Supporting Information

Electrochromic Devices Based on Ultraviolet-Cured Poly(methyl methacrylate) Gel Electrolytes and Their Utilisation in Smart Window Applications

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Experimental

Fully polymerised electrolyte sample for scanning electron microscopy (SEM) was prepared as follows. N_2 purged 0.5 mL of liquid electrolyte in a transparent vial was subjected to UV irradiation at a wavelength of 360 nm for 40 min under N_2 atmosphere. As a result, the liquid electrolyte was fully polymerised. To take the electrolyte, the vial was smashed with a hammer, and the obtained sample was sliced with a razor blade.

Characterisation

Scanning electron microscopy (SEM) measurement was conducted by using field emission scanning electron microscope (SU8010, Hitachi, Tokyo, Japan).

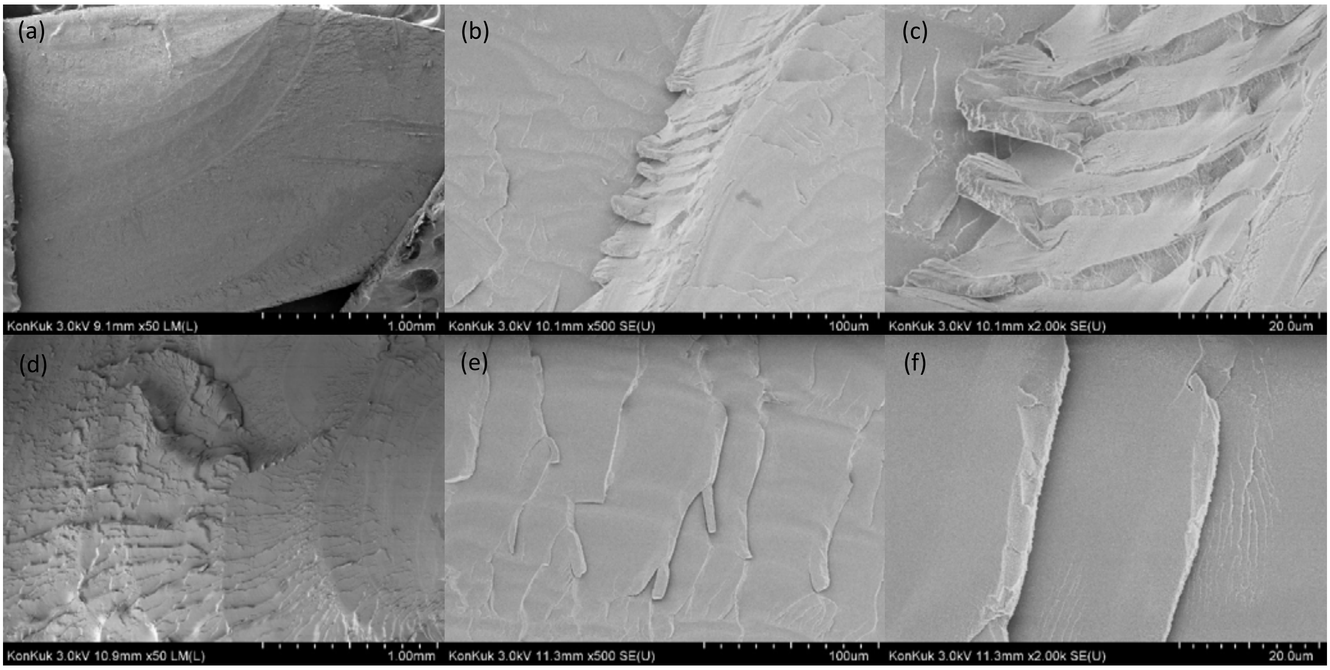

Fig. S1. SEM images of fully polymerised electrolyte under various magnifications of (a and d) 50, (b and e) 500, and (c and f) 2,000. The layer-like texture would result from the sampling process.

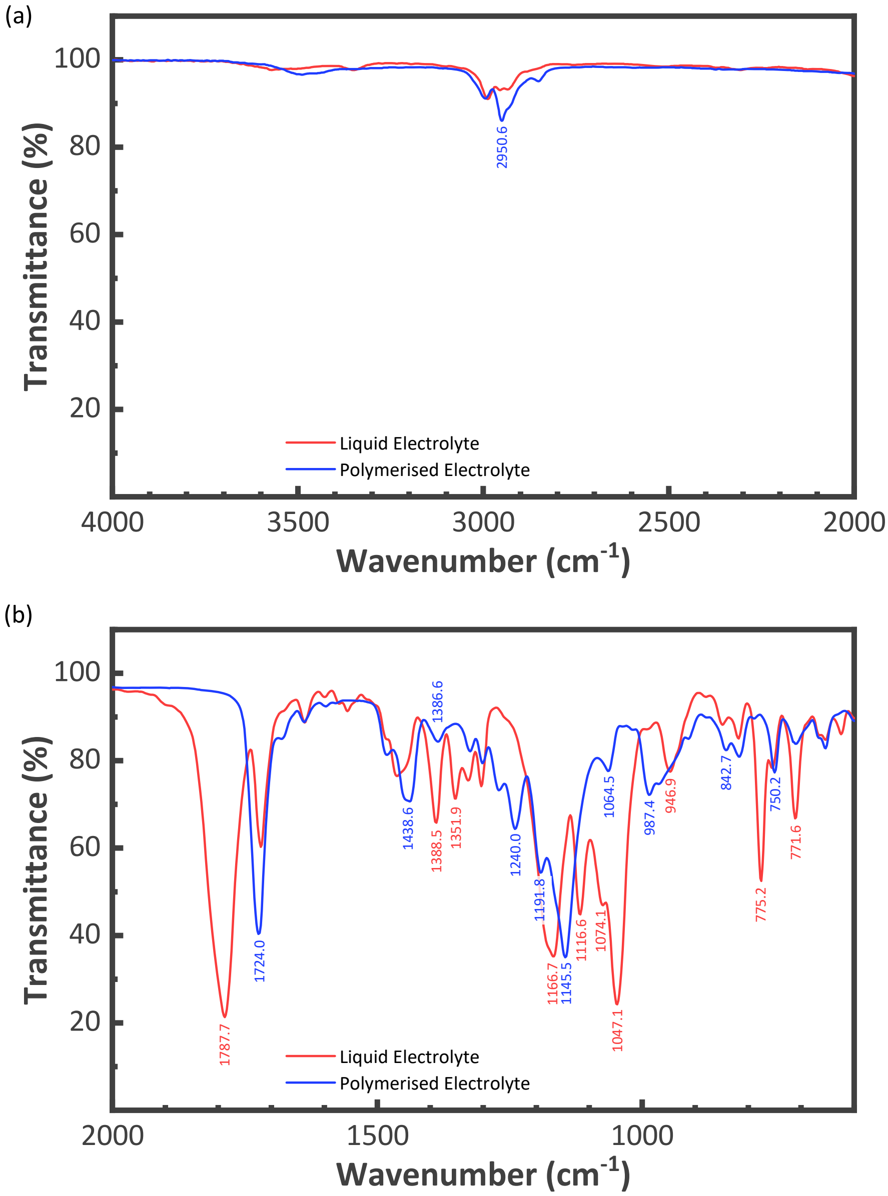

Fig. S2. Fourier-transform infrared spectroscopy results of liquid electrolyte and fully polymerised electrolyte at a wavenumber range of (a) 4000–2000, and (b) 2000–600 cm^{-1}. For liquid electrolyte, peaks at wavenumbers of 1787.7 (C=O), 1388.5 (C–H bend/rock), 1351.9 (H–C–C–H bend), 1166.7 cm^{-1} ($–CH_2$ rock) are characteristic peaks of propylene carbonate (PC), and peaks at wavenumbers of 1116.6 (ring str. + C=O), 1074.1 (ring str.), 1047.1 (ring str. + C–H, C–C), 946.9 (ring), 775.2 (ring bend), 771.6 cm^{-1} (ring str.) are correlated with motions of the ring of PC. Meanwhile, new poly(methyl methacrylate) (PMMA) characteristic peaks arose in the fully polymerised electrolyte at wavenumbers of 1064.5, 987.4, 842.7 cm^{-1} with other PMMA peaks at wavenumbers of 2950.6 (C–H str. of $–CH_2$), 1724.0 (O–C–O), 1438.6 (C–H str. of $–CH_3$), 1386.6 ($–CH_3$), 1240.0 (C–O–C str.), 1191.8 (C–O–C str.), 1145.5 (C–O–C str.), 750.2 cm^{-1} ($–CH_3$).

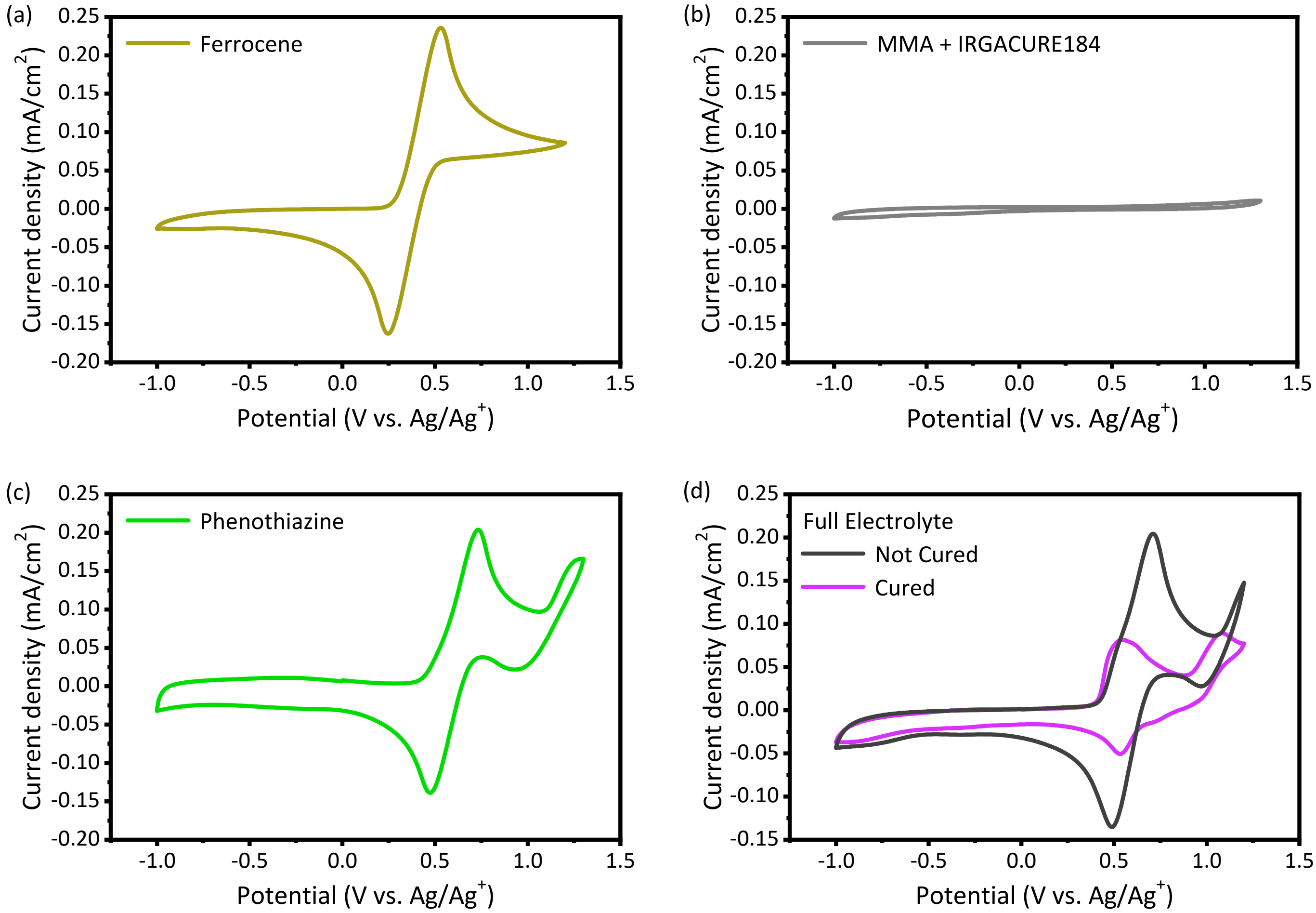

Fig. S3. Cyclic voltammetry results of (a) ferrocene, (b) MMA and IRGACURE 184, (c) phenothiazine, and (d) full electrolyte solutions with and without ultraviolet (UV) curing. The measurements were conducted under a scan rate of 20 mV/s. Solvents were propylene carbonate.

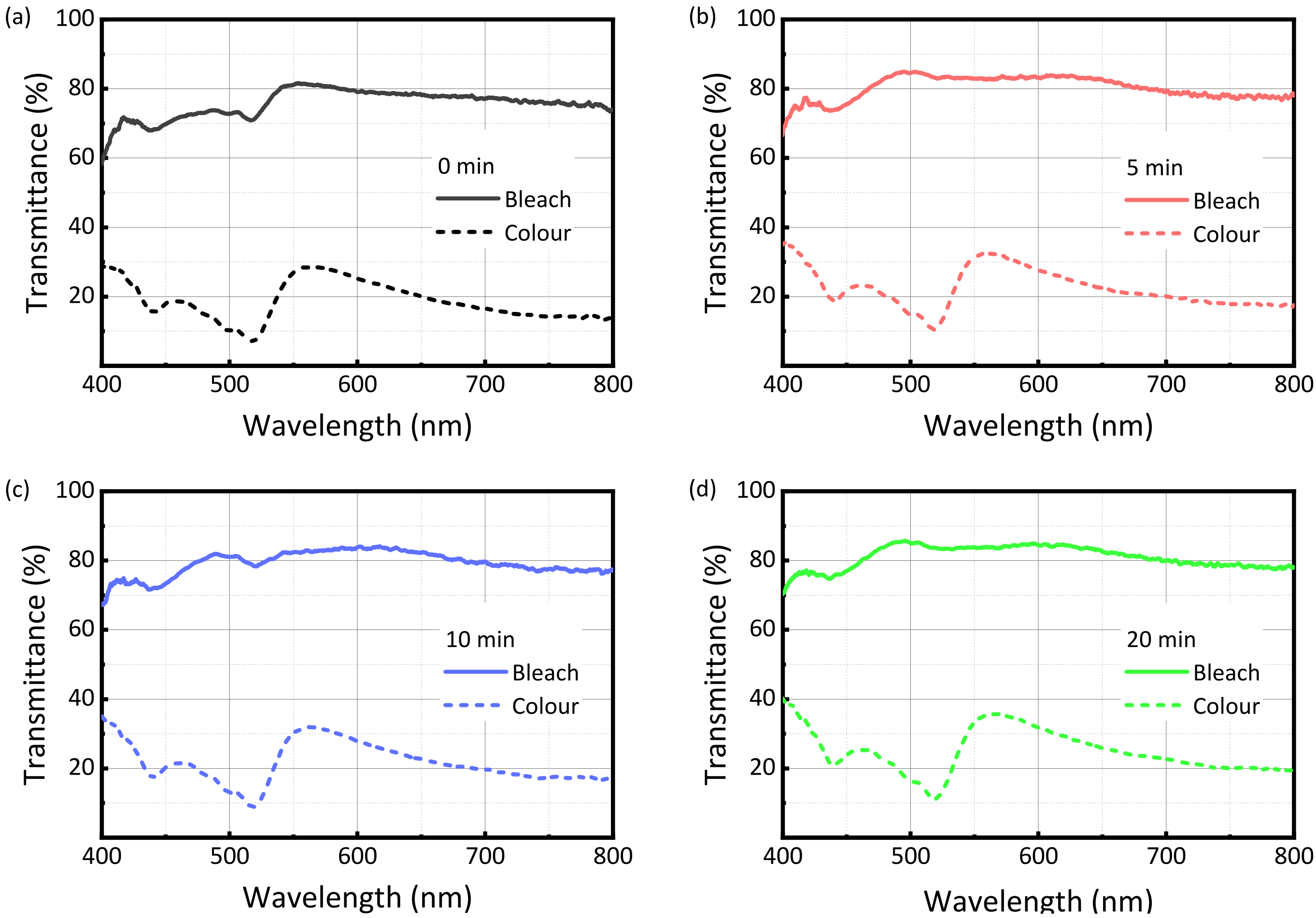

Fig. S4. Optical transmittance spectra of the electrochromic devices (ECDs) with various UV curing time for (a) 0, (b) 5, (c) 10, and (d) 20 min. Top and bottom lines are for the bleached and the coloured states, respectively.

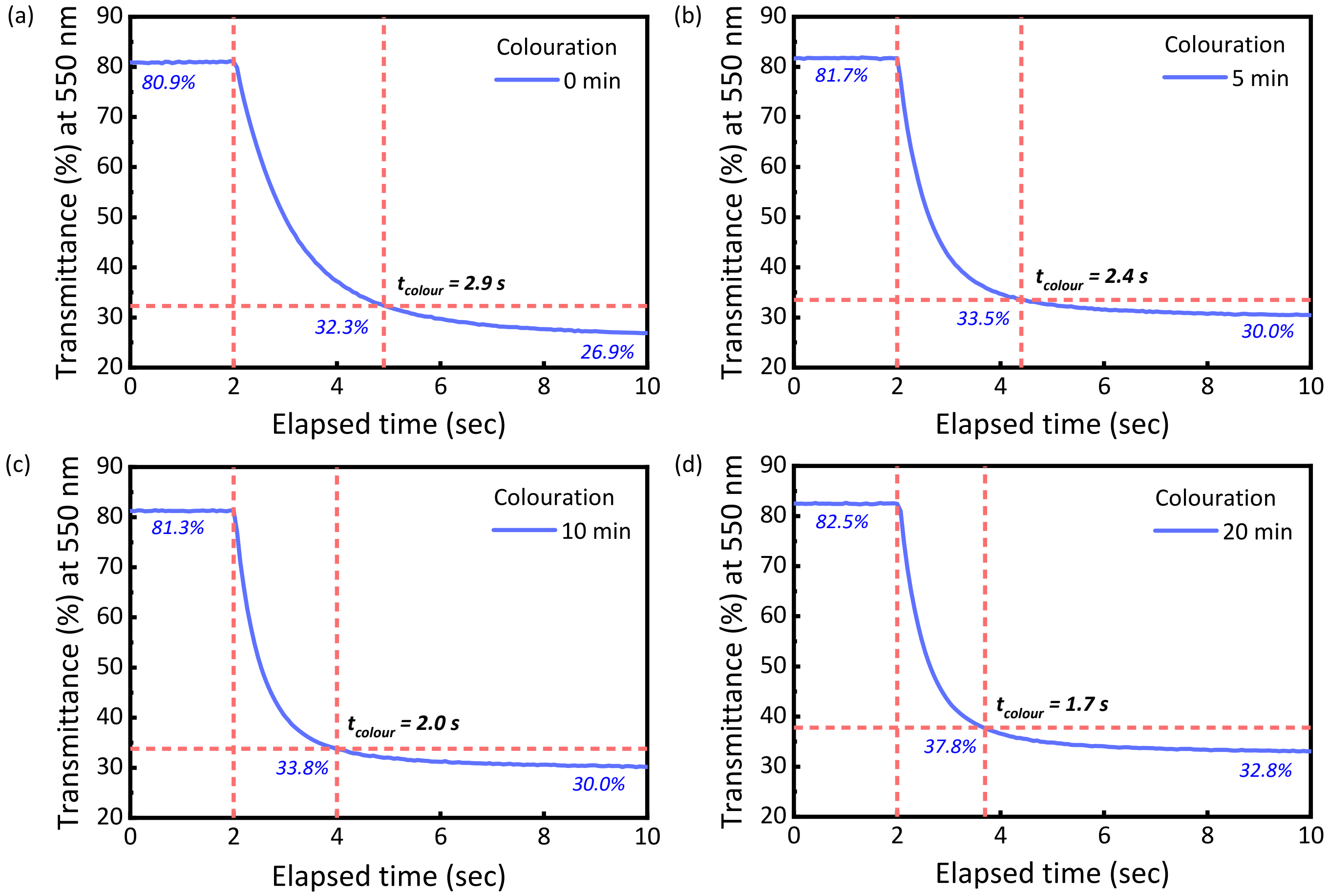

Fig. S5. Detailed dynamic transmittance spectra at a wavelength of 550 nm of colouring half-cycles. The ECDs were UV-cured for (a) 0, (b) 5, (c) 10, and (d) 20 min.

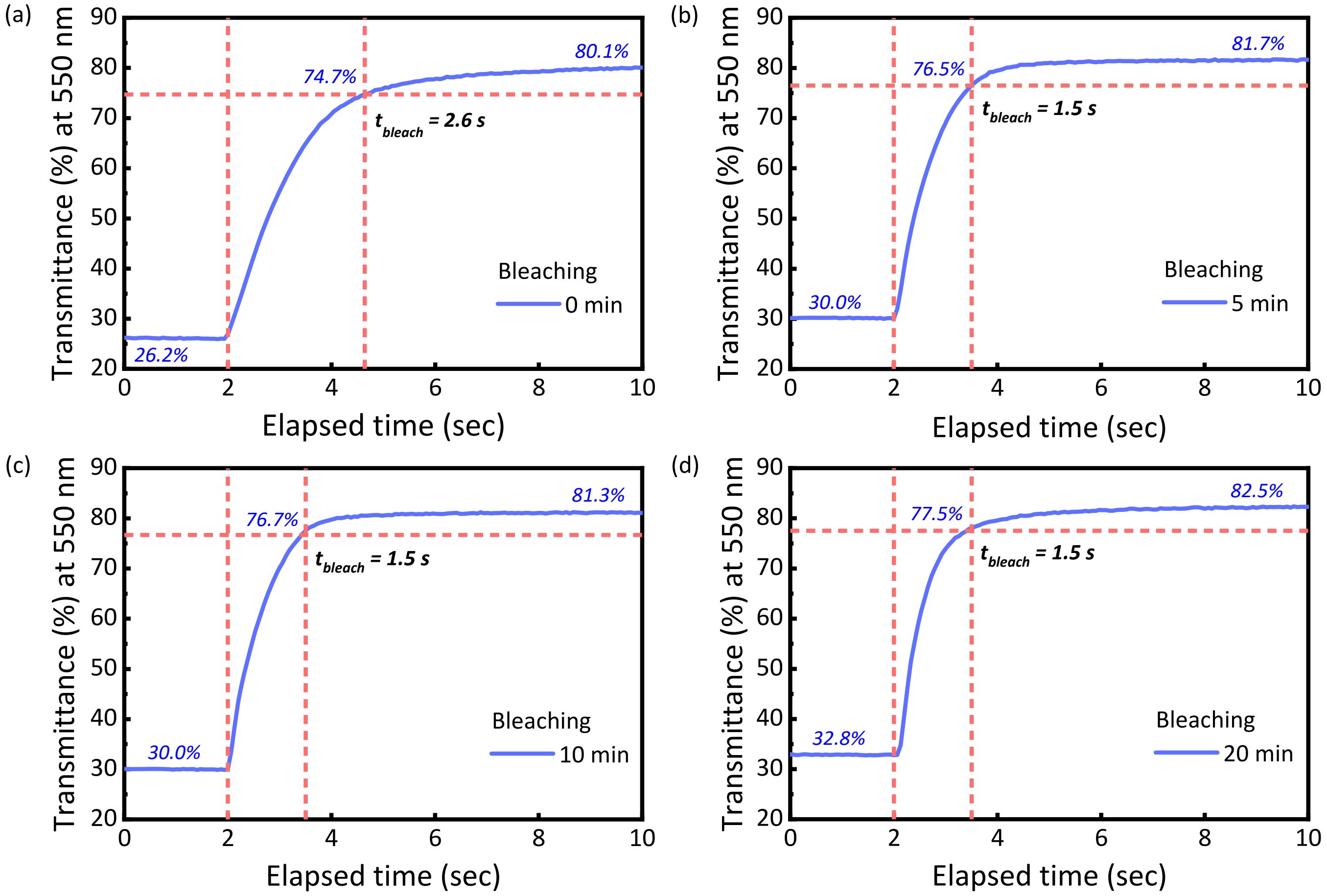

Fig. S6. Detailed dynamic transmittance spectra at a wavelength of 550 nm of bleaching half-cycles. The ECDs were UV-cured for (a) 0, (b) 5, (c) 10, and (d) 20 min.

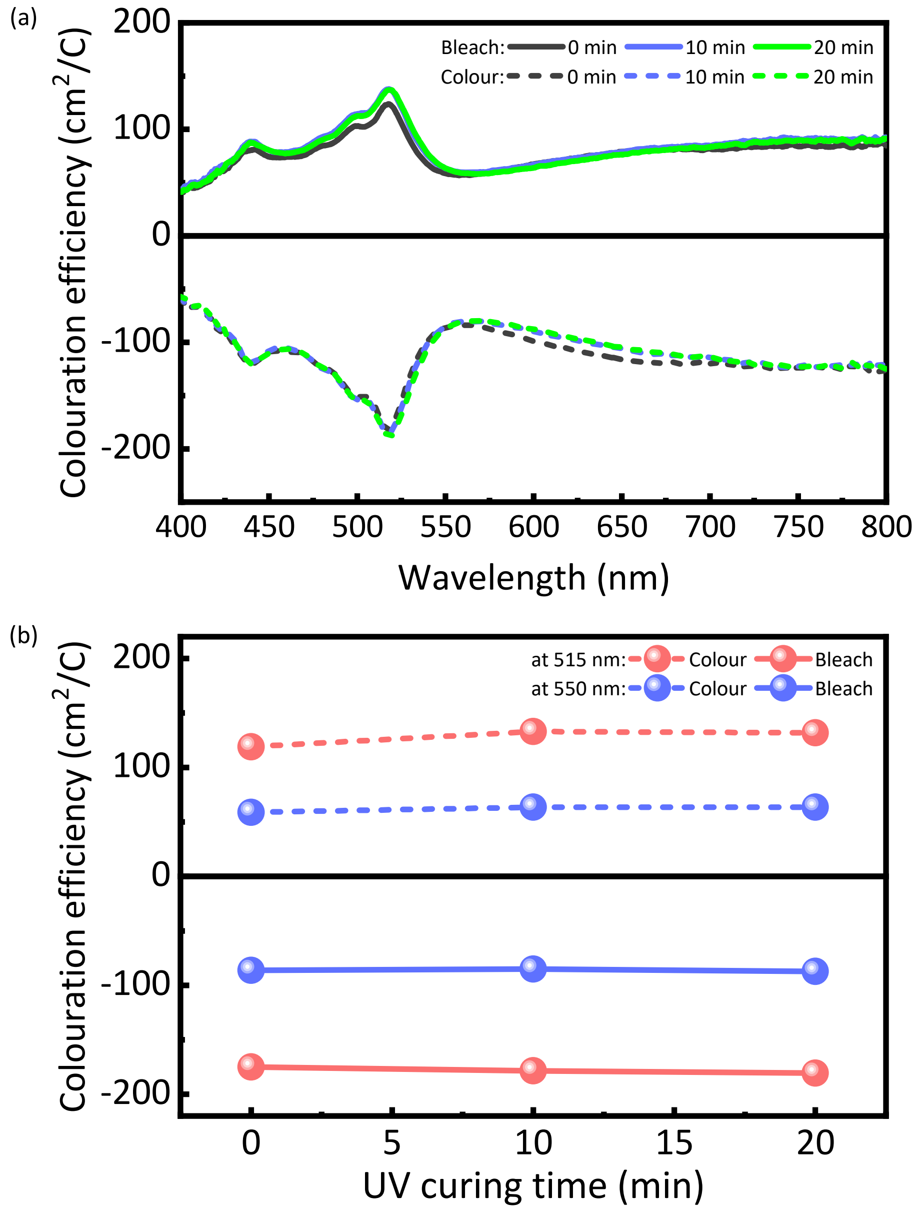

Fig. S7. Colouration efficiencies of the ECDs in (a) the visible region, and (b) specific wavelengths at 515 and 550 nm. Top and bottom lines are for the bleached and the coloured states.

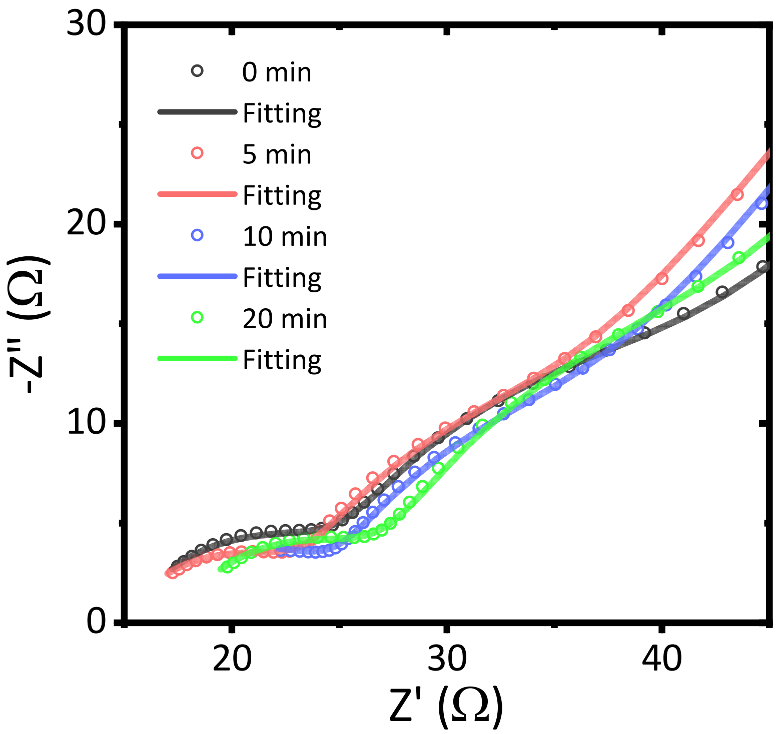

Fig. S8. Magnified high-frequency region of obtained Nyquist plots from electrochemical impedance spectroscopy (EIS) measurement for electrolyte with UV curing time of 0, 5, 10, and 20 min, and those fitting results.

Codings

1. Coding for photocell

```
#define VOLTAGE 60

int LIGHTOUT = A0;

void setup()
{
	Serial.begin(9600);

	pinMode(LIGHTOUT, INPUT);

	pinMode(9, OUTPUT);
	pinMode(10, OUTPUT);
	pinMode(11, OUTPUT);
}

void loop()
{
	int lightLevel = averageAnalogRead(LIGHTOUT);

	Serial.print("Light : ");
	Serial.print(lightLevel);

	if(lightLevel<300) {
		analogWrite(9, 0);
		analogWrite(10, 0);
		analogWrite(11, VOLTAGE);
	}
	else if(lightLevel>300 && lightLevel<700) {
		analogWrite(9, VOLTAGE);
		analogWrite(10, 0);
		analogWrite(11, 0);
	}
	else if(lightLevel>700) {
		analogWrite(9, 0);
		analogWrite(10, VOLTAGE);
		analogWrite(11, 0);
	}
	Serial.println();
	delay(500);
```

```
}

int averageAnalogRead(int pinToRead)
{
	byte numberOfReadings = 8;
	unsigned int runningValue = 0;

	for(int x = 0 ; x < numberOfReadings ; x++)
		runningValue += analogRead(pinToRead);
	runningValue /= numberOfReadings;

	return(runningValue);
}
```

2. Coding for UV photodiode

```
int UVOUT = A0

void setup()
{
	Serial.begin(9600);

	pinMode(UVOUT, INPUT);

	pinMode(5, OUTPUT);
	pinMode(6, OUTPUT);

	Serial.println("ML8511 example");
}

void loop()
{
	int uvLevel = averageAnalogRead(UVOUT);

	Serial.print("UV : ");
	Serial.print(uvLevel);

	if(uvLevel<700) {
		analogWrite(5, 60);
		analogWrite(6, 0);
	}
	else if(uvLevel>700) {
		analogWrite(5, 0);
		analogWrite(6, 60);
```

```
	}

	Serial.println();
	delay(500);
}

int averageAnalogRead(int pinToRead)
{
	byte numberOfReadings = 8;
	unsigned int runningValue = 0;

	for(int x = 0 ; x < numberOfReadings ; x++)
		runningValue += analogRead(pinToRead);
	runningValue /= numberOfReadings;

	return(runningValue);
}

float mapfloat(float x, float in_min, float in_max, float out_min, float out_max)
{
	return (x - in_min) + (out_max - out_min) / (in_max - in_min) + out_min;
}
```

Table S1. Detail summary of EIS results and calculated ionic conductivity values for UV curing time of 0, 5, 10, and 20 min.

	Parameter											
Curing Time	**R_S**	**R_1**	**CPE_1**	**n_1**	**R_2**	**CPE_2**	**n_2**	**R_3**	**CPE_3**	**n_3**	**Z_W**	**σ**
	(Ω)	***(Ω)***	***(Mho·s^n)***	**—**	***(Ω)***	***(Mho·s^n)***	**—**	***(Ω)***	***(Mho·s^n)***	**—**	***(Mho·s$^{0.5}$)***	***(S/cm)***
0 min	15.3	11.4	7.39E-06	0.95	150.4	1.67E-04	0.72	9.3	2.30E-06	0.79	2.12E-03	1.70E-05
5 min	15.0	6.6	5.40E-06	1.04	198.1	1.65E-04	0.76	7.8	3.02E-06	0.77	1.56E-03	1.29E-05
10 min	15.4	6.7	6.26E-06	1.03	209.1	1.68E-04	0.77	8.7	1.75E-06	0.79	1.55E-03	1.23E-05
20 min	17.5	10.2	6.60E-06	1.00	414.1	1.01E-04	0.81	8.6	2.10E-06	0.79	1.20E-03	6.19E-06

Ionic conductivity values (σ) are calculated from equation below:

$$\sigma = \frac{d}{R_{ct} \times A}$$

A distance between electrodes (d) are the same as the thickness of Surlyn adhesive film, and an working area (A) is defined as 2.34 cm^2. As explained in the main article, R_2 values were used as charge transportation resistance (R_{ct}) values.