Closed-loop additive manufacturing fueled by upcycled plastic

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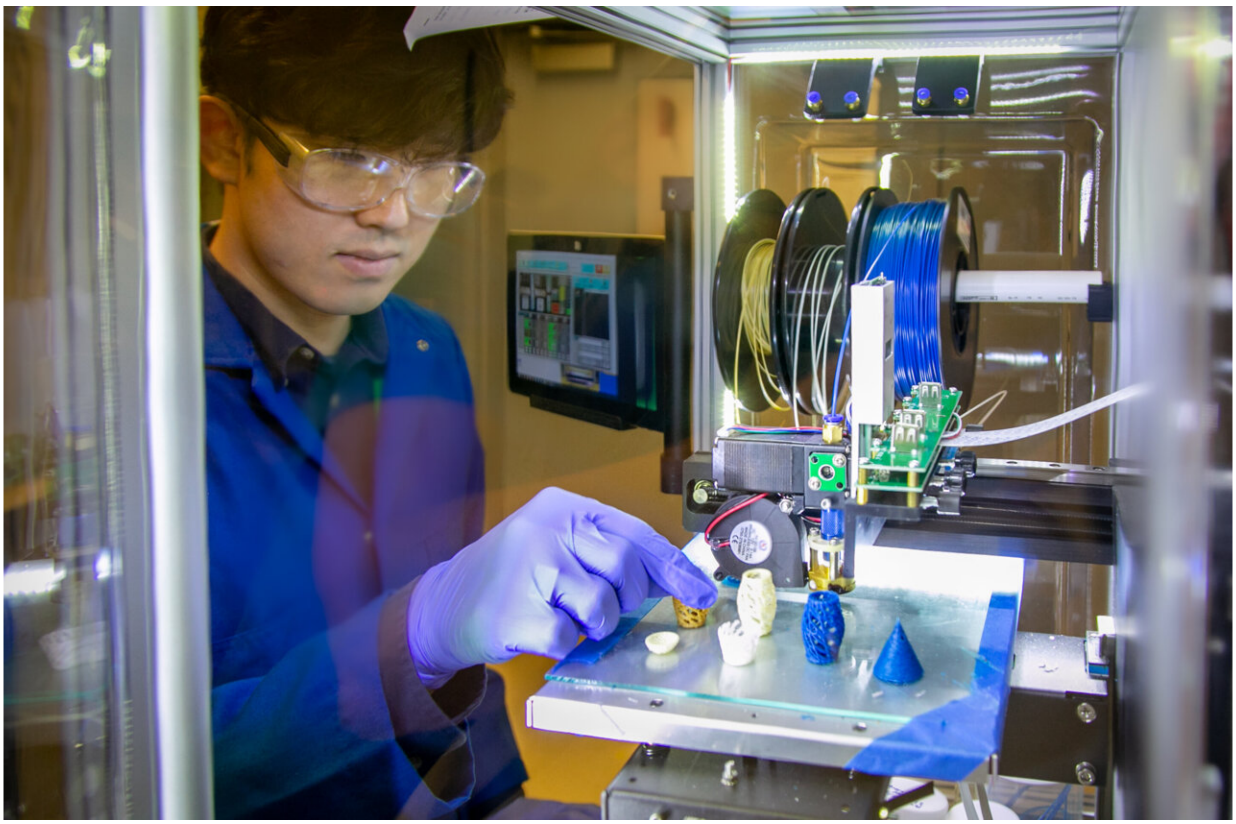

An ORNL strategy for upcycled plastic waste offers printable, high-performance materials to advance additive manufacturing. Credit: Genevieve Martin/ORNL, U.S. Dept. of Energy

Researchers at the Department of Energy's Oak Ridge National Laboratory have developed an upcycling approach that adds value to discarded plastics for reuse in additive manufacturing, or 3D printing. The readily adoptable, scalable method introduces a closed-loop strategy that could globally reduce plastic waste and cut carbon emissions tied to plastic production.

Results published in *Science Advances* detail the simple process for upcycling a commodity plastic into a more robust material compatible with industry 3D-printing methods.

The team upgraded acrylonitrile butadiene styrene, or ABS, a popular thermoplastic found in everyday objects ranging from auto parts to tennis balls to LEGO blocks. ABS is a popular feedstock for fused filament fabrication, or FFF, one of the most widely used 3D-printing methods. The upcycled version boasts enhanced strength, toughness and chemical resistance, making it attractive for FFF to meet new and higher performance applications not achievable with standard ABS.

Polymer upcycling plays an important role in addressing the growing challenge of global plastic waste accumulation. Approximately 400 million tons of plastic waste is generated each year, largely as single-use items that end up in landfills or the environment. Globally, less than 10% of plastic waste is recycled.

"We will need fundamental discoveries to overcome the challenges of increased costs and deteriorating material properties associated with recycling," said lead author Tomonori Saito of ORNL's Chemical Sciences Division. "Our goal was to develop an easily adoptable strategy that reuses plastic waste to create a more valuable material instead of generating fresh plastic."

The team targeted additive manufacturing, which is more resource efficient than conventional manufacturing and can achieve useful and complex 3D structures that would be difficult to produce by molding or casting. FFF makes up the largest share of this industry at nearly 70% of the global market.

"Developing new, recyclable materials with superior properties for FFF creates opportunities to make a big impact on plastic production and expand additive manufacturing capabilities that have the potential to reduce our carbon footprint," said ORNL's Sungjin Kim.

FFF printing requires materials that can be extruded, or pushed, through a heated nozzle to form the threads of 3D structures, built layer by layer, like coiling a rope. As a thermoplastic material that responds to heat, ABS works well for the process because it can flow easily and harden quickly into strong, rigid structures. However, there are inherent weaknesses in the way the threads stack up and bind together. Developing new feedstocks with superior properties could advance high performance applications for FFF, but these have been difficult to design.

The team applied "click" chemistry to convert the chemical makeup of ABS into a vitrimer, a type of polymer that combines the processability and recyclability of thermoplastics with the superior mechanochemical properties of thermosets, such as epoxy, which are typically not compatible with FFF. The synthesis uses widely available medical compounds that are mixed in a single step under mild conditions, followed by curing with heat.

Results show upcycled ABS achieved approximately double the toughness and strength of standard ABS, with enhanced solvent resistance.

Researchers demonstrated the excellent compressive strength of the material with intricate 3D-printed geometric structures modeled on beetle wings.

"Solvent resistance has added value because it allows us to easily separate the modified ABS from mixed, unsorted plastic waste commonly encountered in recycling scenarios," said Saito.

The team dissolved mixed plastic waste in various solvents, and in each experiment upcycled ABS maintained its structure, while all other plastics including ABS completely dissolved.

"The approach is extremely versatile," said Kim. "Recovered upcycled ABS can be reused again and again for FFF with minimal loss of properties. It can also be combined with mixed and standard ABS, and directly printed as a blend."

The multi-pathway approach enables both recycling and upcycling of mixed ABS waste containing any combination of standard, upcycled, or blended ABS. All are compatible with FFF and do not need to be separated prior to reprinting, but separation can easily be performed to offer a selection of useful materials for broad manufacturing applications.

"This effort demonstrates a closed-loop for manufacturing plastic items, potentially with higher value and performance, using only existing plastic waste in one of the most accessible areas of additive manufacturing," said Saito.

More information: Sungjin Kim et al, Closed-loop additive manufacturing of upcycled commodity plastic through dynamic cross-linking, *Science Advances* (2022). DOI: 10.1126/sciadv.abn6006

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