Defense program advances U.S. ceramics industry

A new ceramic production process, in which the Russians currently lead, could be the key to industrial recovery in the American Midwest, reports Robert Gallagher.

Lawrence Livermore National Laboratory (LLNL) is looking for industrial corporations willing to commercialize an efficient, new, productive ceramic production technology developed in Russia and already commercialized there, but mainly in laboratory development in the United States. As a result of Soviet work led by physicist A. G. Merzhanov over the past 20 years, the Russians are ahead of the West and Japan in this area of advanced ceramics.

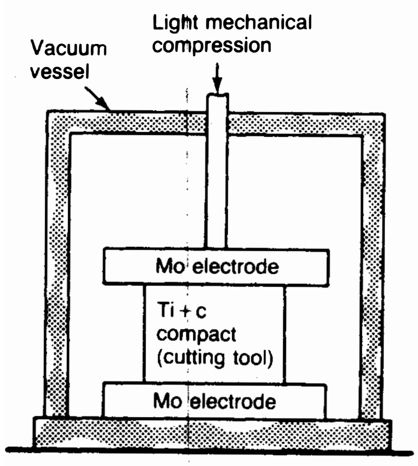
Further research and development is necessary to bring the combustion process into the pilot-plant stage in the United States. A single, large organization such as a consortium of industrial firms, or a state government, could close the Russian lead by sponsoring such research and development, and then, construction of a pilot plant for the new ceramic production technology. It is known in Russia as "self-propagating high-temperature synthesis" (SHS), or as "combustion synthesis" in the United States.

The Defense Advanced Research Projects Agency (DARPA) has asked Livermore to transfer this technology to American industry. In the interview below, Birch Holt, Livermore Laboratory principal investigator of the program in SHS, describes the technology and outlines what might be required to move to pilot-plant operation.

This technology represents a unique kind of opportunity for the states of the industrial Midwest, now almost a wasteland of industrial collapse in steel, auto, railroads, and other industries. Aggressive programs to develop and introduce new technologies like the SHS ceramics production process, or plasma steelmaking, will enable the United States to "leap-frog" over production bottlenecks, and may provide the key to an industrial recovery in states like Illinois, Michigan, Ohio, and Pennsylvania.

Self-propagating high-temperature synthesis involves igniting the energy-producing, exothermic reactions that produce ceramic compounds from mixtures of their elemental constituents. A small amount of energy, for example, about 10 watt-hours for reaction of titanium and carbon, is applied at a high energy-flux density (10^9 watts per square meter)

FIGURE 1
Schematic representation of Soviet pressure-aided densification



through a tungsten wire or with a laser pulse or other means, to ignite the reaction of formation of, for example, titanium carbide from a mixture of titanium and carbon powders. If the powders are pressed into the shape of a desired part, and pressure is applied to the shaped reacting mixture, the result of combustion could be a finished product (see **Figure 1**).

Self-propagating high-temperature synthesis of ceramics takes only minutes, and promises to replace many existing U.S. ceramic production methods for the production of non-oxide ceramics. Many existing methods require days to produce a single batch of powder or parts in an inefficient process little different from baking a cake.

The Russians have been working with the process for about 20 years, and according to DARPA reports, now have nine plants in operation; each produces 1,000 tons per year of titanium carbide, silicon nitride, or supercanthol ($MoSi_2$). The Russians are presently licensing use in the West of a three-reactor unit which produces a continuous average output of 90 kilograms per hour of ceramic powders.

Self-propagating high-temperature synthesis has the unusual characteristic that it releases more energy than it consumes, thousands of times more energy in the process reactors that the Soviets are now retailing. **Table 1** shows the basic physical properties of self-propagating high-temperature synthesis, and some examples of the energy transformation rates of SHS processes (expressed in output per kilowatt hour). These figures represent only those conditions attained so far.

Existing processes for production of non-oxide ceramic powders, such as the Acheson process for production of silicon carbide, use industrial furnaces that are run in batch cycles that take days.

Existing ceramic part production technologies also require several days' process time. The most advanced is known as injection molding. In this process, ceramic powders are molded with a wax into the desired shape; this process takes about a minute. However, the part must then be processed through an oven for three days to remove the wax binder, and then be fired in a high-temperature furnace over a period of two days, before complete.

The production figures of **Table 2,** show that self-propagating high-temperature synthesis is a more advanced method of ceramic powder and part production.

Production of ceramic powders. As with all SHS, one begins with powders of the elements (or compounds) to react, mixed in an appropriate ratio, and placed into a reaction vessel filled with an inert, oxygen-free atmosphere (for production of titanium carbide, a mixture of titantium and carbon powders). Electrical energy (about 10 watt-hours) is applied to a portion of the material near the surface, through a tungsten wire or graphite strip (see **Figure 2**). This small amount of energy excites the exothermic (energy-producing) reaction of formation of the ceramic compound desired. The reaction then propagates completely through the mix. The energy *released* in production of titanium carbide is in the area of 850 watt-hours per kilogram of reacting material.

In addition, metal powders may be reacted with nitrogen to produce nitrides. The Livermore program has made advances in this area that surpass the work done in Russia.

Either way, the resultant ceramic powders can be used for the fabrication of ceramic parts in the conventional way, or for lubricants, electrolytes, and for grinding and polishing applications. The Russians have replaced tungsten with SHS-produced titantium carbide in cutting tools, and also fabricate

TABLE 1
Physical parameters of self-propagating high-temperature synthesis

Basic physical parameters		Output per kwh
Energy flux density applied	2×10^9 W/m$_2$	Potentially infinite
Released (TiC)	10^8 W/m$_2$	
Ignition energy required	7-14 Whrs	
Energy released (TiC)	850 Whrs/kg	
Examples		
Ceramic powders		
Titanium carbide	3 tons (30 kg batches)	
The chemical furnace		
Tungsten carbide parts and titanium carbide powder	0.4 tons (batches of 3 1-kg parts and 1.5 kg powder)	

TABLE 2
Comparison of SHS with other existing ceramic powder and ceramic part manufacturing methods

	EFD (W/m²)	kg/kwh	Process Time
Ceramic powders			
SHS	10^9	3,000	Minutes
Acheson process	NA	2	3 days[2]
Ceramic parts			
SHS	10^9	10,000	Minutes
Injection molding	10^5	1	5 days

it into substitutes for industrial diamonds. Silicon nitride is used for rocket nozzles and ceramic auto parts, and supercanthol for high-temperature industrial heating elements.

Finished parts. In this application, the elemental powders are pressed into the shape of a final product, as is done in powder metallurgy. The ignition energy is then applied to the surface of the shape. The application of pressure to the combusting material is presently required to prevent the product from being porous. With this technique, up to 96% theoretical densities have been achieved for titanium carbide at Livermore. Also at the national lab, solid aluminum nitride has been produced in an SHS developed there.

Parts produced with SHS include titanium nickelide wire, plate and tubes used aboard Soviet aircraft for fuel and air lines; this material is produced in batches of hundreds of kilograms.

The ideal SHS parts production technology would not require application of pressure. Livermore scientists are also working on ways to achieve this.

Gasless combustion castings. Ceramic castings is the latest technology development in self-propagating high-temperature synthesis, and is still in the research and development stage in Russia. In this process, SHS ignition of a mixture of a metal oxide, aluminum powder, and carbon, results in the reduction of the metal oxide by the aluminum, and the combustion of the reduced metal with the carbon, to produce a carbide. The process produces a slag of aluminum, in molten form. Of course, this slag can itself be a useful material.

FIGURE 2
Schematic representation of Soviet synthesis of titanium carbide powder