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ABSTRACT: An off-the-shelf, moisture-responsive, acetate-backed adhesive tape is investigated as a commercially available smart material for fabricating low-cost, multifunctional, humidity-responsive millimeter-scale structures. Laser ablation is used for cutting and thinning-down the tape to enhance its response. Water-submerged cantilevers show a radius of curvature of 3 mm or lower (for laser-thinned cantilevers). Additionally, their humidity response is a function of the angle between the longitudinal axis of the cantilever and polymer orientation. A cut angled at 80° with respect to this orientation results in a tip rotation of up to 25°, enabling the formation of bending cantilevers with twisting behavior. The tape cantilevers are further functionalized with magnetic nanoparticles and used to create four-finger grippers that close underwater within minutes and can sample 100 μL of liquid. A cyclic humidity monitor is also fabricated using a tape strip that walks unidirectionally on a ratchet-shaped surface upon exposure to humidity variations. © 2013 Wiley Periodicals, Inc. J. Polym. Sci., Part B: Polym. Phys. **2013**, *51*, 1263–1267

KEYWORDS: cellulose acetate; hygroscopic; laser ablation; stimuli-sensitive polymers; swelling

INTRODUCTION Commercially available products offer a stable platform upon which new technologies can be developed. Such common items are often taken for granted or dismissed in favor of more complex emerging materials. Mass-produced items offer many economic, engineering, and environmental advantages including a refined fabrication process that is low in cost, guarantees a high degree of consistency, and has a minimized carbon footprint. Although these materials are often optimized for a single purpose, they sometimes exhibit unintended extraordinary physical or chemical properties. The close inspection of such coincidental properties from a material and microfabrication point of view can reveal an enhanced toolset of new raw materials that can be used to create more complex technologies while maintaining a low cost. Recent research has endorsed this view. A good example is the use of paper as a versatile platform for fabrication of actuators,[1] sensors,[2] and microfluidic systems.[3–5] Others include repurposed ball-point pens,[6] bakers' yeast,[7] and commercial graphite paper[8] to create novel technologies that can be readily adopted in laboratories around the world.

Another such low-cost multifunctional material is cellulose acetate (CA)-backed adhesive tape, whose ubiquitous availability often results in it being taken for granted. Aside from excelling at its primary intended purpose (i.e., to adhere surfaces together while appearing translucent), the tape possesses various other appreciable qualities such as low cost, flexibility, and hygroscopicity.[9–11] Like many novel environmentally responsive polymeric materials,[12–18] a cellulose acetate-backed tape offers the possibility to fabricate soft and flexible structures exhibiting large strains, high fracture strength, and inherent vibration damping, but its hygroscopic behavior renders it especially useful as a moisture controlled transducer.

In this article, we present a method of fabricating cellulose acetate-based, water-responsive actuators. Previous efforts in the development of humidity sensors[10,19,20] or moisture-triggered actuators[21,22] have been geared toward the synthesis of engineered hygroscopic polymers with enhanced sensitivity such as polyethylenimine/poly(acrylic acid) films and polyaniline membranes.[23,24] Despite their large humidity response, such materials can be difficult to handle and/or require complicated fabrication processes.[23,25] Our approach, in contrast, is based on a commercially available adhesive tape, 3M's MagicTape™ (MT), which offers an inexpensive bilayer platform comprising a matte-finish hygroscopic acetate layer[26] lined with an adhesive on one side. As the adhesive absorbs water at a much lower rate than CA, the differential swelling of the layers results in a bending moment that deforms the structure (Supporting Information Fig. S1). This effect can be exploited to fabricate millimeter-scale

Additional Supporting Information may be found in the online version of this article.

actuators. Various microstructures can be carved out of the cellulose acetate/tape bilayer by using laser machining. This fabrication method offers the capabilities of rapid prototyping and batch processing without the need for complex cleanroom processes.[27] Additionally, the precise control of the laser beam position and power makes it possible to selectively ablate regions of the tape to smaller thicknesses. The ablation does not change the surface energy of the tape, but the resulting lower thickness allows for increased curvature of the tape (uniform ablation) as well as hinge-like bending (selective ablation). The fabricated devices can then be further functionalized by the incorporation of other material (e.g., ferrofluid) onto either layer to enhance their capabilities.

Two illustrative applications of hygroscopic bilayers are presented. The first is a four-fingered, magnetically functionalized, claw-like structure that is fabricated from tape cantilevers. Upon contact with water, it closes to trap a specific volume of fluid for potential bio/chemical analysis and is collected with a small permanent magnet. An alternate application depicts a cantilever walking along a ratcheted surface in response to changes in relative humidity; such a structure is suitable for measuring the frequency of humidity variations in environments having cyclic humidity levels.

The MT structures exhibit a strong response to humidity. Figure 1 shows the deflection versus relative humidity for MT cantilevers of various ablated thicknesses. For working humidity levels of 55–80%, the devices display an almost linear response for any given tape thickness. The deflection is greater for thinner tapes than thicker ones, as expected by cantilever beam deflection theory. When submerged in water, the cantilevers curl toward the adhesive side with a radius of curvature of 3 mm (for original thickness of 30 μm) or lower (for thinner ablated ones).

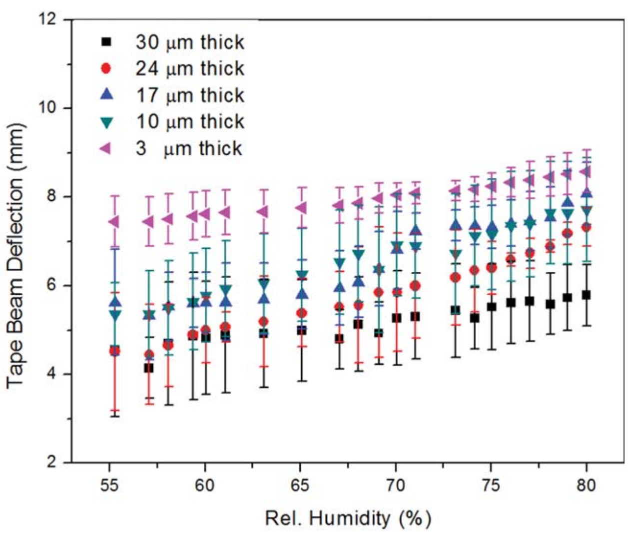

FIGURE 1 Cantilever beam deflection as a function of relative humidity for MagicTape^{TM} devices of various thicknesses. Variations in thickness were achieved by laser ablation.

SEM and goniometry measurements are used to evaluate the effect of laser-ablation on the surface of the tape. The SEM images (Supporting Information Fig. S2, with inset photographs of goniometric measurements) of the original and ablated tape show a more porous surface morphology for ablated samples; however, contact angle measurements reveal similar wetting properties for both surfaces. The original tape and the ablated one both show a water contact angle close to 90°. The unchanging contact angle suggests that in the process of laser-ablation, the tape does not alter its surface energy and can be safely utilized for creating thinner films.

Further control over bending/actuation direction of the MT structures is possible by varying the angle at which the sample is cut from the original tape. Our experiments reveal that cantilevers cut along the width of the tape always curve about their transverse axis, suggesting an existing grain or polymer chain orientation perpendicular to such axis (i.e., parallel to the tape length). Such orientation can be attributed to applied stresses during the manufacturing process of the tape. As is the case with hygroscopic fibrous materials such as paper, it is more energetically favorable for the parallel fibers to expand away from each other than to elongate along their chain axis; this energetic asymmetry results in anisotropic bending.[28,29] A grain orientation in the same direction is visible with optical photography, Figure 2(a), but the matte finish of the tape allows only faint visibility. Closer inspection of the CA surface, Figure 2(b,c), reveals a roughness with feature size similar to that of the grain, enough to behave as noise, masking the visibility of the grains. This is confirmed in Figure 2(d), which shows a more clearly visible grain pattern on the CA substrate when viewed from the bottom (adhesive) side of the tape. For this image, the adhesive layer is removed, but some adhesive residues on the CA are still visible in the photograph. The resulting constant direction of curvature of the tape allows us to impart rotational displacement capabilities to cantilevers by simply machining them from the tape at an angle (θ) to the tape width (Supporting Information Fig. S3). The degree of rotation in a 100% humidity environment increases from 0 to 25° as θ (the angle between the longitudinal axis of a cantilever and the width of the original tape) is varied from 0 to 80°. Such rotational control can be exploited for more complex three-dimensional (3D) actuation of tape-based devices.

Tape ablation via laser machining also provides the ability to selectively thin down regions of a cantilever and create hinges. This effect is demonstrated in Figure 3(a–d) with cantilevers featuring zero, one, two, and three ablated regions. The ablated regions have a higher curvature due to their smaller thickness. Furthermore, the ablated regions of various geometries can be used to impart additional out-of-plane or torsional deflections. As an example, Figure 3(e) shows the altered tape deflection caused by ablating a parallelogram shape instead of a rectangle as in (a–d).

As an illustrative application, MagicTape^{TM} was used to fabricate 3D claw structures via laser machining, as

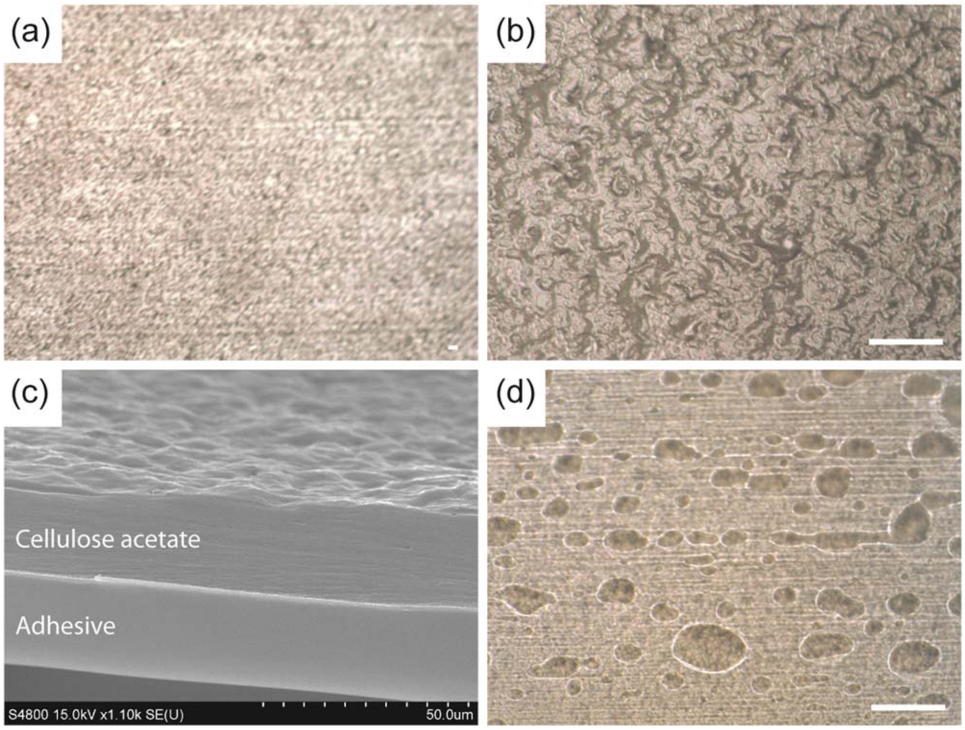

FIGURE 2 (a) An optical image of the cellulose acetate backing of 3M MagicTape™ reveals a native grain orientation, faintly visible as horizontal lines. The grain is parallel to the length of the tape. (b) Closer inspection reveals a rough matte surface that masks the visibility of the grain orientation. (c) An SEM image of the cross section of the tape shows its bilayer composition and confirms a rough cellulose acetate surface. (d) An optical image from the bottom (adhesive side) of the tape reveals a more pronounced grain orientation. The adhesive is removed, but some residues remain. Scale bar: 50 μm.

described in the Experimental section. The claw device consists of two rectangular MT strips (2×19 mm^2) attached to each other to form a cross whose four arms fold inward upon submersion in water and return to normal when dried. The structure's reversible gripping actuation can be used to entrap suspended particles or to collect a sample volume of close to 100 μL of an aqueous solution. To impart magnetic functionality, the laser-machined structures are loaded with ~2.4 mg of oil-based ferrofluid (on their non-CA side) by stamping technique. The ferrofluid-functionalized water-

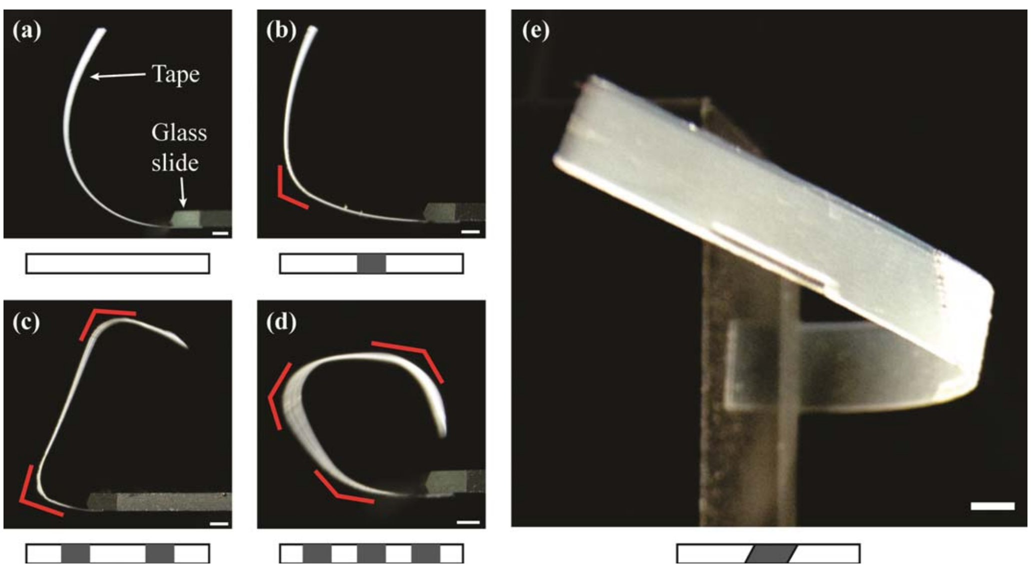

FIGURE 3 Selective ablation of tape regions imparts hinge-like behavior to a tape cantilever. The number of hinges corresponds to the number of ablated regions: (a) no ablation; (b–d) one, two, and three ablated regions, schematically illustrated below each photograph. (e) Skewed ablation regions allow three-dimensional motion in response to humidity. Scale bar: 1 mm.

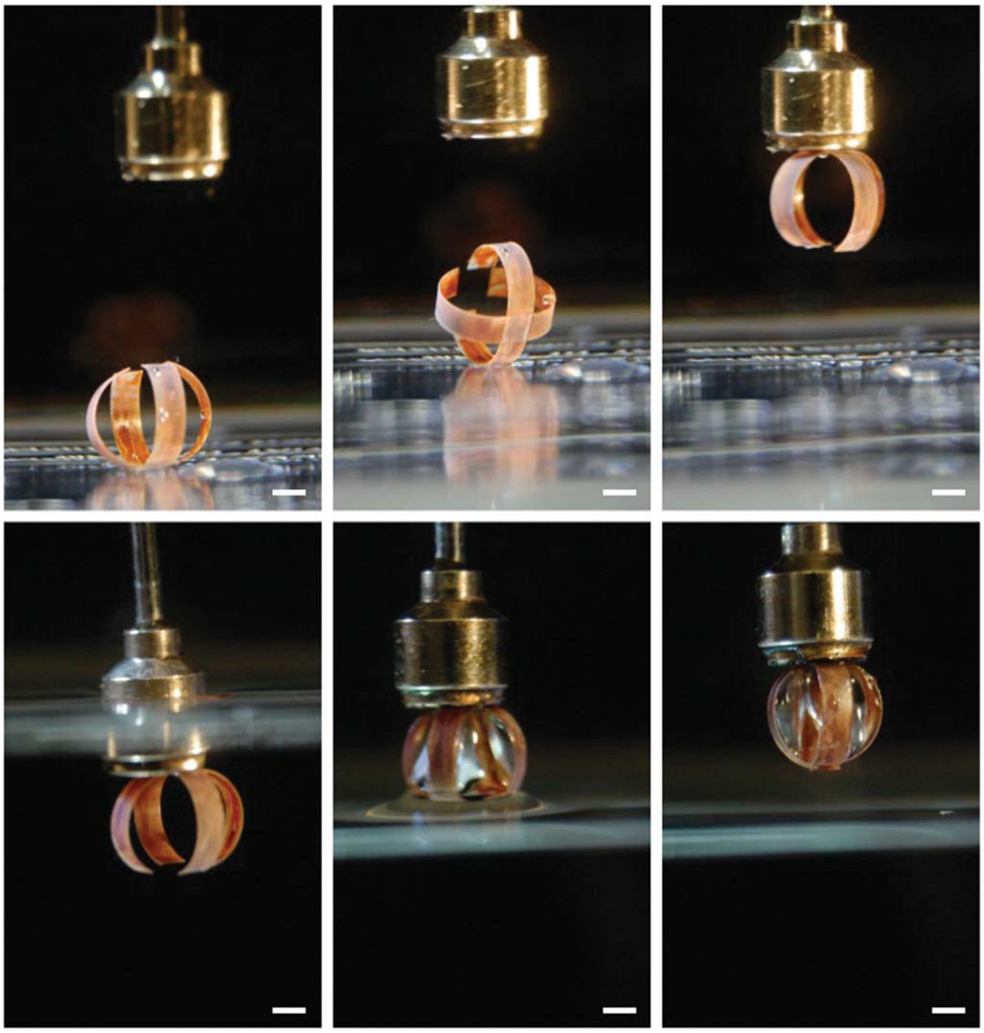

FIGURE 4 A ferrofluid-coated gripper (MT structure) is shown closing underwater and being collected with a small magnet to sample the aqueous solution. (Top three shots are underwater; bottom three show the air/water interface). Scale bar: 2 mm.

triggered-actuators can be suspended and distributed throughout a water reservoir and subsequently collected using a small permanent magnet and analyzed for chemical or biological contaminations. Figure 4 shows a demonstration of this concept with a ferrofluid-coated MT gripper closing underwater (top panel) and being retrieved with a small magnet (bottom panel showing the sample at air/water interface).

In an alternate application (Supporting Information Fig. S4), a tape cantilever is shown to walk along a ratcheted surface in response to a cyclic relative humidity. The surface is laser-machined out of acrylic. A tape cantilever, with its ends lightly coated in cellulose powder, is placed on the ratcheted surface, and the local relative humidity is alternated between 22 and 100%. The asymmetric forces at the tape ends cause the tape to move along the surface. Such a response can be applied for use as a humidity cycle counter or, if one end is anchored, as a sensor of maximum humidity.

In this work, we present low-cost millimeter-scale hygroscopic structures fabricated out of a commercially available adhesive tape (3M MagicTape^{TM}) (MT) that can be further functionalized with magnetic nanoparticles. The structures are conveniently machined by laser for rapid batch fabrication of complex designs. Structures consist of a hygroscopic cellulose acetate-based backing layer attached to an adhesive layer that is less conducive to water diffusion, creating a bending moment upon exposure to humidity. Cantilevers of MT are exposed to various degrees of humidity to characterize the bending. Selective laser ablation of the cantilevers allows for the formation of thin, high-curving tape regions that behave like hinges. In addition to cantilever deflection, the MT devices are also shown to exhibit a rotational displacement that is a function of the angle between the longitudinal axis of the cantilever and the transverse axis of the original tape. Ferrofluid-functionalized MT cantilevers are subsequently used to create other structures, including a magnetically controlled, four-arm claw that can be used for sampling aqueous solutions of biological or chemical components. A tape cantilever was also demonstrated to walk along a ratcheted surface in response to a cyclic humidity environment. Together, the strong humidity response of MagicTape™, its ubiquity, and its ease of processing for making multiple structures render it a valuable material for low-cost humidity-responsive microsystem fabrication.

EXPERIMENTAL

Fabrication

The 3M MagicTape™ (MT) structures were fabricated as follows (Supporting Information Fig. S5). The MT was first mounted onto a PDMS substrate for machining into various shapes using a laser. The PDMS provided a surface with a weak adhesion to the tape, enabling the tape to remain flat while machining but allowing its easy removal after fabrication. Laser machining was performed using a CO_2 laser cutting and engraving system [Universal Laser System, Professional Series, maximum power 60 watts, maximum speed 2 mm/ms, wavelength 10 μm, Continuous Wave (CW) mode] that allows a minimum feature size of 62 $\pm$ 1 μm.[5] A power level of 60% and speed of 100% were used for cutting the tape structures. For selective thinning of the tape (from its original 30 μm down to 24, 17, 10, and 3 μm), a decreased power level (5–20%) was used. Rectangular strips of MT were cut and used as building blocks for more complex structures for 3D actuation. An acrylic ratcheted surface was also laser-machined and used as an asymmetrical surface for the walking-tape humidity sensor.

Characterization

The bending response of the tape to humidity was characterized by cantilever beam deflection measurements. A 2 $\times$ 19 mm^2 MT cantilever was attached to a rigid platform and placed in a chamber with controllable humidity (55–80%) while its maximum deflection was measured. This was repeated for cantilevers of various (ablated) thicknesses. The response time of the tape was measured by exposing a cantilever to water vapor and measuring the time required for a full deflection.

The effect of laser ablation on the tape surface was evaluated by goniometric measurements and by SEM. The goniometric measurement consisted of depositing 1 μL droplets of water on the tape samples using a microliter pipette and capturing the droplet profile with a DSLR camera. The contact angle was then measured from the images using photograph-processing software. The SEM images were captured with a Hitachi S-4800 Field Emission SEM.

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