ME-412 : Measure of work fracture using analog electronics on polyacrylamide hydrogel

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The present study experimentally investigated the mechanical properties of a polyacrylamide hydrogel. More precisely, the aim is to measure the work of fracture W_f of an hydrogel with a specific chemistry. Work of fracture is defined as the work per unit volume required for the material to fail. This value can be deduced after a proper tensile test and its resulting loading curve. To obtain this curve, an analog circuit amplifying a Wheatstone bridge contained in a load cell is developed. Then, a tensile test was performed. The chemistry of the studied hydrogel made it difficult to break the samples in a tensile test. Thus, over nine experiments, only one was fractured. This does not provide a reliable value of W_f. An analysis of enhancements for this experiment was performed and multiple ways to improve the results are given.

Hydrogel is a gel in which the swelling agent is water. It is made of a three-dimensional cross-linking of hydrophilic polymers. Hydrogels are mainly used in the biomedical field for drug absorption or burns treatments. They showcase interesting mechanical properties such as their response to high strain or the influence of the cross-linking concentration on the Young's modulus of the hydrogel. Those changeable mechanical properties make hydrogel very appealing in the biomedical field. Determining the mechanical characteristics of hydrogels can be challenging, particularly because of their distinct mechanical behavior when compared to other conventional engineering materials. Polyacrylamide (PAAm) hydrogels are characterized by their long chain lengths and a capacity for preserving their shape and mechanical strength even while imbibing water and ions. Multiple studies have been done on the subject in order to link the cross-linking concentration of the hydrogel on its mechanical properties [1] or on the influence of the water concentration on mechanical properties. This paper wishes to focus on polyacrylamide hydrogel.

Although studies have been carried out on all types of hydrogel, the study of polyacrylamide hydrogel is limited. One of the few papers on the subject [2] aims at studying the mechanical properties of a reversible, DNA-crosslinked polyacrylamide hydrogel. This paper therefore aims at studying a regular type of polyacrylamide hydrogel and answering this question: is it possible to link the mechanical properties of the material to the notions of cross-linker-to-monomer ratio and percentage of weight monomer per water?

In order to answer this question, a tensile test was conducted on hydrogel samples with a known composition using a load cell and an amplifying circuit. The hydrogel studied in this paper has a cross-linker to monomer ratio C equals to $C = 0.153\%$ and a percentage of weight of monomer in water Φ equals to $\Phi = 13.7\%$. This paper will go through the building of said circuit, the processing of the data acquired as well as the results obtained from the tensile tests, and a comparison of those results to similar studies to assess the validity of the experiment made. The paper concludes with a discussion of the results and the challenges faced during testing. The results obtained in this paper are the stress-stretch curve of the hydrogel and thereafter the Young's modulus E of the material as well as its work of fracture W_f.

A load cell is a transducer that converts a force into an electric signal. The load cell works using the strain gauge principle, typically on a Wheatstone bridge[3]. When a force is applied to the load cell, the conductive elements inside it deform slightly causing a change in the strain gauge electrical resistance. This change is proportional to the load, therefore it is possible to quantify the amount of force applied by measuring the difference in voltage before and after the load is applied. This output is a voltage difference expressed in millivolts (mV). To get this measurement, a specific electric circuit is needed to ensure a stable input voltage and to amplify the output voltage of the load cell. This whole setup is a tensile test that allows to get the stress-stretch curve.

The output voltage of the load cell is related to a force in Newton using a calibration. Figure 1 shows the fitting curve obtained from the calibration of the load cell coupled to the circuit. The measurement of the work fracture of the polyacrylamide hydrogel is then made based on this calibration. Regarding the calibration different screws of different known weights were used. Based on the force generated by the weight of the screws the resulting voltages were collected. Then, by applying a linear regression, the linear relation between force and voltage was obtained with a mean absolute percentage error of MAPE = 2.1544%. Thus, the linear regression is an acceptable approximation for this calibration.

$$F = 0.2344 * V - 0.9637 \tag{1}$$

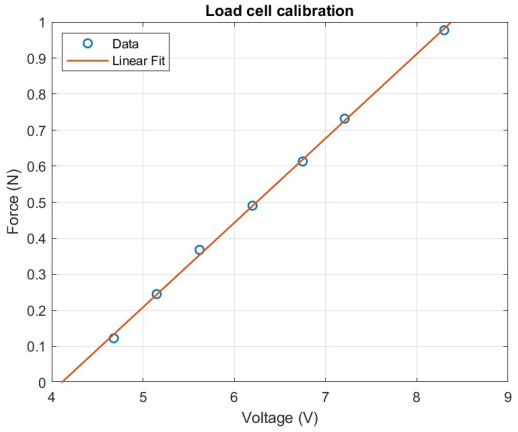

FIG. 1: Load cell calibration using the output voltage from the load cell amplifier circuit. The measurements were made using masses of known weight (blue circles) and reading the amplified output voltage from the load cell. The results can them be fitted using a linear fit (red curve) with MAPE = 2.1544%.

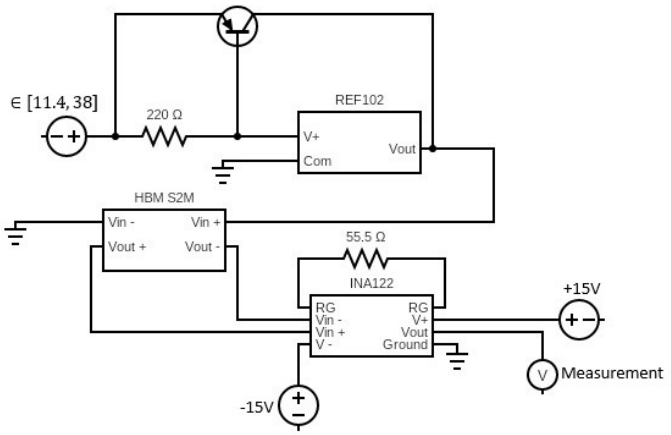

FIG. 2: Electrical circuit to amplify the output voltage of the load cell (HBM S2M 20N) using REF102 to provide a 10V voltage input coupled to a PNP transistor (PNP 2N2905) and a 220Ω to provide enough current to the in-amp and INA 122 as an amplifier with a gain of 3608.6 between the input voltage from the load cell and the output voltage of the amplifier. A symmetrical power supply is used ($\pm 15V$) to power the instrumental amplifier.

The load cell used in this experiment is the HBM S2N 20N load cell [4]. The load cell requires a steady input voltage of $10V$. In order to provide it, the REF102 device is used as it outputs a more stable voltage of $10V$ than a general-purpose power supply and it is also designed for low noise [5]. The overall resistance of the load cell is 300Ω and if a $10V$ voltage is supplied, it will draw a current of $I = V_{input}/R_{total} = 33mA$. A transistor PNP 2N2905 [6] and a 220Ω resistor is added to the REF102 in order to provide this current as the REF102 on itself can only provide up to $10mA$. Drops in voltage in the load cell are therefore avoided.

The load cell outputs a voltage difference in mV that is connected to the amplifier. The amplifier used in this experiment is the INA 122 [7]. To define the gain of the amplifier, a gain resistor R_G of 55.5Ω is chosen. Thus, the gain of the amplifier is $G = 5 + \frac{200k}{R_G} = 3608.6$. To power up the amplifier, a symmetrical tension source is used from a power supply: $\pm 15V$. This reduces the offset of the output voltage of the amplifier. Figure 2 shows a schematic representation of the electrical circuit built for this experiment.

Multiple samples were tested in which the initial length of the different samples as well as their width are different. Table I summarizes the different parameters used for each test. The setup is a tensile test with the load cell mounted on it. In order to measure the traction force the circuit is linked to an oscilloscope and a power supply as well as with the load cell. Figure 3 shows a schematic representation of the experiment setup.

Now that the experiment setup has been laid out, the results of the experiment are going to be developed. The output from the experiment is the displacement of the load cell and the voltage output from the load cell.

Test number \ Sample parameter	Width [mm]	Initial lenght [mm]	Thickness [mm]
1	12.31	2.86	0.58
2	12.80	2.73	0.42
3	12.61	1.87	0.46
4	5.1	1.87	0.34
5	12.7	1.90	0.46
6	5.5	1.90	0.38
7	5.55	1.91	0.33
8	12.4	1.91	0.44
9	5.4	1.91	0.35

TABLE I: Summary of the hydrogel parameters : sample width, thickness and initial length corresponding to the gap between the two grips at $t = 0$ that have been changed throughout the different tests.

A *MATLAB* script converts the voltage output into a force in Newton using the relation described in equation 1. One of the challenges faced was to synchronize the two outputs from the oscilloscope and the load cell as they were on different timelines. The first change in the voltage output was therefore considered as the start of the displacement of the load cell.

Since the purpose of the experiment is to extract the mechanical properties of the hydrogel. It is important to understand what the physical properties of the hydrogels are in order to choose the best model. This paper decided to use a Neo-Hookean model to fit the stress-stretch as the samples stretch in an elastic manner up to 8 times their original length without breaking [1]. The assumption that there is no side traction on the sample allows the model described by equation 2 to be implemented into *MATLAB*. Hydrogel being an incompressible material reduces the number of parameters to be found to one: the shear modulus μ [8].

$$\sigma = 2 * C_1 * (\epsilon - \frac{1}{\epsilon^2}) \tag{2}$$

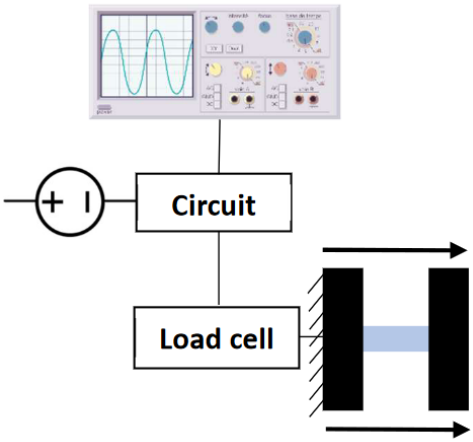

FIG. 3: Experiment setup with the load cell mounted on the tensile test. The hydrogel is tested in traction in the tensile test. The amplifying circuit is the link between the load cell, the oscilloscope, and the power supply. A computer collects the data from the displacement of the tensile test while the oscilloscope collects the data from the load cell through the amplifying circuit.

Test number	MAPE [%]	C_1	E [Pa]
1	13.54	645.6	3.8736e3
2	19.14	722.1	4.3326e3
3	20.77	487	2922
4	47.89	741.9	4.4514e3
5	1931	462.3	2.7738e3
6	65.37	362.5	2175
8	11.61	378	2268
9	37.66	77.81	466.86

TABLE II: Table summarizing the value for C_1 for the different samples tested according to a Neo-Hookean model as well as the Young's modulus computed using equations 2 and 3. The Mean absolute percentage error (MAPE) is shown for each fitting. It has been computed for the section of the curve exhibiting Neo-Hookean behavior. Samples 5 and 6 have really high error percentages that are explained later in this paper.

The factor $2 * C_1$ in equation 2 is the shear modulus μ of the material. The shear modulus is related to Young's modulus by equation 3 for an incompressible material. Young's modulus E can therefore be found by inverting equation 3. Table II summarizes the value of C_1 for the different samples based on the Neo-Hookean model implemented in *MATLAB* as well as the Young's modulus E derived from those values.

$$\mu = \frac{E}{2(1+\nu)} = \frac{E}{3} \tag{3}$$

Figures 4 and 5 shows the comparison between the fitting curve and the original data. One can observe that the fitting of the curve for samples 5 and 9 does not exactly match with the data extracted from the experiments. The possible explanations for this inconsistency are developed later in this paper. On the other hand, the model fits well for most of the samples. The Mean absolute percentage error has been computed for each sample fitting and are summarized in the first column of table II. This metric for error is not scale dependant and therefore allows this paper to compare the different datasets of the different samples. A MAPE greater than 10% but less than 25% indicates low, but acceptable accuracy and MAPE greater than 25% very low accuracy, so low that the forecast is not acceptable in terms of its accuracy [9]. The tensile test using sample 7 failed due to possible mishandling. Since these results were not relevant, they are not included in this analysis.

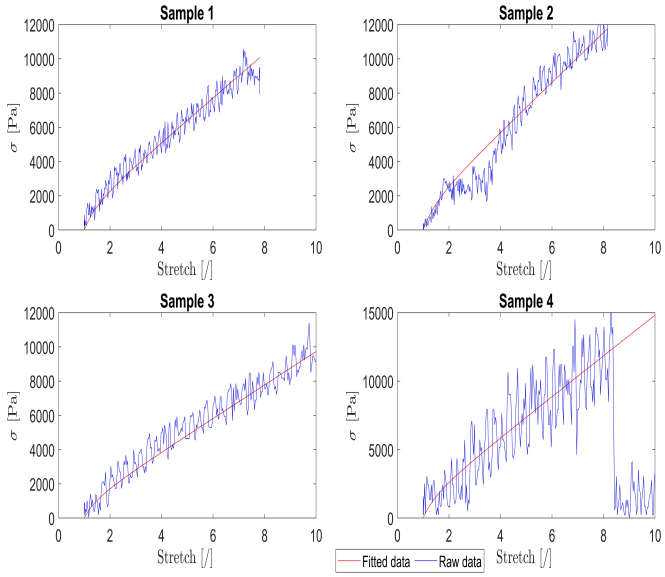

FIG. 4: Curve fitting of the stress-stretch curve for samples 1,2,3 and 4. In red is the result of the fitting and in blue is the original data. The fitting is based on a Neo-Hookean model. The model used matches with the data and allows to extract the shear modulus μ of the sample from the stress-stretch curve.

Out of the nine samples tested, only one fractured. The work of fracture W_f in $[J/m^3]$ can be defined by summing the area under the stress-stretch curve as described in equation 4. The stress σ is computed as the force over the initial cross-section area of the sample A_0 while the stretch ϵ is defined as the current length of the sample divided by the initial length.

$$W_f = \int_{\epsilon} \sigma(\epsilon)\, d\epsilon \tag{4}$$

Figure 6 shows the stretch stress curve for the sample that broke. Using *MATLAB*, the work of fracture was found to be equal to $W_f = 5.54e+04\ J/m^3$.

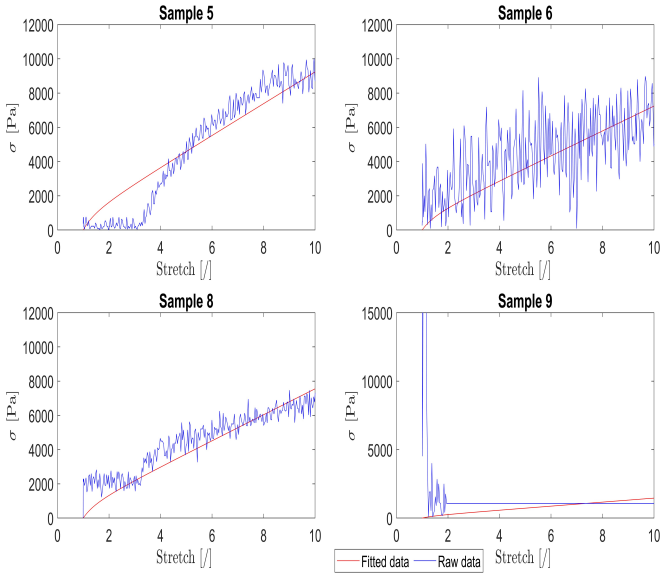

FIG. 5: Curve fitting of the stress-stretch curve for samples 5,6,8 and 9. In red is the result of the fitting and in blue is the original data. The fitting is based on a Neo-Hookean model. The model used does not match all the data from the different samples. The shear modulus μ extracted from those graphs is therefore to be taken cautiously.

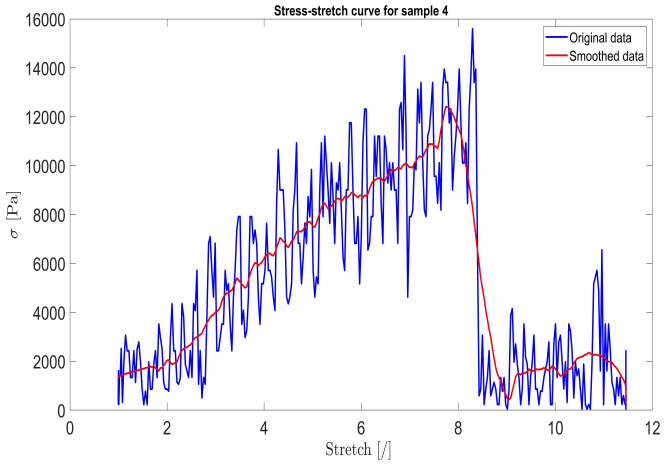

FIG. 6: Stress - stretch curve for sample four. The red curve is the smoothed data for a better computation of the work of fracture. The Fracture happened for a stretch of around 8 times the original displacement and is characterized by a drop in the stress curve. The work of fracture is computed as the area under the blue curve between the start of the stretch to the failure of the sample.

An analysis of the predicted theoretical results will allow to observe if the results obtained from the experiments differ from the theory and why this might be the case. The shear modulus equation from the theory of entropic elasticity gives :

$$\mu = \phi^{\frac{1}{3}} * N * k * T \tag{5}$$

Where ϕ represents the volume fraction of polymer which in this case is equal to 13.7%, $k * T$ is the temperature in the unit of energy, N is the polymer segments' number divided by dry polymer's volume.

$$N = \frac{2 * C}{V} \tag{6}$$

The volume of monomer unit is the fraction of the mass per monomer per the density per monomer unit which equals $V = 1.04 * 10^{-28} m^3$. C as mentioned earlier has a value of 0.153%. Considering $k = 1.38 * 10^{-23} J/K$ and a room temperature of $T = 300K$ the equation 5 gives :

$$\mu = 62.8\ kPA \tag{7}$$

By obtaining a value for the shear modulus we estimate the theoretical value of the elastic modulus using equation 8.

$$E = 3 * \mu = 3 * \phi^{\frac{1}{3}} * N * k * T = 188.4\ kPA \tag{8}$$

This estimated Young's modulus's value E is much larger than the measured value for any samples. Indeed there is approximately a two orders of magnitude's difference. These types of disparities have been observed in [1]. Indeed, this study has shown a large difference between theoretical and experimental values for both work of fracture and extensibility. The measured values were both smaller than the theoretical and such discrepancies could be explained in part by network imperfections, such as chains of different monomer's number, thus disproportionate chain lengths. According to [10], the modulus should be close between theoretical predictions and experimental measurements. It is important to note that theoretical estimation make the assumption that all polymer chains in the network are carrying load and thus contribute to the shear stress. However, not all polymer chains in a real network are carrying stress. There are only the cross-linked polymer chains that play a part in the shear modulus. This could explain why the estimated modulus is higher than the measured modulus.

Multiple problems have arisen during the experiment that have impacted the results presented above. The following section will discuss them with the goal of obtaining more accurate data in future experiments. First, the major problem encountered was in the gripping of the samples. Indeed, samples were fastened at both sides of the tensile test using 3D-printed magnetic grip but when the load was applied, the samples slipped. Because this issue had a negative impact on the measurements, multiple solutions were tried. Binder clips were added at each attachment of the sample to minimize the slip of the samples. In addition, the hydrogel samples are sitting in an aqueous solution before being taken out to be measured and tested. This adds to the slip factor of the samples and they were therefore dabbed with a tissue before being put in the grips. However, none of those solutions worked as the samples continued to slip.

It is important to note that the tests were made with the samples having a low cross-linker-to-monomer molar ratio C of 0.153% which gives a polymer network of dense entanglements and long polymer segments. That could explain why the samples did not break because dense entanglement leads to high young's modulus and toughness. Tests could be made with different ranges of C's value to observe potential fluctuation of the modulus with the changes in C. The value of the water-to-monomer molar ratio W also impacts the entanglement's density and thus the modulus. A large W ratio will give scattered monomers in the precursor. These monomers then create a scattered entangled polymer network that makes the hydrogel capable of imbibing a large water quantity and thus swells more. This kind of network should lead to a smaller stiffness because of the low entanglement number. After drawing these observations, the samples tested could potentially have a large W ratio and that would be the reason why the slipping of the samples increased. Nevertheless, the literature is not very clear about what is a large ratio, in which order or magnitude, and according to [1], it is more frequent to operate this kind of experience with large W ratio. For the samples used in this experiment the monomer weight percentage content of the hydrogel was 13.7%. In order to compute the water-to-monomer molar ratio W a mass of $m_t = 100g$ of total mixture is assumed. The molar mass of water is $M_w = 18\ g/mol$ and that of hydrogel is $M_h = 71.08\ g/mol$. Masses of monomer and water are respectively $m_m = 13.7g$ and $m_w = 86.3g$.

$$
\begin{aligned}
m_m &= m_t * \phi = 100 * 0.137 = 13.7g \\
m_w &= m_t - m_m = 100 - 13.7 = 86.3g \\
Moles_w &= \frac{m_w}{M_w} = \frac{86.3}{18} = 4.79\ mol \\
Moles_m &= \frac{m_m}{M_h} = \frac{13.7}{71.08} = 0.193\ mol \\
W &= \frac{Moles_w}{Moles_m} = \frac{4.79}{0.139} = 34.46
\end{aligned}
$$

The computations leads to an approximate water-to-monomer molar ratio of $W = 34.46$.

Experiments and tests are also made under the assumption that the hydrogels are in an equilibrium state by being submerged in water. However for the measurement of each sample, hydrogels were placed between two plates for measurements and stayed out of their aqueous environment for a couple of minutes. The disruption of the equilibrium state may change hydrogel's mechanical properties. Indeed, as the hydrogel deflates when dehydrating the hydrogel becomes stiffer because of its modulus is strongly influenced by water content. In order to counteract this problem the samples were put back in the water before making the test. In fact the mechanical properties changes above-mentioned are often reversible and thus hydrogel can swell and deflate in reaction to alterations in their surroundings. However, by putting them back in the water the problem of slipping arose again. To overcome it another gripping setup should be designed which would reduce slippage. To improve the gripping method specialized grips with serrated or textured surfaces could be used to offer an improved grip on the hydrogel. Implementing soft jaws on the designed grips may help to distribute the load uniformly and thus avoid slippage.One other easier improvement could be to change the layer of the non-slip material on the gripping surfaces with another non-slip material to see if friction between the grip and the sample is larger.

The rate of stretch could also be studied to see if it plays a crucial role in the results but a paper has shown that the stress-stretch curves were very similar in a range upon 0.05/s to 1/s [10]. Nevertheless, it is relevant to note that hydrogel usually manifests rate-dependent mechanical behavior. Indeed, deformation properties such as stiffness may change based on the strain rate. The hydrogel may get stiffer when stretched rapidly and softer when stretched slowly. Hydrogel are considered viscoelastic materials thus it often exhibits both elastic and viscous responses to deformation. The strain rate could affect the proportion between these two. Increasing the strain rate may lead to enhanced viscous behavior thus getting higher energy dissipation and less elastic recovery. On the contrary slower strain rate accentuates elastic response.

Considering an ideal polymer network, the work of fracture is computed using the covalent energy density, eV, which is approximately $1e-19\ J$, divided by the volume allocated per monomer, V, which is roughly $1e-28m^3$. Consequently, the work of fracture for a perfect network is around $1e9\ J/m^3$, representing a difference of five orders of magnitude with our experimental results. This significant discrepancy is in line with the disparities observed in the modulus.

The difference observed between theoretical and experimental results is too significant to not take into account possible experimental errors discussed above. Regarding the work of fracture obtained from this experiment, nothing can be concluded as only one of the sample broke and therefore the value obtained could be an outlier as no repeatability of the results could be proven.

In conclusion, the work of fracture obtained is $W_f = 5.54e4\ J/m^3$. However, since other mechanical properties of the hydrogel such as the Young's modulus are not coherent with theoretical analysis and only one sample was capable of producing this result, it cannot be accepted as the answer to the initial question. This experiment should be repeated, taking into account the improvements described above.

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