Transparent Ceramics Spark Laser Advances

Livermore's Solid-State Heat Capacity Laser is a showcase for transparent ceramics that offer clear advantages over traditional crystals.

HARD, brittle, and heat- and corrosion-resistant, ceramics have served society for thousands of years. Ceramics are made by shaping and then firing a nonmetallic material, such as clay, at a high temperature. The earliest ceramic products were pots and bricks prepared by forming a wet clay mixture and then drying and firing it in an open hearth. The result was a hardened substance that was impervious to heat, resistant to wear, and opaque—perfect for baking, cooking, and building.

Today, because of their formability, electrical insulating quality, heat-resistance, and robustness, modern ceramic parts are ubiquitous in the aerospace, electrical, and automotive industries. They are used not only

Technician Kurt Cutter works on the Solid-State Heat Capacity Laser (SSHCL)—the most powerful diode-pumped, solid-state laser in the world, capable of generating 25,000 watts of light for up to 10 seconds.

for bricks and pottery but also for products ranging from golf clubs to rocket nozzles. Even the medical industry has embraced ceramics as prostheses for bones and teeth.

In the 1960s, scientists at General Electric (GE) discovered that under the right manufacturing conditions, some ceramics, especially aluminum oxide (sometimes known as alumina or sapphire), could be made translucent. These translucent materials were transparent enough to be used for containing the electrical plasma generated in high-pressure sodium street lamps. During the past two decades, additional types of transparent ceramics have been developed for applications such as nose cones for heat-seeking missiles, windows for fighter aircraft, and scintillation counters for computed tomography scanners.

In the early 1970s, during the first part of a 33-year career at GE, Livermore physical chemist Thomas Soules pioneered computer modeling of light transmission through translucent ceramic alumina. His model showed that microscopic pores in ceramic, mainly trapped at the junctions of microcrystalline grains, caused light to scatter and prevented true transparency. The volume fraction of these microscopic pores had to be less than 10^{-5} for high-quality optical transmission; in other words, the density had to be 99.99 percent of the theoretical perfect crystal density. "Although achieving this density is a tall order," says Soules, "the model showed no intrinsic reason preventing someone from making a truly transparent ceramic."

Recently, Japanese scientists have developed techniques to produce ceramic parts that rival the transparency of traditional crystals (grown from a single seed) and exceed a single crystal's fracture resistance and robustness of manufacturability. In particular, scientists at the Japanese firm Konoshima Ltd., a producer of ceramic construction materials and industrial chemicals, have been looking for markets for their transparent ceramics.

Livermore researchers realized that these ceramics might greatly benefit high-powered lasers used in the National Ignition Facility (NIF) Programs Directorate. In particular, a Livermore research team began to acquire advanced transparent ceramics from Konoshima to determine if they could meet the optical requirements needed for Livermore's Solid-State Heat Capacity Laser (SSHCL). (See *S&TR*, April 2002, pp. 19–21; October 2004, pp. 8–9.) Researchers have also been looking at and testing possible applications of these remarkable materials for use in other Livermore lasers. Potential applications include scalable components and advanced drivers for laser-driven fusion power plants.

The transparent ceramics furnished by Konoshima are well suited to amplify the laser light of the SSHCL, the most powerful diode-pumped, solid-state laser in the world. As in other solid-state lasers, the heart of the SSHCL is a solid, transparent insulator "doped" with a small amount of an optically active ion, in this case, neodymium ions. The insulator's neodymium ions are "pumped," that is, raised to an excited state either by gas-filled flashlamps or, in the case of the SSHCL, arrays of high-power laser diodes. Some of the energy stored in the excited neodymium ions is released when the laser beam passes through the insulator. In this way, the insulator amplifies the input pulse to the required power and energy while maintaining the original beam's spectral and temporal characteristics. In its current configuration, the SSHCL has four transparent ceramic insulators, called amplifier slabs, measuring 10 by 10 by 2 centimeters that are pumped by 16 arrays of battery-powered laser diode bars.

The SSHCL uses the world's largest laser-quality transparent ceramic amplifier slabs, which measure 10 by 10 by 2 centimeters.

Ceramics Fill Critical Need

The SSHCL's amplifier slabs were originally made of crystalline gadolinium–gallium–garnet doped with neodymium ions (Nd:GGG). However, obtaining the large boules has proved difficult. The vendor that formerly supplied the slabs was no longer able to manufacture the required size.

"Growing large boules of highly refractive single-seed crystals is difficult," says Soules. "Although it is being done on a regular basis for some materials, such as sapphire, it is still as much an art as a science." Ceramic neodymium-doped yttrium–aluminum–garnet (Nd:YAG) seemed an ideal solution to the production problems associated with crystals and also offered several advantages. "We wanted to exploit the power of ceramics to advance our laser programs as well as produce amplifiers to replace the unavailable crystal amplifiers," says Soules.

Researchers working with the transparent ceramics include Soules, electronics engineers Mark Rotter and Scott Fochs, laser technicians Balbir Bhachu and Kurt Cutter, mechanical technician Charles Parks, and mechanical engineer and SSHCL project leader Bob Yamamoto. The Livermore researchers worked with Konoshima to obtain ceramic slabs for the SSHCL. "Konoshima developed a process to produce amplifier slabs of unprecedented size and thickness," says Yamamoto.

The SSHCL requires slabs that are 2 centimeters thick, but the original production method used by Konoshima, called slip casting, is only effective for slabs 1 centimeter thick and smaller. The solution developed at Konoshima entails firing two 1-centimeter-thick slabs at a lower temperature and then vacuum sintering and bonding the two pieces using hot isostatic pressing (HIP). In this process, the two slabs are heated to high temperatures in a furnace under a pressure of several hundred megapascals of argon gas. (See the box on p. 16.) The result is the world's largest laser-quality transparent ceramic Nd:YAG slabs. When viewed from a certain angle, the bond joining the two pieces is barely visible and does not cause any loss of transmission.

The ceramic slabs are sent from Konoshima to Baikowski-Japan for polishing. Once at the Laboratory, the slabs are covered with a two-layer coating developed at Livermore, which reduces surface reflection of both the laser beam and the laser diode-pump light to very low amounts.

"Transparent ceramics allow us to scale the SSHCL in a simple manner," says Yamamoto. He explains that other laser systems increase power by adding laser beams. However, the SSHCL remains a single-aperture system regardless of power. "We can increase laser output power linearly in one of three ways: add more amplifier slabs, increase the cross-sectional area of the amplifier slab, or run diodes at 20-percent duty cycle (the percentage of time the laser operates) instead of 10-percent duty cycle. Adding slabs is the easiest way. It's similar to going from a four-cylinder engine to six to eight."

With the transparent ceramic slabs in place, the SSHCL can generate 25,000 watts of light for up to 10 seconds at 10-percent duty cycle. The SSHCL is pulsed, turning on and off 200 times per second to generate a beam that can penetrate a 2.5-centimeter-thick piece of steel in 2 to 7 seconds depending on the beam size at the target. The system recently achieved 67,000 watts of average power with five ceramic slabs for short fire durations. The laser, which is powered by batteries, was conceived as part of the U.S. Army's program to develop directed-energy technologies to defend against missiles, mortar shells, and artillery. Unlike chemical lasers designed for the same purpose, an SSHCL is small enough to be installed on a transport vehicle or helicopter.

An SSHCL can also be used to clear land mines. Its pulses can dig through several centimeters of dirt to expose and neutralize a mine. The SSHCL team received a 2004 R&D 100 Award from *R&D Magazine* for developing this concept. (See *S&TR,* October 2004, pp. 8–9.)

Suppressing Unwanted Light

The transparent ceramics are also being used to suppress amplified spontaneous emission (ASE), which is light emitted spontaneously in all directions by the excited neodymium ions. The presence of ASE degrades laser performance. The method used to remove ASE from the Nd:GGG slabs entailed diffusion-bonding sections of cobalt-doped GGG to the edges of the crystal slab. This process was expensive, time-consuming, often unreliable, and not possible for ceramic Nd:YAG. The SSHCL team worked with Steve Letts in Livermore's Chemistry and Materials Science (CMS) Directorate to bond ASE-absorbing materials to the ceramic slabs with specialized epoxies.

The most promising approach for suppressing ASE is to partially sinter four separate ceramic pieces of YAG doped with samarium (Sm:YAG) and then co-sinter them to the ceramic Nd:YAG in a manner similar to that used to create the thick amplifier slab. The continuous piece that is formed frames the edges of the ceramic Nd:YAG slab with ceramic Sm:YAG. The team worked with Konoshima to design the Sm:YAG pieces and develop this method. (See the figure on p. 14.)

The ceramic Sm:YAG edge-cladded slab has been installed in the SSHCL and has been shown to be effective in reducing ASE. What's more, the edge cladding does not absorb any diode-pump light, which helps limit wavefront distortion by minimizing heating.

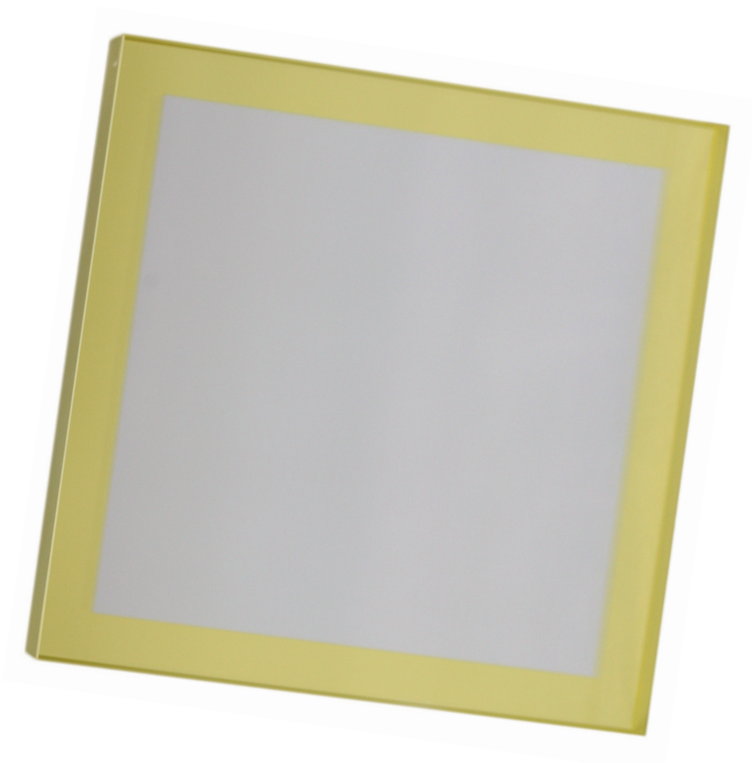
"Making ASE suppression an integral part of the slab and having it be transparent to the pump light had not been done before," says Soules. "Our design also eliminates a lot of manufacturing and reliability problems we encountered with previous approaches and opens up design

The SSHCL can penetrate a 2.5-centimeter-thick piece of steel in 2 to 7 seconds. In the sample shown, the time span of the entire shot was just 10 seconds.

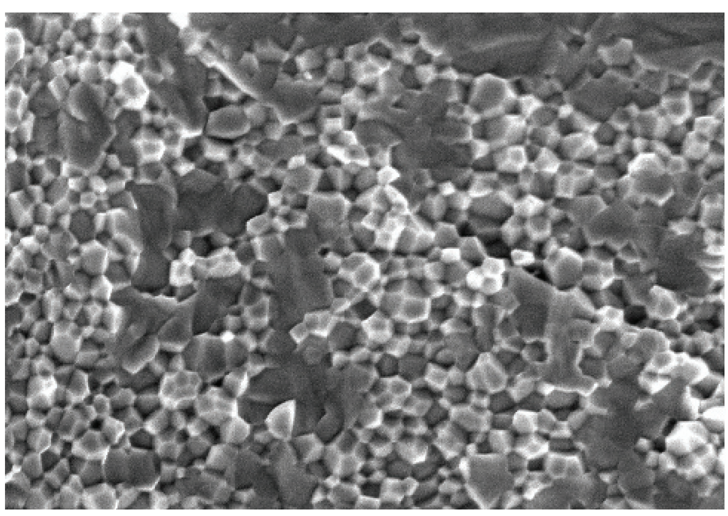
possibilities such as scaling to larger apertures by incorporating ASE suppression into the amplifier slab."

Clear Advantages

Tests show that the transparent ceramics exceed specifications. The amount of scattered light, for example, is similar to that measured from single crystals of Nd:GGG or Nd:YAG. The ceramic slab contains tens of thousands of boundaries between microcrystallites, or "grain boundaries," in the path of the laser light. However, the laser light passing through doesn't "see" the many grain boundaries that measure less than 1 nanometer wide. "The performance of transparent ceramic slabs in the SSHCL is astounding, easily meeting or surpassing the performance of the crystal Nd:GGG slabs," says Soules.



The most promising approach for suppressing amplified spontaneous emission entails co-sintering four separate ceramic pieces of yttrium–aluminum–garnet (YAG) doped with samarium to the amplifier slab of neodymium-doped YAG, forming one continuous piece.



50 micrometers

This image taken with a scanning electron microscope shows the three-dimensional grain structure of a Livermore transparent yttrium–aluminum–garnet ceramic sample.

In what Soules describes as the "acid test" for optical quality, a laser beam is passed back and forth through the ceramic slabs, and any distortions in the wavefront are measured. The ceramic slabs have passed this test with no more wavefront distortion than that expected from polishing.

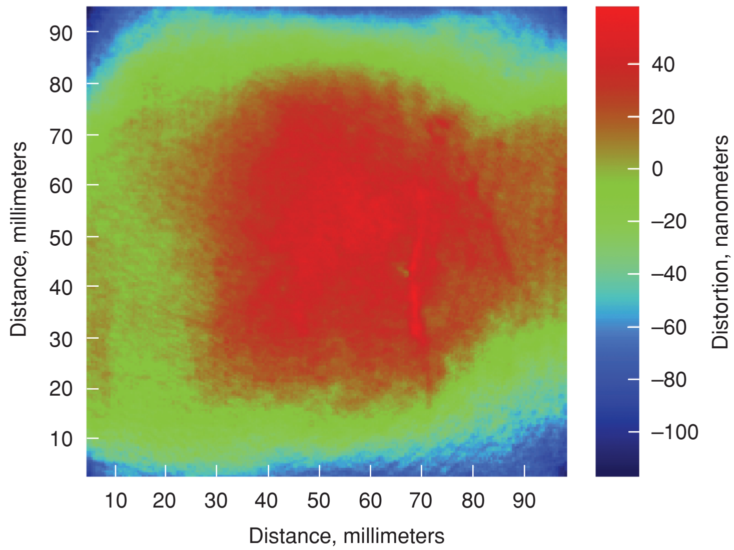
The Livermore team has found that amplifier slabs made from transparent ceramics offer several advantages over those produced from crystals. Perhaps most important is that these slabs can be obtained regularly, on time, and without unexpected additional costs. Ceramic materials are also more easily fabricated into large sizes for greater power, and ceramics can be made any size and shape. "We're only limited by the size of the sintering furnace," says Soules. The time required to produce the slabs from start to finish is much shorter than the time to grow crystal boules—days instead of weeks. In addition, multiple samples can be fired in one furnace at the same time.

Ceramic slabs are also tougher than single-seed crystal slabs and much less apt to undergo a catastrophic fracture. When a crystal slab fractures, the fracture can "run," extending some distance from the original crack and often branching or making a random turn into the center of the crystal to relieve stress. Because cracks are impeded by grain boundaries, ceramic fractures don't run as easily or randomly. Ceramics also measure lower residual stress, which is stress that resides in a material after it has been manufactured. Significant residual stress distorts the laser beam and can make the material more susceptible to cracking.

Ceramics can accommodate higher concentrations of dopants (rare-earth ions such as neodymium), which could permit pumping at wavelengths that might otherwise be impractical. Dopant concentrations are highly homogeneous in ceramics and can be controlled precisely. In crystals, dopants tend to segregate toward the bottom of the growing boule.

Ceramics also offer the possibility of novel composite structures. For example, a single slab could have an "active" layer of YAG doped with neodymium ions and another layer composed of YAG doped with chromium ions. Such a design is called a passive Q-switch, which turns on the laser after saturation. The different materials would be co-sintered to produce a single integral structure in the same way that the team co-sinters the Sm:YAG ASE suppressors. Another possible approach is to embed different powders with the same host before sintering the slab to create a gradation of neodymium ions or incorporate the passive Q-switch.

"We're making tremendous progress in understanding, using, and manufacturing transparent ceramics," Yamamoto says. "We need slabs that are very pure because contaminants absorb heat and become hot spots. The quality we're seeing is outstanding."



Wavefront distortion through the Konoshima ceramic is extremely low, as seen in the test results from passing a laser beam back and forth through the ceramic amplfier slab. The distortion at the edges is caused by polishing.

Livermore researchers have produced 15-millimeter-diameter samples of transparent ceramic yttrium–aluminum–garnet. A sample exhibits excellent optical qualities after hot isostatic pressing (right).

On Their Own

Soules is also working on a Laboratory Directed Research and Development–funded project with Joshua Kuntz, Alex Gash, and Richard Landingham from CMS and Kathleen Schaffers from NIF. The researchers are making small samples of transparent ceramics to better understand the key process variables involved. "We don't plan to produce large transparent ceramic slabs for lasers, but we want to understand the critical parameters that affect transparency and then extend the technology to other materials," says Soules.

The team has produced 15-millimeter-diameter samples of transparent Nd:YAG and determined the most important parameters that affect their quality. In making these pieces, the team largely followed the Japanese production methods and used an in-house furnace to vacuum sinter nanopowders. The pieces were then sent out for HIP. Finally, the pieces were returned to Livermore for coating and testing. "It's not an easy process, but once optimized, it's a repeatable process," says Soules. The samples have shown excellent optical qualities.

The team is also exploring new ways to make the initial nanopowders. Borrowing on expertise developed in CMS over the past 5 years, the team is making nanopowders based on aerogel and solgel processing and then sintering them. Another technique being tested uses a combustion process to generate the powders by burning an organic solid containing yttrium, aluminum, and neodymium and then collecting the smoke, which consists of spherical nanoparticles.

In addition, the team is using manufacturing techniques, including Livermore-developed extruding techniques, that allow the transparent ceramic parts to have complicated shapes, such as shells and tubes, for improved coupling to the pump light and for transferring heat more efficiently. Different materials can be co-extruded and then sintered into a single transparent piece. For example, an amplifier slab can be made so that part of the structure acts as a light guide to focus pump light from laser diodes into regions with a high concentration of neodymium ions located in the smaller, central region of the slab.

In collaboration with the University of California at Davis and Stanford University, the team is investigating other ceramic materials for lasers. These include other oxides and fluorides. The team also wants to identify new commercial sources of ceramic Nd:YAG and determine their viability for Livermore lasers. “If laser scientists have a need for a different material, we will try to determine if it is

Making Transparent Ceramics

Transparent ceramics are produced by forming a nanopowder of ingredients into the desired shape, then sintering (heating below the melting point) in a vacuum to form an aggregate of microcrystals that exhibit optical and thermal qualities almost identical to those of a single-seed crystal.

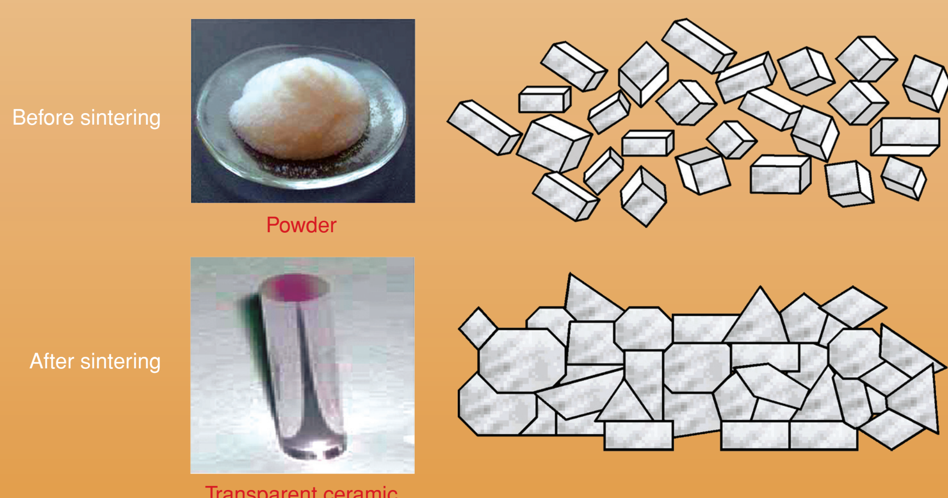
Livermore researchers are experimenting with several methods to make transparent ceramics. Following a recipe similar to that used by Japanese scientists, they begin with a solution of yttrium, neodymium, and aluminum salts and add a solution of ammonium hydrogen carbonate. The precipitate is then filtered, washed, and dried. At this point, the co-precipitated amorphous carbonate is made up of agglomerates of particles measuring about 10 nanometers in diameter. The particles are heated to about 1,100°C to decompose the carbonates and obtain particles of neodymium-doped yttrium–aluminum–garnet (Nd:YAG) measuring about 100 nanometers in size. Highly agglomerated, the particles are treated ultrasonically, and then the large particles are removed to obtain a uniform small size.

In a process called slip casting, a suspension of the fine powder is poured into a plaster of paris mold and allowed to settle. Excess water is poured off, and the mold is set aside to absorb most of the remaining water and dry. The result is a porous structure called a preform structure, which is removed from the mold. The preform still contains many pores and is only about 40 to 45 percent dense.

The preform structure is then fired in a vacuum at high temperature for many hours. This sintering process involves surface atom diffusion, resulting in the particles fusing together and decreasing the total surface energy. Some of the pores are squeezed out, and the structure shrinks but still retains its overall shape. Additionally, many physical and thermal properties undergo dramatic improvements during sintering.

Under a microscope, the structure begins to resemble a three-dimensional mosaic of small, irregularly shaped, and densely packed grains fused together with channels running along grain edges and in regions where the grains do not fit together well. Finally, the channels become narrower and break up into small spherical pores that are usually trapped at corners where more than two grains meet.

Because the sintering process still leaves a few trapped pores, the ceramic parts are sent to a vendor for a 1- to 2-hour treatment in a hot isostatic press. The press drives out the last pores by heating the sample to high temperatures under enormous pressure (several hundred megapascals). Provided that no impurities exist, the remaining trapped pores collapse, and the finished part achieves the greater than 99.99 percent theoretical density required for nearly perfect transparency.



Transparent ceramics are produced by forming a nanopowder of a desired shape, then sintering the sample in a vacuum to form an aggregate of microcrystals.

feasible," says Soules. "We will share technology we develop with industry." He notes that several companies are "jumping in" to provide ceramics for lasers. "We want to work with domestic suppliers as well as maintain strong relationships with our Japanese suppliers."

Looking Ahead

Yamamoto and colleagues are designing a megawatt-class, solid-state ceramic laser that builds on the success of the ceramics in the SSHCL. The new design features 16 ceramic laser slabs measuring 20 by 20 by 4 centimeters.

Transparent ceramics promise to greatly expand the availability of low-cost, high-quality components in much larger sizes than is possible with traditional crystals. For example, many classes of laser designs could benefit from ceramic-based laser structures such as amplifiers with built-in edge claddings. Also, ceramic amplifier slabs could provide more robust and compact designs for high-peak-power, fusion-class lasers for stockpile stewardship and high-average-power lasers for theatre missile defense. Scientists working at NIF, which houses the largest laser in the world, are interested in transparent ceramics because of their high thermal conductivity, high resistance to fracture, and potential damage resistance. Livermore scientists planning the Mercury laser, a large-aperture, high-average-power laser with a high repetition rate, are also interested in the team's work. Mercury is a compact version of a potential prototype for an inertial fusion energy power plant.

"For the first time, laser designers have available gain materials with all the desirable properties of crystals for scaling lasers to high average power," says Bruce Warner, principal deputy associate director for NIF Programs. "At the same time, transparent ceramics maintain all the desirable properties of high-quality laser glass such as that used in NIF—superb optical quality combined with the ability to scale to large apertures. These materials promise to transform the laser landscape and bring us one step closer to realizing laser drivers suitable for the production of energy in laser-driven fusion power plants."

With their optical quality, high thermal conductivity, and manufacturing savings, transparent ceramics are changing the way lasers are being designed and built. Scientists engineering the next generation of lasers have a new class of materials available to expand laser science.

—Arnie Heller

Key Words: hot isostatic press (HIP), laser diodes, neodymium-doped gadolinium–gallium–garnet (Nd:GGG), neodymium-doped yttrium–aluminum–garnet (Nd:YAG), passive Q-switch, samarium-doped yttrium–aluminum–garnet (Sm:YAG), sintering, Solid-State Heat Capacity Laser (SSHCL), transparent ceramics.

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