Electronic Supporting Information (ESI)

Light-stimulated paper/polymer bilayer actuators incorporated with photosensitizers for enhanced actuation

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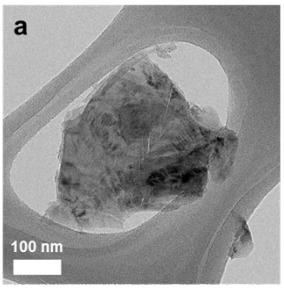
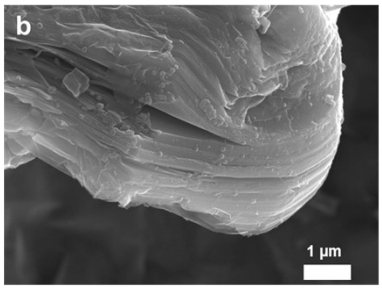
A) Methods

Materials: Methylene blue**,** Agarose, Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), Potassium Chloride (KCl), Ti_3AlC_2, Hydrochloric acid (HCl), and lithium fluoride (LiF) were purchased from Sigma-Aldrich (USA). K19 dye was purchased from Ossila Ltd (Korea). Polyethylene glycol (PEG) was purchased from Yakuri Pure Chemicals Co. Ltd (Japan), and Potassium Chloride (KCl) was purchased from Junsei Chemicals Co. Ltd (Japan). Distilled water was produced by Milli-Q system (Millipore, USA). pE4000 was purchased from CoolLED (UK).

Experimental details:

Synthesis of MXenes





c

El	AN	Series	unn. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
C	6	K-series	42.12	42.12	72.51	2.01
Ti	22	K-series	50.42	50.42	21.77	1.61
Al	13	K-series	7.46	7.46	5.72	0.30
Total:			100.00	100.00	100.00	

Fig. S1 Microscopic images of MXenes (a) TEM image, (b) SEM image, and (c) EDS analysis.

The $Ti_3C_2T_x$ MXenes NSs were prepared by the mild etching method. To synthesize the MXenes NSs, 1 g of Ti_3AlC_2 powder was slowly added into 20 mL of 6 M HCl solution containing 1 g of LiF. The solution was stirred at 35 °C under continuous stirring for 24 h. The black precipitates were centrifuged at 3,500 rpm for 5 min and washed with distilled water until the pH reached to approximately 6. The resulting $Ti_3C_2T_x$ solution was re-dispersed in distilled water by sonication for 1 h. The suspension was carried out in the centrifuge at 3,500 rpm for 1 h to eliminate the un-exfoliated $Ti_3C_2T_x$. The final sample was dried at 80 °C for 10 h.

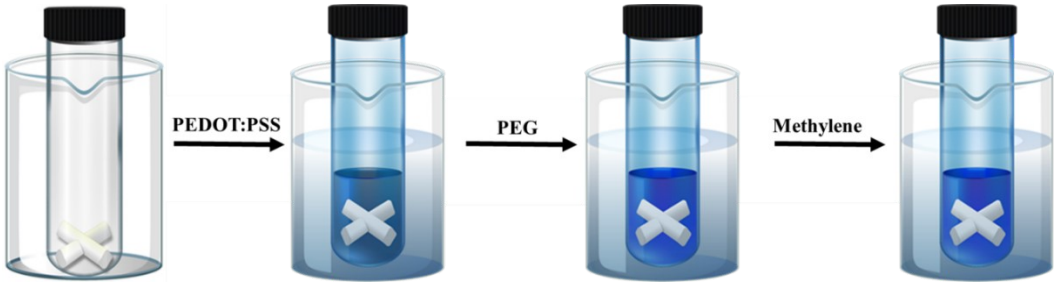


Fig. S2 Stepwise synthesis of the polymer blend.

Synthesis of Polymer Blend

The polymer blend was synthesized in a glass vial filled with water to maintain a uniform temperature. 5 mg of Agarose was taken with 10 ml of PEDOT:PSS was taken in a 10 ml of glass vial in stirring mode at 90 °C overnight. Once agarose gets completely dissolved in PEDOT:PSS, 500 µl of PEG is added and left for stirring for another 2 hr. 0.1 M concentration of MXenes solution was sonicated for 15 min at 30 °C, of which 200 µl was added to the polymer blend and stirred for another 2 hr. This is followed by the addition of 50 mg of methylene blue dye and left overnight stirring. Finally, 5 mg of KCl is added and stirred for another 2 hr, making sure the polymerization is fully completed. This leads to the formation of the polymer blend.

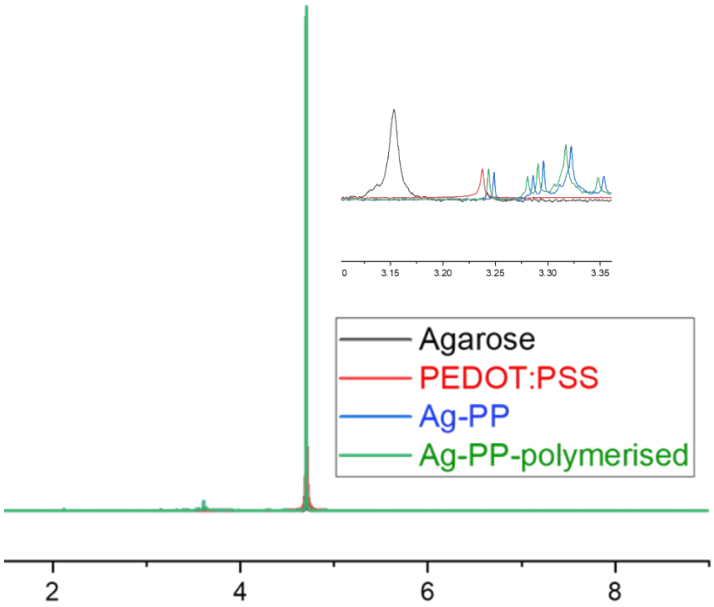


Fig. S3 ^{1}H NMR of the polymer blend and its individual components. Inset showing the expansion of regions from 3 to 4 ppm.

Electrochemical analysis of the polymer blend

The electrochemical analysis of the developed polymer blend was conducted under a three-electrode system composed of the ITO as a working electrode, Ag/AgCl as a reference electrode, and Pt as a counter electrode. The electrochemical signals were analyzed using an electrochemical analyzer, CHI-660E (CH Instruments, Inc., USA). Cyclic voltammetry (CV) analysis was conducted under a potential range from 0.6 V to -0.2 V, a scan rate of 0.05 V/s,

and a sample interval of 0.001 V to investigate the electrochemical properties of the synthesized polymer blend.

Measurement of the photocurrent generated by the polymer blend

The photocurrent generated by the polymer blend was measured through chronoamperometry under the same experimental conditions as CV. For the light source, the pE-4000 illumination 5 system (CoolLED, UK) was used as a light-emitting diode (LED) illuminator for the irradiation of the light with a power output of 60 W. Experiments to measure photocurrent were conducted in a dark room to avoid interference from undesired light sources.

Comparison of variation in the paper/polymer blend in the presence of light

a

	Distance (mm)	Velocity (mm/s)
Agarose/PEDOP:PSS	1.44 ± 0.38	0.6
Agarose/PEDOT: PSS/MXene/Mb	9.1 ± 0.82	2.1
Agarose/PEDOT: PSS/Mb	5.4 ± 2.77	1.7
Agarose/PEDOT: PSS/MXene	3.46 ± 0.82	1
Agarose/PEDOT: PSS /PEG/MXene/Mb	18.4 ± 1.14	3.3

b

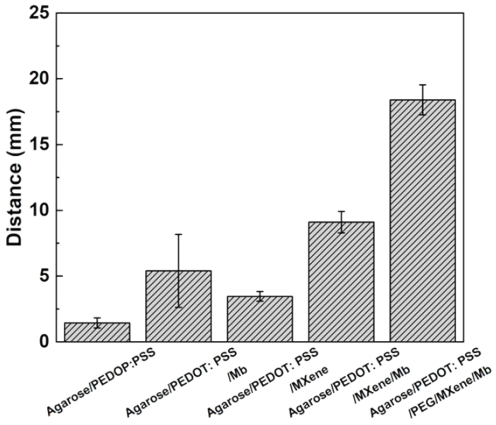


Fig. S4 Comparison of variation in the paper/polymer blend in the presence of light. (a) comparative study, and (b) actuation using different variations in the components used in the polymer blend.

Stimuli source	stimulus	Actuators (Materials)	Degree of Actuation	ref
Electric/Thermal	8V, 0.14 cm^{-1}	PEDOT:PSS/Polypropylene	Bending Angle-360 °	1
Electric	80 °C	PEDDOT:PSS	Bending Angle-80 °	2
Thermal	300 $mVcm^{-2}$	Graphene/Polymer	Light =1.4 cm^{-1} Humidity=1.2 cm^{-1}	3
Thermal	10 V	Inkjet Polymer	Displacement-10 mm	4
Electric	Moisture	Hydrophobic wax	Inchworm-33 µm/s Insect-10.9 mm/s	5
Thermal	80 – 90 V	Polyactide	~50 °	6
Thermal	800 $mVcm^{-2}$	PEDOT:PSS/Mxene	Curvature-1.8 cm^{-1}	7
Light		PEDDOT:PSS/Mxene/dye	3.3 mm/s	This study

Fig. S5 Comparison of characteristics of different paper-based actuators

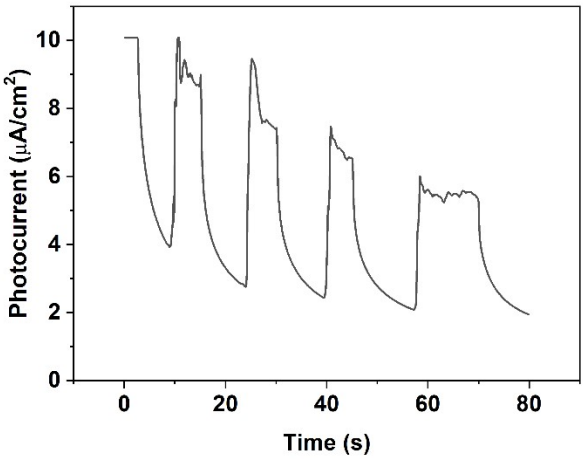


Fig. S6 Photocurrent generated polymer blend on irradiation

Actuation of polymeric blend/paper in the presence on irradiation in terms of speed vs time

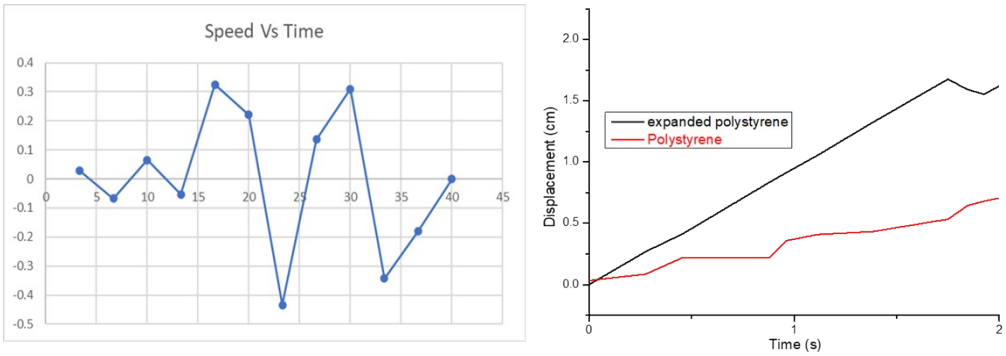


Fig. S7 Actuation of paper/polymer blend on irradiation.

References

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