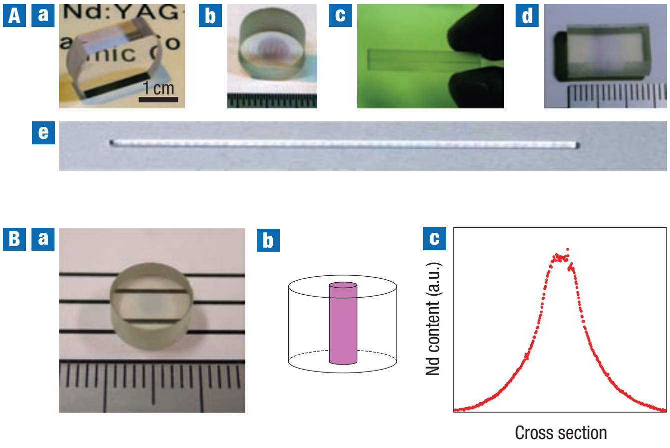

Figure 4 Composite laser elements fabricated by current ceramic technology. **A**, Photos of example structures: **a**, layer; **b**, cylindrical; **c**, waveguide; **d**, gradient; and **e**, fibre cladding–core. Ceramic technology facilitates the fabrication of composite laser elements with complicated configurations and improved functionality, which can not be easily produced by conventional technology[65]. **B**, A cladding–core composite that has a Gaussian distribution of neodymium concentration in the core area. **a**, A photo, **b**, a schematic and, **c**, a plot of the doping profile. By controlling the distribution profile of neodymium ions in the core, the laser oscillation mode can be controlled.

degrade the laser oscillation efficiency, as well as the laser beam quality. Figure 5b shows the heat distribution of a conventional Nd:YAG single crystal that has uniform doping (top panel) and advanced Nd:YAG ceramic rods that have a smooth gradient configuration (bottom panel) during edge-pumped laser operation[67]. Thermal distribution in terms of infrared intensity in the laser gain media was qualitatively observed by a thermo-viewer during laser operation. In the case of single-crystal media, very high heat generation was concentrated near the edge regions. In contrast, it was clear that the ceramic composite element suppressed such sudden heat generation thanks to the gradient distribution of neodymium ions. The effect of design on the heat distribution of gain media during laser operation is under investigation, and will be reported in the near future.

TECHNOLOGICAL GOALS

One of the technical issues associated with conventional melt–growth single crystal technology is the difficulty of achieving heavy and homogeneous doping of laser active ions with a small segregation coefficient in the host materials[28,29]. For instance, the segregation coefficient of neodymium ions in a YAG host single crystal is 0.2 and only about 1at% of the neodymium ions (where at% means atomic per cent) can homogeneously dissolve in the host crystal. Even if heavily doped Nd:YAG single crystal could be grown, it would be hard to obtain optical-grade quality. The grown crystal rod may contain facets and a concentration difference of neodymium ions in the length direction of the crystal rod. Such technical issues have not yet been solved.

In 2005, fabrication of Nd:YAG single crystal, a single-crystal microsphere, and single-crystal composite by means of sintering were reported[68]. The principle of fabrication of the ceramic from a single crystal by sintering is shown in Fig. 6a. An Nd:YAG ceramic that has a relatively small grain size is bonded with a seed (single crystal). Under high-temperature treatment, absorption (merging) of small grains of the ceramic into the seed crystal occurs. In solid-state crystal growth (SSCG), when the surface energy of the single crystal is sufficiently smaller than the surface energy of the small grains of ceramics, continuous grain growth occurs and polycrystalline ceramics transform into single-crystal materials. (Hereafter this type of single crystal is referred to as 'SSCG single crystal'.)

The microstructure of the middle step of the crystal-growth process for heavily doped Nd:YAG and the appearance of the SSCG single crystal are shown in Fig. 6b,c. The lasing performance of pore-free ceramics and SSCG single crystal (both with

Table 1 Characteristics of composite laser elements fabricated by current ceramic technology.

	(Layer, Fig. 4Aa)	(Cylindrical, Fig. 4Ab)	(Waveguide, Fig. 4Ac)	(Gradient, Fig. 4Ad)	(Fibre cladding-core, Fig. 4Ae)
Power and efficiency (for samples with a high Nd^{3+} content)	Very good	Very good	Very good	Very good	Very good
Beam quality (e.g. low thermal lensing, low thermal double refraction)	Very good	Very good	Very good	Very good	Very good
Beam mode control	Same as conventional	Good *	Good *	Same as conventional	Very good
Beam pattern control	Same as conventional	Good	Good	Very good	Same as conventional
Functionalization (e.g. laser element with *Q*-switch)	Very good	Good	Same as conventional	Same as conventional	Good
Miniaturization of laser oscillator	Good	Good	Good	Good	Very good

* Downsizing the core is necessary.

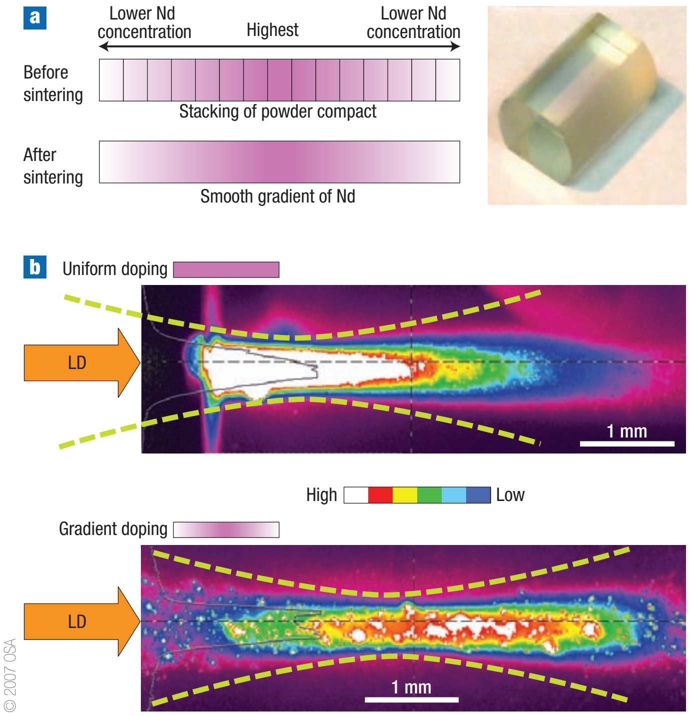

Figure 5 Gradient-distribution ceramic laser composite. **a**, A schematic showing the distribution of neodymium ions before and after sintering. **b**, A comparison of the thermal distribution for uniform and gradient neodymium distribution. the images show the thermal distribution during edge-pumped laser operation using a laser diode (LD) of conventional Nd:YAG single crystal with uniform doping of neodymium, top panel, and advanced Nd:YAG ceramics with smooth gradient doping of neodymium, bottom panel. Concentrated heat generation can be controlled by gradient doping. Reprinted with permission from ref. 67.

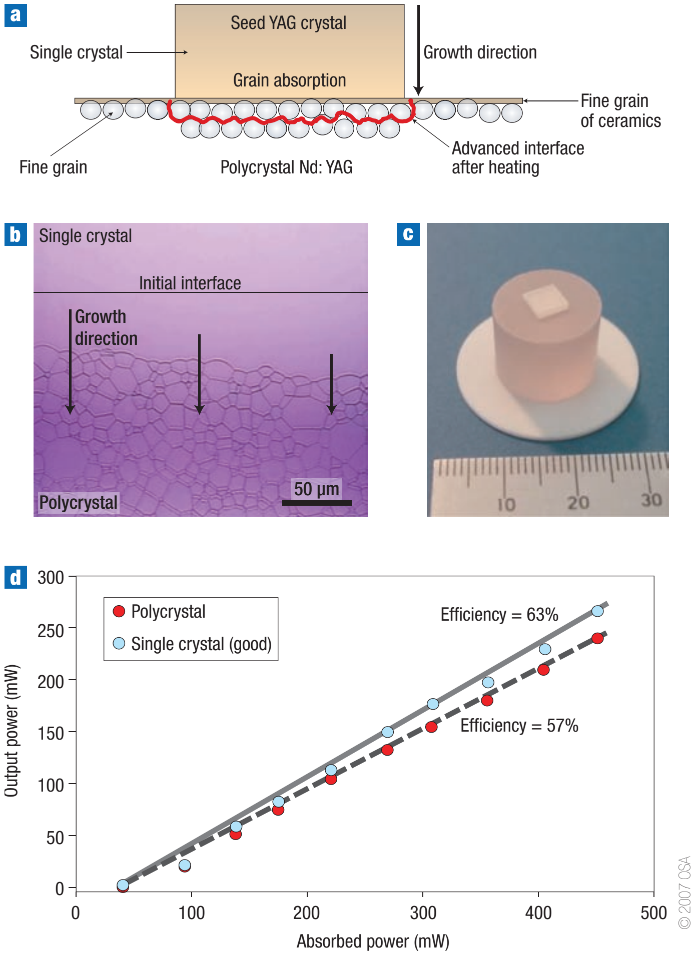

Figure 6 The growth of SSCG single crystal and its laser performance. **a**, The principle of single-crystal growth by the solid-state crystal growth (SSCG) method. **b**, A cross section (reflecting-polarized image) of Nd:YAG ceramics at the middle step during crystal growth[66]. Single-crystallization occurred in the solid state, but scattering sources, such as residual pores and double refractions, are hardly observed. **c**, Appearance of Nd:YAG single crystal by the SSCG method. The crystal growth rate was about several hundreds to thousands of micrometres per hour. **d**, The laser performance of SSCG Nd:YAG single crystal in comparison with that of the polycrystalline Nd:YAG ceramics with the same composition (2.4at% Nd:YAG). Parts b and c are reprinted with permission from ref. 66. The SSCG single crystal has higher laser oscillation efficiency because of the absence of grain boundaries.

2.4at% Nd:YAG composition) are shown in Fig. 6d. Although the SSCG single crystal was not tested for laser damage, improvement of laser conversion efficiency owing to the absence of grain boundaries was clearly shown.

The SSCG single crystal fabricated by sintering does not have optically inhomogeneous parts, such as a core or facets, as found in the conventional single crystal fabricated by melt–growth. In addition, it has a low dislocation density. In polycrystalline ceramics, large numbers of dislocations (defects) exist at grain boundaries, and this may cause damage during ultrahigh-power laser operation or long-term operation. However, the production of SSCG single crystal by sintering can provide fabrication of high-performance gain media, exceeding the polycrystalline ceramic gain media, and allow fabrication of single crystals that cannot be easily achieved by conventional melt–growth technology, such as heavily doped Nd:YAG, composite single crystal and single-crystal microspheres.

So far the crystal structure of ceramic lasers has been limited to cubic structures as a result of the polycrystalline microstructure of ceramics. However, the sintering fabrication technology for single crystals can provide not only cubic laser gain media, but also hexagonal and tetragonal laser gain media. Fabrication of single crystals by sintering began with the growth of single-crystal ferrite[69,70], but the fabrication of laser-grade single crystals was not achieved. Now, SSCG technology has shown potential for the fabrication of laser bulk crystals, composite single crystals and several tens of micrometre-size microspheres for whispering-gallery-mode lasers.

Another key technology that is as important for single-crystal fabrication is that of nanostructured ceramics. For example, alumina with a hexagonal crystal system has grain sizes ranging from a few micrometres to several tens of micrometres. Such alumina ceramics can not be used in laser applications because of their very low in-line transmittance and very large double refractions. However, if nanostructured alumina ceramics can be fabricated with a grain size sufficiently smaller than the wavelength of laser oscillation, the in-line transmittance can be drastically increased, enabling nanostructured alumina ceramics for future laser generation.

LIMITATIONS AND SOLUTIONS

Despite all the advantages of ceramic technology listed above, the existing ceramic technology is not yet perfect. Remaining challenges include efficient production of large-scale laser gain media for the development of high-power lasers, stable laser operation, formation of grain boundaries that are highly resistant to laser damage, and development of a novel technology that can provide laser gain media with other microstructures besides cubic crystals.

Because particles of submicrometre- to nano-sized raw materials are used for fabricating ceramic materials, the handling process is difficult. In particular, in the fabrication of ceramic laser gain media, preparation of powders with almost perfect packing is a critical process. Packing with low homogeneity causes voids (residual pores, which are scattering sources) in the sintered product. However, it is very difficult to prepare a powder without any inhomogeneous parts. Therefore, further innovative technologies are required for the fabrication of laser gain media on a scale as large as glass laser gain media. There are two ways to overcome this issue: one is to establish a precise method of moulding, and another one is to bind together small-sized laser gain media to obtain a large-size laser gain medium.

Ceramic is formed with grains of different crystal orientations. Therefore impurities can easily accumulate, and the dislocation density, which is a crystal defect, is extremely high at grain boundaries. When the grain boundary of ceramics is observed using optical tomography, very strong local scattering at grain boundaries can be seen. To improve the laser conversion efficiency and optical damage, grain boundaries where optical scattering occurs must be completely eliminated.

Crystals other than cubic crystals have birefringence, which causes extremely high scattering losses in ceramics. Fabrication of nanostructured materials or fabrication of SSCG single crystal is very important for the development of promising laser materials, to meet the customer's requirement and to expand the range of customer choice.

FUTURE OUTLOOK

The development history of ceramic lasers at a global level is still very short, less than ten years. Ceramic technology originated in Japan and still prevails internationally, but recently successful laser oscillation using ceramic laser gain media has also been reported from the USA and China. The successful development of ceramic lasers is also expected in Europe and other parts of Asia.

Fundamentally, ceramic lasers are not merely a potential replacement for the conventional single-crystal laser. Until now, lasing properties that cannot be achieved by conventional single-crystal technology have been realized using ceramics, though the field is still at the research and development stage. It took about 20 years to commercialize melt–growth single-crystal materials for industrial applications, and similarly, some time may elapse before commercially viable ceramic lasers can be obtained.

Possible future applications for ceramic lasers include environmental measurements, high-speed metal machining (for example, cutting and welding), cutting-edge medical devices for surgery and diagnostic tools, laser guidance systems, RGB light sources for projectors and laser television, and laser drivers for nuclear fusion. In fact, some of these applications are already being tested for product development.

In the near future, ceramic technology is anticipated to create innovative technology on various passive optics, such as infrared windows, lenses, prisms, and active optics such as nonlinear optics for laser wavelength conversion, scintillators for detecting radioactive rays, and optical shutters and isolators that require optical polarizing and modulation. Moreover, the use of ceramics is envisaged to spread to other technologies in piezoelectric and pyroelectric materials, such as $Pb(ZrTi)O_3$ (that is, PZT), $BaTiO_3$ and $PbTiO_3$, and engineering ceramics that require high-strength and fracture toughness, such as ZrO_2 and Si_3N_4, which are beyond the field of optics.

The content of scattering sources in traditional ceramics has hindered their applications in optics. Thus, it is imperative to challenge the theoretical limit for achieving the best ceramic technology that can produce perfect ceramic microstructures. Once this is achieved, the fabrication methods for perfect material microstructures can be extended to other technologies.

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