Electronic Supplementary Material (ESI) for Soft Matter.

Triple crosslinking conductive hydrogels with digitally printable and outstanding mechanical stability for high-resolution conformable bioelectronics

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Figure S1. Schematic representation of the hydrogel synthesis process.

Figure S2. The synthesis process of the hydrogel via ultra-power probe sonication results in the hydrogel formation within a few seconds, (a) the synthesis process and schematic representation, and (b) unsuccessful molding.

Figure S3: The particle size distribution of the Ga-dispersed in glycerol solution obtained by ultra-power probe sonication and long-term ultrasound routes in the water bath.

Figure S4. The IR spectra of the samples.

Figure S5. SEM images of the laponite-involved samples accompanied by their mapping and EDX spectra.

Figure S6. Maximum strain at break (%) for different batches of the hydrogels obtained from the stress-strain curves. The deviation is obtained by repeating each test 5 times for each sample.

Figure S7. Electromechanical behaviour of the samples, (a) resistance (R) versus time for 100% strain and 100 cycles for the 0.1g-involved laponite sample, (b) Resistance versus strain for the laponite-involved samples, and (c) resistance versus cycle number for the same sample.

Table S1: Some extra experimental works and their corresponding results.

Table S2: The IR spectra characteristics of the as-prepared hydrogels at different conditions.

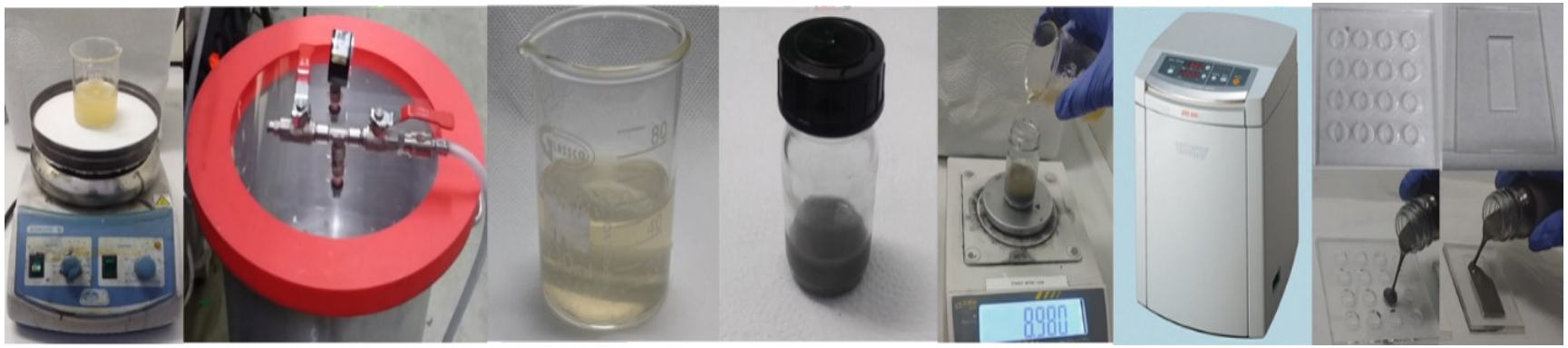


Figure S1. Schematic representation of the hydrogel synthesis process. The AAm was dissolved in water and BIS and other components were added to prepare one part. Then, it was degassed in a vacuum chamber, and a transparent solution was obtained. Then, it was mixed with Ga-dispersed in glycerol solution and mixed at 2000 rpm for 3 min, and cased in the desired mold.

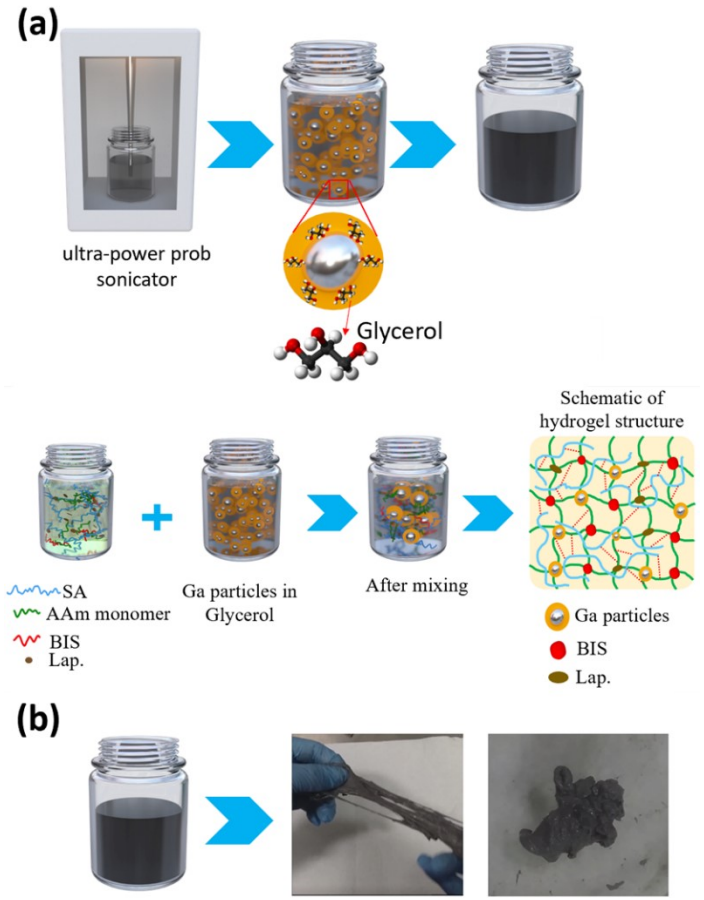


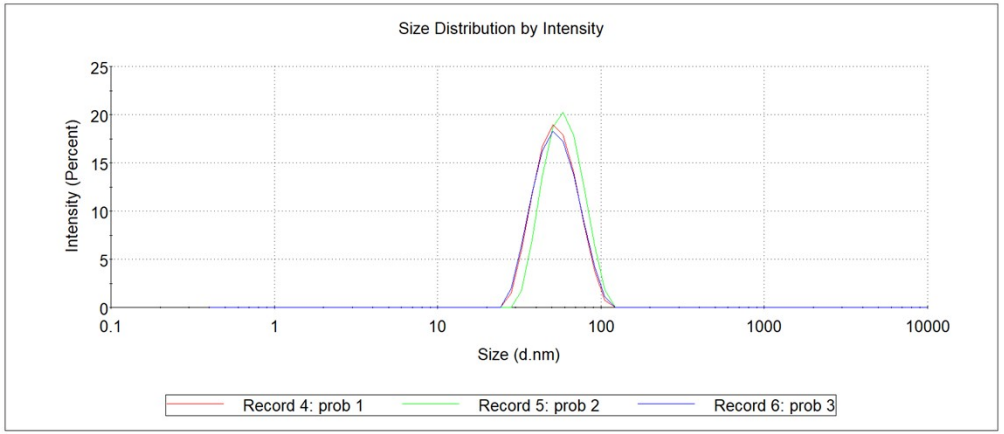
Figure S2. The synthesis process of the hydrogel via ultra-power probe sonication results in the hydrogel formation within a few seconds, (a) the synthesis process and schematic representation, and (b) unsuccessful molding.

Sample Name:	prob 1		
SOP Name:	mansettings.nano		
File Name:	particles.dts	**Dispersant Name:**	glycerol
Record Number:	4	**Dispersant RI:**	1.475
Material RI:	1.59	**Viscosity (cP):**	1412.0000
Material Absorbtion:	0.010	**Measurement Date and Time:**	6 de setembro de 2022 14:41:39

Temperature (°C):	24.9	**Duration Used (s):**	80
Count Rate (kcps):	183.3	**Measurement Position (mm):**	0.45
Cell Description:	Disposable sizing cuvette	**Attenuator:**	1

			Size (d.nm):	**% Intensity:**	**St Dev (d.nm):**
Z-Average (d.nm):	67.36	**Peak 1:**	54.95	100.0	15.65
PdI:	1.000	**Peak 2:**	0.000	0.0	0.000
Intercept:	0.324	**Peak 3:**	0.000	0.0	0.000

Result quality : **Refer to quality report**



Sample Name:	bath 1		
SOP Name:	mansettings.nano		
File Name:	particles.dts	**Dispersant Name:**	glycerol
Record Number:	1	**Dispersant RI:**	1.475
Material RI:	1.59	**Viscosity (cP):**	1412.0000
Material Absorbtion:	0.010	**Measurement Date and Time:**	6 de setembro de 2022 14:27:21

Temperature (°C):	25.0	**Duration Used (s):**	60
Count Rate (kcps):	496.0	**Measurement Position (mm):**	0.85
Cell Description:	Glass cuvette with round aperture	**Attenuator:**	2

			Size (d.nm):	**% Intensity:**	**St Dev (d.nm):**
Z-Average (d.nm):	172.3	**Peak 1:**	74.79	100.0	15.10
PdI:	1.000	**Peak 2:**	0.000	0.0	0.000
Intercept:	0.507	**Peak 3:**	0.000	0.0	0.000

Result quality : **Refer to quality report**

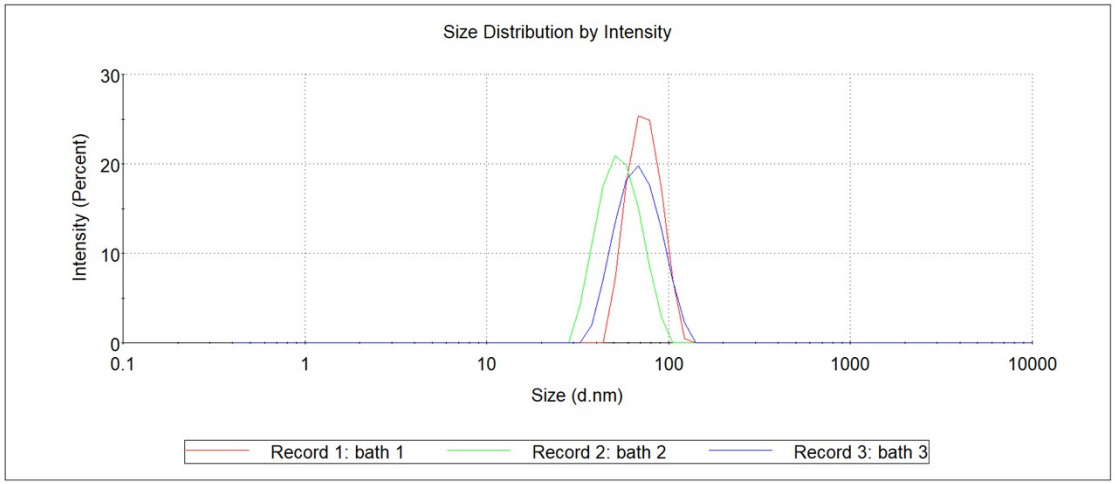
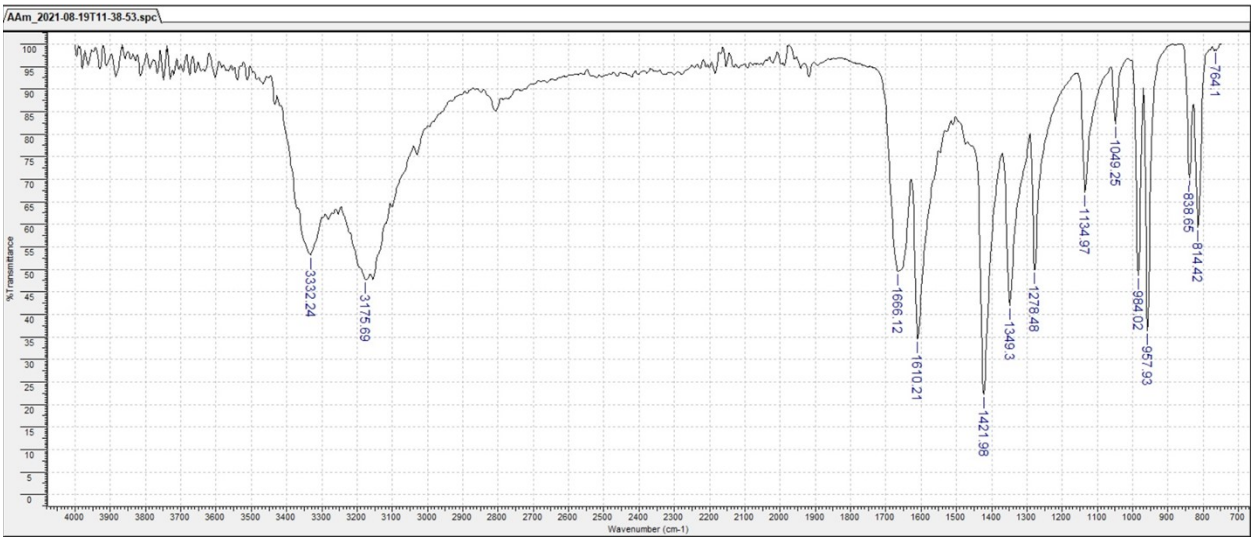
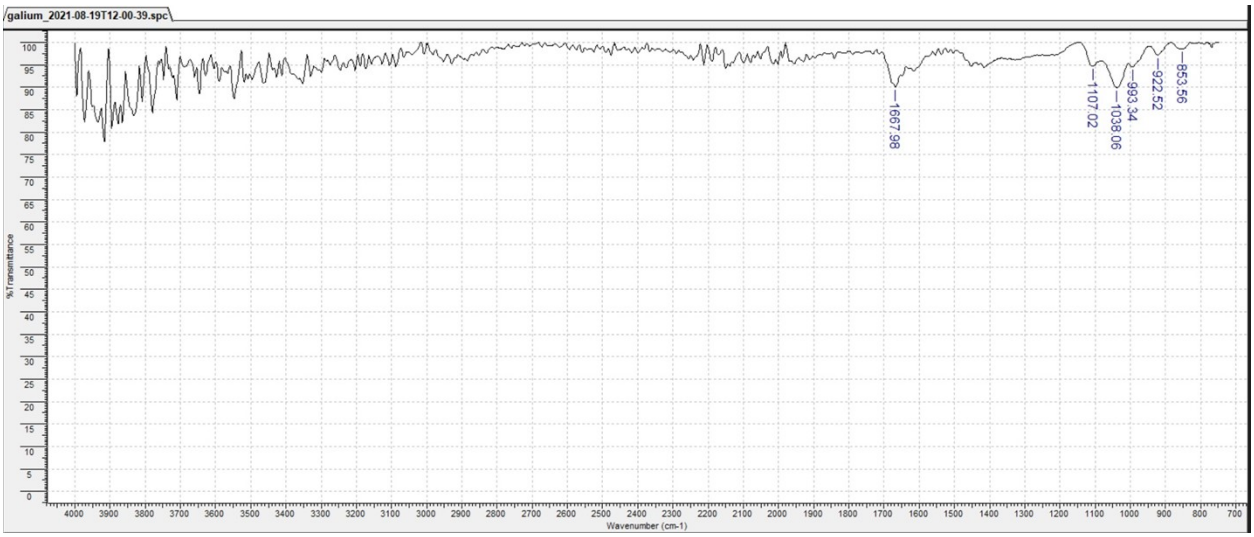


Figure S3: The particle size distribution of the Ga-dispersed in glycerol solution obtained by ultra-power probe sonication and long-term ultrasound routes in the water bath.

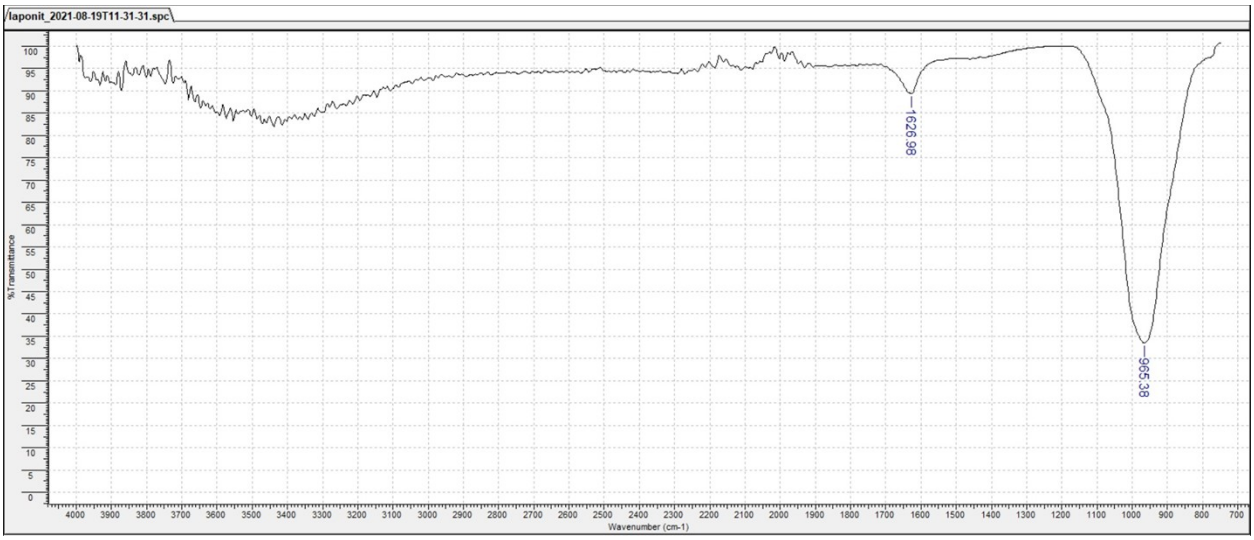
AAm



Ga



Laponite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%Transmittance
Wavenumber (cm-1)
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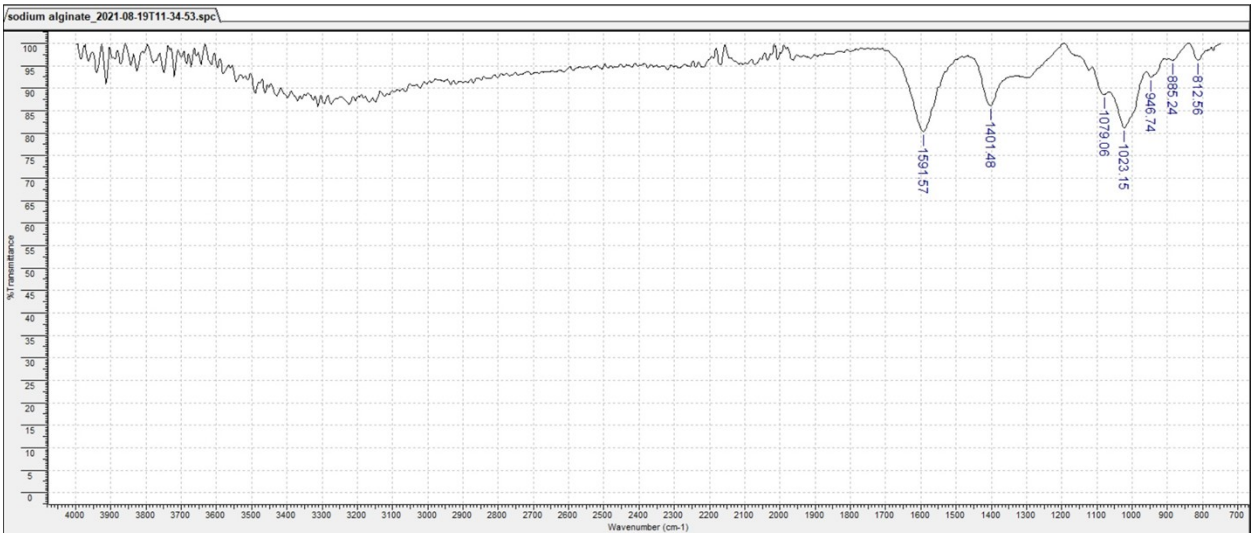
965.38

SA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Wavenumber (cm-1)
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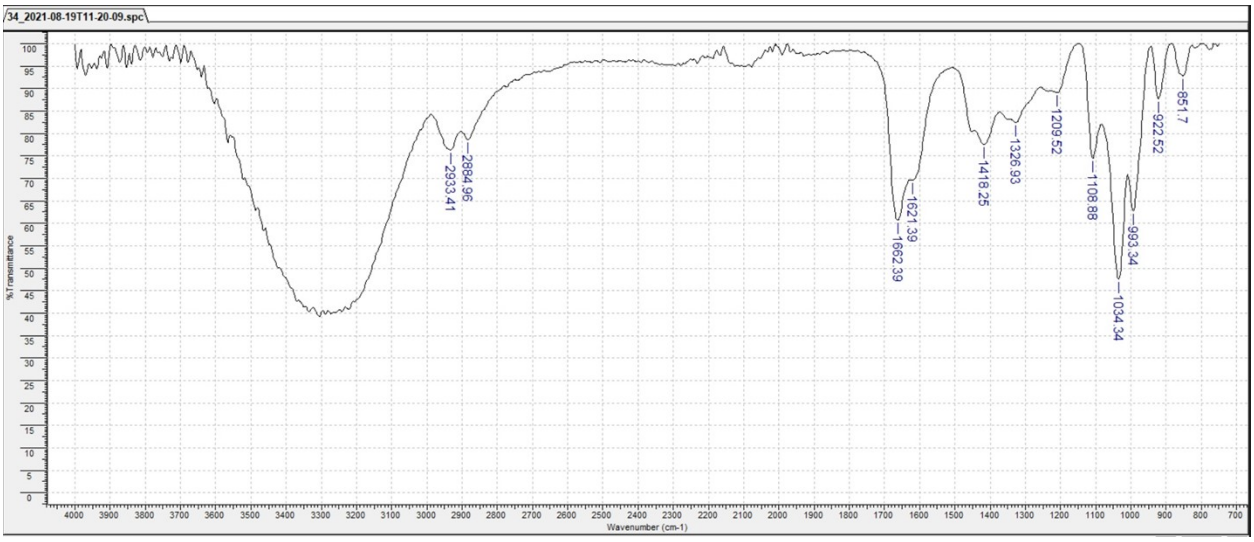
1023.15
946.74
885.24
812.56

0.2 g Ga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%Transmittance
Wavenumber (cm-1)
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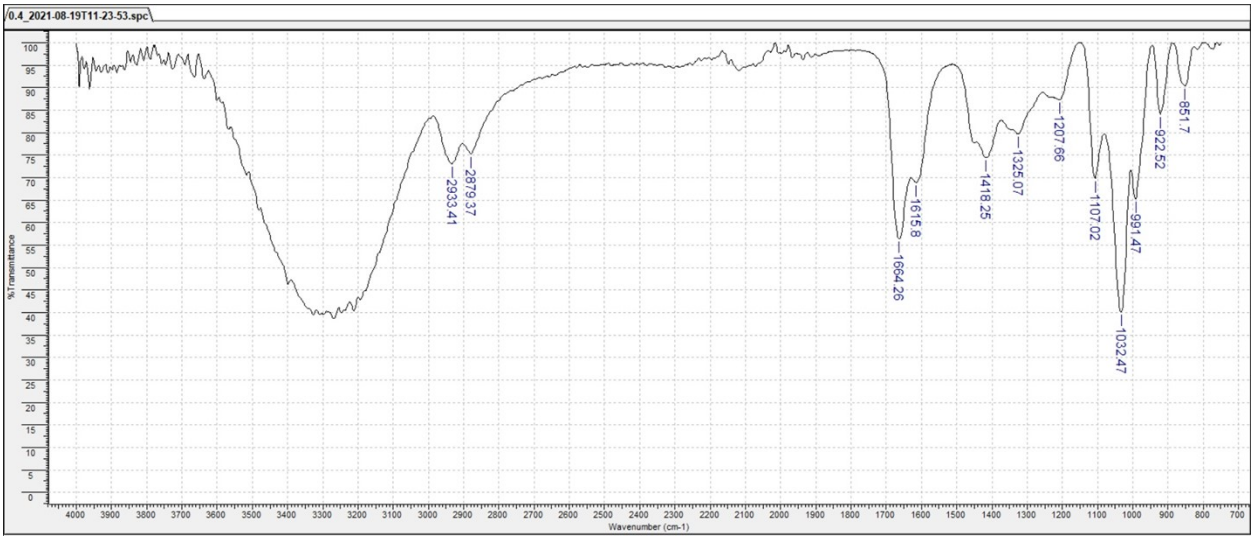
2884.96
1662.39
1621.39
1418.25
1326.93
1209.52
1108.88
1034.34
993.34
922.52
851.7

0.4 g Ga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1325.07
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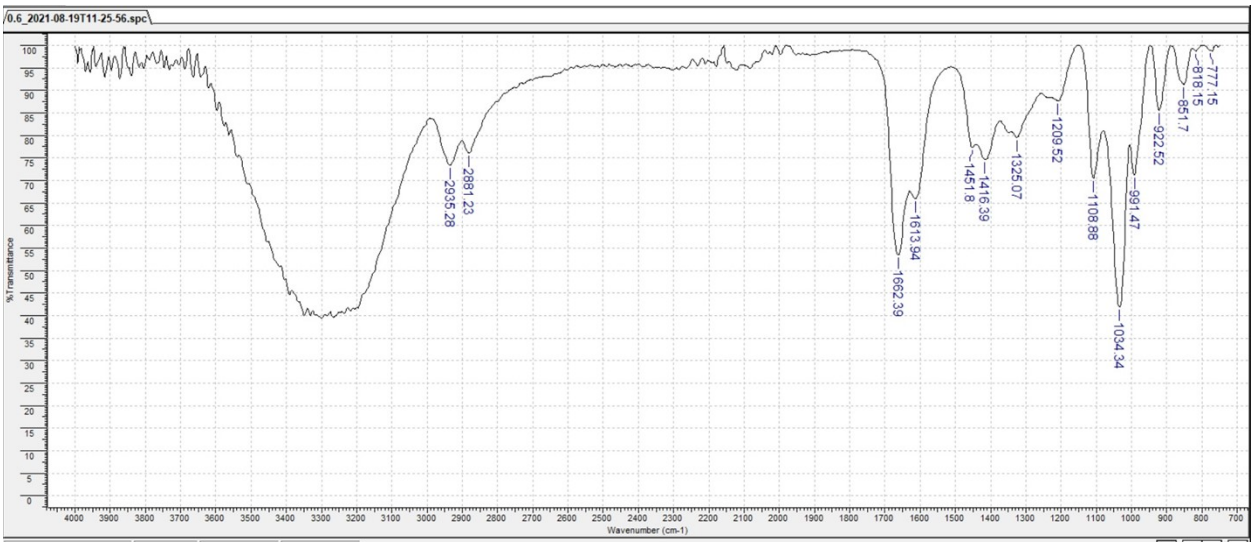
1107.02
1032.47
991.47
922.52
851.7

0.6 g Ga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1451.8
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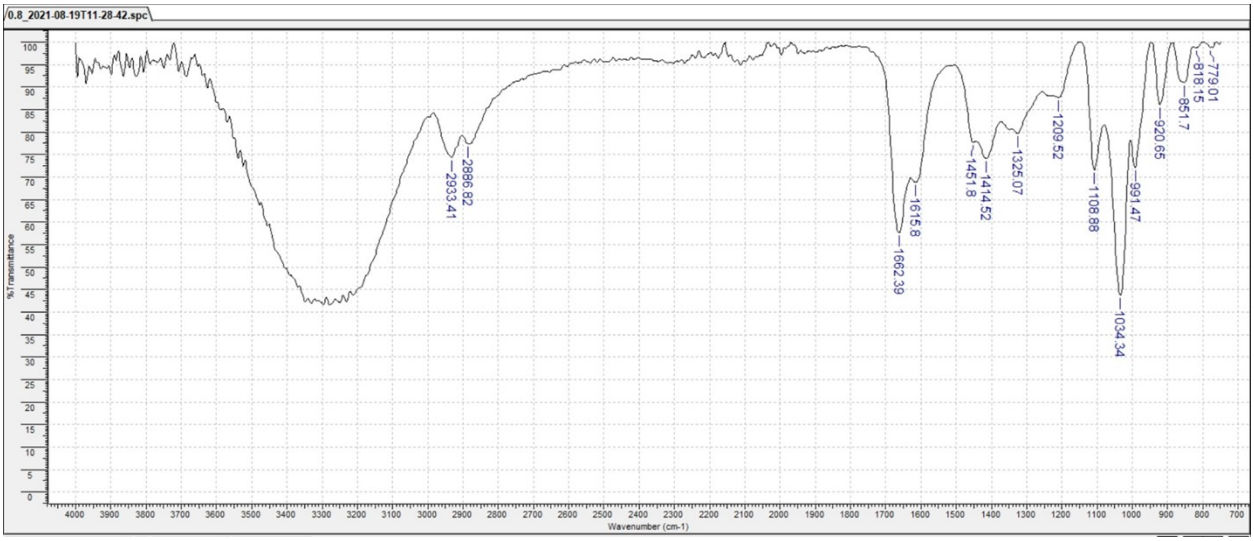
1108.88
1034.34
991.47
922.52
851.7
818.15
777.15

0.8 g Ga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%Transmittance
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1662.39
1615.8
1451.8
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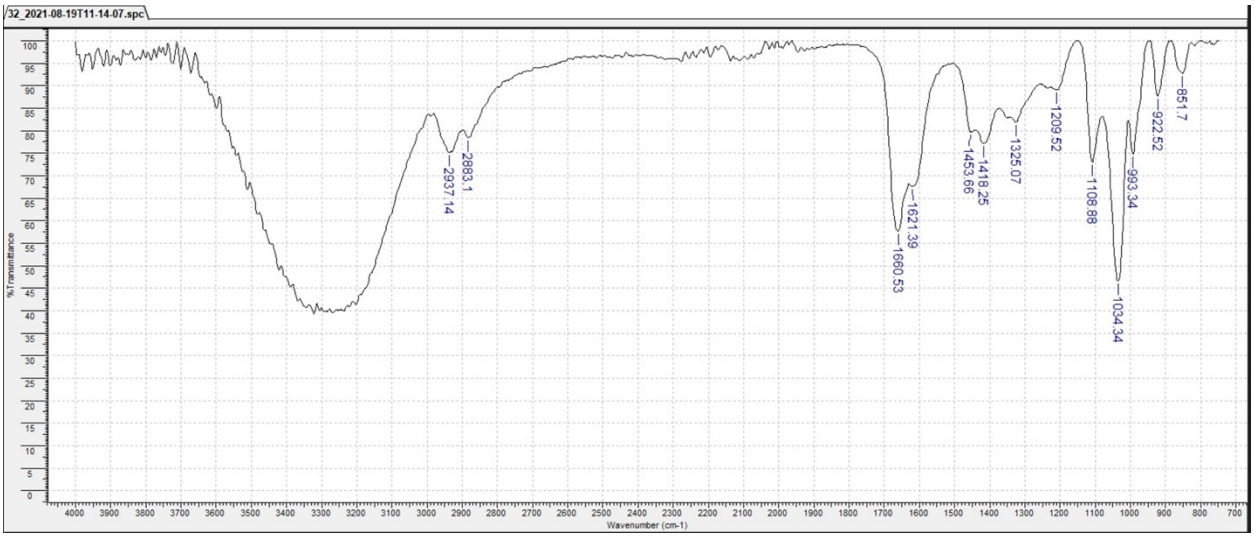
1209.52
1108.88
1034.34
991.47
920.65
851.7
818.15
779.01

1 g Ga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1325.07
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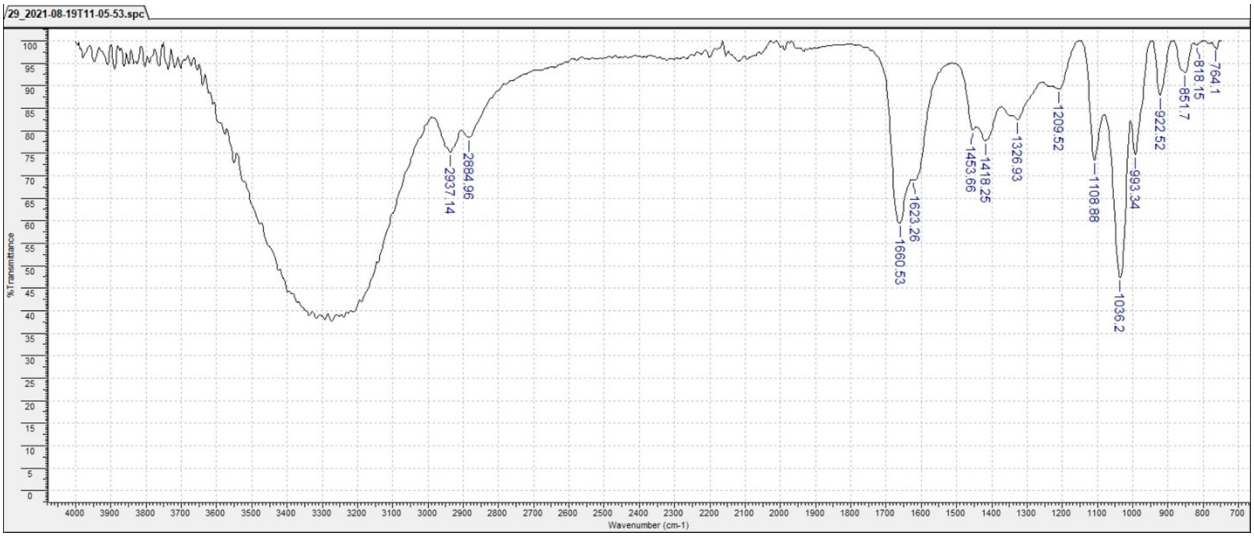
1108.88
1034.34
993.34
922.52
851.7

0.1 La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%Transmittance
Wavenumber (cm-1)
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1453.66
1418.25
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1108.88
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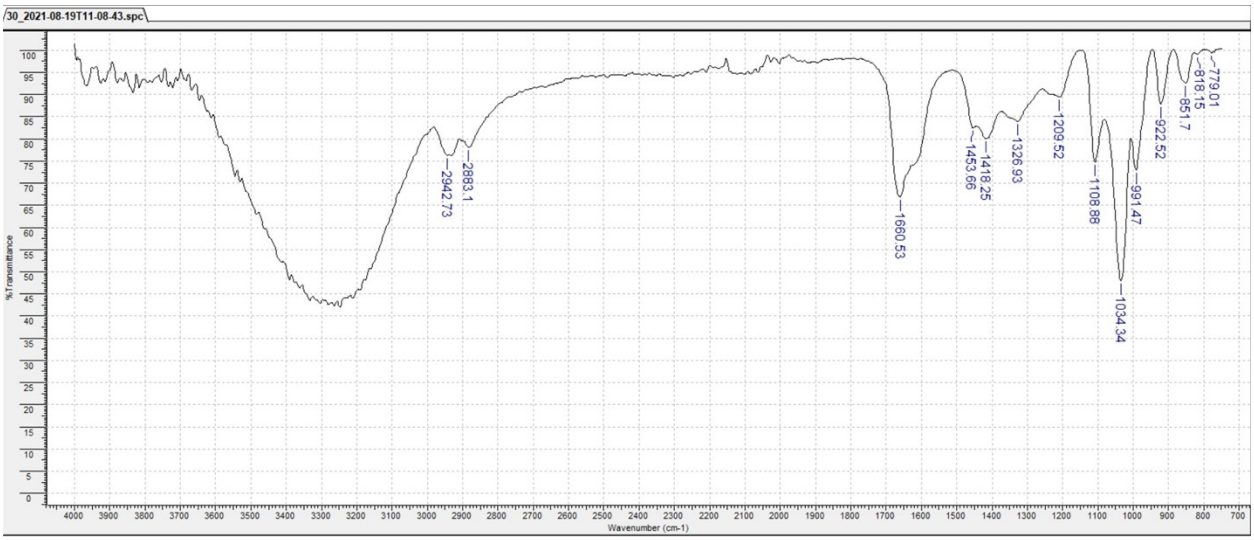
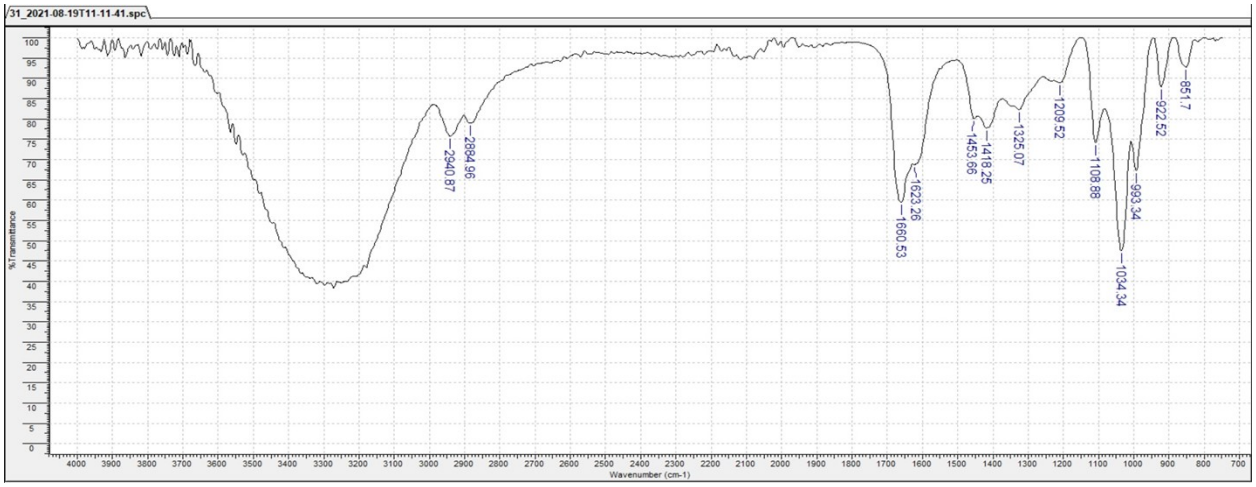
993.34
922.52
851.7
818.15
764.1

0.2 La



0.3 g La



0.4 g La

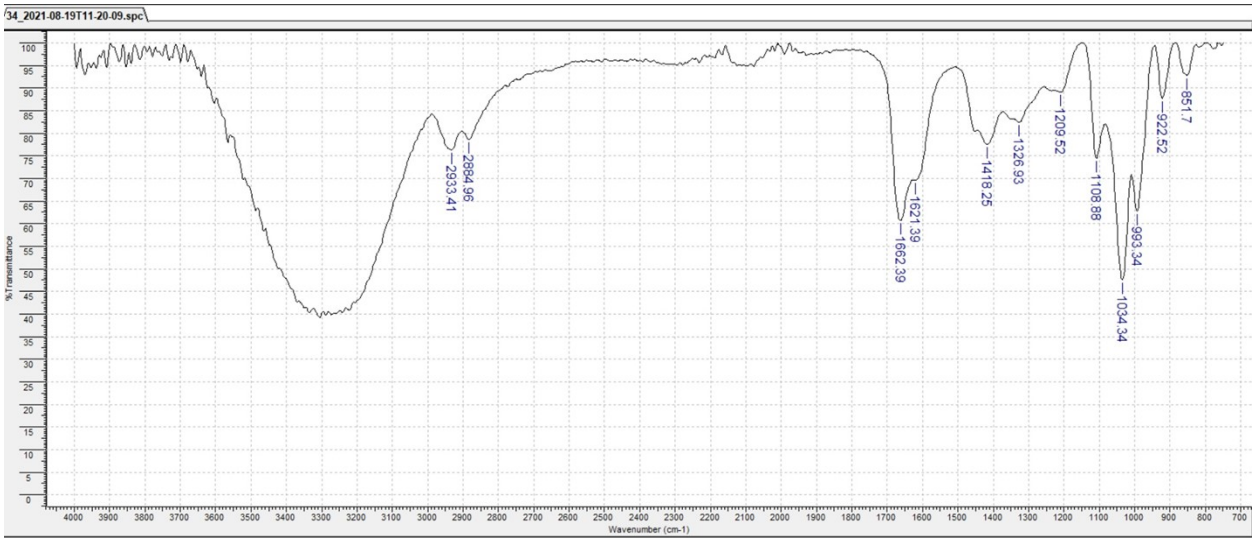
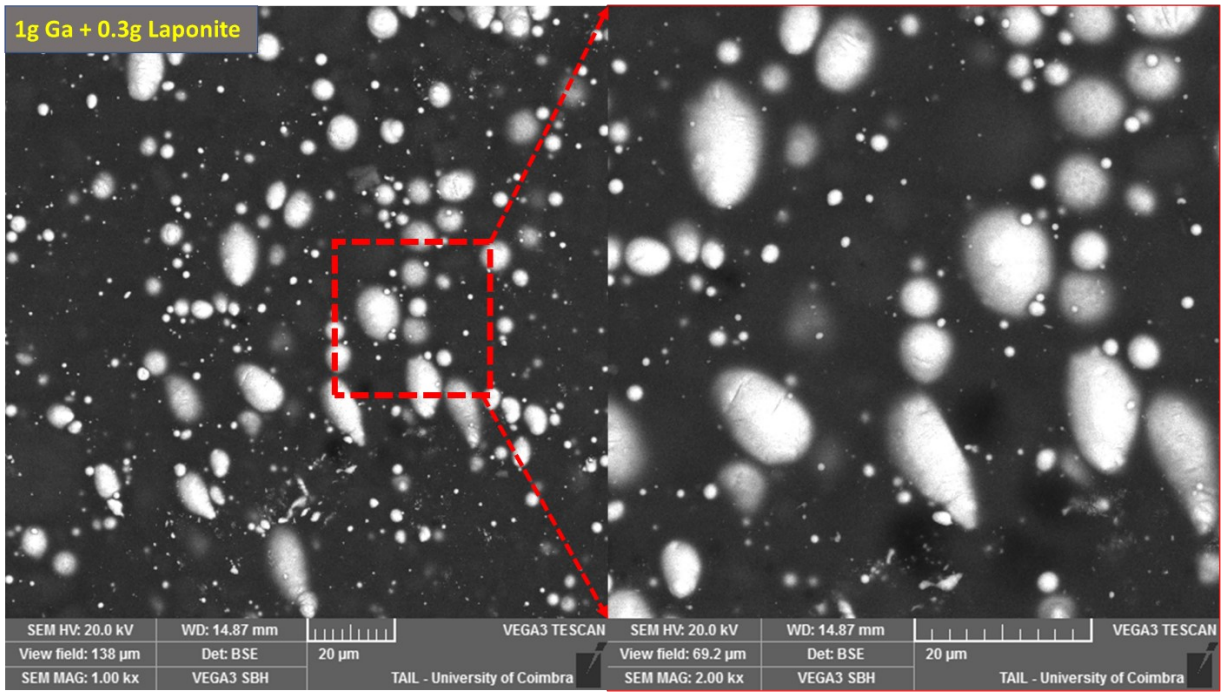
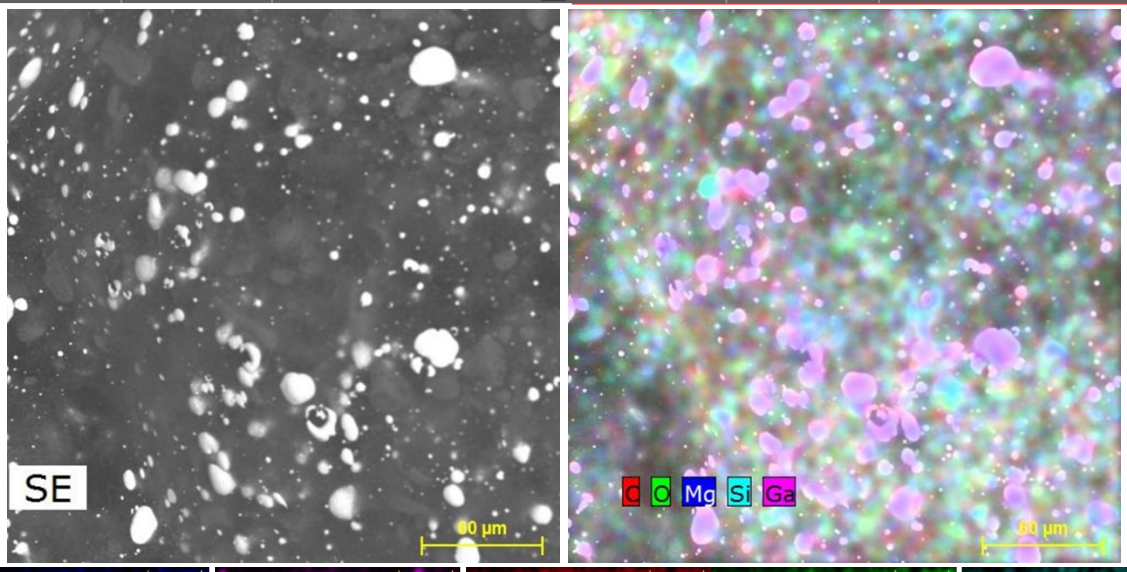
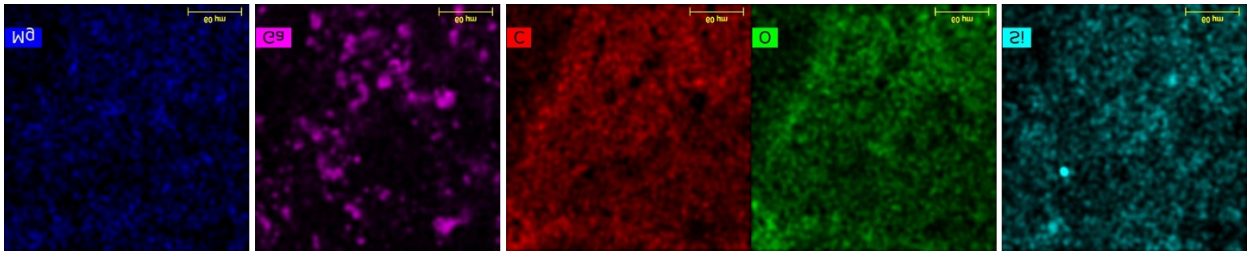
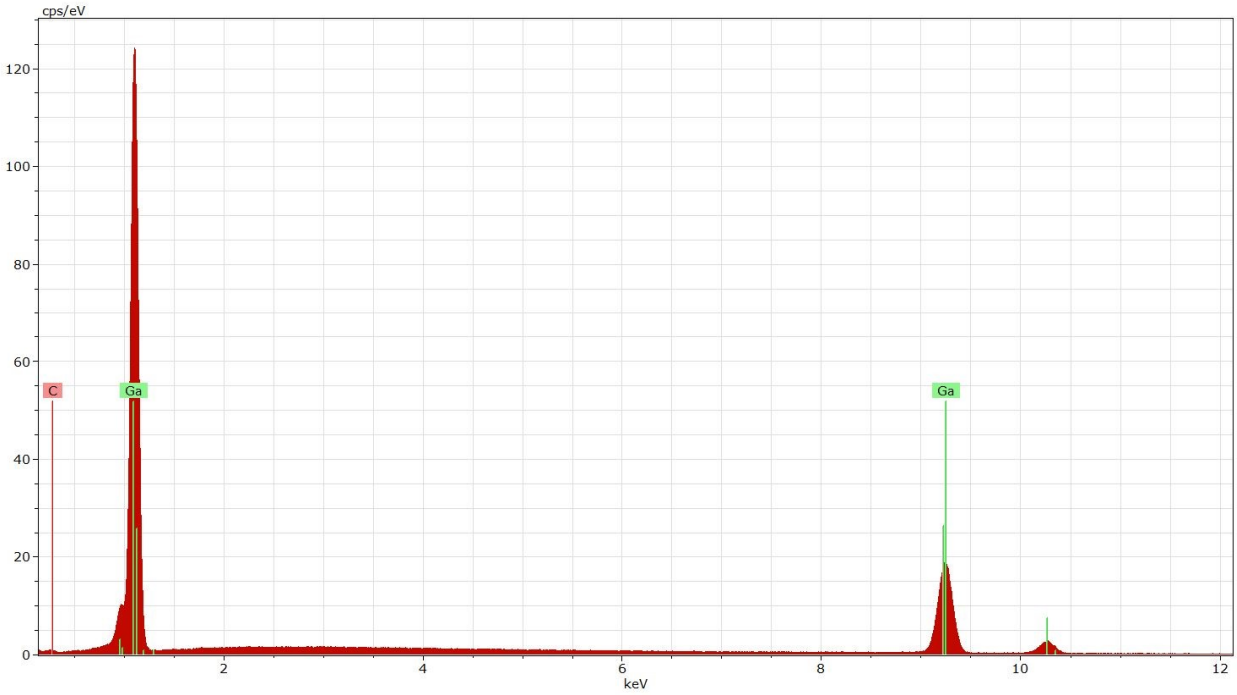


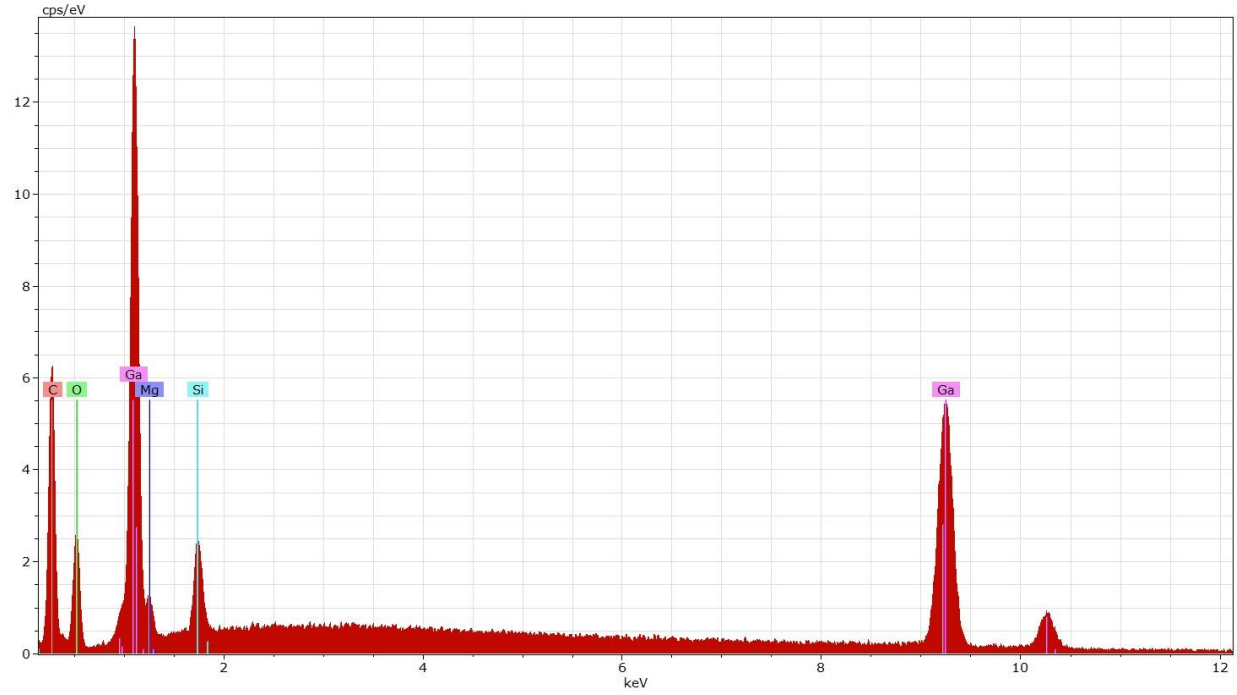
Figure S4. The IR spectra of the samples.


1g Ga + 0.3g Laponite
SEM HV: 20.0 kV
WD: 14.87 mm
VEGA3 TESCAN
View field: 138 µm
Det: BSE
20 µm
SEM MAG: 1.00 kx
VEGA3 SBH
TAIL - University of Coimbra
SEM HV: 20.0 kV
WD: 14.87 mm
VEGA3 TESCAN
View field: 69.2 µm
Det: BSE
20 µm
SEM MAG: 2.00 kx
VEGA3 SBH
TAIL - University of Coimbra


SE
60 µm
C O Mg Si Ga
60 µm




cps/eV
120
100
80
60
40
20
0
C
Ga
Ga
2
4
6
8
10
12
keV


cps/eV
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10
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6
4
2
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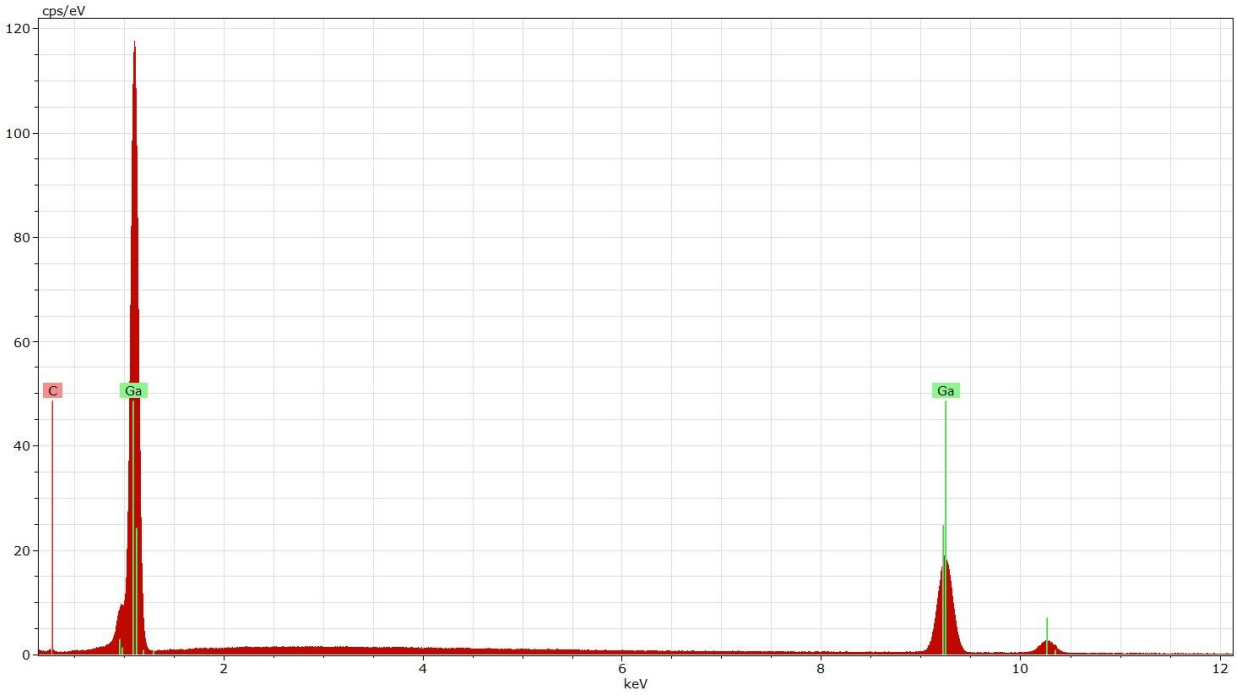
C
O
Ga
Mg
Si
Ga
2
4
6
8
10
12
keV


cps/eV
120
100
80
60
40
20
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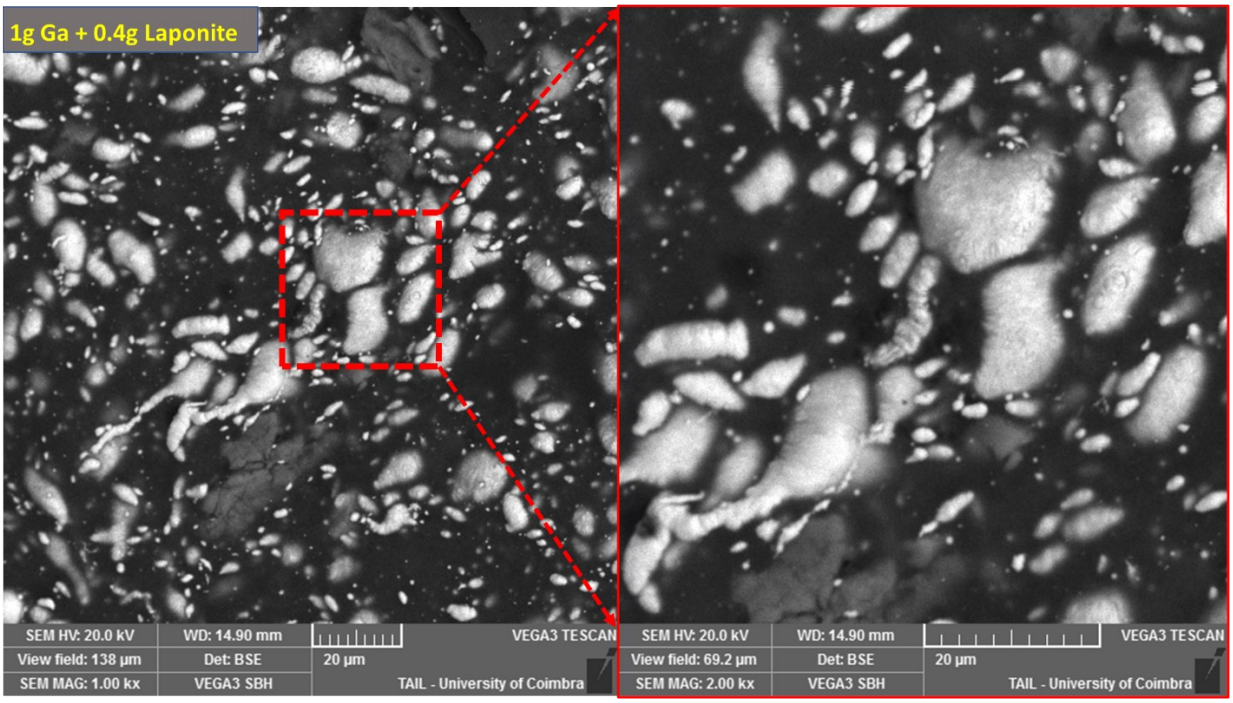
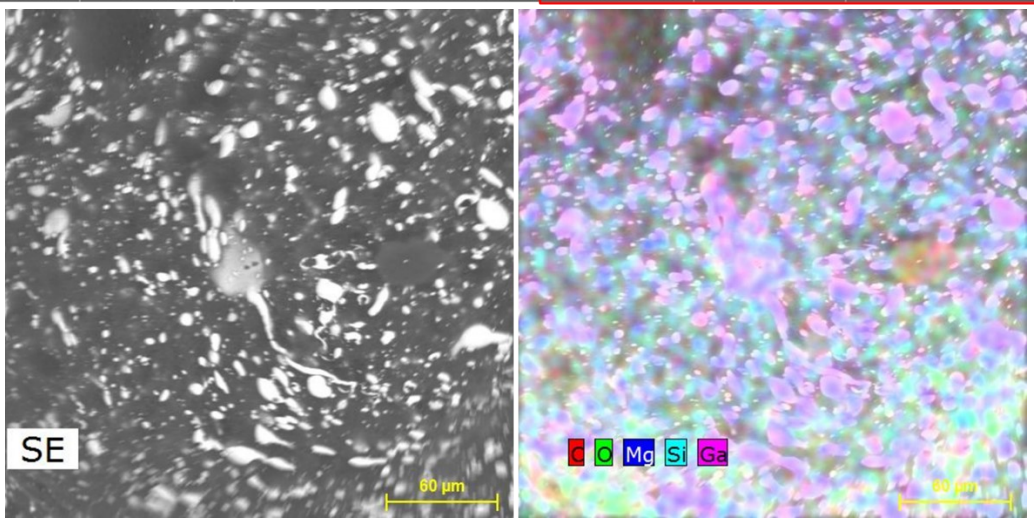
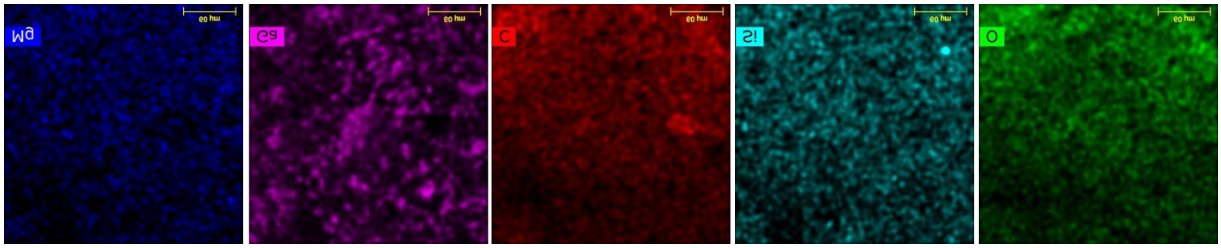
C
Ga
Ga
2
4
6
8
10
12
keV


1g Ga + 0.4g Laponite
SEM HV: 20.0 kV
WD: 14.90 mm
VEGA3 TESCAN
View field: 138 µm
Det: BSE
20 µm
SEM MAG: 1.00 kx
VEGA3 SBH
TAIL - University of Coimbra
SEM HV: 20.0 kV
WD: 14.90 mm
VEGA3 TESCAN
View field: 69.2 µm
Det: BSE
20 µm
SEM MAG: 2.00 kx
VEGA3 SBH
TAIL - University of Coimbra


SE
60 µm
C O Mg Si Ga
60 µm


Mg
Ga
C
Si
O

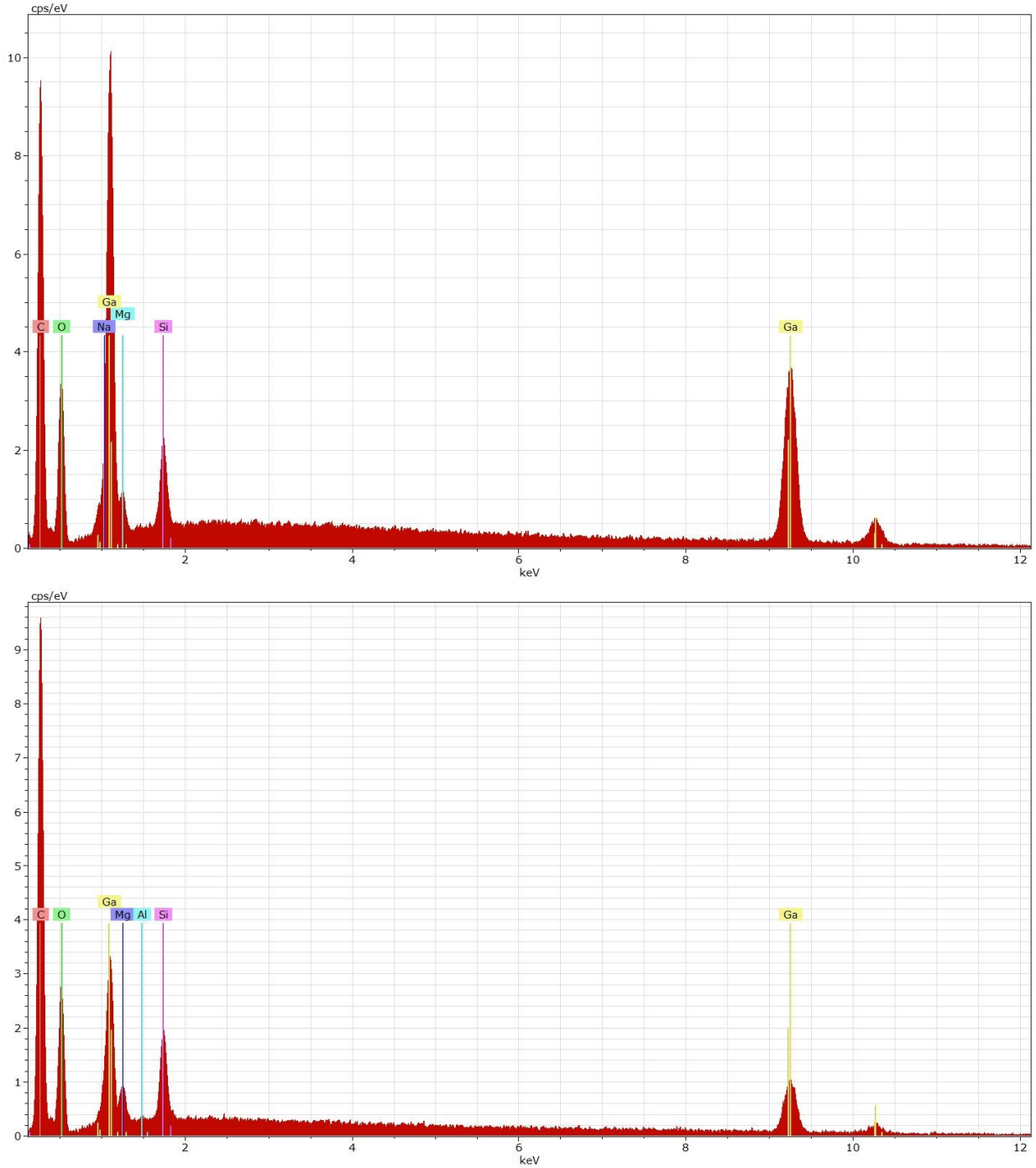


Figure S5. SEM images of the laponite-involved samples accompanied by their mapping and EDX spectra.

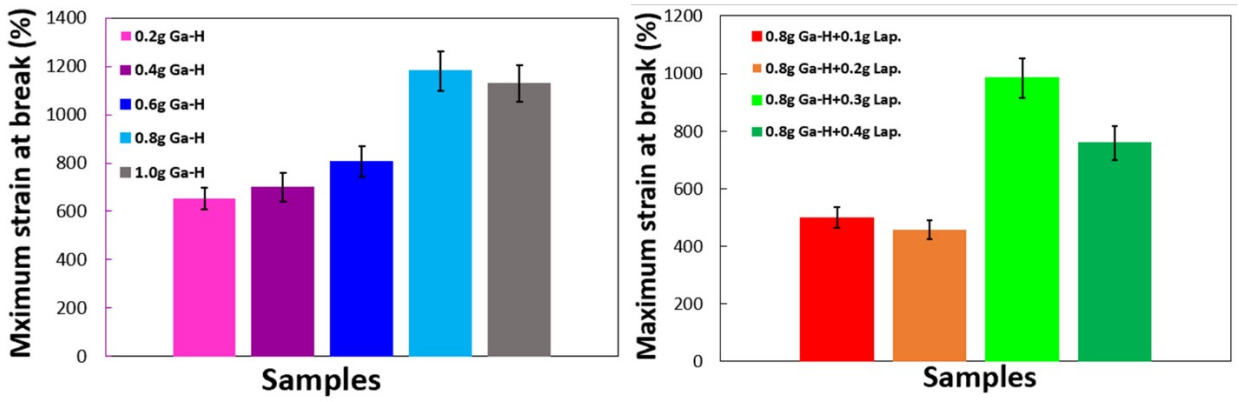


Figure S6. Maximum strain at break (%) for different batches of the hydrogels obtained from the stress-strain curves. The deviation is obtained by repeating each test 5 times for each sample.

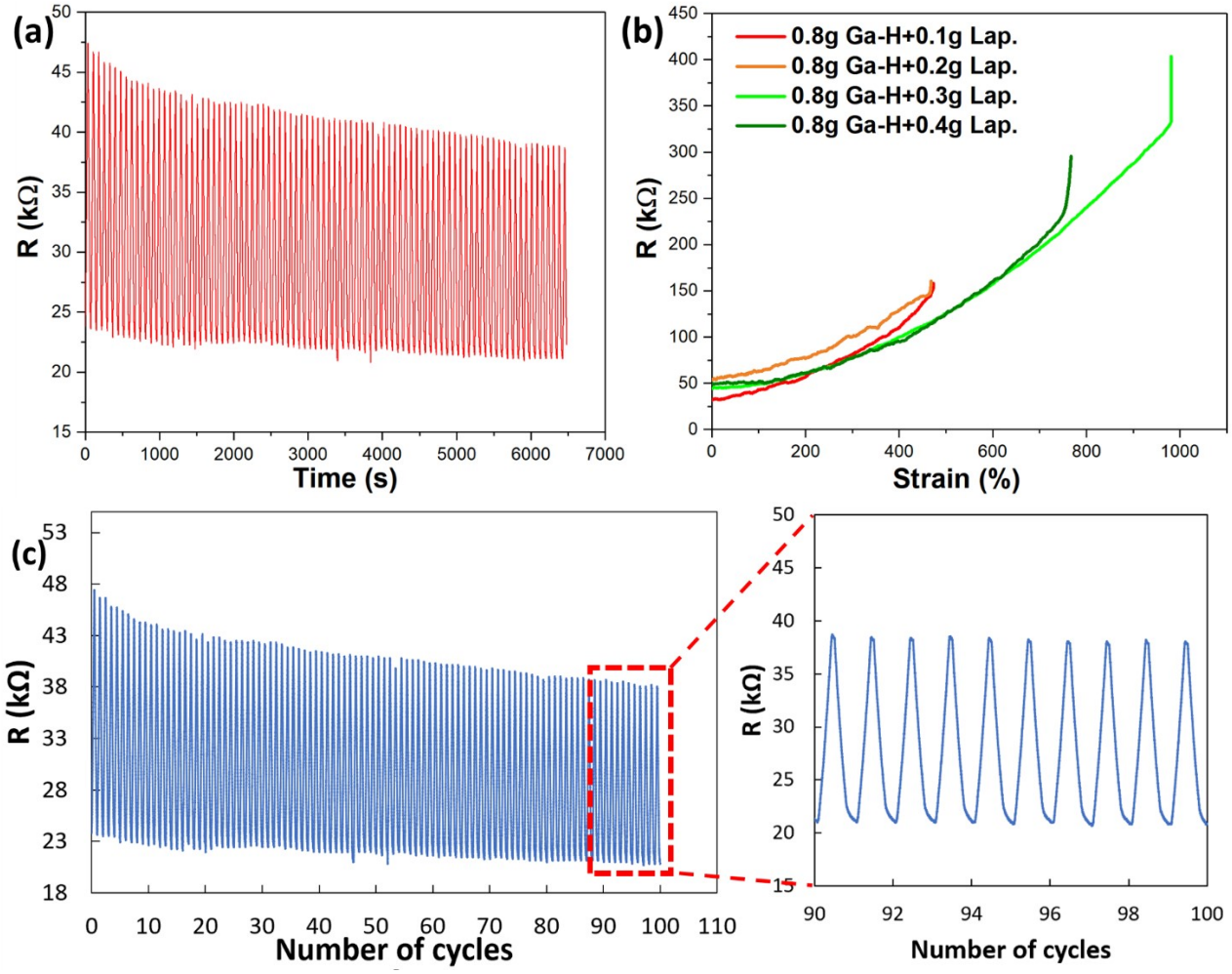


Figure S7. Electromechanical behaviour of the samples, (a) resistance (R) versus time for 100% strain and 100 cycles for the 0.1g-involved laponite sample, (b) Resistance versus strain for the laponite-involved samples, and (c) resistance versus cycle number for the same sample.

N	Part A					Part B			Mixing							
	H2O (g)	Alginate (g)	Acrylamide (g)	BIS (g)	Inhibitor (g)	**H2O (g)**	**Glycerol (g)**	**Ga (g)**	**Part A (g)**	**Part B (g)**	**Mix Part A and B**	**Dark**	**Degassing Part A**	**Degassing Part B**	**Cool Down Part A and B**	**Obs.**
1 Control Hydrogel	25	1,464	9	0,012	0	0	8	1	9	9	3 min 2000RPM	No	Yes	No	Yes	Solid in around 2 min Small liquid layer on top Syringe gets Hot for some minutes After 2 Hours (Sticky and stretchable)
2	25	1,464	9	0,012	0	8	0	1	9	9	3 min 2000RPM	No	Yes	No	Yes	Solid in around 60 min Liquid layer on top Syringe gets a little Hot After 2 Hours (Low adhesion but stretchable)
3	25	1,464	9	0,012	0,09	0	8	1	9	9	3 min 2000RPM	No	Yes	No	Yes	Solid in around 10 min Some liquid layer on top Syringe gets a little Hot After 2 Hours (High Sticky and stretchable)
4	25	0	9	0,012	0	0	8	1	9	9	3 min 2000RPM	No	Yes	No	Yes	Solid between 1h to 8h!? A Lot of liquid layer on top Syringe is cold Hydrogel on the bottom Seems Adhesive, Stretchable
5	25	1,464	0	0	0	0	8	1	9	9	3 min 2000RPM	No	Yes	No	Yes	Didn't made this test
6	25	1,464	9	0,012	0,09	0	8	1	9	9	**Slowly by Hand**	**Yes**	Yes	No	Yes	1 day pre-solution used. Result Very pasty hydrogel
7	25	1,464	9	0,012	0	0	8	1	9	9	**Slowly by Hand**	**Yes**	Yes	No	Yes	Measured spontaneous increase of temperature from 19C to 35C in few minutes after mixing Result to a Hydrogel

Table S1: Some extra experimental works and their corresponding results.

																equal to the control hydrogel (1)
8	5	0	9	0,012	0	0	8	1	3,5	9	3 min 2000RPM	No	No	No	No. Because acrylamide solution precipitates	Apparently generate small amount of heat but I am not sure Became very hard hydrogel after 1h30 hour. The solution gradually become more solid with time The hydrogel is sticky but because is too solid seem less adhesive
9	5	0	9	0,012	0,09	0	8	1	3,5	9	3 min 2000RPM	No	No	No	No. Because acrylamide solution precipitates	Hydrogel between 4 and 7 hours Slowly recover to original shape when stretched Is Sticky Seems uncured on the surface but is very resistant.

The given experiment in Table S1 indicates some interesting results as follows:

- Sonication of gallium with acrylamide, water, and BIS at 50 ˚C results in an obtainment of the sticky and transparent hydrogel.
- Sonication of acrylamide, water, and BIS at 50 ˚C remains clear liquid. This indicates that the use of Gallium is necessary for the "Polymerization" of acrylamide.

Other central findings of the investigation in terms of the synthesis method could be:

- It is possible to make hydrogel with acrylamide +BIS + Ga (No alginate), and the resulting hydrogel is still sticky. Although it seems that adding alginate makes the process faster. Using alginate accelerates the reaction and heat generation.

- Probably Ga ions are responsible for the reticulation because in 1 experiment, we sonicated gallium with acrylamide, water, and BIS at 50 ˚C, and the result was a transparent hydrogel (this probably means that there are little amount of gallium ions in the solution, which makes crosslinking possible, and at the same time doesn't allow color change).
- Either temperature or light is not responsible for starting the reaction, because we mixed cold solutions by hand at dark and the resulting hydrogel is the same as with light and mixed by the mixer (Thinky ARE-250).

Table S2: The IR spectra characteristics of the as-prepared hydrogels at different conditions.

Ga	1667.98	1107.02	1038.06	993.34	922.52	853.56								
AAm	3332.24	3175.69	1666.12	1610.21	1421.98	1349.3	1278.48	1134.97	1049.25	984.02	957.53	838.65	814.42	764.1
	NH2-antisymmetric streching	NH2-symmetric streching	C-O double bond-streching	Nh2-Deformation	-Ch2 Double-deformation	C-N streching	-Ch **rocking**	NH2 **rocking**	-CH2 **rocking**	-CH **wagging**	-CH2 **wagging**	C-C streching	**C-C s**	**C=O wa, CH=CH wa**
Laponite	3880	3450	1626.98	965.38										
	shoulder at approximately 3,665 cm-1, assigned to the Mg-OH stretching vibration of the magnesium ion present in the octahedral sheet of the clay	broad H–OH stretching	bending adsorption bands	Si–OH stretching of silanol groups present on the edge of the clay										
SAlg	1591.57	1401.48	1079.06	1023.15	946.74	885.24	812.56							
	asymmetric COO stretching vibration of the carboxylate salt group on the polymeric backbone	symmetric COO stretching vibration of the carboxylate salt group on the polymeric backbone		stretching vibration for COC groups	For the polysaccharides, the peak is present at a wavelength of 1180-953 cm-1 from the vibration of C-C, stretching of C-O, and the C-H bond bending mode.									
0.2ga	2935.28	2881.23	1660.53	1621.39	1453.66	1418.25	1326.93	1209.52	1108.88	1034.34	991.47	922.52	851.7	
0.4Ga	2933.41	2879.37	1664.26	1615.8	1418.25	1325.07	1207.66	1107.02	1032.47	991.47	922.52	851.7		
0.6Ga	2935.28	2881.23	1662.39	1613.94	1451.8	1416.39	1325.07	1209.52	1108.88	1034.34	991.47	922.52	851.7	818.15 777.15
0.8Ga	2933.41	2886.82	1662.39	1615.8	1451.8	1414.52	1325.07	1209.52	1108.88	1034.34	991.47	920.65	851.7	818.15 779.01
1 Ga	2937.14	2883.1	1660.53	1621.39	1453.66	1418.25	1325.07	1209.52	1108.88	1034.34	993.34	922.52	851.7	
La 0.1	2937.14	2884.96	1660.53	1623.26	1453.66	1418.25	1326.93	1209.52	1108.88	1036.2	993.34	922.52	851.7	818.15 764.1
La 0.2	2942.73	2883.1	1660.53	1453.66	1418.25	1326.93	1209.52	1108.88	1034.34	991.47	922.52	851.7	818.15	779.01
La 0.3	2940.87	2884.96	1660.53	1623.26	1453.66	1418.25	1325.07	1209.52	1108.88	1034.34	993.34	922.52	851.7	
La 0.4	2933.41	2884.96	1662.39	1621.39	1418.25	1326.93	1209.52	1108.88	1034.34	993.34	922.52	851.7		