

# Characterization of Hydrogel Curing Methods for Manufacturability

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**Abstract**—In the field of soft robotics, hydrogels possess material properties that allow them to function as both soft strain sensors and electrodes for dielectric elastomer actuators. However, there is still much that needs to be understood about the curing process of hydrogels and the resulting material characteristics before devices can be reliably manufactured. In this study, we investigated the effect of curing temperature on the as-cured material properties of acrylamide-based hydrogels hydrated with lithium chloride and magnesium chloride salt solutions. Samples were cured at room temperature, 60°C and 100°C, and the resulting changes in mechanical stiffness, rehydration rate, and electrical conductivity were measured. We found that stiffness decreases with increasing curing temperature, and the rehydration rate and electrical conductivity had an erratic relationship with curing temperature. These hydrogels were also shown to function as strain gauges. To determine under what conditions hydrogel would spontaneously cure at room temperature, uncured hydrogel was deposited in drops between 1 L and 0.5mL. We found that uncured hydrogel did not spontaneously cure when placed in volumes smaller than 20 L at room temperature. These samples did, however, cure at elevated temperatures. These experiments show that hydrogels have material properties and curing times that can be tuned depending on the needs of the manufacturing process, and that conductive hydrogels retain their required functionality after prolonged use at ambient temperatures.

*Keywords:* hydrogel, dielectric elastomer actuator, strain gauge, curing process, soft robotics

## I. INTRODUCTION

In recent years, interest has been growing around the idea of soft robotics and compliant electronics[1][2][3][4][5]. The desire for wearable electronics, lighter and more efficient actuators, and simplified robot construction are some of the many reasons this field continues to grow [2][6][7][8]. In order for this field to develop further, two capabilities in particular must progress. Soft robots, like all robots, require sensing and actuation in order to function. However, many of the devices currently available require multiple complicated steps to produce, resulting in an increase in variation, production time, and failure[9][10]. These limitations make the design process slower due to longer iteration times. In this paper, we evaluate hydrogels as a possible solution due to their ability to be used in both sensors and actuators. Their versatility in soft robotic devices means that finding a way to manufacture devices using hydrogel would improve the production of devices such as strain gauges and dielectric elastomer actuators[7][11][12]. If successful, these technologies would allow for the development of novel electronics and robotics that are presently impossible.

Hydrogels show promise in the area of soft robotics due to their electrical conductivity and flexibility[11][12]. In the area of soft strain gauges, two sensing methods have been demonstrated based on changes in resistance and capacitance[7][11]. Resistive sensors operate based on the fact that the sensor geometry changes as strain is applied, causing the resistance to change. The resistance was calculated in the same way reported by Keplinger, et al[13]. The resistance is characterized according to:

$$R_0 = \frac{\rho L}{A} \quad (1)$$

$$R = \frac{\rho L \lambda}{\frac{A}{\lambda}} \quad (2)$$

where  $R_0$  is initial resistance,  $\rho$  is the resistivity of the material,  $L$  is the length of the sample,  $A$  is the cross-sectional area,  $R$  is the resistance, and  $\lambda$  is the stretch. These equations show that the relationship between  $R_0$ ,  $R$ , and  $\lambda$  is  $R/R_0 = \lambda^2$ . This means that as the hydrogel stretches, its resistance will increase.

Hydrogels are also used in soft actuators as compliant electrodes on dielectric elastomers[12][15]. The theory behind dielectric elastomer actuators is reported by Pelrine, et al.[14]. Dielectric elastomer actuators operate primarily as flexible capacitors, with compliant electrodes on the top and bottom of a nearly incompressible dielectric film. When a voltage is applied across the dielectric, the top and bottom electrodes are forced towards each other due to the electrostatic attraction of opposite charges. Because the dielectric is incompressible, the force causes the dielectric to move in the remaining two directions due to conservation of volume as shown in Figure 1. This movement results in the actuation of the dielectric elastomer. Chen, et al. was able to create a dielectric elastomer using hydrogels that showed areal

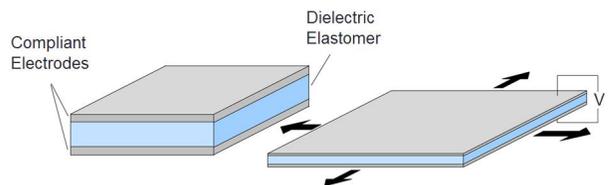


Fig. 1. As a voltage is applied across the dielectric, the top and bottom electrodes attract. This motion causes the dielectric to compress in one direction, and expand in the others [14]

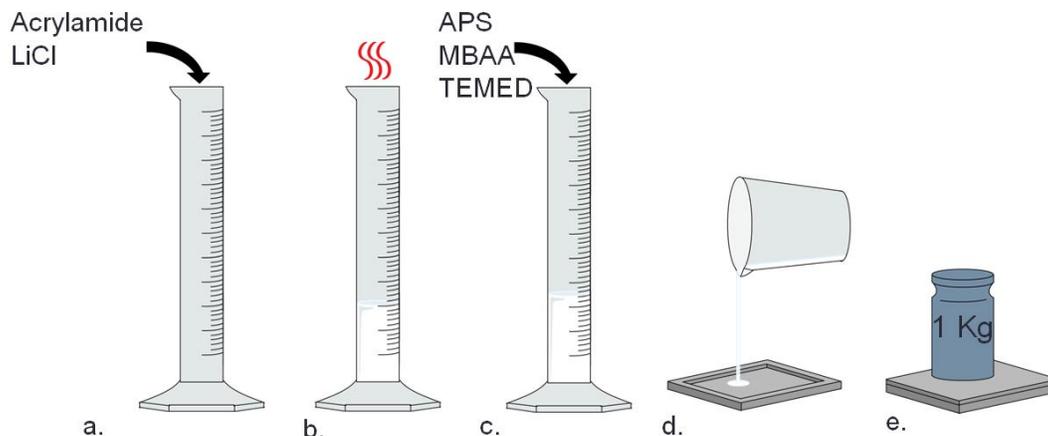


Fig. 2. a) Acrylamide and lithium chloride are placed in the cylinder, water was added until completely dissolved. (b) We wait 5 minutes for the heat from the lithium chloride to subside. (c) APS, MBAA, and TEMED were added to the solution and mixed. (d) For the stiffness testing and rehydration testing, the solution was poured into a 3in x 2in x 0.06in container and placed at room temperature, 60°C and 100°C. Electrical resistance testing procedures are shown in figure x. (e) weight was applied until cured to prevent leakage.

strains of up to 134% when a voltage was applied across the actuator[12].

Although there have been successful devices created in the past, further characterization of hydrogel is necessary before these devices can be manufactured and handled reliably. If a hydrogel is to be useful, it must be robust in a manufacturing environment. In such an environment, the hydrogel will likely encounter elevated temperatures to quicken the curing process. Hydrogel will also be stored in the manufacturing equipment as the process is occurring, possibly in tubes or nozzles. This means hydrogel must be able to retain its functionality when heat is applied and be able to remain uncured for extended periods of time in order to be manufactured into operational devices.

In order to further characterize hydrogels for manufacturing, experiments were conducted to assess the functionality of devices and the ability of the hydrogel to stay uncured during processing. Hydrogel samples were cured at room temperature, 60°C and 100°C, and tested for stiffness, water retention in an ambient environment, and electrical conductivity. We found that devices cured at elevated temperatures showed no major changes in electrical or physical functionality, meaning that hydrogels can be cured quickly in manufacturing processes. We also found that uncured hydrogel at room temperature does not spontaneously cure

## II. EXPERIMENTAL

### A. Hydrogel Fabrication

Acrylamide hydrogel was synthesized according to the procedure reported by Chen, et al.[12]. Acrylamide and lithium chloride (LiCl) were completely dissolved in water, with concentrations of 2.2 M and 8 M respectively. After the solution was cooled to room temperature, N,N- Methylenebisacrylamide (MBAA) , N,N,N,N-tetramethylethylenediamine (TEMED), and ammonium persulphate (APS) were added to act as a crosslinker, accelerant, and thermo-initiator. These components were added with a

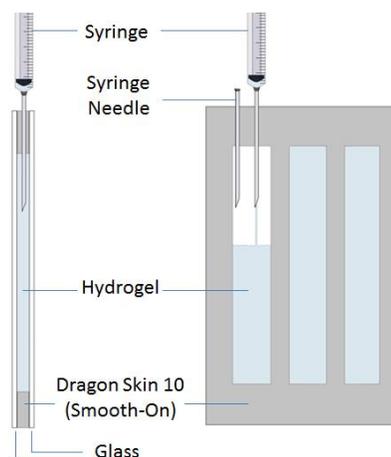


Fig. 3. Mold used to create samples for electrical resistance testing. Two glass slides and a Dragon Skin 10 (Smooth-On) frame were layered as shown, with uncured hydrogel injected into the air pockets. Not shown are binder clips used to clamp the layers together and prevent leakage. The molds shown are 6cm x 1cm x 0.25cm.

weight ratio of 0.06 wt.%, 0.05 wt. %, and 0.17 wt.% respectively. The resulting solution was then poured or injected into a mold, and left to cure at room temperature, 60°C and 100°C as shown in Figure 2.

For stiffness testing and rehydration testing, samples were cured using the method shown in (d) and (e) of figure 2. A second method to produce samples was developed in order to limit the waste of uncured hydrogel, and was used for the testing of electrical resistance. This method is shown in Figure 3. Here, uncured hydrogel was injected into the mold using a syringe while a second syringe needle was used to vent air. After completion, these assemblies were cured at room temperature, 60°C and 100°C.

### B. Mechanical Stiffness Testing

Hydrogel samples were placed in saturated LiCl solution. These samples were allowed to hydrate for one day, after

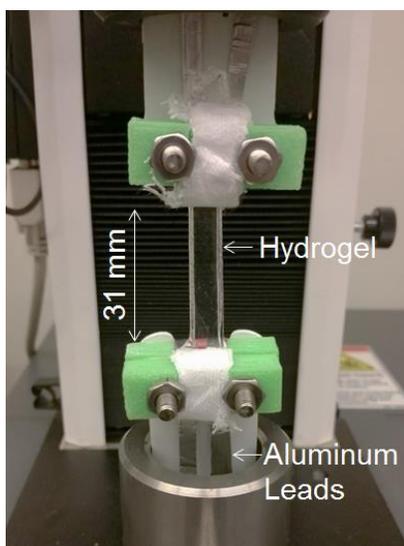


Fig. 4. A Hydrogel 1cm x 6cm is attached to the Instron using 3D printed clamps. For both top and bottom clamps, one side is wrapped in absorbent cotton to increase grip, the other is wrapped in two aluminum foil strips to be used as electrical leads.

which they were cut into 0.5 inch x 2.5 inch strips using a VLS 2.30 Laser Patterning System (Universal Laser Systems, Inc.). These samples were then wrapped in cotton strips on either end with 2cm of hydrogel in between them, then taped and fastened between two plastic holders. These samples were placed in an Instron 3345 configured with a 50N load cell and extended until failure. This was repeated for six samples cured at room temperature, six at 60°C, and five at 100°C.

### C. Rehydration Testing

Four samples 1cm x 1cm in size were cut from hydrogels cured at room temperature, 60°C and 100°C created using the method shown figure 2. Four additional samples were created using 3.56 M MgCl<sub>2</sub> as a substitute for LiCl. These samples were left to dry in an ambient environment for 5 days before cutting, and one day after cutting. Then, they were weighed and placed in deionized water. These samples were then removed from the water, dried, and weighed periodically for the next four hours, and once the next day. The LiCl samples cured at room temperature were dried by a paper towel tap on each side of the square, and the remaining samples were dried by shaking for three seconds.

### D. Electrical Resistance Testing

Three samples each were cured at room temperature, 60°C and 100°C in molds as shown in Figure 3. These samples were then placed directly into the clamp shown in Figure 4. This configuration was then placed in an Instron and extended to strains of 0%, 100%, 200%, and 300%. At each of these strains, a function generator supplied a voltage with a frequency of 10 Hz, 100Hz, and 1000Hz at an amplitude of 0.5 V.

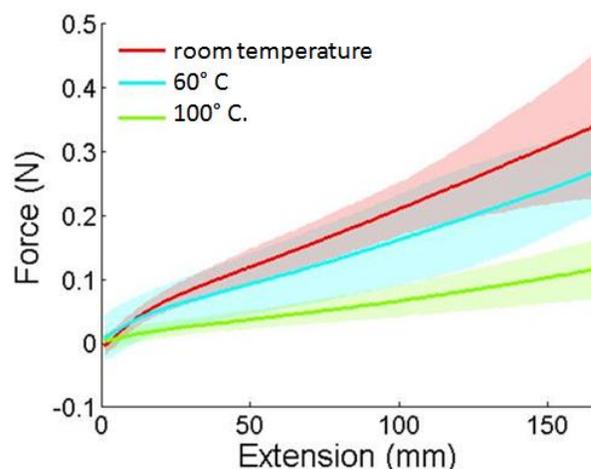


Fig. 5. This figure shows the average forces exerted on hydrogel samples under increasing strain for samples cured at room temperature, 60°C and 100°C. Six samples were used for both room temperature and 60°C, and five were used for 100°C.

## III. RESULTS

It was found that stiffness decreases as curing temperature increases, as shown by Figure 5. The Young's modulus was found to be  $2.368 \pm .589$  kPa when cured at room temperature,  $1.809 \pm .663$  kPa at 60°C, and  $.7494 \pm .0951$  kPa at 100°C. These numbers were found by using a linear model between the extensions of 25mm and 125mm. The results found in this experiment are close to the results reported by Chen et al, who found Young's modulus for samples cured at 50°C to be 1.8 kPa for small strains. Errors are at 95% confidence.

There appears to be no relationship between curing temperature and rehydration rates at this time, possibly as a result in the variation in preparation methods between samples. In addition to changes in drying methods before weighing the samples, the lithium chloride samples cured at 100°C

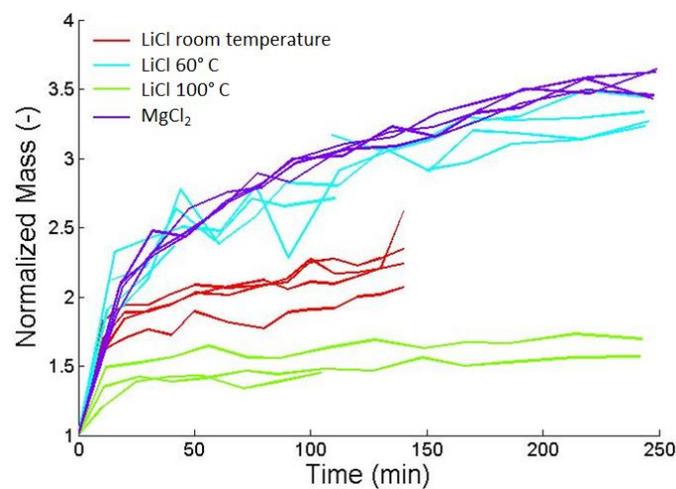


Fig. 6. Normalized mass with respect to the initial mass of rehydrating hydrogel over time. Hydrogels were left at room temperature and humidity for 5 days and then cut into 1cm by 1cm squares, which were placed in water at t=0.

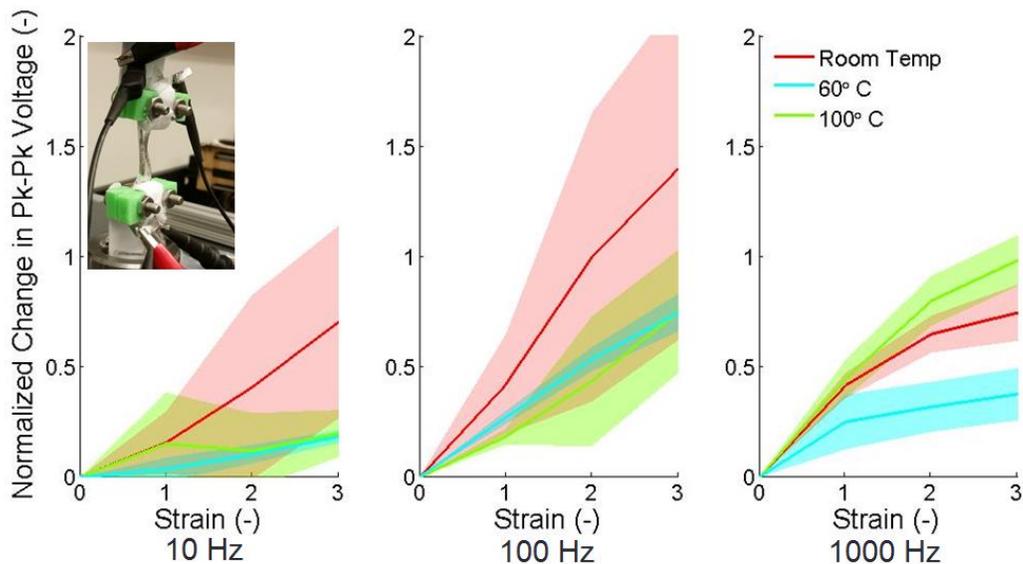


Fig. 7. This figure shows the normalized change in peak-to-peak voltage across the hydrogel for increasing strain with respect to the initial peak-to-peak voltage. The input voltage was provided using an AC voltage supply at the frequencies 10Hz, 100Hz, and 1000Hz. The clouds show 95% confidence. The inset picture shows the configuration of the hydrogel sample in the Instron.

were dehydrated completely before cutting in an attempt to quicken the preparation time of the samples. As this was not desired, it was rehydrated and prepared again with the other samples. The methods do not allow for the quantitative analysis between samples, although qualitatively it can be seen in Figure 6 that lithium chloride preforms better at retaining water than magnesium chloride. This is likely due to the fact that magnesium chloride is not as hydroscopic as lithium chloride. Further analysis is required to understand what is occurring in the lithium chloride hydrogels.

The experiment conducted on the hydrogel to find the effects of curing temperature on resistance showed no definitive relationship, although all samples showed an increase in resistance as a function of strain for the 100 Hz and 1 kHz frequencies. Figure 7 shows that the 1kHz frequency has a relatively small error, and so has potential to be used as a strain gauge.

Uncured hydrogel placed at room temperature showed no signs of curing in volumes less than 10 L. Samples that had

shown the beginning stages of curing, specifically in the volumes of 10 L and 20 L, eventually returned to an uncured state over time. Samples between the volumes of 50 L and 500 L were able to cure in their core, and then remained in that state for the next 24 hours. When placed on a hot plate, uncured samples were able to cure normally. These results can be seen in Figure 8.

#### IV. CONCLUSION

These results show that this hydrogel has the potential to be used in manufacturing processes while still retaining its ability to stretch, retain water, and conduct electricity. The results from the stiffness testing suggest that it is possible to modify the stiffness of hydrogels as needed by changing the temperature at which they are cured. The ability to retain water does not have a predictable relationship with curing temperature at this time, although this testing shows that hydrogel samples are able to adequately retain water within the temperature range observed. Electrical resistance also shows no relationship with curing temperature. The hydrogels did, however, show increases in resistance with increasing strain, meaning they could be used as strain gauges. These experiments also show that by decreasing the volume of hydrogel stored, spontaneous curing can be limited within a device. Once the manufacturing of hydrogels is successful, devices such as strain gauges and dielectric elastomer actuators can be created with less variation and wasted materials. As a result, soft robots as a whole will have more reliable devices that can be created with little human interference. In the future, in order to create hydrogels that are easier to manipulate when cured, there should be testing conducted to characterize the adhesive qualities of hydrogel on different surfaces.

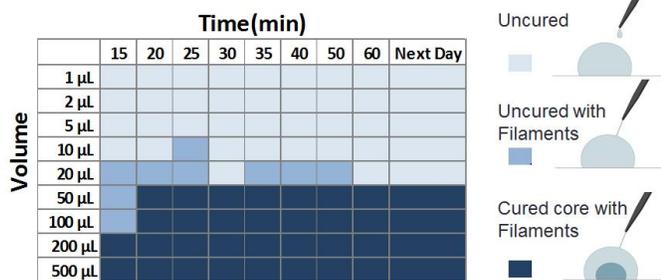


Fig. 8. Different volumes of hydrogel were placed in a plastic container at room temperature and left to cure over time. When a similar process was conducted in an incubator at 60°C, all samples were cured within 50 minutes.

## V. ACKNOWLEDGMENTS

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