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An elastic biopolymer, resilin possesses exceptional qualities such as high stretchability and resilience. Such attributes are utilized in nature by many species for mechanical energy storage to facilitate movement. The properties of resilin are attributed to the balanced combination of hydrophilic and hydrophobic segments. To mimic the properties of resilin, we developed a hydrogel composed of hydrophilic acrylic acid (AAc) and methacrylamide (MAM) chains and hydrophobic poly(propylene glycol diacrylate) (PPGDA) chains. The gel was produced through free-radical polymerization in 0.8 M NaCl solutions using KPS as an initiator. In these gels, AAc and MAM can form hydrogen bonds, whereas the association between PPGDA chains can lead to hydrophobic domains. The PPGDA concentration affects the level of hydrogen bonding and gel mechanical properties. Tensile experiments revealed that the elastic modulus increased with a higher PPGDA concentration. Retraction experiments demonstrated increased velocity and acceleration when released from a stretched state with increasing PPGDA concentration. Swelling and deswelling of gels in saline solutions led to a change in mechanical properties and retraction behavior. This study shows that the stretchability and resilience of these hydrogels can be adjusted by changing the concentration of hydrophobic components.

Introduction

An elastic biopolymer, resilin, present in cuticles of many insects and other arthropods, displays remarkable properties such as high stretchability and resilience, which are exploited for mechanical energy storage to facilitate walking and jumping, defense mechanisms, and feeding.[1–3] Such interesting mechanical properties of resilin have been attributed to the balanced combination of hydrophilic and hydrophobic segments.[1,4–6]

The properties of hydrogels can be tuned by incorporating hydrophobic components and varying the ratio of hydrophilic and hydrophobic components. For example, hydrogels with poly(vinyl alcohol) and poly(ethylene-co-vinyl alcohol), a co-polymer containing hydrophobic ethylene and hydrophilic vinyl alcohol segments, behave like semi-crystalline polymers.[7] Hydrophilic segments resembling amorphous regions aid with water absorption and viscous behavior of the gel, and the hydrophobic segments resembling the crystalline regions provide strength.[7] Hydrophobic interactions in hydrogels prepared from hydrophobically modified polyacrylamide,[8] hydrophobically modified poly(acrylic acid),[9] and copolymerization of hydrophilic N, N-dimethyl acrylamide and hydrophobic 2-(N-ethyl perfluoro octane sulfonamido) ethyl acrylate[10] have been shown to provide additional energy dissipation mechanism leading to improved toughness and stretchability. The presence of hydrophobic components has been shown to increase the resilience of the hydrogels, as observed in natural resilin.[6,8,9,11] These types of stretchable and tough gels have gained attention for applications in wound healing, tissue culture, prosthetic devices, soft robots, artificial skin, flexible sensors, and wearable devices when paired with electronic components.[1,12–15]

Some of the small organisms, such as mantis shrimp and locusts, utilize energy storage and release mechanisms to achieve rapid movement, often defined as a power amplification process.[16] To investigate whether hydrogels can mimic such power amplification behavior, we can capture how fast the gels can retract when released from a stretched state. These retraction experiments provide insights into how fast these materials can release the stored energy. Engineering materials with high retraction velocity will be helpful in mimicking biological performances.

One of the simplest methods to prepare hydrogels with both hydrophilic and hydrophobic components is micellar copolymerization of hydrophilic and hydrophobic monomers through free radical polymerization.[8,17] The literature reports on synthesizing hydrogels with micellar hydrophobic domains, which form physical crosslink points in the network, have shown promising results in improving hydrogels' mechanical

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Electronic Supplementary Information (ESI) available: Tensile testing set-up; Change in gel transparency with PPGDA concentration; Images capturing the tensile behavior of 0% gel; Determination of tensile modulus of gels; Retraction experimental set-up showing marked lines for 1% gel; Displacement vs. time from retraction experiment; Tensile properties of the gels after swelling/ deswelling. See DOI: 10.1039/x0xx00000x properties.[17,18] However, most of the gels synthesized through this method involve physical crosslinking. In contrast, we have synthesized a chemically crosslinked network consisting of hydrophobic domains and chains forming hydrogen bonding.

Here, we synthesized hydrogels by copolymerizing hydrophilic monomers acrylic acid and methacrylamide, and hydrophobic poly (propylene glycol diacrylate) or PPGDA. We previously studied the effects of the concentration of hydrophilic monomers (AAc and MAM) on the stretchability and modulus of the hydrogels in which the hydrophobic polymer concentration was maintained constant.[6] We also investigated the retraction behavior of these gels when released from the stretched state, a behavior not widely investigated for hydrogels.

The current work aims to understand the effects of varying PPGDA (hydrophobic component) concentrations on tensile properties, swelling behavior, and retraction behavior. We have fixed the total AAc and MAM concentration of 37% (g/mL), which displayed better properties than lower concentrations. The PPGDA concentrations of 0.1, 0.3, 1, and 3% with respect to total monomer (AAc and MAM) concentration were considered. Hydrogels were also prepared without adding any hydrophobic components for comparison. The swelling and deswelling in saline solutions with different salt concentrations were conducted. This study provides insights for developing highly stretchable and resilient hydrogels having potential biological applications (physiological solution with low salinity) and seawater applications (high salinity environment).

Experimental section

Materials

For the gel synthesis, sodium dodecyl sulfate (SDS), acrylic acid (AAc), methacrylamide (MAM), potassium persulfate (KPS), and poly (propylene glycol) diacrylate (PPGDA) were purchased from Sigma Aldrich. Sodium chloride (NaCl) was purchased from Fisher Scientific. The molecular weight of PPGDA was 800 g/mol. The purity of the reactants was 98.5, 99, 98, 99, and 99.9% for SDS, AAc, MAM, KPS, and NaCl, respectively. NaCl was dissolved in Millipore deionized water with a resistivity of 18.2 mΩ to prepare solutions with molarities of 0.1, 0.8, and 4.0 M. All reactants were used as received.

Hydrogel synthesis

The gelation was conducted in 0.8 M salt solution. The total monomer (AAc + MAM) content was fixed at 37 g per 100 mL of saline solution for all gels prepared here. The AAc to MAM ratio was 4:1 wt/wt. The concentration of PPGDA was varied and was estimated with respect to the monomer concentration. First, 7.00 g of SDS was added to 83 mL of saline solution, and the solution was heated to 55 °C using a water bath until a clear solution was obtained (≈30 min). In a separate container, 7.40 g of MAM was added in 28 mL of AAc at room temperature (RT≈22 °C) and stirred for 15 min. Separately, 0.07 g of KPS was added to 17 mL of saline solution and stirred for 5 min or until dissolved at RT. Later, the SDS solution was taken out of the water bath, and the desired amount of PPGDA was immediately added dropwise, followed by stirring for ≈5 min. Next, the mixture with AAc and MAM was added dropwise to the PPGA-containing solution and stirred for 5 min. In this mixture, the KPS solution was also added dropwise and stirred for ≈5 min. Immediately after, the final transparent solution was poured into an airtight 2 L bottle and placed at 75 °C for 2 h. After 2 h, as the gel sheet formed, the heat source was turned off to allow the water bath to cool for the next 24 h. For retraction experiments, samples were prepared in 5 mm diameter glass tubes, following the same protocol discussed above. All the mixing processes were performed using magnetic stirrers at 220 rpm. The amount of PPGDA added was 37, 110, 366, and 1099 µL to obtain gels with 0.1, 0.3, 1, and 3% of PPGDA concentration (wt%), respectively. Note that the gels are named throughout this paper according to their PPGDA content (wt %).

Water content measurement

The water content of the samples was measured by determining the change in sample weight after drying in an oven for 3h at 110 °C. The samples were weighed before and after drying at room temperature. The water content in the sample was then calculated as

$$\text{wt\% water} = \frac{\text{weight of water}}{\text{total weight of the gel}}\,\text{x}\,100$$

Tensile experiments

A custom-built set-up was used to perform the tensile tests on the gels. The set-up included a moving stage (M414.3PD, Physik Instruments), a capacitance-based position sensor (Micro-Epsilon, DT 6220), and a monochrome camera (Grasshopper3, Point Grey Research Inc.). All these components were synchronized and controlled by a custom-built NI LabVIEW program. For these experiments, dogbone-shaped samples were cut from the gel sheets using a 3D-printed stamp. The length, breadth, and thickness of the dogbone sample's gauge region were 4.2, 4.2, and 9.5mm, respectively, as used by Mozer et al[19] and in our previous study.[6] The gel sample was held by supporting pins and clamps attached to the top and bottom bars of the stage to avoid slippage of samples (Fig. S1). The selected dimensions and clamping mechanism ensured sample failure in the gauge region without a significant slippage from the clamps holding the samples. The moving stage travels vertically upwards to stretch the gel at a prescribed velocity. The strain rate was estimated from this prescribed velocity. The camera captured images at 16 fps to track the displacement of the 3 marked lines at the gauge region of the gel to estimate the stretch values (λ). The distance between the lines during stretching was measured using a custom-built image-processing program in MATLAB.[20,21] The measured distance normalized by their initial distances provided the λ values. The corresponding strain was calculated as $\epsilon = \lambda - 1$. While stretching the gel, the position sensor measured the displacement of a cantilever with known stiffness to measure force. The measured force was divided by the initial cross-sectional area of the sample gauge region to estimate the nominal stress (σ). All experiments were performed at room temperature.

Retraction experiments

A gel string with a diameter of 4 mm and a length of 45 mm marked with seven equidistant lines was used for gel retraction experiments. The sample was clamped at both ends of the tensile testing instrument and stretched to a pre-determined stretch. The string was then cut with scissors adjacent to the bottom clamp and allowed to retract. The retraction process was recorded using a high-speed camera (Miro M310, Phantom) at ≈14000 fps. The images were analyzed using a custom-developed image-processing program in MATLAB to track the distance between the lines.

Swelling / deswelling experiments

Samples were swelled in 0.1M saline solution and were deswelled at 4M saline solution. The samples were swelled/deswelled until an equilibrium weight gain/loss was reached, ensured by weighing the samples at regular intervals. Swelling or deswelling capacity was calculated as:

$$\%\ \text{swelling or deswelling} = \left(\frac{\mathrm{W_{final}} - \mathrm{W_{initial}}}{\mathrm{W_{initial}}}\right) \text{x } 100$$

Where, $\mathrm{W_{initial}}$ and $\mathrm{W_{final}}$ are the initial and equilibrium weights of a sample, respectively. These swelled and deswelled samples were subjected to tensile and retraction experiments using the set-up described above. Before tensile testing, the changed dimensions of the samples were measured to calculate the cross-sectional area.

Results and discussions

Synthesis of hydrogels

Hydrogels were formed through free-radical copolymerization of hydrophilic monomers, AAc and MAM, and PPGDA. PPG or poly (propylene glycol) present in PPGDA has been considered to be the hydrophobic component at room temperature, as reported in the literature.[22] The structure of the synthesized gels is difficult to determine. However, the proposed structure for the gels can be found in our previous report.[6] We hypothesize that the PPG chains in the gel aggregate to form micellar domains in the presence of the anionic surfactant sodium dodecyl sulfate (SDS).[23–26] Salt ions in the saline solution (reaction media) assisted in stabilizing the SDS-stabilized micelles.[23–26] In addition, the carboxylic groups of AAc and amide groups of MAM chains can form hydrogen bonding. Gels of AAc and acrylamide (AM) with N, N-methylenebisacrylamide (MBAA) as crosslinker have been reported in the literature.[27–29] In contrast, in our case, we have not used any crosslinker, but randomly polymerized AAc and MAM chains are connected to each other by PPG chains. However, for 0% gel, i.e., without PPGDA, the gel formation was mostly via hydrogen bonding between AAc and MAM.[30]

The hydrogel prepared without PPGDA, the hydrophobic component, was opaque. A large number of hydrogen bonds between AAc and MAM can cause heterogeneity in the structure, leading to opaqueness in the gels.[31] With increased PPGDA concentration, a decrease in opacity was observed, and the 3% gels were transparent (Fig. S2). Previously, gels prepared with AAc and acrylamide (AM) and a hydrophobic monomer octylphenol polyoxyethylene ether acrylate (OP7-AC) displayed this kind of phase transition, from opaque to transparent with the addition of hydrophobic monomer.[32] Adding a hydrophobic component impeded the hydrogen bonding formation between AAc and MAM, leading to transparent samples.[32] Note that the large number of hydrophobic aggregates can render a gel opaque. We hypothesize that transparent gel samples, as observed in our case, even with increased PPGDA concentration, indicate that the hydrophobic aggregates are not large enough or do not have a significant concentration to render the gels opaque.

Water content

The water content of the as-prepared hydrogel without PPGDA (0%-gel) was ~50% (Fig. 1). It was observed that the increase in hydrophobic concentration led to decreased water content in the hydrogels. We have attributed this to the addition of the hydrophobic component in the structure.

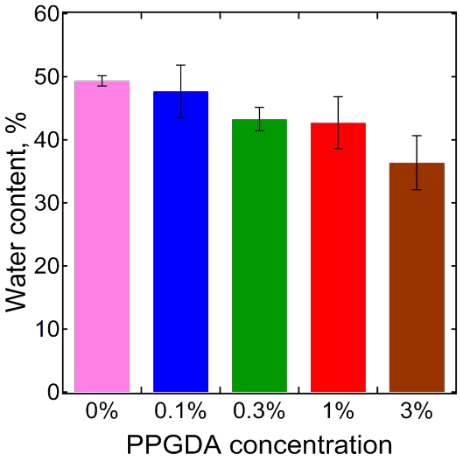

Fig. 1 Hydrogel water content as a function of PPGDA concentration. Experiments were repeated at least three times and the average values are shown with one standard deviation.

Mechanical properties

Tensile testing experiments were performed at a strain rate of 0.048 s^{-1} on the gels with varying PPGDA concentrations, from 0% to 3%. Nominal stress vs. strain results are shown in Fig 2A. 0% gel exhibited a distinctly different mechanical response compared to the PPGDA-containing gels (Fig. 2A). Here, with increasing strain, the stress linearly increased to a maximum value (yield stress), followed by a long decay until sample failure took place at a large strain, > 10 (Fig. 2A-B). Such long decay was manifested by the sample thinning out before failure during the tensile testing (Fig. S3). The failure stress value has been found to be low. This long stress decay and a gradual decrease in nominal stress can be attributed to the gradual dissociation of hydrogen bonds present in the system.

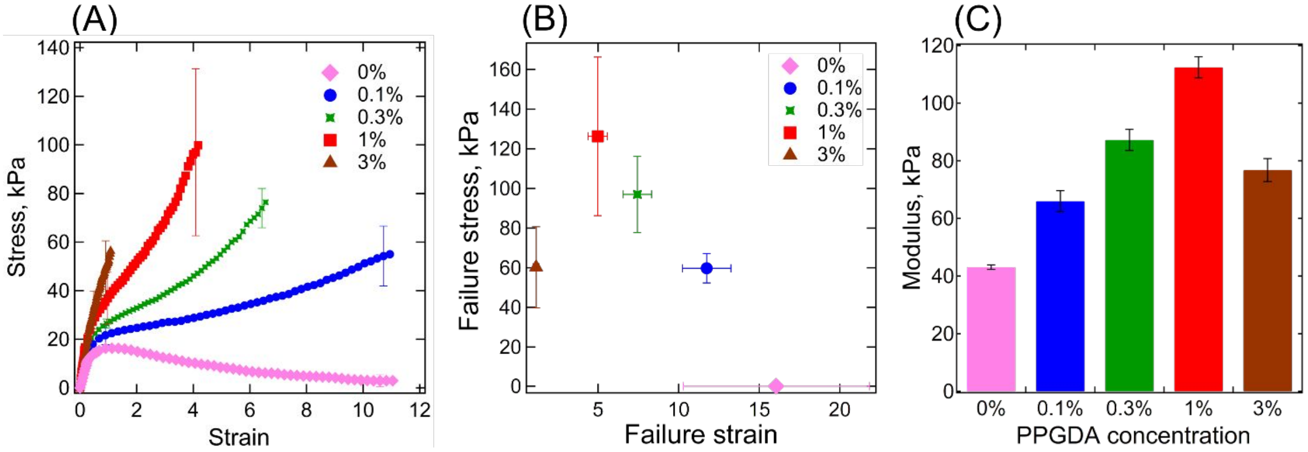

Fig. 2 Results from tensile experiments for 0, 0.1%, 0.3%, 1%, and 3% gels. (A) Nominal stress as a function of strain; (B) Failure stress and failure strain; (C) Young's modulus of the gels obtained by finding the slope of the linear region (up to strain of ~0.3) (see Fig. S4). All experiments were repeated at least three times, and the average values are shown with one standard deviation.

In contrast, gels with 0.1, 0.3, and 1% PPGDA displayed an S-shaped curve typically observed for elastomers.[33,34] The increase in PPGDA concentration exhibited a decrease in failure strain, however, the tensile strength (failure stress) increased. The failure stress and failure strain both decreased for 3% gel. With the lowest failure strain of ~1, the tensile test data for this gel was similar to a brittle solid. The higher standard deviation in the failure stress values, particularly for 1% PPGDA, likely originated from the small variation in the synthesis process involving a higher concentration of hydrophobic component.

The stress-strain response for 0% gel was similar to that reported in the literature for MAM-AAc gels, synthesized without crosslinker, but in the presence of catalyst tetramethyl ethylenediamine (TMEDA), except for the absence of strain-softening behavior observed in our case.[35] Those gels display mostly constant stress values after yield stress.[35]

The mechanical properties of hydrogels were influenced by the amount of water present in the gels.[36–39] With decreasing water content in the gels, the number of elastically active chains per unit volume increased, leading to higher elastic modulus.[40] [18,41] Further, the addition of PPGDA increased the number of connecting chains and, therefore, the crosslinking density. These resulted in a higher modulus and failure stress. Similar behavior was observed when gels were prepared by micellar copolymerization of acrylamide (AM) and octylphenol polyoxyethylene ether (OP-4).[17,18,41–43] However, when the PPGDA concentration was increased beyond a certain limit, we hypothesize that the PPGDA chains might not have been effectively incorporated into the network, as the SDS content was kept constant, leading to a lower modulus.

The gel mechanical responses with respect to strain rate were studied by performing the tensile experiments at a higher strain rate of ≈0.48 s^{-1}. Fig. 3 compares the results for two strain rates 0.048 and 0.48 s^{-1}. For both these strain rates, the gels did not show a significant strain-rate dependence for low strain values (<1), whereas differences were observed for higher strain values. Particularly, when the strain rate was increased, the gels attained higher stress values at larger strain, including higher failure strain. The strain rate dependence was more prominent for lower PPGDA concentration, viz., for 0.1% gel compared to 0.3% gel. It has been shown previously that strain-rate dependence was mostly absent in 1% gel.[6] We have attributed this behavior to the presence of hydrogen bonding in low PPPGDA-containing samples, as discussed above.**[44]** At lower strain values, the dissociation of hydrogen bonding was minimal, leading to similar stress-strain responses for different strain rates.**[45]**

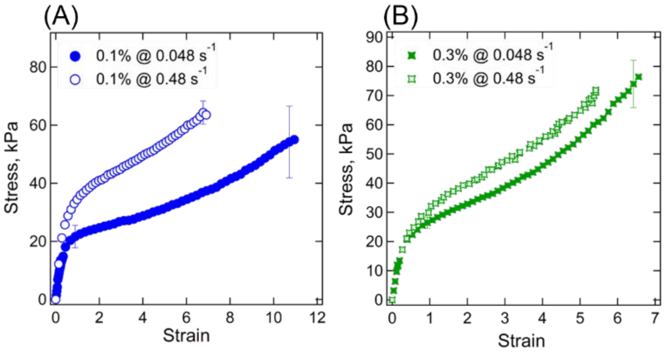

Fig. 3 Effect of strain rate on the tensile properties for (A) 0.1% gel and (B) 0.3% gel. Experiments were repeated at least three times, and the average values are shown with one standard deviation.

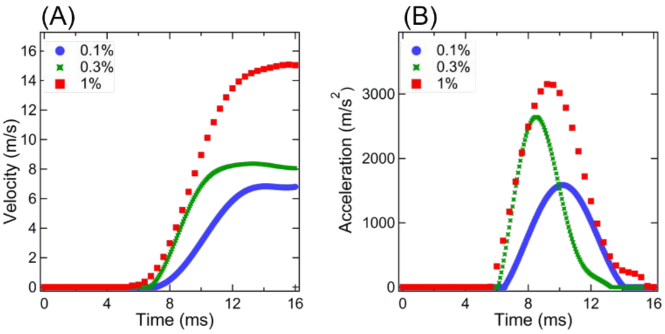

Fig. 4 Retraction behavior of the gels with different PPGDA concentrations from a stretched state are shown for one representative run: (A) velocity (B) acceleration of Line 1 (see Fig. S6 for position vs time data) as a function of time. The samples were released from λ = 6.

Retraction Behavior

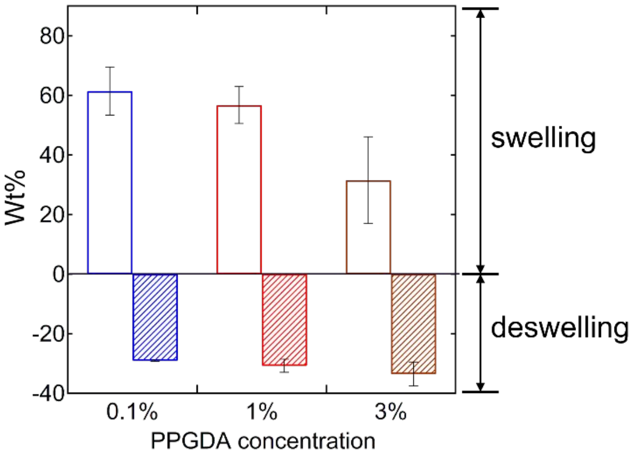

Fig. 5 Swelling and deswelling of 0.1, 1 and 3% gels in 0.1M and 4.0M NaCl solutions, presented in terms of wt%. Experiments were repeated at least three times, and average value is shown with one standard deviation.

The retraction experiments were performed for 0.1, 0.3, and 1% gels, where the samples were stretched to λ ≈6 and were subsequently released. Results from our previous work have shown that the retraction velocity and acceleration increase with the increase in the applied stretch.[6] For 1% sample, the maximum attainable stretch before failure was 6, and we kept that as the basis for comparison between samples. Retraction experiments could not be performed on 3% gel because of its very low stretchability and brittle nature. Further, gels prepared without PPGDA could not withstand the stress from clamping to the geometry, making the retraction experiments extremely challenging. Fig. S5 displays the image of a sample with lines that were tracked for estimating the retraction velocities and acceleration. While retracting, the string slacked as soon as it crossed its initial length. The displacement vs. time data from the time the string was snapped to the slacking was used for analysis. Fig. S6 shows the position of the lines as a function of time obtained by analyzing the images. The tracking of each line on the sample captures three regions: (i) a horizontal region, which indicates that the line remains stationary; (ii) steep decay with a constant slope, indicating a constant velocity; and (iii) a region in between, capturing the acceleration from static condition to a constant velocity.[6,16,46–48] The displacement data were fitted using a fifth-order Fourier series (Fig. S6). Here, the position (f) versus time (t) data can be mathematically represented as $\mathrm{f(t)} = \mathrm{a}_0 + \sum_{\mathrm{i}=1}^{5} \mathrm{a_i} \cos{(\omega \mathrm{t})} + \sum_{\mathrm{i}=1}^{5} \mathrm{b_i} \sin{\omega t}$, where a_0, a_is, b_is, and ω are the fitting parameters.[6] The function obtained from the fitting was then differentiated to obtain a function for velocity, which was further differentiated to obtain a function for acceleration.

Fig. 4A and 4B display the estimated velocity and acceleration from one representative run for each gel. An increase in velocity and acceleration of the gels was observed when the PPGDA concentration was increased upto 1%. The maximum velocity and acceleration were achieved by the gels prepared with 1% PPGDA and the values were ~16±1.1 m/s and 3300±390 m/s^2, respectively. An increase in PPGDA concentration led to higher elastic modulus, thereby increasing the amount of stored elastic energy, which was reflected in higher retraction velocity and acceleration.[48]

Swelling and deswelling behavior

Swelling and deswelling experiments were performed to determine the stability of the gels in saline solution in different ionic strengths and the corresponding change in mechanical properties. The 0.1, 1, and 3% gels were submerged in 0.1 and 4 M of NaCl solutions. These concentrations were chosen to understand the behavior of gels when the external solution has an ionic strength less than and greater than the synthesis condition of 0.8M, respectively. It was noticed that the sample volume increased (swelled) in 0.1M and decreased (deswelled) in 4M NaCl solutions, respectively. This behavior was due to the osmotic pressure caused by the difference in ion concentration in and outside the gel samples.[49] The percentage of swelling and deswelling in those solutions for the samples with different PPGDA concentrations is shown in Fig 5. An increase in the PPGDA concentration led to a decrease in swelling capacity. Along with the added hydrophobicity, the increase in crosslinking density caused by the addition of PPGDA can cause decreased swelling capacity.[50] The ionic nature of AAc also plays a role in altering the swelling behavior of the gels. Previous studies have shown that NaCl can reduce water intake.[49,51,52]

The mechanical properties of the gels after swelling and deswelling were measured using tensile testing. The stress vs. strain data are plotted in Fig. 6, whereas the elastic modulus values are presented in Fig. 7. The tensile properties are summarized in Table S1. After swelling in 0.1M NaCl solution, the hydrogels displayed a similar behavior as as-prepared samples irrespective of the PPGDA concentration, except for a slight decrease in the extensibility and failure stress for the low PPGDA concentrations, i.e., 0.1 and 1%. The swelled 3% gel did not display a change in extensibility and failure stress from the as-prepared state. The modulus of the samples decreased with swelling for all PPGDA concentrations (Fig. 7).

The deswelled samples displayed significantly different mechanical properties compared to the as-prepared samples. Particularly, hydrophobic concentration played a significant role. For all cases, the deswelling resulted in an increase in modulus and failure stress. For 3% gel, a substantial increase in stretchability compared to the as-prepared sample was observed, whereas the stretchability decreased for 0.1% gel (Fig.6). However, the stretchability of all deswelled samples was similar for all three PPGDA concentrations.

During swelling, water molecules enter the gel network, moving the crosslinking junctions apart. This stretching can increase the modulus, but at the same time, the reduction in chain density will decrease the modulus. The net effect is the reduction in modulus during swelling, as observed for other gels.[53–56] Also, the pre-stretching in chains during swelling can impact (lower) the failure strain in the subsequent mechanical tests. The 3% gel did not manifest a significant change in failure

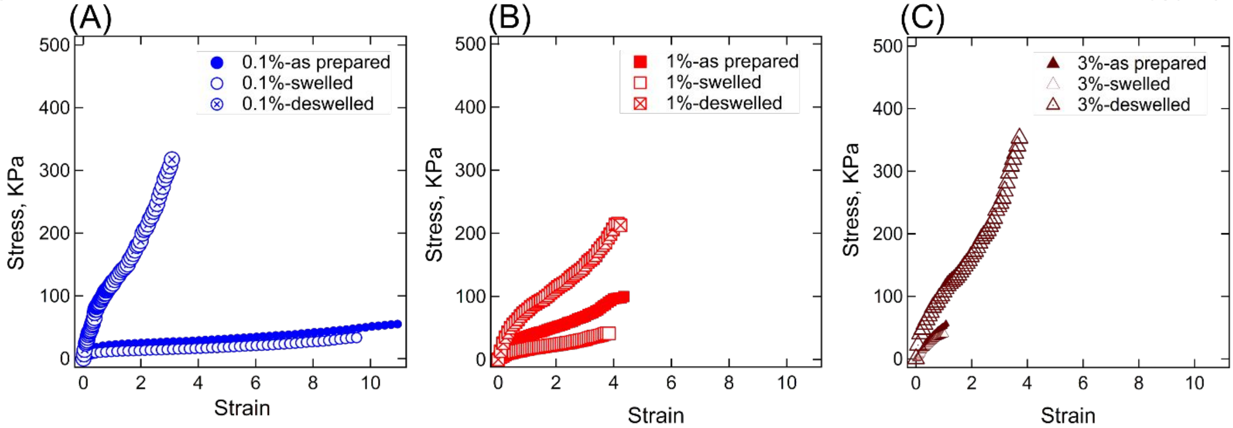

Fig. 6 Results from tensile tests capturing stress vs. strain for (A) 0.1%, (B) 1% and (C) 3% gels swelled / de-swelled in 0.1M and 4.0M NaCl solutions. The results for as-prepared samples are also shown. Experiments were repeated at least three times and average value is shown with one standard deviation.

strain due to the small swelling capacity compared to other gels. Similarly, in the deswelled samples, the polymer chains were in a collapsed state. Also, due to deswelling, the number of crosslinks per unit volume increased, and the corresponding moduli.

The stretchability of the deswelled sample displays some dependency on the strength of the hydrogen bonds and the amount of water present in the gels. As discussed above, there was less hydrogen bonding between AAc and MAM in 3% gel, leading to the availability of the carboxylic acid groups, which could ionically interact with salt ions.[52] In 4.0 M NaCl solution, salt ions further screened the interactions between the carboxylic and amide groups from AAc and MAM. This could lead to the formation of ion pairs between salt ions and the polymers, causing them to form a coiled structure.[52,57] The chain coiling was further enhanced by the lower water content in 3% gel. The highly coiled structure led to higher extensibility in comparison to its as-prepared state.[52] In fact, this is the only sample for which deswelling led to increased extensibility compared to their as-prepared state.

As the tensile properties of the swelled and deswelled samples changed from as-prepared gels, we also studied the retraction behavior of these samples. A comparison of how the retraction behavior varies for as-prepared samples and a de-swelled sample with comparable modulus is shown in Fig. 8. Specifically, we considered an as-prepared 1% gel and deswelled 3% gel, which displayed a high stretchability. Both the samples were released from the stretched state of λ= 6. The retraction of the as-prepared sample was very rapid, as the gel string retracted completely in about 20 ms. In contrast, for the deswelled gel sample, the complete retraction took about 300 ms. As discussed above, we hypothesize that the salt ions in the deswelled gel can form complexation with AAc, which can restrict the chain motions, resulting in slower retraction. Note that stretchability measured from tensile experiments was not affected due to the presence of salt, as the strain rate for the tensile tests was much slower.

In contrast, similar to tensile test data, the swelled samples did not show a significantly different retraction behavior compared to the as-prepared sample. For example, for 1% gel and $\lambda = 3$, the retraction velocity for the as-prepared and swelled samples were 6.58±0.6 and 5.86±0.8 m/s, respectively. Similarly, the accelerations were 1096±305 and 1813±68 m/s^2, respectively. The structure of hydrogels and the change of their structure with PPGDA concentrations and with swelling and deswelling need to be further investigated using the scattering-based technique, which will be further investigated in a future study.

Conclusions

Here, we report that the mechanical properties of the hydrogels consisting of hydrophilic AAc and MAM and hydrophobic PPGDA can be tuned by altering the concentration of hydrophobic polymer. The tensile properties of hydrogels prepared without hydrophobic PPGDA were dictated by the hydrogen bonding between the AAc and MAM. We observed that the addition of hydrophobic polymer improved the fracture stress and the modulus of the gels. The tensile modulus and failure stress of the gels increased with an increase in hydrophobic concentration up to a certain limit, and a further increase in hydrophobic content caused a reduction in the

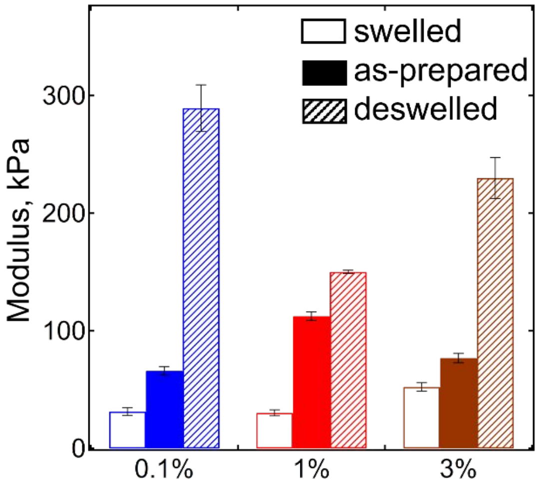

Fig.7 Young's modulus of the swelled and deswelled 0.1, 1 and 3% gels obtained by finding the slope of the linear region (up to strain of ~0.3). All experiments were repeated at least three times and the average value is shown with one standard deviation.

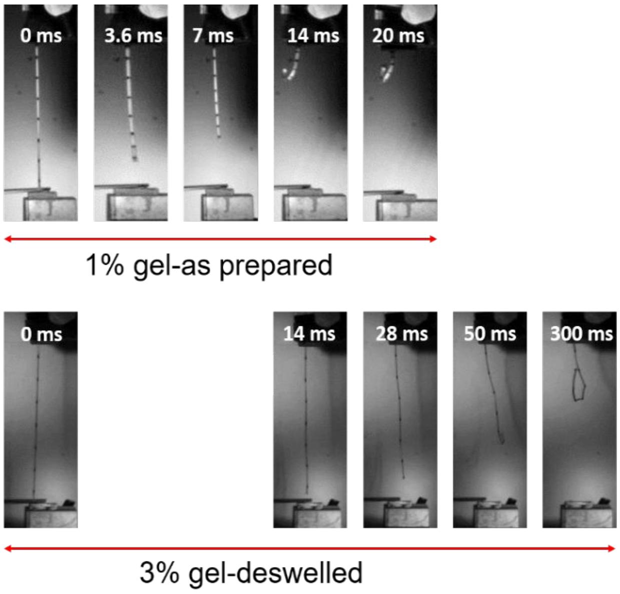

Fig.8 Retraction frames as a function of time for as-prepared 1% gel and deswelled 3% gel.

strength of the material. Resilin in various species allows them power amplified activities because of its high resilience. Our gels, consisting of hydrophobic components similar to resilin have high resilience, are capable of fast retraction from the stretched state. The free retraction velocity and acceleration released from a stretched state were found to be directly dependent on the elastic modulus of the gels. The hydrogels displayed improved strength, modulus, and stretchability after deswelling when exposed to high salt concentrations. The results obtained can be helpful in providing insights to develop new materials for a wide range of applications, especially in low-salinity environments, such as physiological conditions and high-salinity seawater environments.

Author Contributions

A.V. and S.K. designed the experiments and conducted data analysis and interpretation. A.V. conducted the experiments. S.K. conceptualized and oversaw the research. R.M.B.P conducted preliminary experiments. S.M. designed the tensile set-up and framework for tensile and retraction analysis. K.E. assisted in performing tensile and retraction experiments and data analysis. The manuscript was written by A.V. and S.K. and included feedback from all authors. All authors have approved the final version of the manuscript.

Conflicts of interest

There are no conflicts to declare.

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