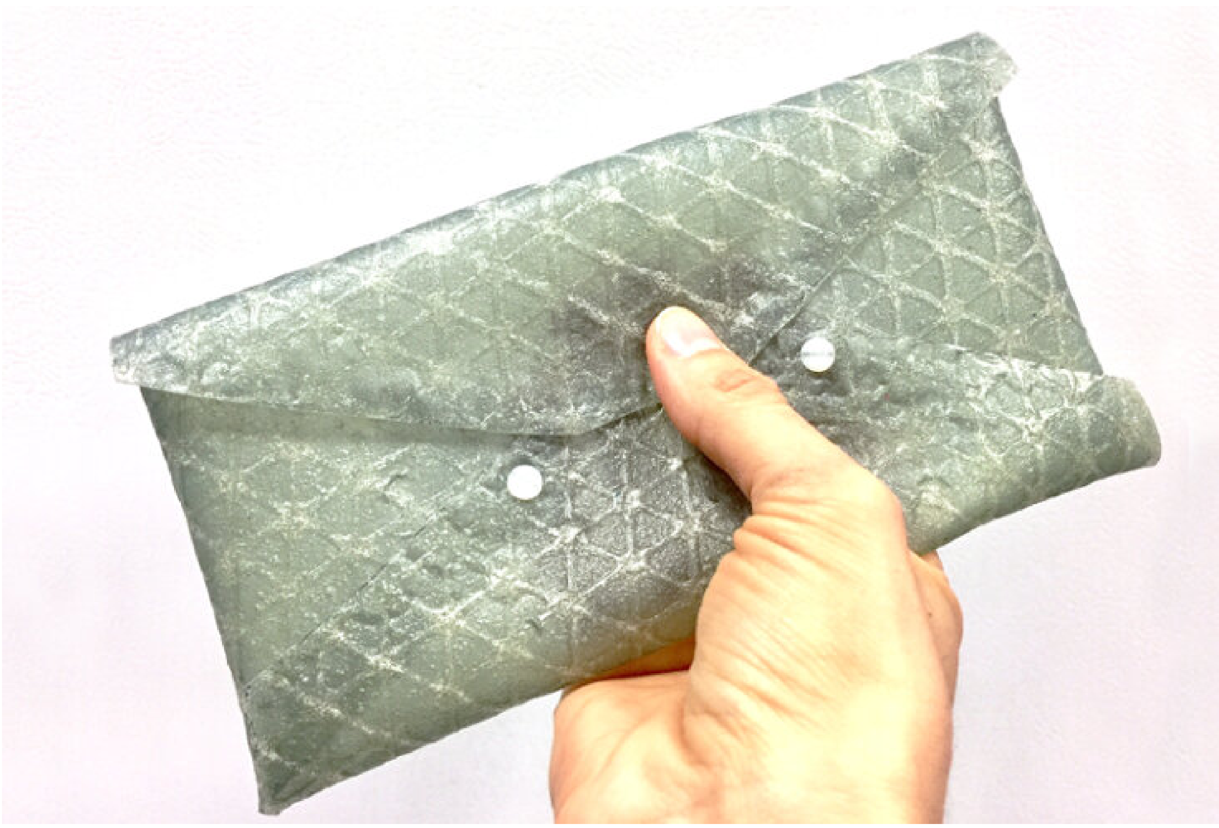
Researchers create leather-like material from silk proteins

May 5 2021



A small purse made of silk leather with attached hardware demonstrates its utility in manufacturing products. Credit: Laia Mogas Soldevilla

Leather is an ever growing multi-billion dollar industry requiring more than 3.8 billion bovine animals—equal to one for every two people on earth—to sustain production each year. And while the products—clothing, shoes, furniture and more—can be quite elegant and durable, the environmental impact of leather production has been severe, leading to deforestation, water and land overuse, environmental pollution, and greenhouse gas emissions.

Researchers at Tufts University School of Engineering set out to find an alternative to leather, with similar texture, flexibility and stiffness, yet focused on materials that are sustainable, non-toxic, and friendly to the environment. It turns out, we have been wearing that material all along—it's silk, but instead of weaving the silk into fabric, the Tufts engineers were able to break down the fibers from silkworm cocoons into their protein components, and re-purpose the proteins to form the leather-like material. The process for making silk-based leather is described in a study published in the journal *Materials & Design*.

The silk-based leather can be printed into different patterns and textures, has similar physical properties to real leather, and can withstand the folding, piercing, and stretching typically used to create leather goods, including the ability to stitch together pieces of material and attach hardware such as rivets, grommets, handles and clasps.

"Our work is centered on the use of naturally-derived materials that minimize the use of toxic chemicals while maintaining material performance so as to provide alternatives for products that are commonly and widely used today," said Fiorenzo Omenetto, Frank C. Doble Professor of Engineering at Tufts School of Engineering, director of the Tufts Silklab where the material was created, and corresponding author of the study. "By using silk, as well as cellulose from textile and agricultural waste and chitosan from shell-fish waste, and all the relatively gentle chemistries used to combine them, we are making progress towards this goal."

There is of course already an existing portfolio of alternative leathers developed by industry and the research community, with a focus on using agricultural byproducts or regenerated materials that have a reduced impact on the environment and animal raising. These include leather-like materials made from petroleum (polyurethane leather or 'pleather'), tree bark, pineapple husks, plant oils, rubber, fungi, and even from cellulose and collagen produced by bacterial cultures.

The silk-based leather made at Tufts offers some unique advantages to all of these approaches. In addition to being derived from dissolving silk fibers, manufacturing is water based, using only mild chemicals, conducted at room temperature, and producing mostly non-toxic waste. The silk leather material can be fabricated using computerized 3D layering with the ability to create regular micropatterns that can tune the material's strength and flexibility, print macropatterns for aesthetics (e.g. a basket weave) as well as non-regular geometrical patterning to mimic the surface texture of real leather. The resulting materials, like leather, are strong, soft, pliable, and durable, and like natural leather, they are biodegradable once they enter the waste stream.

In fact, the silk-leather products could be re-dissolved and regenerated into its gel-like stock matter to be re-printed into new products

The process of making the silk leather starts with silk fibers that are commonly used in the textile industry. These fibers are made up of silk fibroin protein polymers, and they can be broken down to its individual protein components in a water-based slurry. A base layer of chitosan containing a non-toxic plasticizer glycerol and dye is printed by extrusion through a tiny bore nozzle onto a surface to provide flexibility and strength to the material. Chitosan is itself derived from natural sources such as the shells of crabs lobsters and shrimp. A layer of silk fibroin combined with plasticizer and a thickener (from vegetable gum) is printed on top of the base layer.

Extruding the fibroin slurry through the printer nozzle creates shear forces that may contribute to arranging the proteins in a way that that strengthens the material, making it ductile rather than brittle, and mimics the natural extrusion that occurs in the silk gland of a worm or spider. Changing the printed pattern of the silk layer can provide a range of appearance, tunable strengths and other physical qualities.

The printing method, also referred to as "additive manufacturing" is known to be very conservative in the use of materials and waste produced compared to other methods like injection molding or subtractive manufacturing (like carving or shaving from a block).

The Silklab at Tufts has developed a wide range of other products from silk, from implantable medical devices to architectural materials that can sense and respond to the environment by changing color. In fact, much of the technology that has been developed in the lab to derivatize the silk proteins can be applied to the silk-based leather, including attaching and embedding molecules that can sense and respond to the surrounding environment.

"That's the advantage of using silk protein over other methods—it has a well-established, versatile chemistry which we can use to tune the qualities of the material and embed smart elements like sensing molecules," said Laia Mogas-Soldevila, former research fellow in the Silklab, currently assistant professor of Architecture at University of Pennsylvania Stuart Weitzman School of Design and first author of the study. "So while there may be many options for leather-like materials, silk-based leather has the potential to be most amenable to innovative designs."

More information: L. Mogas-Soldevila et al, Additively manufactured leather-like silk protein materials, *Materials & Design* (2021). DOI: 10.1016/j.matdes.2021.109631

Provided by Tufts University