Thermoresponsive Hydrogels with Improved Actuation Function by Interconnected Microchannels

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1. Introduction

Soft robotic strategies are required in many applications, ranging from robot–human interactions to gripping soft objects. [1,2] As the interest in soft robotics grows, so does the demand for materials, which fulfill special requirements regarding their mechanical properties, the way of inducing actuations, and their compatibility with different environments. [3] A low and adjustable stiffness, fast rate of response to external stimuli, and distinctive volume or shape change are examples for desirable properties of materials used in soft robotics.

Stimuli-responsive hydrogels are important in soft actuators research, as they change volume in response to environmental factors. Thermoresponsive hydrogels, such as poly(N-isopropylacrylamide) (pNIPAM), typically have slow response rates and exert comparably weak forces, which usually limit their use as artificial muscles. Herein, it is shown that the incorporation of interconnected microchannels into the pNIPAM hydrogel by a template-assisted approach leads to a significant increase in both the response rate and the volume change. For a microchannel density of only 5 vol%, a volume reduction of 90% is achieved, compared with only 12% for the bulk material, while material stiffness of the swollen hydrogels remains unchanged. By tailoring the channel density and the stiffness of the material, it is further possible to adjust the response rate and the exerted stroke force in an actuation setting. It is shown in a demonstrator gripper setup driven by the pNIPAM-based artificial muscle that the performance of the gripper is strongly improved by the microengineered material compared with conventional bulk pNIPAM. The strategy of incorporating microchannels into the pNIPAM hydrogel provides a practical approach for the future use of volume phase transition-based responsive materials in soft robotic applications.

Stimuli-responsive hydrogels are highly attractive candidates for such materials, due to their soft constitution and their ability to respond to environmental changes. Among such hydrogels, thermoresponsive poly(N-isopropylacrylamide) (pNIPAM) is a well-studied material, which changes its volume as a function of temperature. Its lower critical solution temperature (LCST) is at 32 °C and determines the start of phase transition from hydrated extended polymer chains below the LCST into dehydrated collapsed coils above the LCST. [4–6] In this way, pNIPAM-based hydrogels reversibly shrink upon dehydration at temperatures above the LCST by expelling water and swell via rehydration below the LCST.

A variety of pNIPAM-based soft actuators that are folding or bending have been developed, as well as soft walkers, which can be stimulated by light. [7,8] The possibility of miniaturizing pNIPAM hydrogel structures and even the fabrication of multicomponent materials have been used by Nishiguchi et al. using an in-gel direct laser writing process. [9] Hippler et al. presented complex 3D architectures consisting of pNIPAM heterostructures exhibiting large-amplitude and...
complex actuation responses.\textsuperscript{[10]} However, applications based on pNIPAM structures that exert tensile or pushing forces to move external objects are rarely investigated, due to the limitations of pure pNIPAM hydrogels regarding their fragility and their marginal response to external stimuli. Many approaches of enhancing these shortcomings are associated with chemical modifications of the material, such as designing nanocomposites, copolymerization, or interpenetrating polymer networks.\textsuperscript{[11]} In turn, these changes in material chemistry are often accompanied by undesired changes in the hydrogel properties, such as elasticity, optical transparency, or a shift in LCST.\textsuperscript{[12]}

In this work, we propose a novel microengineering approach to 3D-structured pNIPAM hydrogels on the microscale by introducing a random network of interconnected microchannels into the hydrogel. By this microstructural modification, we demonstrate that the thermoresponsive shrinking of the material occurs at a considerably higher rate and that much stronger force strokes are possible compared with bulk, unstructured pNIPAM. Moreover, a temperature-driven gripper has been designed that closes its arms above the LCST and holds soft objects. Hence, our novel material provides a new strategy to generate higher forces with chemically unmodified pNIPAM, thus making this material suitable for actuator devices. From a broader perspective, this strategy can be a starting point for next-generation soft robotic devices driven by thermoresponsive artificial muscles.

2. Results and Discussion

Thermoresponsive hydrogels were microengineered by a template-assisted approach described in other studies.\textsuperscript{[13,14]} Briefly, pNIPAM-based hydrogel precursor solutions were infiltrated into highly porous, sintered 3D scaffolds consisting of micrometer-sized zinc oxide tetrapods. These scaffolds are referred to as templates and the diameter of the tetrapod leg structures is in the order of 2–3 μm (Figure 1, 2).\textsuperscript{[15]} After polymerization, zinc oxide is chemically dissolved so that a replica of the template is generated in the hydrogel, that is, a hydrogel with interconnected hollow microchannels, which is shown in Figure 1. The microchannel density in the hydrogel can be adjusted by the volume-to-mass ratio used in the molding process of the zinc oxide templates, ranging from microchannel densities of 2 to 11 vol%. However, the zinc oxide templates can in principle be fabricated with porosities between 50 and 98 vol%. Thus, even higher microchannel densities are possible.\textsuperscript{[16]}

The template-assisted approach presented in this work was chosen due to the ease of sample preparation and the flexibility to adjust the shape and density of the zinc oxide templates, which determine the geometry of the final hydrogel samples. In addition, this method was intended to result in an increase in the hydrogel surface area, which is directly related to the flow of water in and out of the hydrogel during external temperature variations, as well as to prevent the formation of a dense skin layer during dehydration. Hence this method appears promising to increase the responsivity of pNIPAM hydrogels in a controlled manner.

To analyze the performance of our microengineered thermoresponsive hydrogels in soft robotic devices, several material properties have to be investigated. In a first step, we have studied the microstructure of the hydrogels after treatment with tannic acid and osmium tetroxide by scanning electron microscopy (SEM) (Figure 2). The characteristic tetrapodal structure of the dissolved zinc oxide microparticles is clearly visible from the surface morphology of the microengineered pNIPAM shown in Figure 2a. These results demonstrate the successful replication of the zinc oxide tetrapod structure into microchannels, where even the rippled surface structure of the zinc oxide tetrapods is transferred to the hydrogel surface (Figure 2c,d).\textsuperscript{[17]} It should be mentioned that the drying process of the hydrogels during SEM sample preparation may change the microchannel sizes compared with the hydrated state, which does not allow for a quantitative analysis of microchannel sizes.\textsuperscript{[18]} Further details on shape and size of the sacrificial zinc oxide microparticles are shown in Figure S1 in the Supporting Information.

An important step for investigating the material’s thermoresponsive behavior is the characterization of thermally induced swelling and shrinkage at the LCST, as this change in volume finally determines the stroke length of the actuation. The latter is a crucial parameter for using the material in soft robotic devices.

Due to the amphiphilic nature of pNIPAM side chains [−CONH/−CH(CH₃)₂], a temperature-dependent hydrophilic/hydrophobic balance is established, which controls the amount

Figure 1. Sketches and photographs of bulk pNIPAM (left) and microengineered, channel-containing pNIPAM (right). The microengineered material undergoes a significantly larger volume change upon phase transition at the LCST compared with the bulk material and it also reacts considerably faster.
of water stored in the pores of the hydrogel matrix.\textsuperscript{[19]} The temperature-dependent dynamic shrinking and swelling characteristics of the different hydrogel matrix compositions and microchannel presence are shown in Figure 3, where we show the normalized sample weight as a function of temperature and time. Unless stated otherwise, the microengineered hydrogels contain 5 vol\% of microchannels, which resulted in reliable sample preparation with good reproducibility.

During the first 10 min in a heat bath at 42 °C, a reduction of sample weight can be observed as a result of the dehydration and shrinkage of the hydrogel matrix above the LCST (32 °C).\textsuperscript{[20]} In contrast, during the subsequent 10 min of cooling in a water bath at 22 °C, the weight of the material increases, due to the rehydration and swelling of the gels at temperatures below the LCST. The curves show several systematic differences: The weight loss of the samples increases with decreasing crosslinker concentration. After 10 min of dehydration at 42 °C, highly crosslinked pNIPAM (hard) containing microchannels goes down to 13% of the initial weight, whereas the material with the lowest amount of crosslinker (soft) reduces its weight to 6%. This can be explained by a decrease in gel hydrophilicity, resulting from a higher crosslinker content, which densifies the polymer network and reduces the overall number of free hydratable pNIPAM side chains.\textsuperscript{[21]}

Most notably, there is a significant difference in the dehydration curves of the samples with and without microchannels. Whereas for medium-crosslinked bulk pNIPAM the weight is reduced to 88%, the same material containing microchannels has a weight loss down to 10%. This corresponds to a 7.5-fold larger weight loss for the microengineered material compared with the bulk material, even though only 5% of volume was removed from the microengineered hydrogel by introducing the microchannels. As the weight change is strongly associated with a release of water to the outside of the material, this large weight loss can be attributed to the interconnected hollow microchannels, which increase the surface area of the hydrogel and provide a channel network accelerating the water release. This result is remarkable, as our strategy is solely based on a microstructural modification of the hydrogel material without a chemical modification of the pNIPAM matrix. It is well known that chemical modification strategies can increase pNIPAM weight losses, for example, when hydrophilic poly(vinyl alcohol) (PVA) is incorporated into the pNIPAM matrix.\textsuperscript{[22]}

In our experiments, the swelling rate was in general lower than the shrinking rate, and also the absolute volume change was smaller for reswelling compared with the initial shrinking. This material behavior prohibits the hydrogels to reach their initial state within the time limits of a typical experiment, but it can be achieved by rehydrating for 24 h (Figure S3 and Video 1, Supporting Information). This hysteretic behavior of pNIPAM has been reported before and is assumed to be attributed to additional hydrogen bonds formed between side groups of the pNIPAM chains in the collapsed state, causing a delay in chain dissociation upon swelling.\textsuperscript{[23]}

The low reswelling speed of the microengineered material may originate from the collapse of microchannels during dehydration, which does not mitigate the flow of water into the hydrogel anymore and compromises applications in actuators. Depa et al. introduced filler-stabilized pores into nanocomposite pNIPAM hydrogels, which prevent the pore collapse during dehydration and result in fast de- and reswelling properties.\textsuperscript{[24]}

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**Figure 2.** Scanning electron microscopy images of microengineered pNIPAM that prove the existence of microchannels in the hydrogel. a,b) The tetrapodal structure of dissolved zinc oxide particles in the surface of a sliced sample. c,d) The rippled surface morphology of the tetrapodal zinc oxide microparticles that is replicated in the microchannels that is visible.
To investigate in detail the influence of microchannels on the shrinking kinetics, experiments were conducted as a function of microchannel density (Figure 3b). We show that the shrinking rate significantly depends on channel density and increases with increasing channel density. This is again explained by a larger surface area and the support of water release by a higher number of microchannels. The minimum weight reached after dehydration at 42 °C for 10 min is, within the detection limits of our method, equal for all microchannel densities.

Whereas the shrinking rate and swelling rate are measures for how fast the hydrogel shrinks or swells, the swelling ratio describes how much water is stored in the polymer matrix relative to the dry polymer weight at a certain temperature. To determine the hydrogels’ swelling ratios as a function of temperature, the samples were equilibrated in water at different temperatures for 24 h and weighed afterwards. Swelling ratios were calculated by dividing the weight of stored water by the dry polymer weight, according to Equation (1).

\[ \text{SR} = \frac{w_{\text{wet}} - w_{\text{dry}}}{w_{\text{dry}}} \]  

(1)

The results are shown in Figure 3c,d for hydrogels with and without microchannels, respectively. The swelling ratios decrease with increasing temperature and, as expected, the largest difference is around the LCST of 32 °C, where the phase transition occurs. A systematic trend of reduced swelling ratio with increasing crosslinker concentration occurs for both hydrogels with and without microchannels. A lower crosslinker concentration increases the gel hydrophilicity and the size of polymer matrix pores and increases the amount of stored water inside the hydrogel. This results in higher swelling ratios compared with higher crosslinker concentrations. In addition, the formation of a dense skin layer upon dehydration is reported for bulk pNIPAM hydrogels, which attenuates the release of water and explains the differences in swelling ratios for microengineered and bulk pNIPAM above the LCST.

In case of the microengineered hydrogels, this effect is superimposed by a mitigation of water outflow by microchannels, which leads to lower swelling ratios above the LCST compared with the bulk material. The results shown in Figure 3c,d are comparable with the work of Depa et al., who characterized nanocomposite pNIPAM hydrogels with respect to the swelling ratio.

Interestingly, samples that contain microchannels have a slightly higher swelling ratio below the LCST than the hydrogels without microchannels. It can be assumed that the microchannels are filled with water below the LCST, which increases the
overall amount of water in the gel and in turn increases the swelling ratio compared with the bulk material.

From these experiments it can be concluded that the integration of an interconnected network of microchannels in the hydrogels leads to a significant increase in shrinking rate and overall potential for fast water release from the hydrogel polymer matrix. This strategy makes the material attractive for applications in actuation and soft robotics, as a high extent of volume change and a fast response rate are desirable and are currently a significant limitation of pNIPAM-based actuator systems. The integration of an interconnected network of microchannels in the hydrogels leads to a significant increase in shrinking rate and overall potential for fast water release from the hydrogel polymer matrix. This strategy makes the material attractive for applications in actuation and soft robotics, as a high extent of volume change and a fast response rate are desirable and are currently a significant limitation of pNIPAM-based actuator systems. To prove that our microengineered hydrogel is better suitable for such applications than bulk pNIPAM, we characterized both materials mechanically, including cyclic compression tests and stroke force experiments (Figure 4). From the force–distance curves recorded with the mechanical testing device, stress–strain curves were calculated to account for the different geometries of the tested materials (Figure 4a,b). These stress–strain curves show a slight hysteresis for all material compositions, which can be explained by the intrinsic viscoelasticity of the hydrogels. Furthermore, the initial loading curves are different from the curves recorded in the following cycles and a drift between the cycles is visible. Both effects might be related to small amounts of water being squeezed out of the gels during compression. However, all loading and unloading curves have similar slopes, allowing for the calculation of the elastic compressive modulus for each cycle from the initial slopes of the loading curves by a linear fit up to 5% strain. In general, the samples with higher concentrations of crosslinkers have higher slopes in the loading curves compared with those with a lower concentration of crosslinker.

Figure 4c shows the elastic compressive modulus as a function of compression cycle for microchannel-containing hydrogels. The individual modulus values are very similar, except for the initial compression cycle. This indicates a reproducible cyclic deformation of the material and allows for the calculation of an average elastic compressive modulus for each material composition (Figure 4d). Clearly, the elastic compressive modulus increases with increasing crosslinker concentration. This observation is attributed to a higher network density of the polymer and less water in the gel for higher crosslinker concentrations. These values of elastic compressive modulus are in agreement with Haq et al., who reviewed the mechanical properties of native pNIPAM hydrogels and list modulus values between 4 and 81 kPa.

The ability of generating a force onto external objects is a critical requirement for the application of soft materials in robotic systems. For this reason, both microengineered and bulk hydrogel samples were tested for their ability to apply forces...
against a force sensor. Materials in the shrunk state were clamped between a force sensor and a fixed glass substrate and the force associated with their swelling after addition of cold water was recorded. **Figure 5b** shows this stroke force as a function of time. All samples show an increase in force over time, but the stroke force applied by the hydrogels is highest for the lowest microchannel density. In contrast, bulk hydrogel only exhibits a comparably low stroke force, which is negligible compared with the force exerted by the microengineered hydrogels.

In general, the microchannel-containing hydrogels exert higher forces compared with the bulk material, as the volume increase upon swelling is higher for increasing microchannel densities, as shown in **Figure 3a**. In contrast, a higher density of microchannels also makes the hydrogel less resistant to pressure, due to an increase in sample porosity.\(^{[28]}\) Hence, this experiment shows that a low density of microchannels enables the hydrogel to exert stroke forces against external objects, making them much more attractive for soft robotic applications than bulk pNIPAM.

To illustrate the actuation capabilities of the microengineered hydrogels designed in this study in a simple demonstrator, a temperature-controlled gripper was developed and tested. The gripper consists of two 3D-printed polylactic acid (PLA) arms, which are connected by a joint. A cylindrical hydrogel sample is glued in between, as shown in **Figure 5c,d**, and serves as an artificial muscle. When transferring the gripper from a water bath at 22 °C into a water bath at 42 °C, a significant contraction of the microengineered hydrogel is observed, which results in a closing movement of the gripper, so that a piece of sponge can be held and even be deformed. In contrast, if the bulk hydrogel is used as an artificial muscle in the gripper, the gripper does not close and the sponge is neither held nor deformed. The latter is the result of the low volume change and the weak force generation of the bulk hydrogel. Further, it is possible to actuate the microengineered soft gripper repeatedly, as shown in **Figure 5e**. A density of 5 vol% microchannels was used as the material of choice for the presentation of the microengineered soft gripper.

**Figure 5.** Stroke force experiments and application in a soft gripper. a) Sketch of the experimental setup, where the sample was pressed onto a glass substrate with a stamp in the shrunken state at 42 °C. Afterwards, the sample was cooled down to 22 °C in the presence of water and pressed against the force sensor. b) Average curves from three identical samples per material show the temporal change in force resulting from the swelling of the hydrogels clamped below the force sensor (light-colored areas around curves show error bar ranges). In contrast to bulk pNIPAM, microengineered pNIPAM exerts significant forces, which are a function of microchannel density. c,d) Temperature-controlled grippers, which are actuated by bulk and microengineered hydrogels (see red arrows), respectively. The latter contains 5 vol% of microchannels and is made of the pNIPAM (hard) material. The microengineered hydrogel can close the gripper and deform a piece of sponge, while the bulk material cannot move the gripper arms. e) A series of cyclic actuation of the microengineered soft gripper is shown for ten cycles and demonstrates repeatability of actuation (see Supporting Information, Video 2).
gripper, as this material exhibited the best tradeoff between mechanical stability and stroke force generation. Our microengineered material can be implemented in a variety of soft robotic gripping devices. Scanning electron microscopy showed the successful implementation of microchannels into the hydrogel and we demonstrated that the thermoresponsive behavior of the materials is a function of microchannel density. Importantly, in our system, the chemistry of the hydrogel matrix remained unchanged; only the microstructure was engineered. It turns out that our material exhibits a 7.5-fold larger weight reduction compared with the bulk pNIPAM material and a significantly enhanced change in volume. In addition, the response to cyclic mechanical compression and the ability of the material to exert stress during swelling show that our microengineered hydrogels are attractive candidates for applications in soft actuators, which is finally proven in the design of a temperature-driven soft gripper. Taken together, this work provides a novel strategy for improving the actuation properties of pNIPAM-based hydrogels without the need for chemical modifications, thus providing the starting point for novel applications of these materials in artificial muscle systems and devices, where a high volume contraction of a thermoresponsive material is required.

3. Conclusion

In this work, we investigated the potential of microengineered hydrogels for achieving improved actuation in soft robotic gripping devices. Scanning electron microscopy showed the successful implementation of microchannels into the hydrogel and we demonstrated that the thermoresponsive behavior of the materials is a function of microchannel density. Importantly, in our system, the chemistry of the hydrogel matrix remained unchanged; only the microstructure was engineered. It turns out that our material exhibits a 7.5-fold larger weight reduction compared with the bulk pNIPAM material and a significantly enhanced change in volume. In addition, the response to cyclic mechanical compression and the ability of the material to exert stress during swelling show that our microengineered hydrogels are attractive candidates for applications in soft actuators, which is finally proven in the design of a temperature-driven soft gripper. Taken together, this work provides a novel strategy for improving the actuation properties of pNIPAM-based hydrogels without the need for chemical modifications, thus providing the starting point for novel applications of these materials in artificial muscle systems and devices, where a high volume contraction of a thermoresponsive material is required.

4. Experimental Section

Preparation of Polymer Precursor Solutions: N-isopropylacrylamide (NIPAM) and N,N,N',N'-tetramethylethylenediamine (TEMED) were purchased from Sigma Aldrich (Taufkirchen, Germany). N,N'-methylenebisacrylamide (BIS) and ammonium persulfate (APS) were purchased from BioRad (Hercules, California, USA). All chemicals were used without further purification.

Hydrogels were synthesized by free radical polymerization. For each sample composition, two sealable vials were prepared, one containing aqueous solutions of NIPAM (monomers), BIS (crosslinker), and APS and another one containing an aqueous solution of TEMED. The different compositions of the precursor solutions are shown in Table 1.

Both vials were purged with nitrogen for 10 min while being magnetically stirred over an ice bath at 500 rpm to ensure homogeneous mixing. Subsequently, the aqueous TEMED solution was transferred into the first vial and stirred for a few seconds before the polymer solution was immediately used for molding.

| Sample composition | m (NIPAM) [mg] | m (BIS) [mg] | m (APS) [mg] | V (TEMED) [μL] | V (H₂O) [μL] |
|--------------------|----------------|--------------|--------------|----------------|-------------|
| pNIPAM (soft)      | 600            | 5            | 10           | 23             | 4600        |
| pNIPAM (medium)    | 600            | 10           | 10           | 23             | 4600        |
| pNIPAM (hard)      | 600            | 15           | 10           | 23             | 4600        |

Preparation of Zinc Oxide Templates: Tetrapodal zinc oxide (t-ZnO) microparticles were prepared by flame transport synthesis, as described in other studies. The harvested t-ZnO powder was pressed into cylindrical templates with both a diameter and height of 6 mm. Templates with densities of 0.2, 0.3, 0.4, and 0.6 g cm⁻³ were fabricated by adjusting the mass of the used t-ZnO powder. Subsequently, the prepared templates were sintered at 1150 °C for 5 h to create a network of interconnected ZnO tetrapods.

Preparation of PNIPAM Hydrogels With and Without Microchannels: To prepare bulk hydrogel samples without microchannels, the cylindrical wells (d = 8 mm and h = 12 mm) of a PTFE mold were filled with 750 μL of the polymer precursor solution. Polymerization occurred at room temperature (22 °C) for 2 h and the cured samples were stored in distilled water at room temperature. Over a period of 5 days, the water was exchanged daily to remove any unreacted molecules.

Hydrogel samples containing interconnected microchannels were produced based on previous publications. In brief, t-ZnO templates were infiltrated with the polymer solution and subsequently cured for 6 h at room temperature. Afterwards, the templates were immersed in hydrochloric acid (0.5 M) for at least 24 h until the zinc oxide was dissolved, leaving hollow microchannels, which resembled the previous zinc oxide structure. Then, the acid was replaced by distilled water and the hydrogels were washed with regular water exchange for 5 days. This concentration of hydrochloric acid did not have any significant influence on the hydrogel properties (see Figure S5 in the Supporting Information).

The detailed sample compositions denoted as “soft,” “medium,” and “hard” depending on the concentration of crosslinker are shown in Table 1. Further, hydrogels containing microchannels were termed “channel pNIPAM” in contrast to those without channels being termed “bulk pNIPAM.” The microchannel density or hydrogel porosity \( P \) is directly related to the density of the zinc oxide templates and can be calculated by Equation (2), where \( V_{\mathrm{ZnO}} \) is the volume occupied by the zinc oxide microparticles and \( V_{\mathrm{template}} \) is the total volume of the cylindrical templates. The porosities used in this study are shown in Table 2.

\[
p = \frac{V_{\mathrm{ZnO}}}{V_{\mathrm{template}}} = \frac{P_{\mathrm{ZnO}}}{V_{\mathrm{ZnO}}}
\]

Scanning Electron Microscopy: Pieces from equilibrated pNIPAM hydrogel samples were incubated overnight in 5% tannic acid (Sigma, Darmstadt), washed with deionized water, and stained with 1% aqueous OsO₄ (Science Services, München) for 4 h. Tannic acid increased the density of the used t-ZnO powder. Subsequently, the prepared templates were sintered at 1150 °C for 5 h to create a network of interconnected ZnO tetrapods.

Swell Tests: To investigate the thermoresponsive behavior of the different pNIPAM hydrogel samples, swell tests were conducted at temperatures above and below the LCST of pNIPAM. For a period of 10 min, each sample was placed in a beaker with distilled water at 42 °C, while being regularly taken out each minute to record the sample weight on an analytical balance (Kern, d = 0.1 mg). A prewetted paper tissue was used to remove any excess water from the sample surface.

| \( \rho \) (template) [g cm⁻³] | \( P \) (vol%) | \( \rho \) (zinc oxide) [g cm⁻³] |
|-----------------------------|--------------|-----------------------------|
| 0.2                         | 3.6          | 5.61                        |
| 0.3                         | 5.3          | 5.61                        |
| 0.4                         | 7.1          | 5.61                        |
| 0.6                         | 10.7         | 5.61                        |
Subsequently, the same procedure was repeated in another beaker containing distilled water at 22 °C.

Swelling Ratios: Swelling ratios were determined at several temperatures below and above the LCST for analyzing the ability of the hydrogels to absorb water. For this purpose, each sample was placed into a sealed vial filled with distilled water. All vials were placed into an oil bath and equilibrated for 24 h at each specific temperature, before the sample weights were measured. For determining the dry polymer weight, the samples were dehydrated for 24 h in an oven at 60 °C. Swelling ratios were calculated by the following equation, where \( w_{\text{wet}} \) and \( w_{\text{dry}} \) correspond to the wet and dry sample weights, respectively.

\[
_SR = \frac{w_{\text{wet}} - w_{\text{dry}}}{w_{\text{dry}}} 
\]

**Cyclic Compression Tests:** To investigate the mechanical properties of the samples, including the elastic compressive modulus, compression tests were conducted by a Zwick/Roell universal testing machine (2.5 kN Zwicki, Ulm). The machine was equipped with a 5 N load cell and a cylindrical steel stamp with 15 mm in diameter. Each hydrogel sample was placed into a beaker filled with distilled water at 22 °C, which was mounted to a steel holder. First, the stamp was lowered onto the sample surface until an initial load of 10 mN was reached. Afterwards, force–displacement curves were recorded in ten consecutive cycles of compressing to a strain of 10% and unloading to the initial force at a speed of 5 mm s \(^{-1}\). Sample dimensions were measured with a caliper prior to testing.

**Stroke Force Tests:** The potential ability of the hydrogel material to generate a force upon swelling was analyzed using the same Zwick/Roell machine as mentioned earlier. After shrinking the samples upon dehydration in 42 °C warm water for 10 min and subsequent clamping at 10 mN between the steel stamp and glass bottom of an empty beaker, the samples were hydrated by filling the beaker with 22 °C cold water. Simultaneously a force–time curve was recorded, while the stamp was kept in its position and the hydrogel samples started swelling.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

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**Data Availability Statement**

Data available on request from the authors.

**Keywords**

hydrogels, microengineering, porous materials, responsive materials, soft actuators

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