

# Powering Next-Generation Energy Storage Devices



**W**ORLDWIDE demand for electronics is surging. So, too, is reliance on the energy storage components that power them. Shrinking device sizes coupled with the need for longer charge life and better performance have

spurred research efforts into battery designs that not only meet escalating benchmarks but do so economically and with fewer harmful byproducts.

To address this growing challenge, a Livermore-led effort drew upon the Laboratory's additive manufacturing expertise to enable effective 3D printing of functional energy storage devices. Specially formulated feedstock materials, Energy Inks, will equip manufacturers with the tools to rapidly explore new designs of batteries and supercapacitors in addition to devices used in catalysis, filtration, and remote sensing.

## Charging Ahead

Energy Inks rely on the increasingly popular process of direct ink writing (DIW), which enables extrusion and deposition of materials including metal, glass, and ceramics to build objects in unparalleled, 3D forms. Applying DIW technology to energy storage devices in particular will allow researchers and manufacturers to optimize components for operation within specific size, performance, efficiency, and cost constraints. "At Livermore, we constantly develop advanced materials with functional and improved properties," says staff scientist Swetha Chandrasekaran, who headed the award-winning Energy Inks

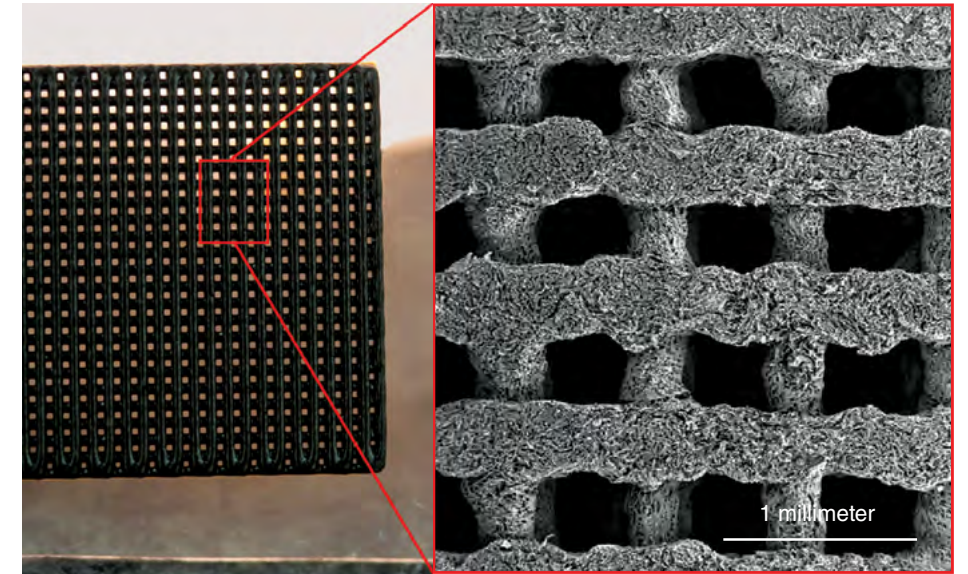
Livermore development team for Energy Inks (from left to right): Marcus Worsley and Swetha Chandrasekaran. (Photo by Garry McLeod.)

project. "Many of these materials are reserved for the Laboratory's mission space, but our project has global applications."

While 3D printing of solid-state electrode architectures has been achieved, Energy Inks introduces direct printing of fully operational components. Printing functional devices such as batteries with DIW requires a carefully designed ink formulation that retains the desired device characteristics both during extrusion and after hardening. Chandrasekaran explains, "Controlling the composition and rheological behavior of the feedstock inks is crucial to forming self-supporting filaments that maintain their shape as they span over previously deposited layers. To fabricate parts from a variety of materials, each ink must be tailored to deliver the optimal combination of rheological properties, such as viscoelastic response and shear thinning." The result is a method that supports customized properties and lowers manufacturing cost.

Support such as that from the Department of Energy's Technology Commercialization Fund (TCF) enabled the team to develop materials that could be made available for academic research institutions and commercial users. "I'm especially thankful to business development experts in the Laboratory's Innovation and Partnerships Office—Genaro Memphin and Annemarie Meike (now retired)—for assisting in the TCF collaboration with industry partner MilliporeSigma," says Chandrasekaran.

MilliporeSigma now offers three Energy Inks in the marketplace. The first, graphene oxide (GO), is used as an electrode material in lithium-ion batteries, supercapacitors, and electrocatalytic devices. Based on GO nanosheets, the ink promises fast charge rates, increased cycle life, and improved gravimetric capacitance for next-generation energy storage devices. In 2020, Chandrasekaran's team collaborated with researchers from the University of California at Santa Cruz to build a graphene aerogel electrode. The component overcame traditional limitations of supercapacitors to achieve the highest areal storage capacitance to date, reaffirming the potential of extrusion-based architectures.



A component printed with Energy Inks graphene oxide product (left) retains the material's characteristic porous quality (right) and related functionality.

## Enabling High Performance

The remaining two Energy Inks are composed of ceramic microparticles to withstand harsh conditions. The team's yttria-based zirconia (YSZ) ink is designed to operate within chemically corrosive, low-pH environments. YSZ's tunable porosity and high surface area make the ink best suited for filtration, catalysis, and thermal insulation. The ink also presents great utility as a retrofitting material in existing energy storage, conversion, separation, and sensing devices. The mechanically robust B<sub>4</sub>C ink, on the other hand, exhibits a unique combination of lightweight and ultrahigh temperature resistance, making it ideal for high-wear components and heat exchangers in settings such as nuclear reactors and lightweight body armor. While robust once printed, both liquid inks exhibit high viscosity and stable dispersion to enable a smooth printing process.

Staff scientist Alyssa Troksa focused on optimizing YSZ ink for use in membranes and filters as well as characterizing its long-term stability. Working alongside MilliporeSigma required regular testing and data sharing of rheological behavior and extrudability to ensure consistent compositional quality. Troksa says the team needed to find a way for ceramic nanoparticles to remain dispersed and suspended over long periods of time. "We tuned the relative concentrations of nanoparticles and polymer binders and additionally adjusted storage conditions and mixing procedures until we found a formulation best suited for printing and performance," she says. Each ink was analyzed to ensure a shelf-life of at least 10 months for inclusion in the MilliporeSigma product catalogue.



Materials scientist James Cahill, who worked on the B<sub>4</sub>C ink design,

describes intricacies of the development process. "The main challenge was ensuring sufficiently high solids loading of particles to enable sintering to near-full density," he says. "However, because solids loading affects viscosity, and therefore printability, the process is full of trade-offs." He says the team achieved between 50 and 60 percent solids loading by volume, which optimized the sintering process while maintaining printability.

With applications as diverse as consumer electronics, transportation, and medical devices, printable energy materials will empower researchers and industries to incorporate novel approaches when conceiving new energy technologies. Energy Inks' customizability and low relative cost to traditional manufacturing techniques present innovators with a powerful tool to help facilitate a broad transition to clean, efficient, and effective energy storage components. "Seeing Energy Inks translated from the benchtop to a commercially available product was extremely fulfilling as a scientist," says Troksa. "We're helping shape and drive growth of the high-performance energy device field. I'm excited to see what people will create with this technology."

— Elliot Jaffe

**Key Words:** additive manufacturing, battery, ceramic, direct ink writing (DIW), energy storage device, Energy Inks, functional device, MilliporeSigma, R&D 100 Award, supercapacitor.

For further information contact Swetha Chandrasekaran (925) 424-6816 (chandrasekar2@llnl.gov).