Editorial

Justin R. Barone PhDp

Professor, Biological Systems Engineering, Virginia Tech, Blacksburg, VA,
 USA (jbarone@vt.edu)

Materials are often thought of as passive participants in their application, simply supporting load and making sure that the object does not break. But in nature, materials do more than just bear load. Cucumber tendrils wrap around objects and grow towards light. Flower petals open and close to reproduce. A jellyfish undulates, relying on elastic recoil to swim efficiently through the ocean. There are many, many more examples throughout biology. All of these phenomena require materials that respond to a stimulus. Green materials are often composed of and/or inspired by natural materials. Creating new stimuli-responsive green materials made from and inspired by natural materials is the focus of the majority of this issue.

In the first contribution on the theme, Seguíne *et al.*¹ describe the four-dimensional (4D) printing of a composite composed of cellulose nanocrystals (CNC) and thermoplastic polyurethane (TPU). 4D printing arises when three-dimensional (3D) printed objects are stimulated to change into a different shape. Flat CNC/TPU composites are softened with water and fixed into a shape that persists when dried. Upon re-stimulating with water, the original flat shape is recovered.

For the second contribution, Hanzly *et al.*² create gelatin bilayers where each layer is of a different cross-link density. The bilayers can exhibit shape change when swollen in water with the amount of shape change dependent on the cross-link density difference between the layers. To demonstrate shape change controlled by a biochemical reaction, starch is added to the lower cross-linked layer to swell it more than with water alone. This increases the bilayer curvature. The original bilayer shape can be recovered when the starch is hydrolyzed

to small sugars with amylase enzyme and the small sugars and additional water diffuse out of the lower cross-linked layer.

Finally, Frost *et al.*³ formulate iron nanoparticle (NP)–polymer composites with the potential to be made into shape memory polymers (SMPs). Unlike the first two contributions to the issue, these materials are stimulated indirectly through heat generation by the NPs when exposed to a magnetic field. The internal heat generation allows the polymer to cross a thermal transition and realize the shape memory effect. Indirect stimulation would allow for the composites to be stimulated once implanted into the body. CNC–sodium alginate (SA) hydrogels are also made that can serve as a proxy for biological tissue. It is shown that heat transfer from the composites to the hydrogel is negligible, indicating that no damage to biological tissue would be expected.

These themed papers demonstrate the unique opportunities available in stimuli-responsive materials, particularly when they are biologically inspired and sustainable.

REFERENCES

1. Seguíne TW, Fallon JJ, Das A *et al.* (2021) Enabling mechanically adaptive 4D printing with cellulose nanocrystals. *Green Materials* 9(4): 146–156, <https://doi.org/10.1680/jgrma.20.00066>.
2. Hanzly LE, Kristofferson KA, Chauhan N and Barone JR (2021) Biologically controlled gelatin actuators. *Green Materials* 9(4): 157–166, <https://doi.org/10.1680/jgrma.20.00069>.
3. Frost BA, Carlton H, Martinez R *et al.* (2021) Controlled shape memory effects of magnetic polymer nanocomposites by induction heating. *Green Materials* 9(4): 167–181, <https://doi.org/10.1680/jgrma.20.00079>.