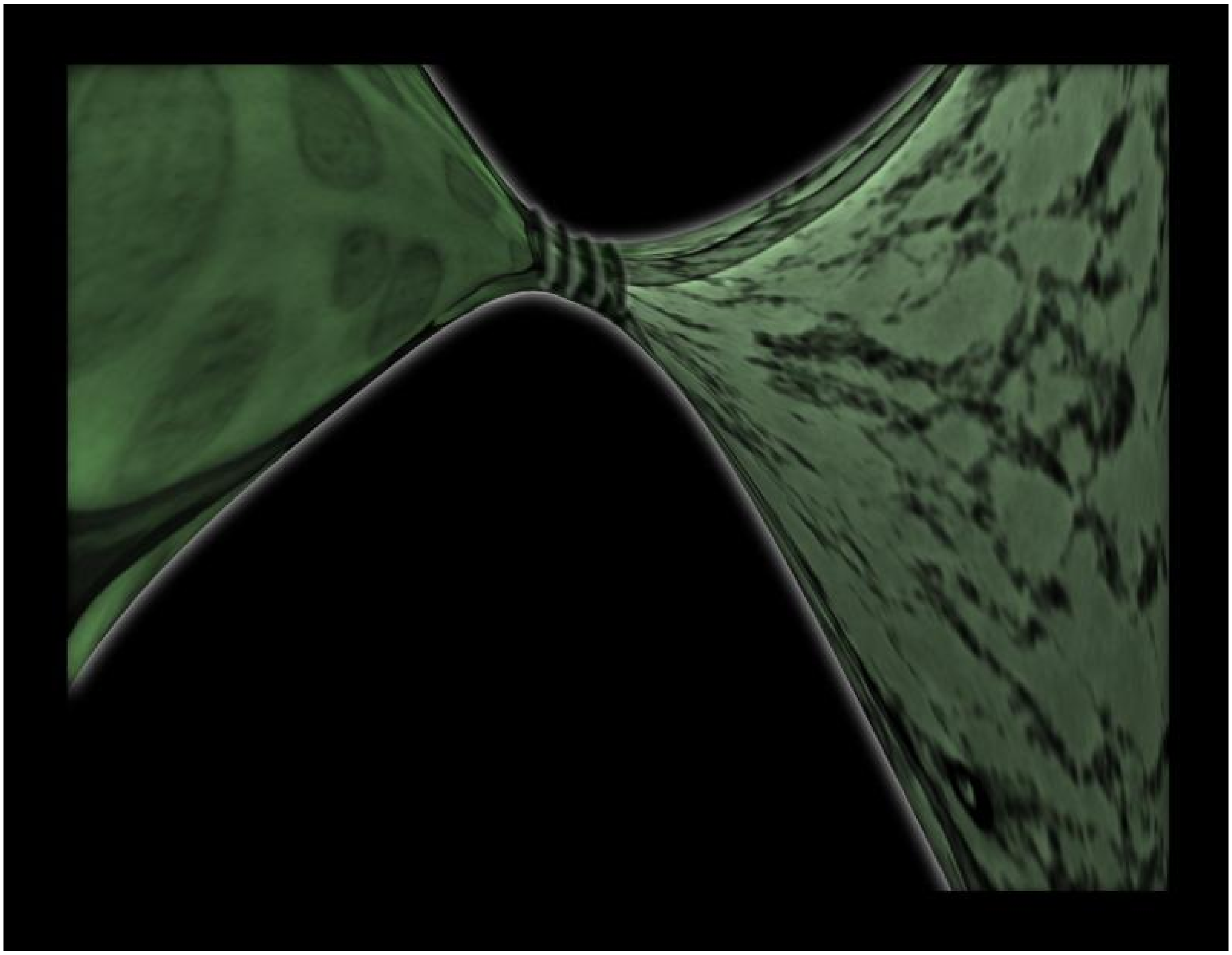
Researchers invent tougher plastic with 50 percent renewable content

March 22 2016, by Dawn Levy

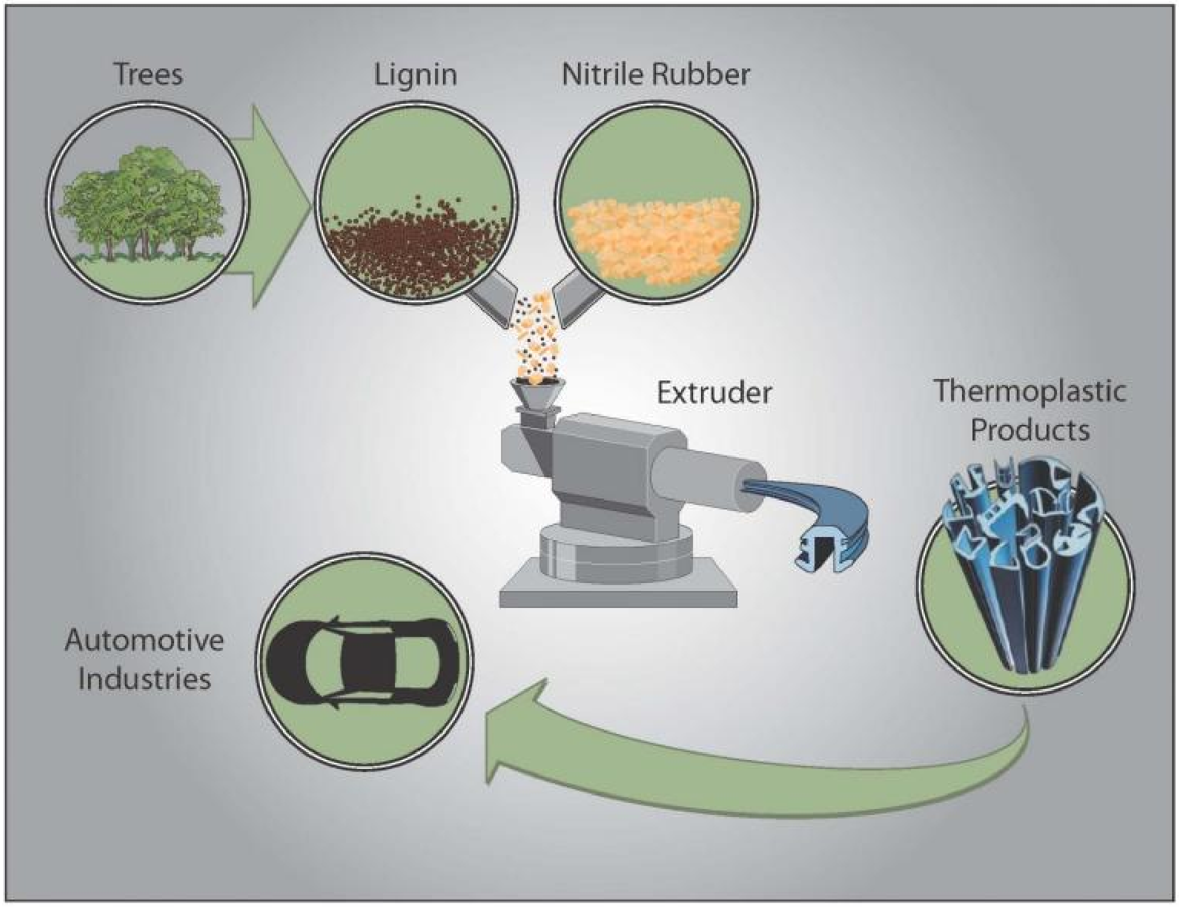


ORNL's tough new plastic is made with 50 percent renewable content from biomass. Credit: Oak Ridge National Laboratory, U.S. Dept. of Energy; conceptual art by Mark Robbins (hi-res image)

Your car's bumper is probably made of a moldable thermoplastic polymer called ABS, shorthand for its acrylonitrile, butadiene and styrene components. Light, strong and tough, it is also the stuff of ventilation pipes, protective headgear, kitchen appliances, Lego bricks and many other consumer products. Useful as it is, one of its drawbacks is that it is made using chemicals derived from petroleum.

Now, researchers at the Department of Energy's Oak Ridge National Laboratory have made a better thermoplastic by replacing styrene with lignin, a brittle, rigid polymer that, with cellulose, forms the woody cell walls of plants. In doing so, they have invented a solvent-free production process that interconnects equal parts of nanoscale lignin dispersed in a synthetic rubber matrix to produce a meltable, moldable, ductile material that's at least ten times tougher than ABS. The resulting thermoplastic—called ABL for acrylonitrile, butadiene, lignin—is recyclable, as it can be melted three times and still perform well. The results, published in the journal *Advanced Functional Materials*, may bring cleaner, cheaper raw materials to diverse manufacturers.

"The new ORNL thermoplastic has better performance than commodity plastics like ABS," said senior author Amit Naskar in ORNL's Materials Science and Technology Division, who along with co-inventor Chau Tran has filed a patent application for the process to make the new material. "We can call it a green product because 50 percent of its content is renewable, and technology to enable its commercial exploitation would reduce the need for petrochemicals."



Equal parts lignin and synthetic nitrile rubber are heated, mixed and extruded to yield a superior thermoplastic for cars and other consumer products. Credit: Oak Ridge National Laboratory, U.S. Dept. of Energy; schematic by Mark Robbins

The technology could make use of the lignin-rich biomass byproduct stream from biorefineries and pulp and paper mills. With the prices of natural gas and oil dropping, renewable fuels can't compete with fossil fuels, so biorefineries are exploring options for developing other economically viable products. Among cellulose, hemicellulose and lignin, the major structural constituents of plants, lignin is the most commercially underutilized. The ORNL study aimed to use it to produce, with an eye toward commercialization, a renewable thermoplastic with properties rivaling those of current petroleum-derived alternatives.

To produce an energy-efficient method of synthesizing and extruding high-performance thermoplastic elastomers based on lignin, the ORNL team needed to answer several questions: Can variations in lignin feedstocks be overcome to make a product with superior performance? Can lignin integrate into soft polymer matrices? Can the chemistry and physics of lignin-derived polymers be understood to enable better control of their properties? Can the process to produce lignin-derived polymers be engineered?

"Lignin is a very brittle natural polymer, so it needs to be toughened," explained Naskar, leader of ORNL's Carbon and Composites group. A major goal of the group is producing industrial polymers that are strong and tough enough to be deformed without fracturing. "We need to chemically combine soft matter with lignin. That soft matrix would be ductile so that it can be malleable or stretchable. Very rigid lignin segments would offer resistance to deformation and thus provide stiffness."

All lignins are not equal in terms of heat stability. To determine what type would make the best thermoplastic feedstock, the scientists evaluated lignin from wheat straw, softwoods like pine and hardwoods like oak. They found hardwood lignin is the most thermally stable, and some types of softwood lignins are also melt-stable.

Next, the researchers needed to couple the lignin with soft matter. Chemists typically accomplish this by synthesizing polymers in the presence of solvents. Because lignin and a synthetic rubber containing acrylonitrile and butadiene, called nitrile rubber, both have chemical groups in which electrons are unequally distributed and therefore likely to interact, Naskar and Chau Tran (who performed melt-mixing and characterization experiments) instead tried to couple the two in a melted phase without solvents.

In a heated chamber with two rotors, the researchers "kneaded" a molten mix of equal parts powdered lignin and nitrile rubber. During mixing, lignin agglomerates broke into interpenetrating layers or sheets of 10 to 200 nanometers that dispersed well in and interacted with the rubber. Without the proper selection of a soft matrix and mixing conditions, lignin agglomerates are at least 10 times larger than those obtained with the ORNL process. The product that formed had properties of neither lignin nor rubber, but something in between, with a combination of lignin's stiffness and nitrile rubber's elasticity.

By altering the acrylonitrile amounts in the soft matrix, the researchers hoped to improve the material's mechanical properties further. They tried 33, 41 and 51 percent acrylonitrile and found 41 percent gave an optimal balance between toughness and stiffness.

Next, the researchers wanted to find out if controlling the processing conditions could improve the performance of their polymer alloy. For example, 33 percent acrylonitrile content produced a material that was stretchy but not strong, behaving more like rubber than plastic. At higher proportions of acrylonitrile, the researchers saw the materials strengthen because of the efficient interaction between the components. They also wanted to know at what temperature the components should be mixed to optimize the material properties. They found heating components between 140 and 160 degrees Celsius formed the desired hybrid phase.

Using resources at ORNL including the Center for Nanophase Materials Sciences, a DOE Office of Science User Facility, the scientists analyzed the morphologies of the blends. Scanning electron microscopy, performed by Chau Tran, explored the surfaces of the materials. Jihua

Chen and Tran characterized soft matter phases using transmission electron microscopy, placing a thin slice of material in the path of an electron beam to reveal structure through contrast differences in the lignin and rubber phases. Small-angle x-ray scattering by Jong Keum revealed repeated clusters of certain domain or layer sizes. Fourier transform infrared spectroscopy identified chemical functional groups and their interactions.

Future studies will explore different feedstocks, particularly those from biorefineries, and correlations among processing conditions, material structure and performance. Investigations are also planned to study the performance of ORNL's new thermoplastic in carbon-fiber-reinforced composites.

"More renewable materials will probably be used in the future," Naskar said. "I'm glad that we could continue work in renewable materials, not only for automotive applications but even for commodity usage."

The title of the paper is "A New Class of Renewable Thermoplastics with Extraordinary Performance from Nanostructured Lignin-Elastomers."

Provided by Oak Ridge National Laboratory

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