MATERIALS AND METHODS

Preparation of Patterned Flat Origami Sheets

The patterned free-standing flat sheets used in the origami-based fabrication method are obtained using *photolithography*. First, SU8-2050 (MicroChem) is spin-coated on a silicon wafer at 1500 rpm and subsequently soft baked for 10 minutes. This soft baking duration is less than the recommended 20 minutes, ensuring a poor adhesion between the SU8 film and the wafer and thus facilitating the later release of the finalized patterned sheets from the wafer. After soft bake, a lithographic photomask that exposes the *entire origami sheets*, including folds *and* faces, is used to irradiate such regions for 40 seconds using a 2 mW/cm^2 UV light source. Multiple origami sheets can be created simultaneously in this step. Following the first exposure, a mask that exposes *only the face regions*, but not the folds, is aligned on top of the first mask to exclusively irradiate the faces for another 40 seconds using the same 2 mW/cm^2 UV light source. This gives a total exposure time of *80 seconds for the faces* and *40 seconds for the folds*. The difference in total exposure time is required to obtain dissimilar modulus between the folds and faces, ensuring a folding response in good agreement with theoretical assumptions. Such assumptions require the folds to be highly flexible (less cross-linked) and the faces to be stiff (more cross-linked). The maximum exposure duration (80 seconds) is also less than the recommended period (120 seconds) for the photolithography process to prevent complete crosslinking during the Post-Exposure Baking (PEB) and therefore prevent adhesion of the sheets to the wafer. The period of the PEB is also reduced from 10 minutes (recommended period) to 5 minutes for the same reason. The patterned sheets are released from the silicon substrate after the PEB by developing using acetone or SU8 developer. The released patterned sheets are allowed to sediment in the developing liquid (acetone or SU8 developer) and are later separated from the developer by carefully draining the developer and cleaning the sheets with isopropyl alcohol (IPA). The patterned flat sheets are recovered from the IPA mixture by drying them on a parchment paper.

Extensions of the Fabrication Method

- **Fabrication of polyhedra anchored to a substrate.** A *third exposure step* can be applied to create bonding between selected faces of the sheets and the silicon substrate. A mask exposing an area inside a single face of each sheet is used to irradiate these regions for additional 40 seconds using a 2 mW/cm^2 UV light source. These regions, exposed to UV light for a total of *120 seconds*, are *bonded to the substrate* while the remainder of the sheets is not. All other fabrication steps remain the same.
- **Fabrication of polyhedra with arbitrarily shaped hole cut-outs in the faces.** Masks covering arbitrary shapes inside faces of the origami sheets are used in the two exposure steps. This leads the blocked regions to receive no UV irradiation and therefore be completely removed from the sheets during development, resulting in the arbitrarily-shaped holes being created at the selected faces of the sheets.

Folding of Polymer Polyhedra Using Capillary Forces

The resulting free-standing patterned SU8 sheets are placed inside a silicone oil bath. Afterwards, a water *droplet* is placed on top of each of the sheets. Once the droplets are placed above each patterned sheet, the oil bath is heated up to 110°C, causing the sheets to become more compliant, increasing their polymer chain mobility, and consequently be folded driven by *capillary forces* induced by the droplets. In that way, the target polyhedral shapes are formed. The initial volume of the deposited droplets is at least 1.5 times larger than the volume of the target polyhedral shape to ensure that the droplets enter in contact with all the faces of the sheet during capillary folding. As the sheet undergo capillary folding, the water droplet is gradually evaporated until its volume is reduced to approximately that of the target shape. This permitted the formation of fully closed polyhedral structures. The obtained polyhedral structures are filtered out after cooling the bath down to room temperature and thereby regain their initial stiffness and decrease their polymer chain mobility. Therefore, at room temperature, the sheets are *permanently locked* in their folded polyhedral shape.

Fabrication of Carbon Polyhedra

If it is desired to convert the polyhedral structures into *carbon*, the polymer sheets locked in their folded shape are washed with IPA and then exposed to UV light for 200 seconds using a 2 mW/cm^2 light source. This additional exposure ensures that the obtained structures are completely cross-linked and will retain their folded shape during pyrolysis. A Lindberg Blue M furnace is used to pyrolyze the polymer sheets at 900°C for one hour to obtain the final carbon structures. The structures are placed on top of a candle soot coated silicon wafer to prevent them from bonding to the substrate during the pyrolysis.

SUPPLEMENTARY RESULTS

Effect of heat treatment of the patterned sheet on the folding response

Heating the patterned sheets for one hour or more prior to placing a droplet on top of them resulted in no discernible folding deformation as shown in Fig. SA1.

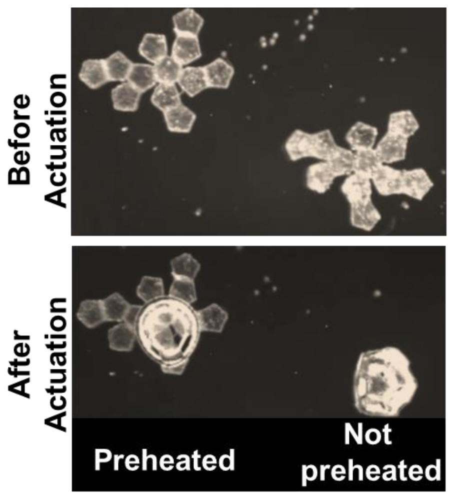

Fig. SA1. Effect of preheating on the folding response of the sheets. The precursor flat sheet (left) that was heated at 110°C for one hour after its patterning, prior to the capillary folding procedure, showed little to no folding as compared to a film that was not heated before the capillary folding procedure (right).

Parametric Study of Folding Deformation

Further representative pictures of the parametric study of the single-fold sheet are shown in Fig. SA2.

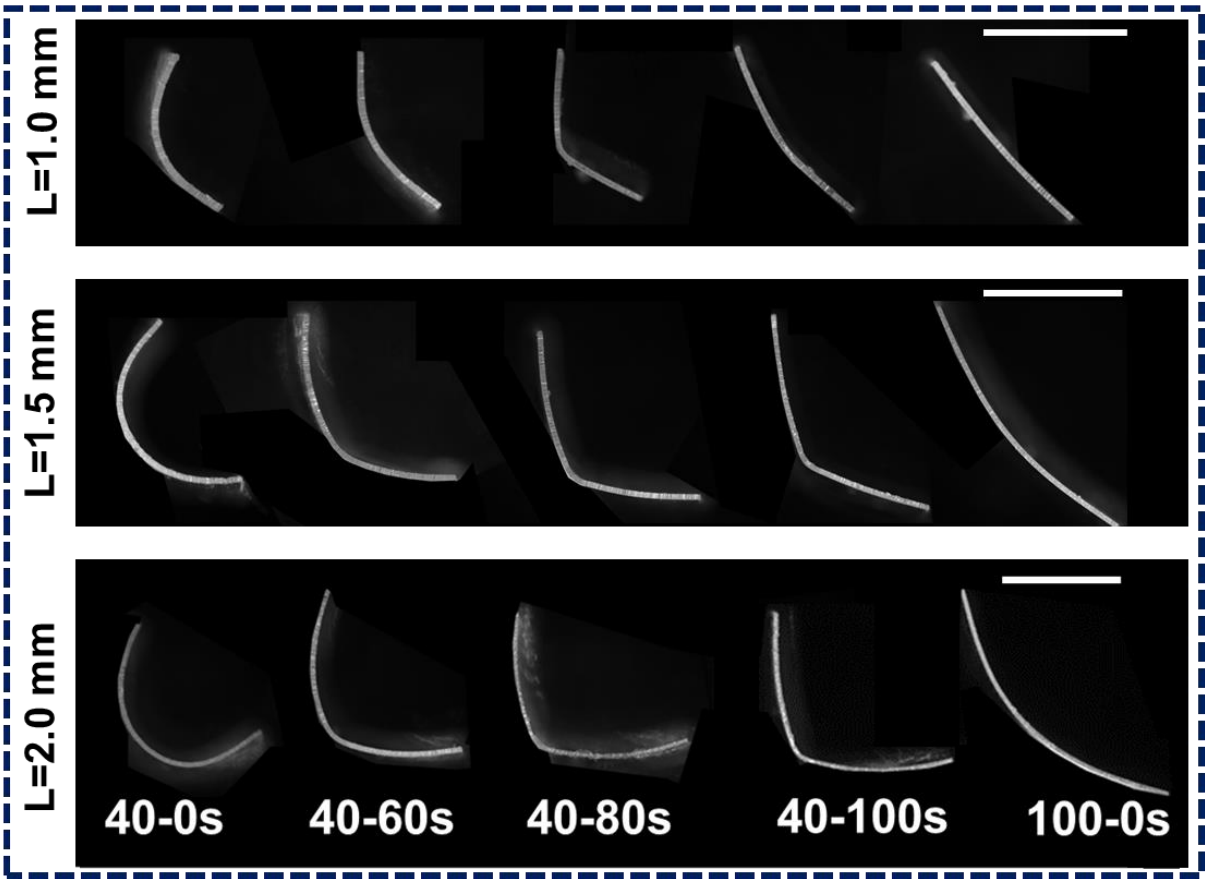

Fig. SA2. Experimental analysis of single-fold sheets. Bending of single-fold sheets of three different sizes subjected to distinct exposure times. The first number denotes the total exposure time for the folds and the second denotes the total exposure time for the faces (e.g., in a 40-60s sample, the folds are exposed for 40 seconds while the faces are exposed for 60 seconds). The exposure intensity was 2 mW/cm^2 for all cases. The scale bars are 1.5 mm long.

Data collected for the generation of the results in Fig. SA2

Table SA1. Angle between faces, curvatures of faces, and curvatures of folds of single-fold sheets with L = 1 mm, L = 1.5 mm and L = 2 mm (refer to Fig. 2).

			Diameter of curvature (mm)			Curvature (1/mm)			Angle between faces (deg)
Size (L in mm)	Exposure 1 (s)	Exposure 2 (s)	Left	Center	Right	Left	Center	Right	
1	40	0	1.960	1.140	1.530	1.020	1.754	1.307	94.710
1	40	0	1.670	1.280	1.930	1.198	1.563	1.036	95.570
1	40	0	1.700	1.120	1.590	1.176	1.786	1.258	93.370
1	40	0	1.590	1.140	1.640	1.258	1.754	1.220	93.530
1	40	0	1.970	1.170	1.780	1.015	1.709	1.124	97.400
Average (L = 1 mm)	40	0	**1.778**	**1.170**	**1.694**	**1.134**	**1.713**	**1.189**	**94.916**
STD			0.157	0.057	0.144	0.098	0.079	0.097	1.480
1.5	40	0	1.680	1.360	1.730	1.190	1.471	1.156	58.930
1.5	40	0	2.020	1.470	1.930	0.990	1.361	1.036	45.280
1.5	40	0	1.860	1.490	1.850	1.075	1.342	1.081	60.020
1.5	40	0	1.720	1.500	1.750	1.163	1.333	1.143	59.600
1.5	40	0	2.220	1.490	1.770	0.901	1.342	1.130	58.560
Average (L = 1.5 mm)	**40**	**0**	**1.900**	**1.462**	**1.806**	**1.064**	**1.370**	**1.109**	**56.478**
STD			0.200	0.052	0.074	0.108	0.051	0.044	5.622
2	40	0	2.580	2.460	3.450	0.775	0.813	0.580	46.450
2	40	0	2.450	1.750	2.160	0.816	1.143	0.926	45.740
2	40	0	1.910	2.340	2.420	1.047	0.855	0.826	33.540
2	40	0	2.890	1.930	2.540	0.692	1.036	0.787	49.880
2	40	0	2.580	1.540	2.500	0.775	1.299	0.800	47.140
Average (L = 2 mm)	40	0	**2.482**	**2.004**	**2.614**	**0.821**	**1.029**	**0.784**	**44.550**
STD			**0.321**	**0.348**	**0.438**	**0.120**	**0.180**	**0.113**	**5.681**
1	40	20	5.580	1.090	5.400	0.358	1.835	0.370	123.160
1	40	20	4.210	1.570	3.430	0.475	1.274	0.583	138.740
1	40	20	6.090	1.520	4.420	0.328	1.316	0.452	128.800
1	40	20	3.350	1.120	3.370	0.597	1.786	0.593	129.420
1	40	20	3.760	1.470	3.350	0.532	1.361	0.597	131.400
Average (L = 1 mm)	40	20	**4.598**	**1.354**	**3.994**	**0.458**	**1.514**	**0.519**	**130.304**
STD			**1.058**	**0.206**	**0.810**	**0.102**	**0.244**	**0.092**	**5.029**
1.5	40	20	5.400	0.590	5.070	0.370	3.390	0.394	84.480

1.5	40	20	4.960	0.790	3.470	0.403	2.532	0.576	88.730
1.5	40	20	4.540	0.520	4.120	0.441	3.846	0.485	85.290
1.5	40	20	4.790	0.650	4.270	0.418	3.077	0.468	95.350
Average (*L* = 1.5 mm)	40	20	**4.923**	**0.638**	**4.233**	**0.408**	**3.211**	**0.481**	**88.463**
STD			**0.314**	**0.099**	**0.569**	**0.025**	**0.478**	**0.065**	**4.285**
2	40	20	3.880	0.510	5.550	0.515	3.922	0.360	76.260
2	40	20	5.220	0.740	4.510	0.383	2.703	0.443	65.450
2	40	20	5.330	0.780	5.080	0.375	2.564	0.394	79.690
2	40	20	5.030	0.640	3.890	0.398	3.125	0.514	80.470
2	40	20	4.310	0.770	4.580	0.464	2.597	0.437	68.380
Average (*L* = 2 mm)	40	20	**4.754**	**0.688**	**4.722**	**0.427**	**2.982**	**0.430**	**74.050**
STD			**0.563**	**0.102**	**0.561**	**0.054**	**0.511**	**0.052**	**6.067**
1	40	40	10.640	0.330	6.990	0.188	6.061	0.286	126.600
1	40	40	6.620	0.480	9.170	0.302	4.167	0.218	145.380
1	40	40	7.380	0.480	7.670	0.271	4.167	0.261	114.190
1	40	40	4.190	0.570	6.740	0.477	3.509	0.297	135.430
Average (*L* = 1 mm)	40	40	**7.488**	**0.512**	**7.176**	**0.294**	**4.152**	**0.288**	**133.014**
STD			**1.520**	**0.132**	**1.386**	**0.043**	**1.142**	**0.058**	**12.809**
1.5	40	40	5.900	0.500	6.150	0.339	4.000	0.325	92.420
1.5	40	40	6.920	0.460	5.390	0.289	4.348	0.371	93.630
1.5	40	40	6.570	0.420	6.590	0.304	4.762	0.303	98.870
1.5	40	40	5.610	0.400	5.300	0.357	5.000	0.377	89.520
1.5	40	40	5.610	0.400	5.940	0.357	5.000	0.337	97.770
Average (*L* = 1.5 mm)	40	40	**6.122**	**0.436**	**5.874**	**0.329**	**4.622**	**0.343**	**94.442**
STD			**0.531**	**0.039**	**0.481**	**0.028**	**0.392**	**0.028**	**3.454**
2	40	40	6.920	0.440	5.950	0.289	4.545	0.336	72.360
2	40	40	5.960	0.220	6.930	0.336	9.091	0.289	71.120
2	40	40	5.100	0.390	7.010	0.392	5.128	0.285	71.020
2	40	40	5.360	0.440	5.640	0.373	4.545	0.355	70.560
2	40	40	5.010	0.420	5.240	0.399	4.762	0.382	82.300
Average (*L* = 2 mm)	40	40	**5.670**	**0.382**	**6.154**	**0.358**	**5.614**	**0.329**	**73.472**
STD			**0.708**	**0.083**	**0.704**	**0.041**	**1.751**	**0.037**	**4.454**
1	40	60	6.850	0.520	8.780	0.292	3.846	0.228	147.440
1	40	60	7.760	0.860	8.720	0.258	2.326	0.229	144.470
1	40	60	14.020	1.340	7.960	0.143	1.493	0.251	152.470
1	40	60	8.140	0.840	10.740	0.246	2.381	0.186	153.100
1	40	60	9.750	1.010	8.700	0.205	1.980	0.230	141.690

Average ($L = 1$ mm)	40	60	**9.304**	**0.914**	**8.980**	**0.229**	**2.405**	**0.225**	**147.834**
STD			**2.538**	**0.266**	**0.930**	**0.051**	**0.787**	**0.021**	**4.437**
1.5	40	60	7.510	0.420	7.290	0.266	4.762	0.274	105.850
1.5	40	60	7.230	0.340	7.010	0.277	5.882	0.285	111.460
1.5	40	60	9.810	0.430	7.370	0.204	4.651	0.271	107.730
1.5	40	60	6.400	0.370	6.010	0.313	5.405	0.333	105.990
1.5	40	60	8.820	0.405	9.480	0.227	4.938	0.211	108.290
Average (L = 1.5 mm)	40	60	**7.954**	**0.393**	**7.432**	**0.257**	**5.128**	**0.275**	**107.864**
STD			**1.264**	**0.037**	**0.542**	**0.039**	**0.500**	**0.025**	**2.262**
2	40	60	6.430	0.460	5.900	0.311	4.348	0.339	65.820
2	40	60	5.650	0.350	8.990	0.354	5.714	0.222	76.490
2	40	60	6.050	0.310	6.310	0.331	6.452	0.317	67.850
2	40	60	15.450	0.330	15.000	0.129	6.061	0.133	71.750
Average ($L = 2$ mm)	40	60	**5.743**	**0.405**	**6.493**	**0.352**	**5.128**	**0.324**	**70.478**
STD			**0.590**	**0.078**	**1.548**	**0.038**	**0.997**	**0.070**	**4.073**
1	100	0	29.630	7.060	13.430	0.067	0.283	0.149	164.170
1	100	0	15.080	9.370	17.760	0.133	0.213	0.113	167.560
1	100	0	26.210	7.310	19.910	0.076	0.274	0.100	168.790
1	100	0	12.040	6.300	10.450	0.166	0.317	0.191	164.790
Average ($L = 1$ mm)	100	0	**20.740**	**7.510**	**15.388**	**0.111**	**0.272**	**0.138**	**166.328**
STD			**7.360**	**1.136**	**3.684**	**0.041**	**0.037**	**0.035**	**1.911**
1.5	100	0	7.330	5.070	10.980	0.273	0.394	0.182	154.110
1.5	100	0	10.050	5.350	12.940	0.199	0.374	0.155	146.000
1.5	100	0	11.800	5.940	20.940	0.169	0.337	0.096	139.660
1.5	100	0	9.600	5.230	7.900	0.208	0.382	0.253	140.320
1.5	100	0	11.420	5.700	21.140	0.175	0.351	0.095	155.240
Average (L = 1.5 mm)	100	0	**10.040**	**5.458**	**14.780**	**0.205**	**0.368**	**0.156**	**147.066**
STD			**1.839**	**0.363**	**4.308**	**0.043**	**0.024**	**0.036**	**5.914**
2	100	0	11.350	4.730	11.680	0.176	0.423	0.171	118.820
2	100	0	7.580	4.280	8.980	0.264	0.467	0.223	111.990
2	100	0	8.690	4.310	9.600	0.230	0.464	0.208	116.730
2	100	0	8.180	4.690	9.600	0.244	0.426	0.208	122.050
2	100	0	13.190	4.440	8.880	0.152	0.450	0.225	122.680
Average ($L = 2$ mm)	100	0	**9.798**	**4.490**	**9.748**	**0.213**	**0.446**	**0.207**	**118.454**
STD			**2.130**	**0.188**	**1.012**	**0.042**	**0.019**	**0.019**	**3.890**

STD: Standard deviation

Effect of Droplet Volume

A study on the effect of droplet volume on the level of folding of single-fold sheets was carried out by placing glycerol (boiling point = 290ºC) droplets of different sizes on top of the sheets followed by heating of the system to 110ºC. Single-fold sheets of $L = 2$ mm (see Fig. 2) were used for these experiments. Droplet sizes varying from $0.5L^3$ to $2.5L^3$ were used to fold the sheets and the folded angle of each of them was measured. While the folding showed consistent results when the droplet volume was larger than L^3, the folding became highly dependent on the location of the droplet on the sheet when the volume is less than L^3.

Data collected for the generation of the results in Fig. 3(a)

Table SA2. Volume of the droplet and corresponding folding angle for single-fold sheets with $L = 2$ mm (see Fig. 2).

Volume	Folding angle (deg)
No droplet	±12.53
	±22.10
	±1.93
Average	0
STD	14.7
$2.5L^3$	62.32
	57.79
	64.85
Average	61.65
STD	2.92
$2.0L^3$	67.52
	73.06
	62.09
Average	67.55
STD	4.48
$1.5L^3$	74.47
	86.36
	86.44
Average	82.42
STD	5.63
$1.0L^3$	134.42

	111.76
	112.95
Average	119.71
STD	10.41
$0.75L^3$	136.31
	141.80
	124.34
Average	134.15
STD	7.29
$0.5L^3$	38.20
	4.61
	9.98
Average	17.60
STD	14.73

STD: Standard deviation

Fabrication of Anchored Folded Shapes

Anchored single-fold sheets are fabricated using a third-step exposure as depicted in Fig. SA3.

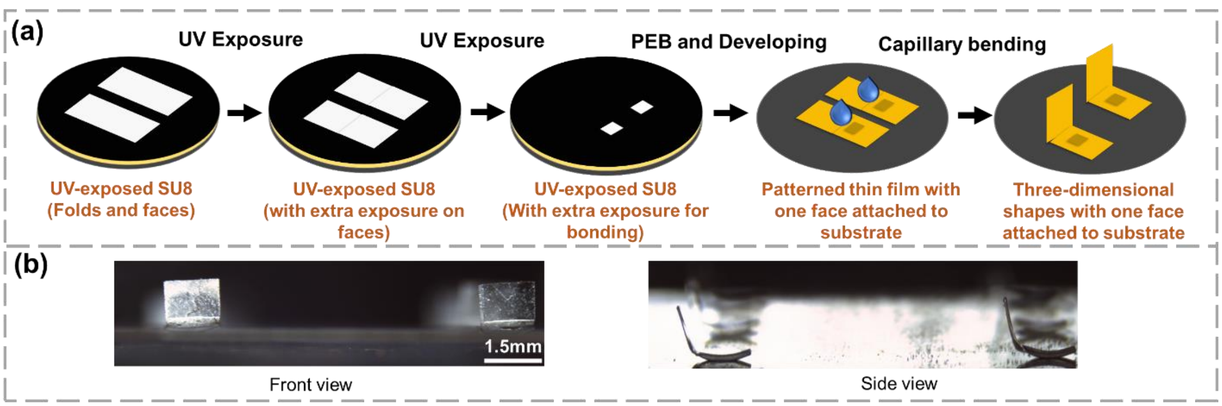

Fig. SA3. Fabrication of origami structures with selected faces attached to a supporting structure. (**a**) Patterning and folding procedure. In addition to the two exposure times required to produce the patterned thin sheets, we perform a third exposure on the regions where bonding to the supporting structure is desired. The sheets are then baked and developed. These anchored sheets are folded to the desired polyhedral shape by using a droplet inside a silicone oil bath at an elevated temperature, just as performed for the free-standing sheets. (**b**) Images of the resulting folding structures from two different views.

Evaluation of storage modulus at temperatures above 90°C

Sheet specimens with a thickness of 55 µm, a value identical to that of the films that were used in the folding experiments, were used to characterize the mechanical properties of SU8. Details of the dynamic mechanical analysis (DMA) tests used for this characterization process are provided in Supplementary Document **B**. A pre-load of 0.0001 N was applied to ensure that the specimens are devoid of any initial buckling. This pre-load force is the lowest that the instrument (TA Instruments DMA Q800) can exert and is constant during the entire temperature sweep study. A time-sinusoidal applied displacement with elongation amplitude of 15 µm was used for data acquisition. Force measurement of the film with this 15 µm elongation amplitude typically enabled data acquisition up to 90ºC. The pre-load prevented the characterization at temperatures higher than 90ºC. This limitation is because the film softens so much at such a high temperature that it experiences a displacement more than the set amplitude because of the pre-load itself. A larger amplitude of oscillation was explored to overcome this measurement limitation. However, an amplitude larger than 15 µm often resulted in breakage of the sheet at lower temperatures and hence was not considered for the characterization.

Therefore, to find the expected values at 110ºC (the temperature at which the capillary folding experiments were performed), the data obtained from DMA experiments at lower temperatures were extrapolated. Typically, storage modulus shows a decreasing slope in the lower temperature range followed by an increase in the slope at the higher temperature ranges (see Fig. 2(**a**)). We fitted the latter region with an expression given by $E' = Ae^{bT}$ (Fig. SA4), where E' is the storage modulus, T is the temperature, and A and b are curve-fitting parameters. The fitted curve is used to extrapolate the data to get a predicted value of the storage modulus values at 110ºC for the films that are made by irradiating for different exposure durations. The extrapolated values of storage modulus at 110ºC are 0.38 MPa, 0.74 MPa, 2.83 MPa, 2.87 MPa and 33.58 MPa for samples with 40 s, 60 s, 80 s, 100 s, and 180 s (completely crosslinked sample) UV exposure durations, respectively. All the fitting parameters and the predicted values of storage modulus at different temperatures are provided in Table SA3.

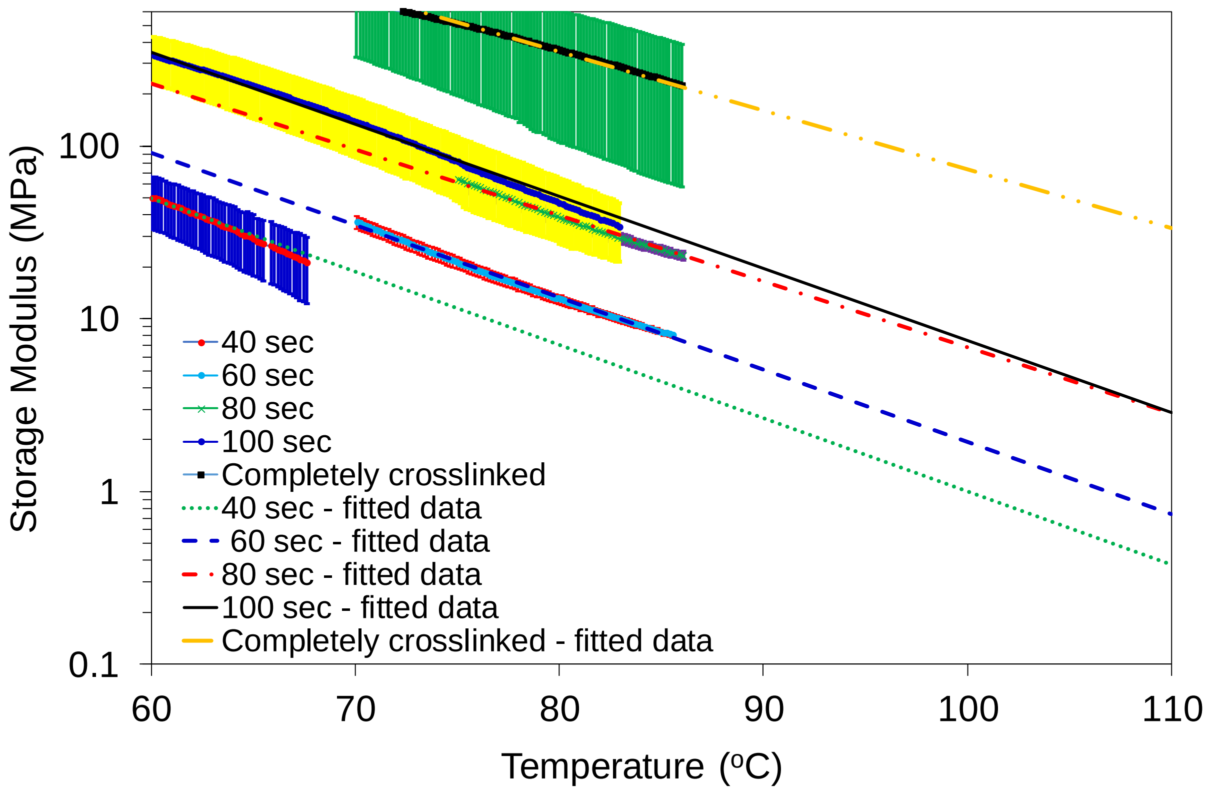

Fig. SA4. Fitting storage modulus data. The storage modulus values (E') obtained from DMA characterization was fitted with an exponential function given by $E' = Ae^{bT}$, where T is the temperature, and A and b are fitting parameters.

Table SA3. Extrapolated storage modulus values at different temperatures through curve fitting. The curves are fitted with a function given by $E' = Ae^{bT}$, where E' is the storage modulus, T is the temperature, and A and b are the fitting parameters. The fitting parameters are also included in the table.

T (in °C)	E' (MPa)	T (in °C)	E' (MPa)	T (in °C)	E' (MPa)	T (in °C)	E' (MPa)	T (in °C)	E' (MPa)
t = 40 s		t = 60 s		t = 80 s		t = 100 s		t = 180 s	
60	49.92	60	91.66	60	229.81	60	349.04	60	1695.99
65	30.62	65	56.60	65	148.05	65	215.94	65	1145.75
70	18.78	70	34.95	70	95.38	70	133.59	70	774.04
75	11.52	75	21.58	75	61.45	75	82.65	75	522.92
80	7.07	80	13.33	80	39.59	80	51.13	80	353.27

85	4.33	85	8.23	85	25.50	85	31.63	85	238.66
90	2.66	90	5.08	90	16.43	90	19.57	90	161.23
95	1.63	95	3.14	95	10.58	95	12.11	95	108.92
100	1.00	100	1.94	100	6.82	100	7.49	100	73.58
105	0.61	105	1.20	105	4.39	105	4.63	105	49.71
110	**0.38**	**110**	**0.74**	**110**	**2.83**	**110**	**2.87**	**110**	**33.58**
Fitting parameters		Fitting parameters		Fitting parameters		Fitting parameters		Fitting parameters	
A	17595.44	A	29831.24	A	44966	A	111034	A	187666
b	-0.09775	b	-0.09642	b	-0.08794	b	-0.09604	b	-0.07844