Supporting Information for

Metal-free electrically conductive bioinspired adhesive polymers

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1. Experimental Procedures

1.1 Materials
All chemicals were purchased from Sigma-Aldrich, Tokyo Chemical Industry America, and VWR and used as received unless stated otherwise. Dialysis pre-wetted spectra/por membranes (1 kD and 10 kD molecular weight cut off) were purchased from Spectrum Labs. Each deionized (DI) water and 1,4-dioxane solvents were degassed prior to use by bubbling a stream of nitrogen gas through the solvent for 20 minutes. 2,2′-Azobis(2-methylpropionitrile) (AIBN) was recrystallized in methanol to remove the inhibitor prior to use. Acetonitrile was dried over a molecular sieve. Polyethylene naphthalate (PET) films (0.01, 0.09, and 0.26 mm thickness), soldering kit, light-emitting diode (LED) bulb, and 9 V battery were purchased from McMaster-Carr, Rocket Department, Inc., Great Value (Walmart Inc.), and Duracell Inc., respectively. The scale for measuring the weight of the bowling ball was purchased from Atomi Inc. Rubber cement (Elmer’s) and glue stick (all purpose glue stick, Elmer’s) were purchased from a local supermarket for comparison of our adhesive polymers to a commercial adhesive source.

1.2 Synthesis
1) N-Methacryloyl 3,4-dihydroxy-L-phenylalanine (MDOPA)
MDOPA was prepared according to the previous reported literature1. Sodium tetraborate decahydrate (19.07 g, 50 mmol, 1.0 equiv.) and 3′(3,4-dihydroxyphenyl)-L-alanine (L-DOPA) (9.86 g, 50 mmol, 1.0 equiv.) were dissolved in 500 mL of DI water and degassed with dry nitrogen gas for 20 minutes. Sodium carbonate (10.6 g) was added into the solution to adjust the pH to 9-10. The solution was degassed for another 20 minutes then followed by cooling to 0 °C in an ice bath. Next, methacryloyl chloride (6.35mL, 65 mmol, 1.3 equiv.) was added in a dropwise fashion into the solution. The pH of the solution was adjusted to 9-10 by adding additional sodium carbonate (5.3 g). That solution was degassed for 20 minutes and stirred vigorously at room temperature (23°C) for 2 hours. The resulting crude solution was acidified using concentrated HCl (25-30 mL) to bring the pH down below 2. The product was isolated using ethyl acetate (4 x 150 mL) extraction and the organic layer was collected. The organic layer was then washed with 0.1 M HCl (2 x 150 mL), brine (2 x 150 mL), and dried over MgSO4. The remaining solvent was evaporated. The obtained product was purified using column chromatography with 5% MeOH/chloroform (TLC Rf = 0.30) as an eluent to achieve
a sticky pale brown MDOPA powder (10.47 g, 78.96 % yield). $^1$H NMR (600 MHz, DMSO-d$_6$, $\delta$ ppm): 1.81 (s, 3H), 2.86 (m, 2H), 4.37 (s, 1H), 5.34 (s, 1H), 5.64 (s, 1H), 6.49-6.63 (m, 3H), 7.94 (d, 1H, $J = 7.68$), 8.72 (s, 2H), and 12.63 (s, 1H).

2) N-Methacryloyl-3,4-bis(tert-butyldimethysiloxy)-L-phenylalanine (MDOPA(TBDMS)$_2$) (1)

MDOPA(TBDMS)$_2$ (1) was prepared according to the previous reported literature$^2$. tert-Butyldimethylsilyl chloride (TBDMS-Cl) (6.78 g, 45 mmol, 3.0 equiv.) was dissolved in 35 mL of anhydrous acetonitrile in a round bottom flask. MDOPA (4 g, 15 mmol, 1.00 eqv.) was then added into the solution, followed by covering the flask and stirred to allow MDOPA to dissolve completely. The solution was cooled to 0°C in an ice bath over 30 minutes. The 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) (6.85 g, 45 mmol, 3.0 equiv.) was then added into the solution in a dropwise fashion and solution was stirred at 0°C for 4 hours. Finally, the reaction was allowed to stir for another 18 hours at room temperature. The crude product was purified by column chromatography using dichloromethane (DCM) followed by 2% v/v methanol/DCM to remove remaining TBDMS-containing impurities and then the product was obtained by using 5% v/v methanol/DCM (TLC $R_f = 0.45$). The solvent was evaporated to obtain the pure product, a pale yellow sticky powder (3.50 g, 46.98 % yield). $^1$H NMR (600 MHz, DMSO-d$_6$, $\delta$ ppm): 0.16 (s, 12H), 0.94 (s, 18H), 1.78 (s, 3H), 2.89 (m, 2H), 4.39 (s, 1H), 5.32 (s, 1H), 5.62 (s, 1H), 6.70-6.76 (m, 3H), 8.01 (s, 1H), and 12.67 (s, 1H). $^{13}$C NMR (150 MHz, DMSO-d$_6$, $\delta$ ppm): -3.76, 18.53, 18.96, 26.21, 35.92, 54.31, 119.86, 120.81, 122.08, 122.69, 131.89, 139.91, 145.03, 146.13, 167.90, and 173.67. HRMS (ESI): calculated. for C$_{25}$H$_{43}$NO$_5$Si$_2$ [M+Na]$^+$ = 516.2572, found 516.2591.

3) Poly(MDOPA(TBDMS)$_2$-co-3-sulfopropyl methacrylate) (Poly MDOPA(TBDMS)$_2$-co-SPM, PMTS) (3)

MDOPA(TBDMS)$_2$ (1) (1.57 g, 3.18 mmol, 1.00 equiv.), SPM (2) potassium salt (4.44 g, 18 mmol, 5.67 equiv.), and AIBN (0.104 g, 0.64 mmol, 0.2 equiv.) were dissolved in 18 mL of degassed DI water/1,4-dioxane (ratio of 1:1 by volume). The solution mixture was degassed by bubbling a stream of nitrogen gas through the solution for 15 minutes. Reaction was then allowed to stir for 16 hours at 65°C. The crude product was purified by dialysis using 10 kD molecular weight cut off membrane in methanol. Methanol was changed every 3 hours and this was repeated four times. Dialysis was repeated in DI water and the water was exchanged four times. The final product was freeze-dried to obtain a white colored polymer (4.62 g, 75.80 % yield).

4) Poly(MDOPA-co-SPM): poly(3,4-ethylenedioxythiophene) (PMS:PEDOT) (5)

PMTS (3) (4.32 g) was dissolved in 162 mL of DI water followed by degassing with nitrogen gas for 20 minutes while being stirred. 3,4-ethylenedioxythiophene (EDOT) (0.54 g) was added into the solution followed by degassing with nitrogen gas for another 20 minutes while being stirred. The solution was then placed inside of the cold room (4°C) and allowed to cool down for 15 minutes while being stirring. Then sodium persulfate (SPS, 1.62 g) and Iron(III) sulfate hydrate (32.4 mg) were added to the solution as an initiator and catalyst, respectively and the reaction solution was vigorously stirred at 4°C for 23 hours. After that the impurities were removed by dialysis using a 1 kD molecular weight cut off membrane in DI water for 1 day to remove the catalyst and initiator. The purified PMTS:PEDOT (4) then underwent deprotection to remove TBDMS by adding concentrated HCl with ratio of 1 mL polymer
solution to 0.2 mL concentrated HCl. The impurities were then removed by dialysis using 10 kD molecular weight cut off membrane in methanol followed by DI water. Methanol and DI water were each replaced every 3 hours and replacement was repeated four times. The dried PMS:PEDOT (5) was obtained (3.51 g) after freeze-drying. It is important to note that gelation occurs when the EDOT is polymerized by oxidative radical polymerization\(^3\) without the protection of the catechol group. It has been reported that semi-quinone radicals are generated by the reaction between the 3,4-dihydroxy-L-phenylalanine (DOPA) and oxidized DOPA\(^4\).\(^6\). Thus, gelation occurs when the catechol-containing monomer is polymerized by oxidative polymerization due to the reaction between semi-quinone radicals in different polymer chains\(^6\). Crosslinking also can occur due to the reaction between catechol radicals generated by the reaction between the DOPA and radical in one polymer chain and propagating radical in another polymer chain\(^7\).\(^8\). Taking these factors into account, the observed that the gelation during the polymerization of EDOT without catechol protection is due to the crosslinking by the semiquinone-radicals and/or catechol radicals.

5) Poly(SPM) (PSPM) (6)
SPM (2) potassium salt (2000 mg, 8.12 mmol, 1.00 equiv.) and AIBN (40mg, 0.24 mmol, 0.03 equiv.) were dissolved in 6.86 mL of degassed DI water/1,4 dioxane (ratio of 1:1 by volume). The solution mixture was degassed by bubbling a stream of nitrogen gas through the solution for 15 minutes. Reaction was then allowed to stir for 16 hours at 65°C. The crude product was purified by dialysis (10 kD molecular weight cut off dialysis tube) in DI water. DI water was changed every 3 hours and repeated four times. The final product was freeze-dried to obtain the white colored polymer (1683.6 mg, 82.83 % yield).

6) PMS (8)
Deprotection of PMTS (3) was carried out by removal of the TBDMS groups using concentrated HCl. 200 mg of PMTS (3) was dissolved in 20 mL of DI water and 4 mL of concentrated HCl. The solution was allowed to stir at 60°C for 12 hours. The deprotected polymer, PMS (8), was then purified by dialysis using 10 kD molecular weight cut off membrane in methanol and followed again by DI water. Methanol and DI water were each replaced every 3 hours and the replacement was repeated four times. PMS (8) was obtained (149.3 mg) after freeze-drying the purified solution.

7) PSPM: PEDOT (7)
PSPM (6) (490 mg) was dissolved in 21 mL DI water followed by degassing the solution with a stream of nitrogen gas for 20 minutes while being stirred. EDOT (70 mg) was then added into the PSPM (6) solution followed by degassing for another 20 minutes while being stirred. The solution mixture was brought into cold room (4 °C) and cooled down for 15 minutes with stirring. SPS (210 mg) as an initiator and iron(III) sulfate hydrate (4.2 mg) as a catalyst are then added into the solution sequentially while being stirred. The reaction was vigorously stirred at 4°C for 23 hours. The product was purified by dialysis using 1 kD molecular weight cut off membrane with DI water for 1 day to remove the impurities. The PSPM:PEDOT (7) was obtained (418 mg, 74.64 % yield) after freeze-drying the solution.

1.3 Characterizations
Nuclear magnetic resonance (NMR) spectra of the samples were recorded on a Bruker Avance III 600 MHz NMR spectrometer. Electrical resistance was measured by Mitsubishi
Chemical Analytech Loresta-GX MCP-T700. Thickness of the polymer coated on the cover glass was observed by FEI Nova 400 nanoSEM. Polymer thickness on the PET film was measured by the Mitutoyo thickness gage. TGA was performed using TA instrument simultaneous thermal analyzer SDT Q600. The adhesion strength test was performed by the Shimadzu EZ-LX tensile-compression tester quipped with 200N force SM-200N-168 transducer. The RMS surface roughness and surface morphology were obtained using AFM (MFP-3D AFM, Asylum Research) machine. Transmittance was recorded using PerkinElmer UV/vis spectrometer (model: Lambda 950). Elemental analysis was performed via Atlantic Microlab, Inc.

1.4 Electrical conductivity measurement

The glass substrate was cleaned with sequential sonication in DI water, acetone, and isopropyl alcohol (30 minutes in each solvent). To measure the electrical conductivity of PSPM (6), PSPM:PEDOT (7), PMS (8), and PMS:PEDOT (5), the 13 mg of dried polymer was dissolved in 400 µL of DI water separately and then the solutions were stirred for 10 min followed by sonication for another 10 minutes. After that, the solutions were stirred for 12 hours at room temperature (23 °C). The polymer solutions were then casted on the pre-cleaned glass substrates and spin-coated at 500 rpm for 10 seconds followed by 1000 rpm for 30 seconds. The spin-coated polymer solutions were dried in ambient air for 1 day.

For the investigation of the electrical conductivity of PMS:PEDOT (5) depending on drying temperatures (Fig. 3b), the 13 mg of dried PMS:PEDOT (5) was mixed with 400 µL of DI water, the solution was stirred for 10 minutes, and then the mixture was sonicated for 10 minutes. After that, the solution stirred for 12 hours at room temperature (23 °C). The polymer solution was spread on the pre-cleaned glass substrate and spin-coated at 1000 rpm for 10 seconds followed by 1500 rpm for 30 seconds. The spin-coated polymer solution was dried at desired temperature for 10 minutes. The applied drying temperatures were 23, 50, 111, 165, 226, and 295 °C. The spin-coated PMS:PEDOT (5) film which was spin-coated at 1000 rpm for 10 seconds followed by 1500 rpm for 30 seconds and then dried at 23 °C for 10 minutes was also used to investigate the RMS surface roughness and surface morphology using the AFM machine.

To study the effects of the glycerol addition on the electrical conductivity of the PMS:PEDOT (5), the 13 mg of dried PMS:PEDOT (5) was put into 400 µL of DI water and then the solution was stirred for 10 min followed by sonication for 10 minutes. The desired amount of glycerol was added to the polymer solution. Next, the mixture was stirred for 12 hours at room temperature (23 °C). The weight ratios between the PMS:PEDOT (5) and glycerol were controlled at 13:0, 13:8, 13:16, and 13:24. The PMS:PEDOT (5) sample which was prepared using the conducting ink of weight ratios (PMS:PEDOT (5) to glycerol) of 13:0, 13:8, 13:16, and 13:24 were denoted as PMS:PEDOT-0%, 38%, 55%, and 65%, respectively. The mixture was then cast on the pre-cleaned glass substrate. After that, the mixture was spin-coated at 1000 rpm for 10 seconds followed by 1500 rpm for 30 seconds. The spin-coated polymer solution was dried at 134 °C for 10 minutes as an annealing process used to dry water and glycerol as well as for a heat treatment. The PMS:PEDOT-38% was used to investigation of the RMS surface roughness and morphology using the AFM.

The direct current electrical conductivity measurements of samples were conducted with the four-probe method using a LORESTA-GX/MCP-T700. The thickness of the polymer layer on the glass was obtained using scanning electron microscopy.
1.5 Adhesion strength test
Adhesive was prepared by mixing 14 mg of polymer with 42 µL of DI water. The wetted polymer was left at 23°C for 29 hours, producing a water-swollen polymer adhesive. The water-swollen polymer was then applied on a 1.4 cm x 0.7 cm rectangle area of 1.4 cm x 9.0 cm sized PET film (0.26 mm thickness) and topped with another PET film (0.26 mm thickness) subsequently (overlapped area of 1.4 cm x 0.7 cm unless otherwise specified). An aluminum block (130 g) was then placed on top of the overlapped area for 20 minutes. The prepared sample was then cured for total 24 hours at room temperature, 23°C, prior to the lap shear test. For the PMS:PEDOT (5) sample, the overlapped area was reduced to 0.7 cm x 0.7 cm (not 1.4 cm x 0.7 cm) due to the maximum allowed force of the tensile tester. To prepare the PMS:PEDOT (5) sample, water-swollen adhesive was first prepared by adding 21 µL DI water to 7 mg of dried polymer. The wetted polymer was left at 23°C for 29 hours, yielding a water-swollen polymer adhesive. The water-swollen polymer was then transferred onto a 0.7 cm x 0.7 cm area of 0.7 cm x 9.0 cm sized PET film and subsequently covered with another PET film (overlapped area of 0.7 cm x 0.7 cm), resulting in one sample. The overlapped area of two samples arranged side by side was compressed under 130 g of weight for 20 min. Then, the prepared samples were cured at room temperature (23°C) for 24 hours prior to lap shear test.

Adhesion strength was measured by conducting a lap shear test using Shimadzu EZ-LX tensile-compression tester machine. The sample was mounted on the lap shear machine grip and pulled apart at speed of 1 mm/min. The data, force vs. displacement, was recorded until the adhesion failure occurred. This testing was repeated at least 4 times and the maximum adhesion strength (MPa) was obtained by dividing the average maximum force (N) by the overlapped area of PET films (m²). The standard error of the mean was also reported.

1.6 TGA
Thermal stability of the conducting polymer adhesive was investigated using TA instrument SDT Q600. Two TA aluminum pans were used as reference and sample pan. Argon (100 mL/min) and air (20 mL/min) were used during the thermal stability test of the polymer. The water content of the samples was removed at 100°C for 20 minutes. Thermal stability was then measured from 100°C to 1000°C at 10°C/minute. The sample mass (mg) vs. temperature (°C) was collected. The sample mass was then normalized with the initial mass to get mass fraction (%) and plotted against temperature (°C).

1.7 Transmittance
The PMS:PEDOT (5) film was prepared by following method. 13 mg of dried PMS:PEDOT (5) was mixed with 400 µL of DI water. Then, the solution was stirred for 10 min followed by sonication for 10 minutes. After that, the solution was stirred for 12 hours at room temperature (23 °C). The polymer solution was cast on the pre-cleaned glass substrate and spin-coated at 1000 rpm for 10 seconds followed by 1500 rpm for 30 seconds. The spin-coated polymer solution was dried at 23 °C for 10 minutes. The PMS:PEDOT-38% film was prepared by following method. 13 mg of dried PMS:PEDOT (5) was put into 400 µL of DI water and then the solution was stirred for 10 min followed by sonication for 10 minutes. After that, 8 mg of glycerol was added to the polymer solution. The mixture was stirred at room temperature (23 °C) for 12 hours. The mixture solution was then spread on the pre-cleaned glass substrate and then spin-coated at 1000 rpm for 10 seconds followed by 1500 rpm for 30 seconds. The spin-coated solution was dried at 134 °C for 10 minutes. The
transmittance of prepared PMS:PEDOT (5) and PMS:PEDOT-38% films were recorded on the UV/vis spectrometer.

1.9 Cytotoxicity test
Cytotoxicity of the samples were assessed using HEK293A cell. Two stock solutions were prepared: 1) a solution containing 13 mg of PMS:PEDOT (5) and 400 µL of DI water; 2) a solution containing 13 mg of PMS:PEDOT (5), 8 mg of glycerol, and 400 µL of DI water. The PMS:PEDOT (5) containing stock solution was prepared by the following procedures. Firstly, the empty vial was sterilized for 1 hour inside the Luzchem LCZ-4X UV-photoreactor equipped with 14 bulbs (8W, 254 nm). The 13 mg of dried PMS:PEDOT (5) was then added to the sterilized vial. After that, 400 µL of DI water was added to the vials and stirred for 10 minutes followed by sonication for 10 minutes. The solution was allowed to stir for 12 hours at room temperature (23 °C). In the case of the PMS:PEDOT (5) and glycerol containing stock solution, empty vials were sterilized by placing them inside Luzchem LCZ-4X UV-photoreactor equipped with 14 bulbs (8W, 254 nm) for 1 hour. 13 mg of dried PMS:PEDOT (5) was transferred to the sterilized vial. The dried polymer was then mixed with 400 µL of DI water. The mixture was stirred for 10 minutes followed by sonication for 10 minutes. 8 mg of glycerol was added to the polymer solution and the mixture was stirred for 12 hours at room temperature (23 °C). Once the two stock solutions were prepared they were sterilized for 30 minutes inside the Luzchem LCZ-4X UV-photoreactor equipped with 14 bulbs (8W, 254 nm). For a quantitative analysis, the same number of cells were plated in twelve 35 mm dishes. The solution containing PMS:PEDOT (3) and DI water was diluted in cell culture medium DMEM containing 10% FBS with 400 µg of PMS:PEDOT (5)/mL and filtered through a 0.22 µm syringe filter for sterilization. In the case of the stock solution consisting of 13 mg of PMS:PEDOT (5), 8 mg of glycerol, and 400 µL of DI water, the solution was diluted in cell culture medium DMEM containing 10% FBS with (400 µg of PMS:PEDOT (5)+250 µg of glycerol)/mL and filtered through 0.22 µm syringe filter for sterilization. Four dishes were treated with 400 µg of PMS:PEDOT (5)/mL for 48 hours and another 4 dishes were treated with 650 µg of PMS:PEDOT-38% (400 µg of PMS:PEDOT (5)+250 µg of glycerol)/mL for 48 hours. The last 4 dishes were treated with the same volume of water and served as the control. After the cells are trypsinized (to detach the cells into suspension) and stained with Trypan Blue, Live cells (unstained cells) from each dish were counted using cover slips and a glass hemocytometer. Live cells do not take up the dye. The average live cell number and standard error of mean of both the control and treatment groups were obtained by using the cell counting result.

1.10 Electrical capability and printed features of the PMS:PEDOT (5)
We first cut the PET (0.09 mm thickness) film to make a long line mask. The patterned PET mask was then placed on the other PET substrate (0.26 mm thickness) and edge of the mask was fixed with scotch tape and then the water-swollen PMS:PEDOT (5) which was prepared for application of PMS:PEDOT (5) as the ECAs to electrically and physically connect the non-flexible printed circuit board (PCB) and electronic components (ECs) (detail procedure at below) was applied on the pattern. We swept down over the pattern using an aluminum bar (130 g). After that, the pattern mask was detached from the PET substrate and the stencil-printed water-swollen PMS:PEDOT (5) line was allowed to dry at room temperature (23 °C) for 24 hours, resulting in the PMS:PEDOT line (3 mm width, 0.05 mm thickness, and 6 cm length). Last, we connected the stencil-printed PMS:PEDOT (5) line to the power source (27
V) and a LED bulb.

1.11 Adhesion capability of PMS:PEDOT (5)

The adhesive was first prepared by mixing 14 mg of dried PMS:PEDOT (5) with 42 µL DI water. The wetted polymer was left at 23°C for 29 hours, producing a water-swollen PMS:PEDOT (5) adhesive. The water-swollen polymer was applied on 1.4 cm x 0.7 cm rectangle area of PET film (0.26 mm thickness) and another PET film (0.26 mm thickness) was subsequently placed on the applied adhesive. An aluminum block (130 g) was then placed on top of the overlapped area for 20 minutes. The prepared sample was cured for total 24 hours at room temperature, 23°C. The 14.78 lb bowling ball was hung up to the one end of the PET films. After that the experimenter lifted up another end of the PET films.

1.12 Application of PMS:PEDOT (5) as the ECAs to electrically and physically connect the non-flexible PCB and ECs

The 1.3 g of dried PMS:PEDOT (5) was first added into empty glue bottle and 4.2 mL of DI water was added. After that, the cap of the glue bottle was closed and sealed with parafilm and polymer was allowed to swell at room temperature (23 °C) for 29 hours. Meanwhile, we inserted through-hole components (THCs) to the PCB. The THCs and PCB were bound by the water-swollen PMS:PEDOT (5) and the applied polymer was allowed to dry at room temperature (23 °C) for 24 hours on the board.

1.13 Application of PMS:PEDOT (5) as the ECA to electrically connect the disconnected electrical lines on the thin, flexible, and low-cost PET substrate

First, the conducting ink consisting of PMS:PEDOT (5), glycerol, and DI water with a weight ratio of PMS:PEDOT (5) to glycerol of 13:8 was prepared. The stock solution consisting of DI water (4 g) and glycerol (0.32 g) was prepared for homogeneous mixture. 2.16 g of stock solution was added to 0.26 g of dried PMS:PEDOT (5). After that, the mixture was vigorously stirred for 24 hours at room temperature (23 °C). Meanwhile, we cut out the PET (0.09 mm thickness) film to create a mask of disconnected lines. The patterned PET mask was placed on the ultrathin (0.01 mm thickness) PET substrate and edge of the mask was fixed with tape, and then the prepared conducting ink was applied on the pattern. We then swept down over the pattern using an aluminum bar (130 g). The patterned mask was detached from the ultrathin PET substrate and the pattern printed ultrathin PET film was placed on a hot plate (134 °C) and was kept at 134 °C for 10 minutes to dry the glycerol, yielding a disconnected PMS:PEDOT-38% lines (0.3 cm width, 0.01 mm thickness). After the glycerol dried, an LED bulb was adhered to the stencil-printed PMS:PEDOT-38% electrical line using water-swollen PMS:PEDOT (5) which was previously prepared for application of PMS:PEDOT as the ECAs to electrically and physically connect the non-flexible PCB and ECs. The applied water-swollen PMS:PEDOT (5) was allowed to dry at room temperature (23 °C) for 24 hours. One more disconnected circuit with a LED bulb on the ultrathin (0.01 mm thickness) PET flexible substrate was prepared for the traditional soldering experiment.

Finally, power (27 V) was applied to the line and the water-swollen PMS:PEDOT (5) was applied on the disconnected area between the lines (Supplementary Movie 2). After confirming that the LED bulb lights up, the applied water-swollen PMS:PEDOT (5) was allowed to dry for 24 hours at room temperature (23 °C) and then power (27 V) was applied to line again (Fig. 4b). In the case of the experiment employing the traditional soldering
technique, the power (27 V) was first applied to the line, and then we applied the solder (LF-99 solder, ELENCO®) on the disconnected area between the lines using the soldering iron (Supplementary Movie 3).
2. Results and Discussion

Figure S1. $^1$H NMR spectrum of MDOPA in DMSO-d$_6$.

Figure S2. $^1$H NMR spectrum of MDOPA(TBDMS)$_2$ (1) in DMSO-d$_6$. 
Figure S3. $^{13}$C NMR spectrum of MDOPA(TBDMS)$_2$ (1) in DMSO-d$_6$.

Figure S4. $^1$H NMR spectrum of PMS:PEDOT (5) in DMSO-d$_6$. 
Figure S5. Force vs. displacement curve of (a) PSPM (6), (b) PSPM:PEDOT (7), (c) PMS (8), (d) PMS:PEDOT (5), (e) PMTS:PEDOT (4), (f) glue stick, and (g) rubber cement.
Figure S6. Photograph of a bowling ball weighed by a balance. A 14.78 lb bowling ball in the photograph was utilized to exhibit the adhesion capability of the PMS:PEDOT (5) (Figure 2e and Video S1).

Figure S7. TGA curves of PSPM (6), PMS (8), and PMS:PEDOT (5).

The PSPM (6), PMS (8), and PMS:PEDOT (5) commonly shows continuous weight loss until 100 °C, corresponding to continuous removal of water in the films. The total amount of water in the PSPM (6), PMS (8), and PMS:PEDOT (5) was revealed as ~10, ~8, and ~16 %, respectively. The PSPM (6) displays significant weight loss at 300-450 °C showing the decomposition. For the PMS (7), after the water removal, significant weight loss occurs at ~180-300 °C and then again ~300-450 °C. This shows that substantial thermal decomposition of polyMDOPA (PMDOPA) first occurs at ~180-300 °C. In the case of the PMS:PEDOT (5), it exhibits a similar thermal decomposition profile to PMS (8). Thus, it was difficult to determine an exact temperature range of PEDOT thermal decomposition. In a previous publication, the first substantial thermal decomposition of pure PEDOT occurs at 225-300 °C and then continues to ~400 °C. The observed similarity between PMS:PEDOT (5) and PMS (8) is due to the overlap of decomposition temperature of PEDOT with that of PMDOPA and PSPM (6).
Compared to the newly synthesized PMS:PEDOT (5), thermal property test of silver containing electronically conducting adhesives has been performed in diverse testing conditions. Weight loss of TGA thermograms below 150 °C represents evaporation of water from the sample. Most silver containing electronically conducting adhesives demonstrates gradual loss of weight starting from 150 °C and then continues until 350 – 400 °C. Herein, the silver fillers also decompose near 400 °C. Overall, our synthesized conducting adhesives possess similar thermal properties to other previously developed silver containing electronically conducting adhesives.

Figure S8. Transmittance and surface resistance ($R_s$) of PMS:PEDOT (5) film. The left inset photograph shows the PMS:PEDOT (5) film placed over the word “chemistry” printed on photo paper. The right inset photograph displays the word “chemistry” printed on photo paper. The bars in the inset photographs represent 0.5 cm.
Figure S9. AFM image of the PMS:PEDOT (5) film (inset: cross section measurement along the white line shown in AFM image).

Supplementary Tables

Table S1. Performance comparison of our ECA with other ECAs in the literature.

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<th>Composition</th>
<th>E.C. (S/cm)</th>
<th>A.S. (MPa)</th>
<th>Reference[a]</th>
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<td>5.0 x 10¹</td>
<td>1.36 x 10⁻¹</td>
<td>41</td>
</tr>
<tr>
<td></td>
<td>Nano carbon black/polyacrylate</td>
<td>4.5 x 10¹</td>
<td>1.12 x 10⁻¹</td>
<td>41</td>
</tr>
<tr>
<td></td>
<td>Expanded graphite/silane polymer</td>
<td>2.4 x 10⁻¹</td>
<td>1.25 x 10⁻³</td>
<td>39</td>
</tr>
<tr>
<td></td>
<td>Short carbon fiber/novolac resin</td>
<td>7.5 x 10⁻⁷</td>
<td>1.61</td>
<td>42</td>
</tr>
<tr>
<td></td>
<td>Carbon fiber/novolac resin</td>
<td>1.5 x 10⁻⁹</td>
<td>1.09 x 10¹</td>
<td>42</td>
</tr>
<tr>
<td>Intrinsically conducting</td>
<td>PEDOT/PMDOPA/PSPM</td>
<td>3.3 x 10⁻¹</td>
<td>1.62</td>
<td>present</td>
</tr>
<tr>
<td>polymer-based ECA</td>
<td>Polyaniline/carbon nanotube/epoxy</td>
<td>4.4 x 10⁻⁶</td>
<td>3.90</td>
<td>6</td>
</tr>
</tbody>
</table>

[a] The references in Table S1 present the references in the main article.
Table S2. Elemental analysis results of synthesized polymers.[a]

<table>
<thead>
<tr>
<th>Sample</th>
<th>C (%)</th>
<th>H (%)</th>
<th>N (%)</th>
<th>S (%)</th>
<th>Theoretical Segmental Ratio of Polymers[b]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMTS (3)</td>
<td>48.3</td>
<td>7.4</td>
<td>0.8</td>
<td>10.2</td>
<td>PMDOPA / SPM (2) 1 / 5.67</td>
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<tr>
<td>Theoretical</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMTS (3)</td>
<td>37.9</td>
<td>5.77</td>
<td>0.74</td>
<td>9.23</td>
<td></td>
</tr>
<tr>
<td>Experimental</td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMS (8) Theoretical</td>
<td>46.1</td>
<td>6.7</td>
<td>0.8</td>
<td>11.7</td>
<td>MDOPA / SPM (2) 1 / 5.67</td>
</tr>
<tr>
<td>PMS (8) Experimental</td>
<td>37.9</td>
<td>6.14</td>
<td>0.85</td>
<td>11.64</td>
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<tr>
<td>PMTS:PEDOT (4)</td>
<td>48.6</td>
<td>7.1</td>
<td>0.7</td>
<td>11.6</td>
<td>PMDOPA / SPM (2) / EDOT 1 / 5.67 / 1.56</td>
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<tr>
<td>Theoretical</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>PMTS:PEDOT (4)</td>
<td>38.54</td>
<td>5.64</td>
<td>0.75</td>
<td>10.38</td>
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<tr>
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<tr>
<td>PMS:PEDOT (5)</td>
<td>46.7</td>
<td>6.4</td>
<td>0.8</td>
<td>13.1</td>
<td>MDOPA / SPM (2) / EDOT 1 / 5.67 / 1.56</td>
</tr>
<tr>
<td>Theoretical</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMS:PEDOT (5)</td>
<td>39.72</td>
<td>5.95</td>
<td>0.8</td>
<td>12.42</td>
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</tr>
<tr>
<td>Experimental</td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

[a] The elemental content was consistent with the expected polymer structure. Experimental results of two atoms, C and H, in PMTS (3) and PMS (8) were less than S compared to the theoretical ratio, because MDOPA and protected MDOPA were included in the produced copolymer slightly less than input ratio of monomers. For PMTS:PEDOT (4) and PMS:PEDOT (5), experimental C and H were less than theoretical value because EDOT was used as a limiting agent in the oxidative polymerization.

[b] The shown ratio is initially added monomer’s molar ratio for polymerizations.
3. References


