

Supporting Information

Effect of gel ageing and electrode corrosion on the performance of direct laser writing carbonization enabled hydrogel-based moist-electric generator

Xuewei Pi, Yanbo Yao*, Dini Qin, and Tao Liu *

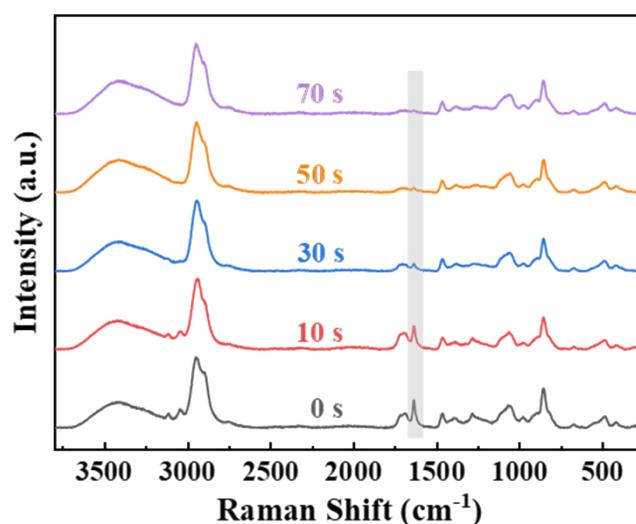


Figure S1. Raman spectra for tracking the polymerization of AA in the solution of PVA/AA/PA (solution aged for 2.5 days) initiated by UV irradiation for different time duration.

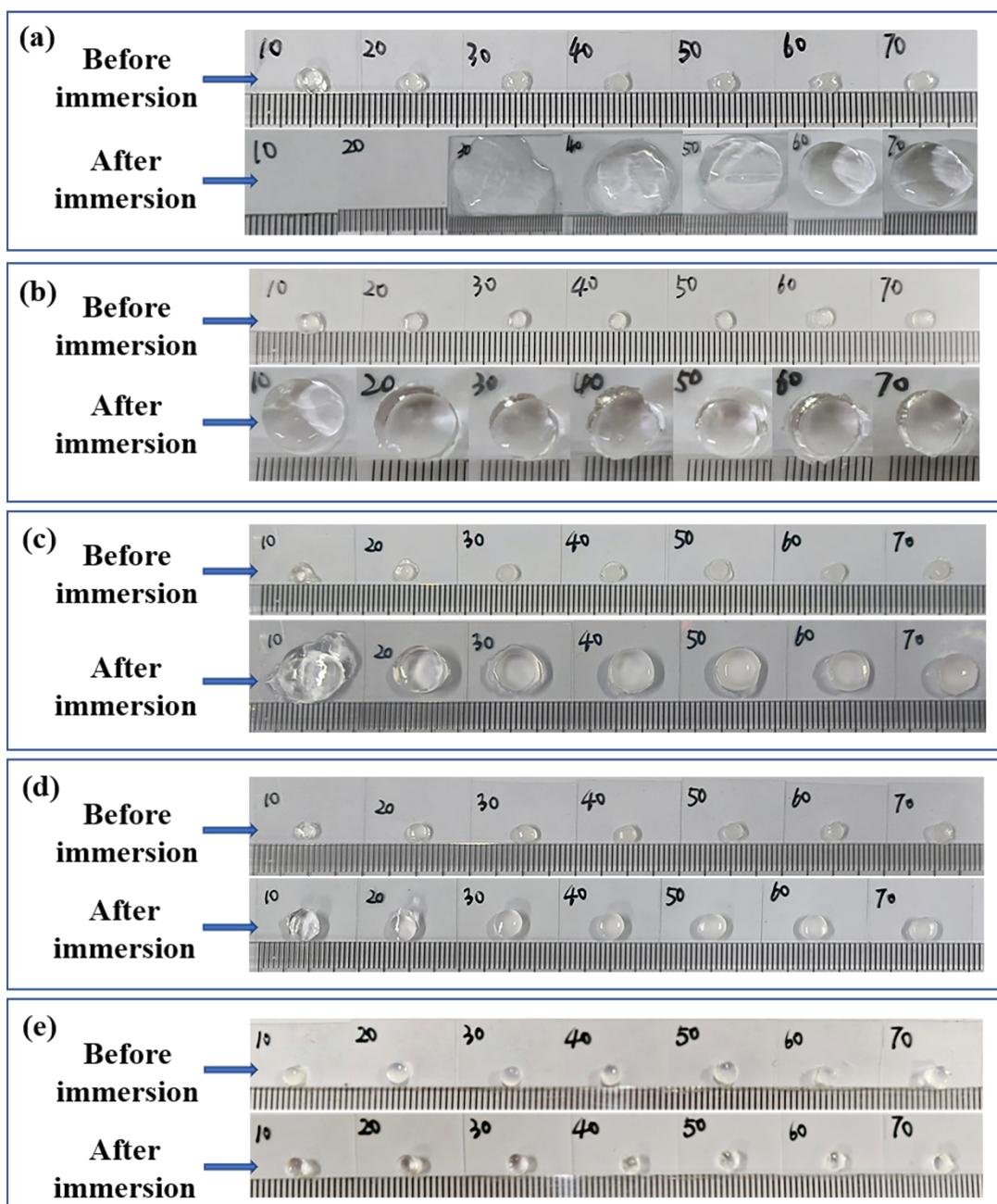


Figure S2. Optical pictures to show the effect of solution and gel ageing on water immersion (24 h) induced swelling for the PVA/PAA/PA hydrogel prepared by UV irradiation for varied time duration (The scale of the ruler is 1 mm; the number labeled in each picture indicates the UV-irradiation time). (a) Solution ageing for 0.5 day without gel ageing; (b) Solution ageing for 2.5 days without gel ageing; (c) Solution ageing for 0.5 day and gel ageing for 24 h; (d) Solution ageing for 2.5 days and gel ageing for 24 h; (e) Solution ageing for 120 days and gel ageing for 0 h.

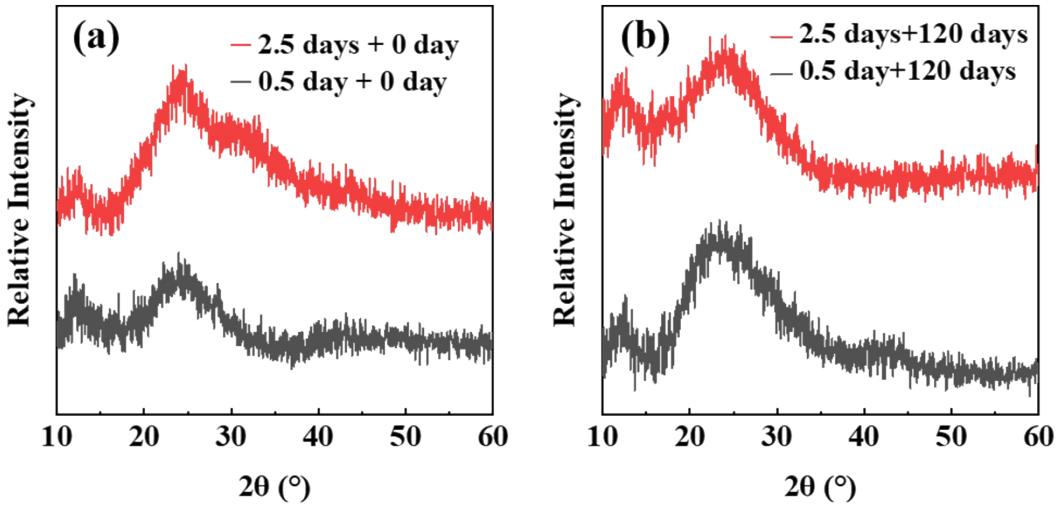


Figure S3. X-ray diffraction pattern of the PVA/PAA/PA hydrogels prepared from the solution aging for 0.5 or 2.5 days upon UV irradiation for 30 s and (a) without and (b) with gel aging treatment.

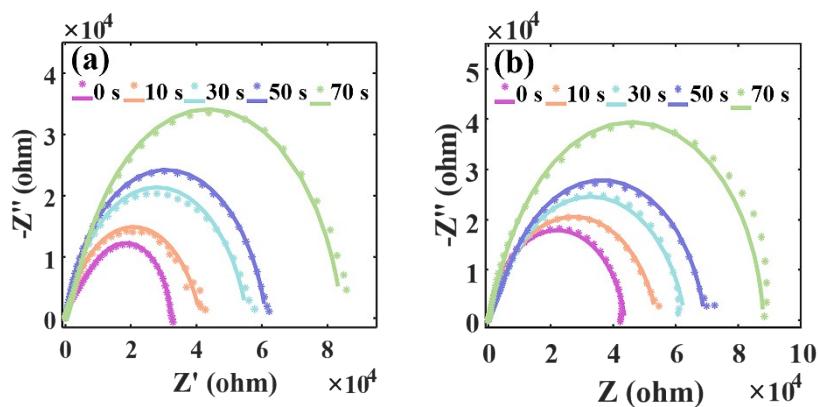


Figure S4. The complex plane impedance diagram of PVA/PAA/PA hydrogel prepared from the PVA/AA/PA solution aged for (a) 2.5 days or (b) 120 days, and UV-exposure for varied time duration (no gel ageing treatment). Scattered data points are the experimentally measured results; and the continuous lines are the equivalent circuit fitting results (See Figure 4a for the circuit diagram).

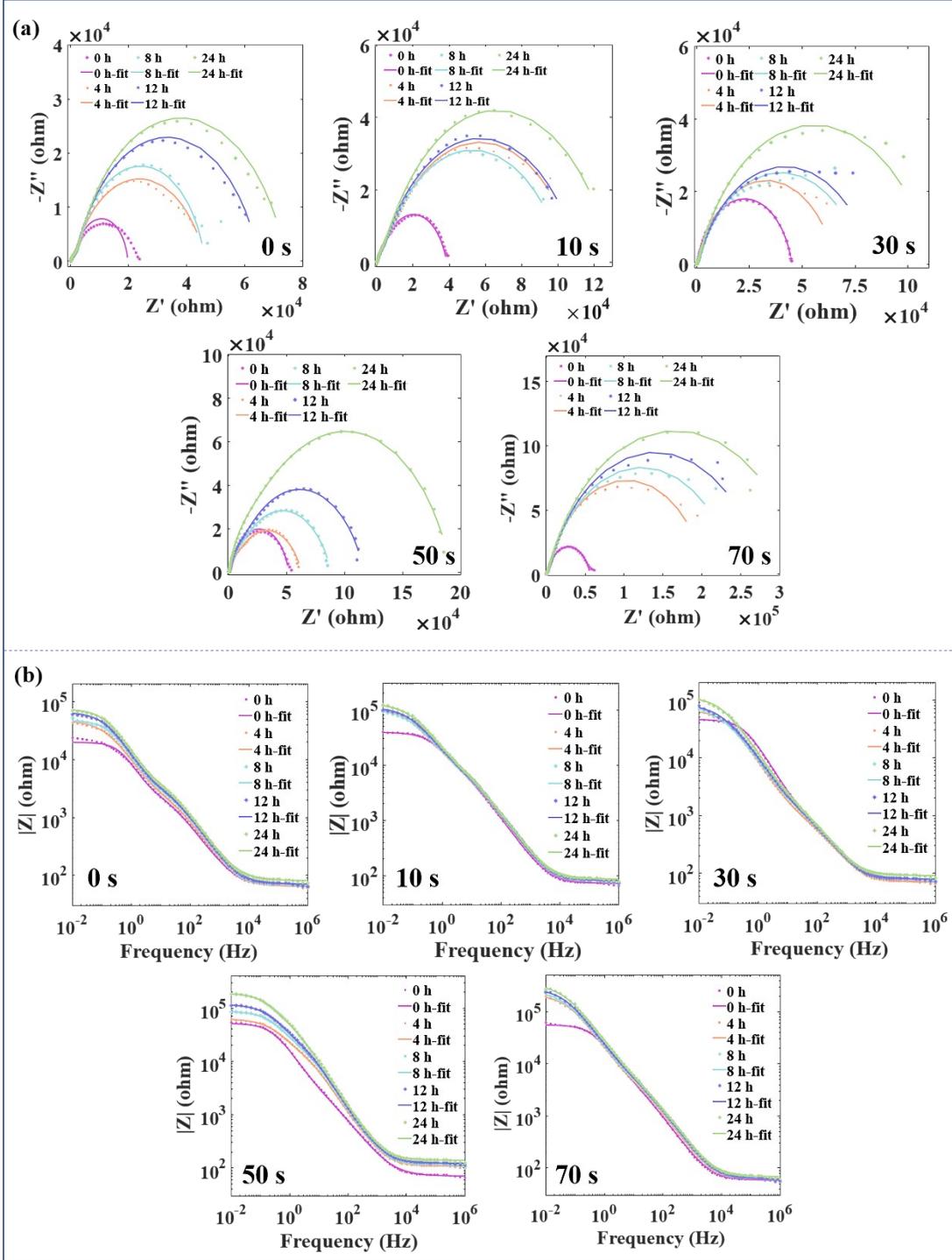


Figure S5. (a) The complex plane and (b) Bode impedance diagrams for the PVA/PAA/PA hydrogels prepared by UV-irradiation for varied time duration (0 s, 10 s, 30 s, 50 s and 70 s) and aged at different conditions. The PVA/AA/PA solution ageing time is 0.5 day; and the PVA/PAA/PA gel ageing time varies from 0 h to 24 h. Scattered data points are the experimentally measured results; and the continuous lines are the equivalent circuit fitting results (See Figure 4a for the circuit diagram).

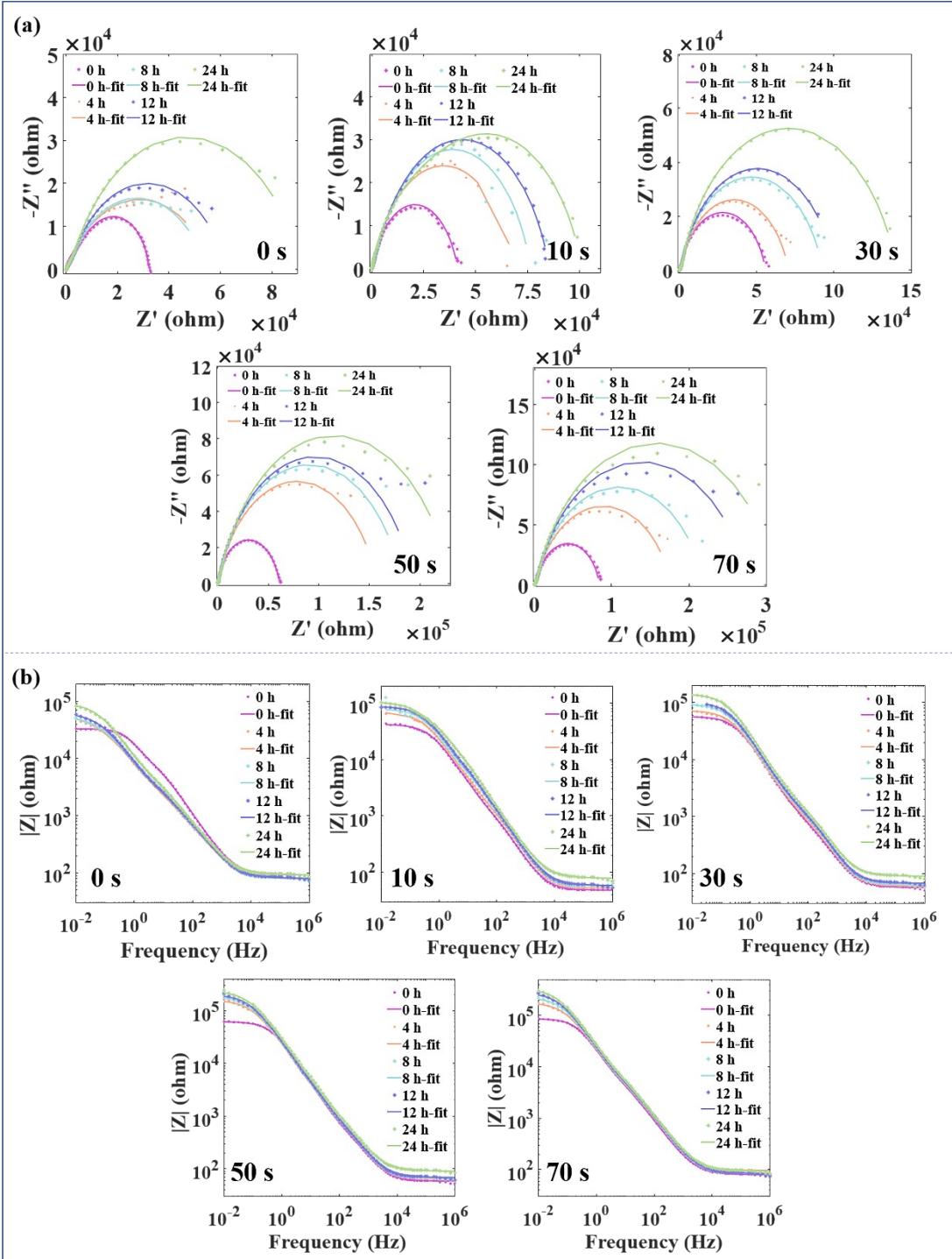


Figure S6. (a) The complex plane and (b) Bode impedance diagrams for the PVA/PAA/PA hydrogels prepared by UV-irradiation for varied time duration (0 s, 10 s, 30 s, 50 s and 70 s) and aged at different conditions. The PVA/AA/PA solution ageing time is 2.5 days; and the PVA/PAA/PA gel ageing time varies from 0 h to 24 h. Scattered data points are the experimentally measured results; and the continuous lines are the equivalent circuit fitting results (See Figure 4a for the circuit diagram).

Table S1. Summary of the equivalent circuit (inset of Figure 4a) fitting results for the experimentally measured impedance spectra shows in Figure S3 – S5.

| Solution Aging Time (d) | UV exposure time (sec) | Gel Aging Time (h) | R3 (ohm) | R1 (ohm) | R2 (ohm) | CPE1-T (F) | CPE1-P | CPE2-T(F) | CPE2-P |
|-------------------------|------------------------|--------------------|----------|----------|----------|------------|---------|-----------|---------|
| 0 | 0 | 0 | 49 | 437 | 19059 | 1.57E-05 | 0.76229 | 2.46E-05 | 0.75932 |
| | 10 | 0 | 68 | 3815 | 18932 | 7.31E-06 | 0.76987 | 1.90E-05 | 0.87104 |
| | 30 | 0 | 57 | 1186 | 40468 | 2.03E-05 | 0.69320 | 1.29E-05 | 0.86426 |
| | 50 | 0 | 57 | 330 | 45824 | 1.26E-05 | 0.84111 | 7.32E-06 | 0.80431 |
| | 70 | 0 | 64 | 811 | 57820 | 1.90E-05 | 0.76557 | 5.36E-06 | 0.85417 |
| | 0 | 0 | 65 | 1036 | 21632 | 1.31E-05 | 0.75020 | 2.63E-05 | 0.76793 |
| | | 4 | 64 | 1630 | 41595 | 1.17E-05 | 0.74084 | 2.34E-05 | 0.79949 |
| | | 8 | 67 | 2008 | 46884 | 9.50E-06 | 0.75093 | 2.00E-05 | 0.82199 |
| | | 12 | 70 | 2128 | 59760 | 8.97E-06 | 0.75152 | 1.83E-05 | 0.82114 |
| 0.5 | 10 | 24 | 80 | 2481 | 68166 | 8.39E-06 | 0.74972 | 1.58E-05 | 0.83032 |
| | | 0 | 70 | 3406 | 36177 | 1.54E-05 | 0.72657 | 1.18E-05 | 0.78694 |
| | | 4 | 77 | 4083 | 102760 | 1.24E-05 | 0.75163 | 1.72E-05 | 0.71450 |
| | | 8 | 77 | 3826 | 96274 | 1.18E-05 | 0.75136 | 1.58E-05 | 0.72313 |
| | 30 | 12 | 79 | 4163 | 103980 | 1.17E-05 | 0.74192 | 1.43E-05 | 0.74265 |
| | | 24 | 83 | 4087 | 124730 | 1.19E-05 | 0.73484 | 1.26E-05 | 0.75334 |
| | | 0 | 70 | 544 | 44561 | 2.04E-05 | 0.70251 | 1.20E-05 | 0.86065 |
| | | 4 | 69 | 536 | 65571 | 2.10E-05 | 0.71729 | 2.75E-05 | 0.77915 |
| 2.5 | 50 | 8 | 75 | 462 | 79094 | 1.84E-05 | 0.76802 | 2.88E-05 | 0.72263 |
| | | 12 | 77 | 401 | 83196 | 1.74E-05 | 0.76519 | 2.53E-05 | 0.73071 |
| | | 24 | 89 | 599 | 110560 | 2.07E-05 | 0.73063 | 2.01E-05 | 0.77271 |
| | | 0 | 69 | 1295 | 50690 | 2.67E-05 | 0.67546 | 1.25E-05 | 0.83742 |
| | 70 | 4 | 105 | 4434 | 58252 | 7.19E-06 | 0.95193 | 1.12E-05 | 0.73516 |
| | | 8 | 114 | 5489 | 83167 | 6.07E-06 | 0.96729 | 8.68E-06 | 0.75226 |
| | | 12 | 121 | 6843 | 110310 | 5.81E-06 | 0.96085 | 7.65E-06 | 0.76202 |
| | | 24 | 138 | 14107 | 177770 | 6.56E-06 | 0.94379 | 5.52E-06 | 0.78862 |
| 2.5 | 0 | 0 | 58 | 1753 | 55190 | 1.34E-05 | 0.76302 | 8.23E-06 | 0.85588 |
| | | 4 | 60 | 2235 | 203480 | 1.15E-05 | 0.76828 | 1.14E-05 | 0.79127 |
| | | 8 | 61 | 1938 | 241780 | 1.12E-05 | 0.77524 | 1.10E-05 | 0.76474 |
| | | 12 | 62 | 2015 | 276980 | 1.19E-05 | 0.76583 | 1.00E-05 | 0.76438 |
| | 10 | 24 | 67 | 1218 | 329190 | 1.19E-05 | 0.78401 | 8.56E-06 | 0.75780 |
| | | 0 | 91 | 7604 | 24634 | 8.30E-06 | 0.75923 | 1.23E-05 | 0.93826 |
| | | 4 | 77 | 641 | 56275 | 3.11E-05 | 0.73974 | 3.50E-05 | 0.66610 |
| | | 8 | 78 | 1233 | 53143 | 2.83E-05 | 0.70100 | 3.16E-05 | 0.70846 |
| 2.5 | 0 | 12 | 80 | 1400 | 61122 | 2.42E-05 | 0.69866 | 3.01E-05 | 0.73530 |
| | | 24 | 91 | 2212 | 89194 | 2.41E-05 | 0.68960 | 2.36E-05 | 0.76762 |
| | | 0 | 48 | 397 | 40949 | 1.01E-05 | 0.84872 | 9.28E-06 | 0.79671 |
| | 10 | 4 | 52 | 499 | 68187 | 8.83E-06 | 0.85104 | 8.89E-06 | 0.77645 |
| | | 8 | 56 | 1993 | 74652 | 1.29E-05 | 0.76689 | 7.63E-06 | 0.80887 |
| | | 12 | 58 | 1984 | 83585 | 1.70E-05 | 0.76301 | 7.03E-06 | 0.79005 |

| | | | | | | | | | |
|------------|-----------|----|-------|--------|----------|----------|----------|----------|---------|
| | 24 | 78 | 25954 | 75286 | 7.62E-06 | 0.76561 | 1.22E-05 | 0.78418 | |
| 30 | 0 | 55 | 525 | 54964 | 1.41E-05 | 0.76048 | 1.04E-05 | 0.83872 | |
| | 4 | 59 | 595 | 70064 | 1.06E-05 | 0.79154 | 1.11E-05 | 0.81439 | |
| | 8 | 63 | 679 | 92093 | 8.73E-06 | 0.80139 | 9.97E-06 | 0.81603 | |
| | 12 | 67 | 722 | 99428 | 8.18E-06 | 0.80219 | 9.05E-06 | 0.82099 | |
| | 24 | 89 | 889 | 139590 | 8.55E-06 | 0.78029 | 7.70E-06 | 0.82166 | |
| | 0 | 57 | 225 | 61237 | 1.41E-05 | 0.78084 | 6.81E-06 | 0.85138 | |
| 50 | 4 | 63 | 263 | 157840 | 8.02E-06 | 0.88339 | 9.23E-06 | 0.79092 | |
| | 8 | 66 | 307 | 182210 | 2.57E-05 | 0.74914 | 8.68E-06 | 0.79293 | |
| | 12 | 68 | 368 | 193800 | 3.46E-05 | 0.71701 | 8.36E-06 | 0.79488 | |
| | 24 | 89 | 416 | 230930 | 4.21E-05 | 0.68892 | 7.61E-06 | 0.78479 | |
| | 0 | 79 | 2465 | 83882 | 1.58E-05 | 0.73593 | 9.36E-06 | 0.86265 | |
| | 4 | 95 | 1924 | 175400 | 1.60E-05 | 0.74821 | 9.88E-06 | 0.81108 | |
| 70 | 8 | 84 | 2183 | 217540 | 1.51E-05 | 0.74487 | 9.47E-06 | 0.81595 | |
| | 12 | 83 | 2295 | 273790 | 1.65E-05 | 0.72846 | 9.07E-06 | 0.81578 | |
| | 24 | 92 | 2777 | 312240 | 1.83E-05 | 0.70821 | 8.49E-06 | 0.82126 | |
| | 0 | 0 | 66 | 43206 | 2.32E-05 | 0.71821 | 5.18E-06 | 0.87944 | |
| | 10 | 0 | 80 | 483 | 1.72E-05 | 0.74696 | 1.16E-05 | 0.83680 | |
| | 30 | 0 | 80 | 2767 | 59885 | 1.29E-05 | 0.74317 | 9.19E-06 | 0.86840 |
| 120 | 50 | 0 | 80 | 4324 | 64662 | 1.11E-05 | 0.75446 | 7.72E-06 | 0.89592 |
| | 70 | 0 | 83 | 6437 | 81217 | 1.48E-05 | 0.74789 | 4.43E-06 | 0.96344 |

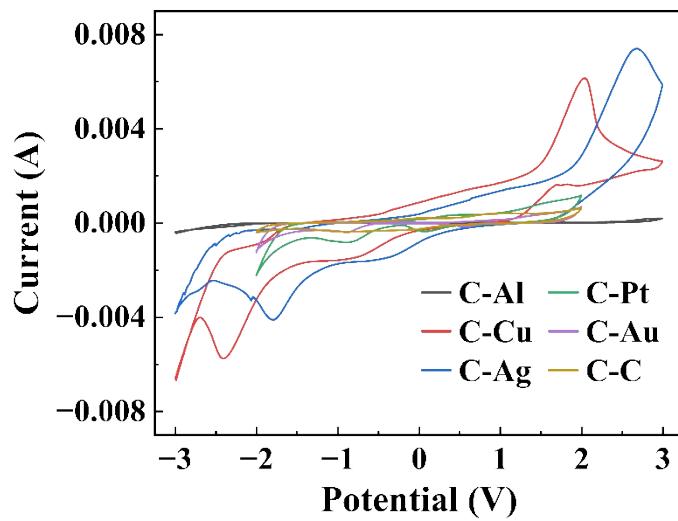


Figure S7. Cyclic voltammograms for different types of metallic electrodes in the solution of PVA/AA/PA obtained in a 2-electrode testing configuration with DLWc carbon as the counter electrode.

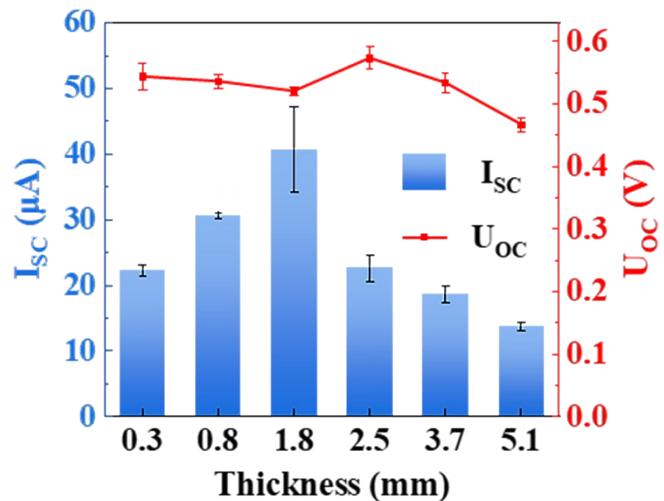


Figure S8. The effect of thickness of the hydrogel active material on I_{SC} and U_{OC} for the DLWc-Hgel-MEG devices assembled from the PVA/PAA/PA hydrogels prepared from the PVA/AA/PA solution aged for 0.5 day and UV-exposure for 10 s without gel ageing treatment and tested at 85 RH%.

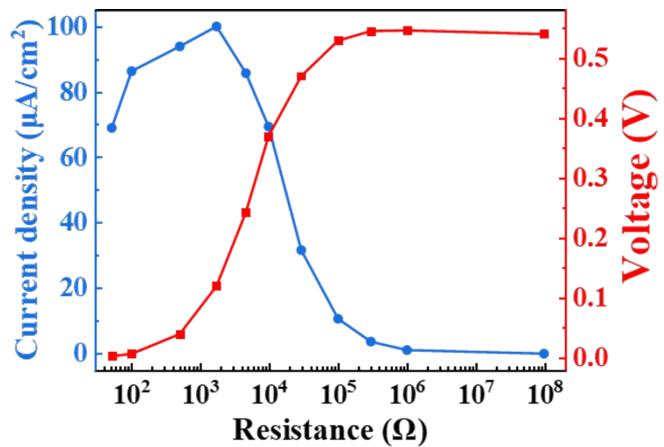


Figure S9. The current and voltage output of a selected DLWc-Hgel-MEG device (assembled using the PVA/PAA/PA hydrogel prepared from the PVA/AA/PA solution aged for 0.5 day and UV-exposure for 10 s without gel ageing treatment) to a resistor load at ambient environment (25°C , $55 \pm 3 \text{ RH}\%$) for evaluating its power transfer capability.

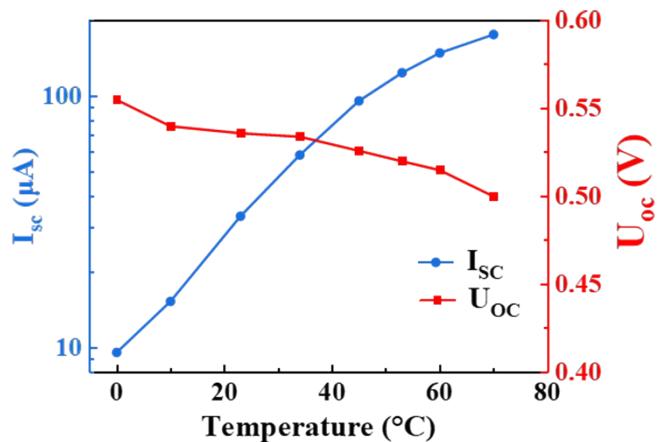


Figure S10. The current and voltage output of a selected DLWc-Hgel-MEG device (assembled using the PVA/PAA/PA hydrogel prepared from the PVA/AA/PA solution aged for 0.5 day and UV-exposure for 10 s without gel ageing treatment) evaluated in a temperature range of $0 - 70^\circ\text{C}$ at $85 \pm 5\% \text{ RH}$.