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Supplementary Information

A Supramolecular Assembly of Cross-linked Azobenzene/Polymers for a High-Performance Light-driven Actuator

Chengqun Qin,[a] Yiyu Feng,[a] Wen Luo,[a] Chen Cao,[a] Wenping Hu,[b] Wei Feng*,[a]

[a] School of Materials Science and Engineering, Tianjin Key Laboratory of Composite and Functional Materials, Tianjin University, Tianjin 300072, China

[b] Beijing National Laboratory for Molecular Sciences, Key Laboratory of Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China

*Corresponding author's Email: weifeng@tju.edu.cn

The self-assembly of AAZO/PDAC was indicated by different solubilities of films in organic solvents. The solubility was investigated qualitatively by adding the film (5 mg) in 5mL water or organic solvents at room temperature.

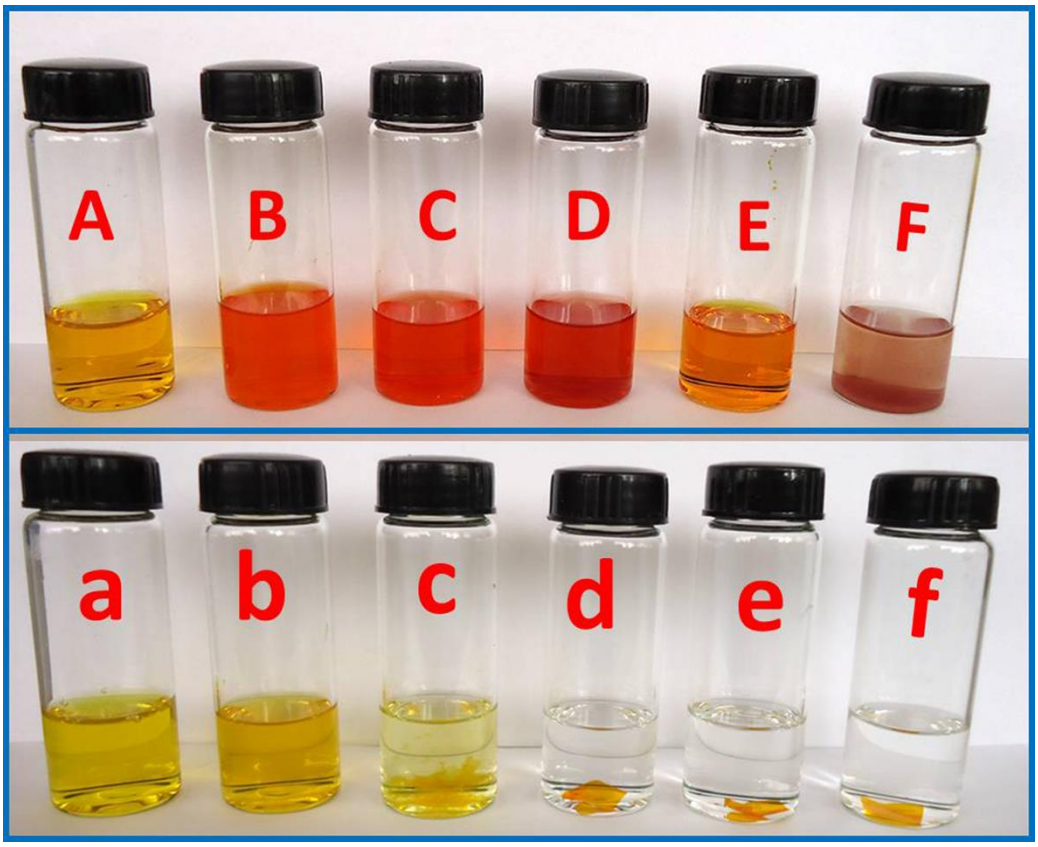


Figure S1. (a) Photographs of AAZO (A-F) and AAZO/PDAC films (a-f) in different solvents: H_2O, methanol, ethanol, DMF, acetone, $CHCl_3$ (from left to right).

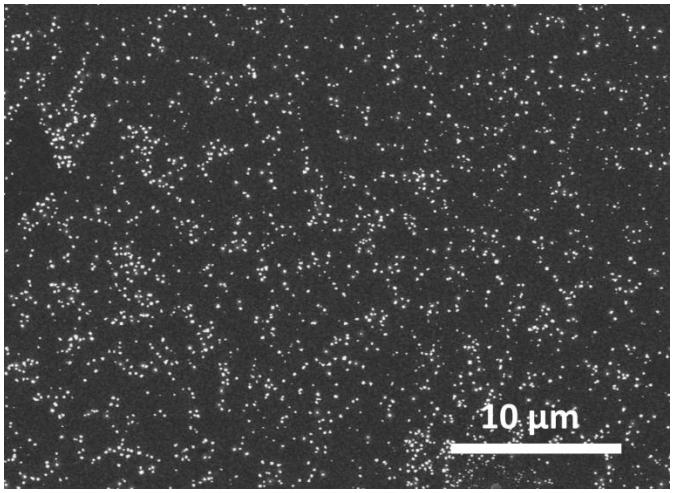
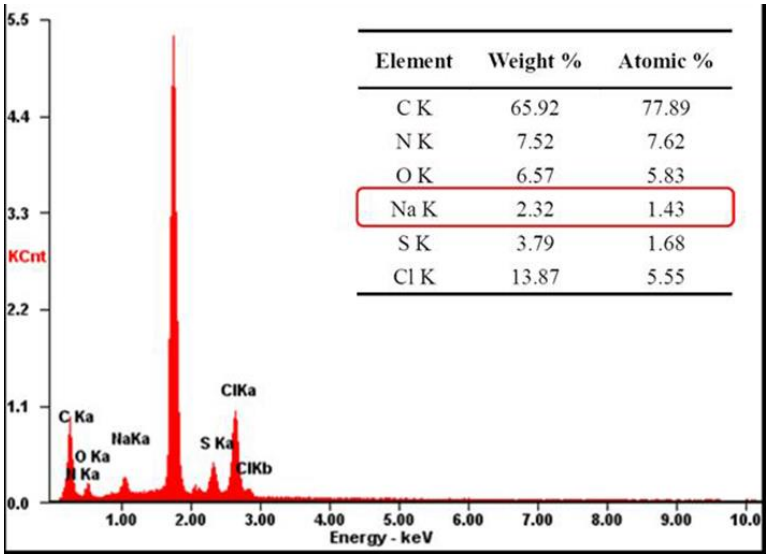


Figure S2. SEM image of a free-standing AAZO/PDAC film prepared by the solution of AAZO/PDAC (1:4.4) without dialysis

The compositions of AAZO/PDAC assembly with a weight ratio of AAZO to PDAC of 1:4.4 without dialysis were determined by an energy-dispersive X-ray spectroscopy (EDS) attached to the field-emission scanning electron microscopy (FESEM, Hitachi S-4800).



Element	Weight %	Atomic %
C K	65.92	77.89
N K	7.52	7.62
O K	6.57	5.83
Na K	2.32	1.43
S K	3.79	1.68
Cl K	13.87	5.55

Figure S3. EDS of the AAZO/PDAC film corresponding to Figure S2.

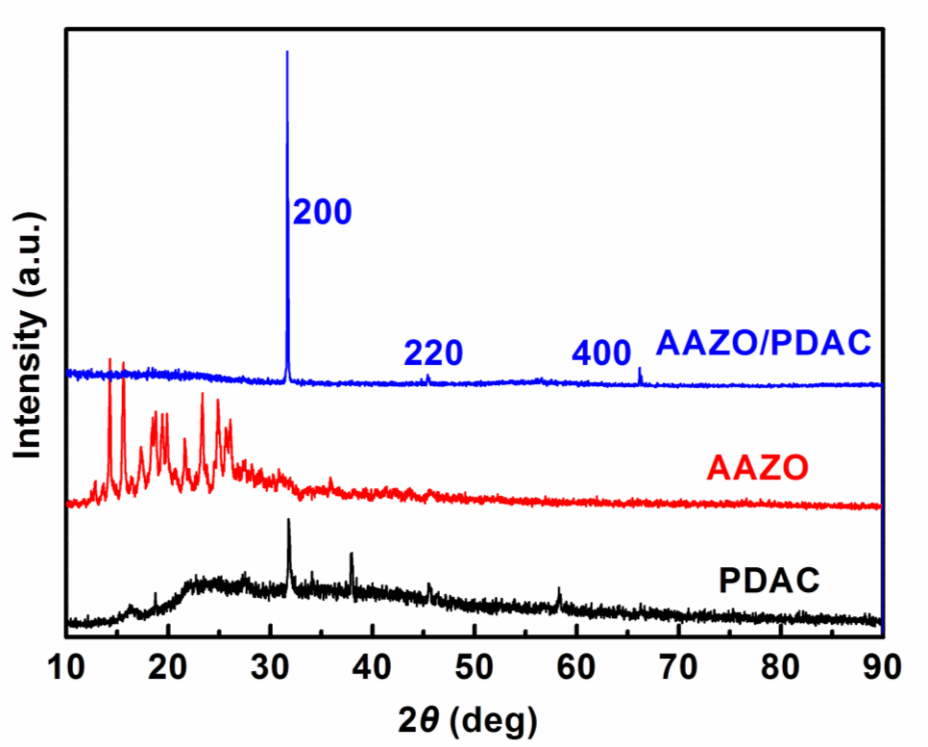
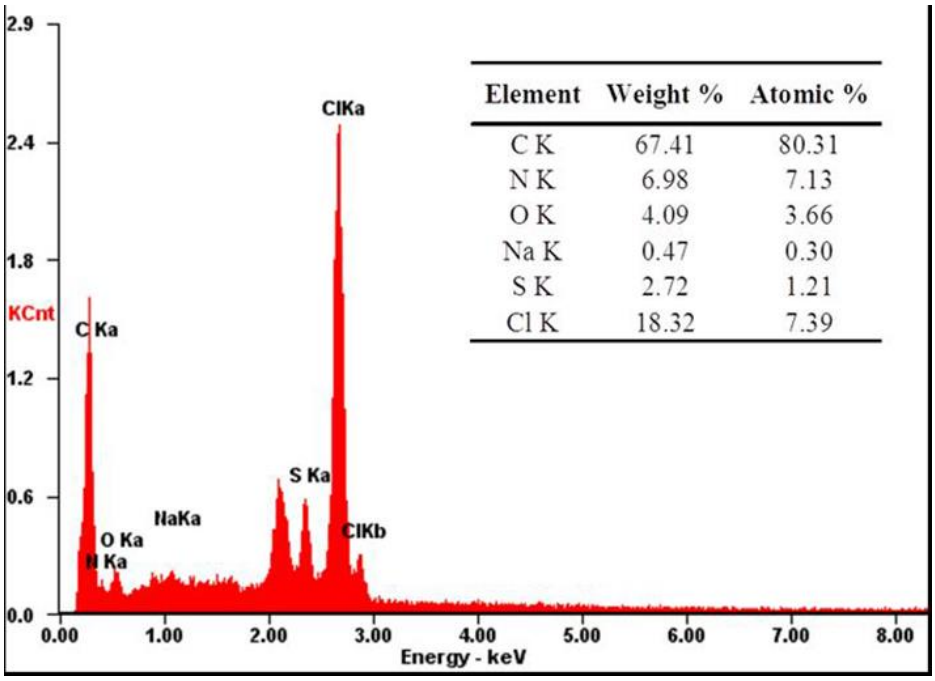


Figure S4. XRD patterns of AAZO, PDAC and the as-prepared AAZO/PDAC film by the solution of AAZO/PDAC (1:4.4) without dialysis.



Element	Weight %	Atomic %
C K	67.41	80.31
N K	6.98	7.13
O K	4.09	3.66
Na K	0.47	0.30
S K	2.72	1.21
Cl K	18.32	7.39

Figure S5. EDS of AAZO/PDAC film with a weight ratio of AAZO to PDAC of about 1:4.4

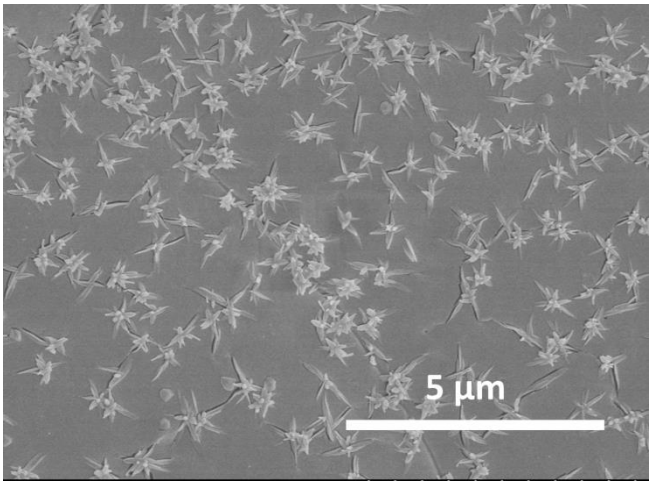


Figure S6. SEM image of a free-standing AAZO/PDAC film with a high weight ratio of AAZO to PDAC of about 1:3.5.

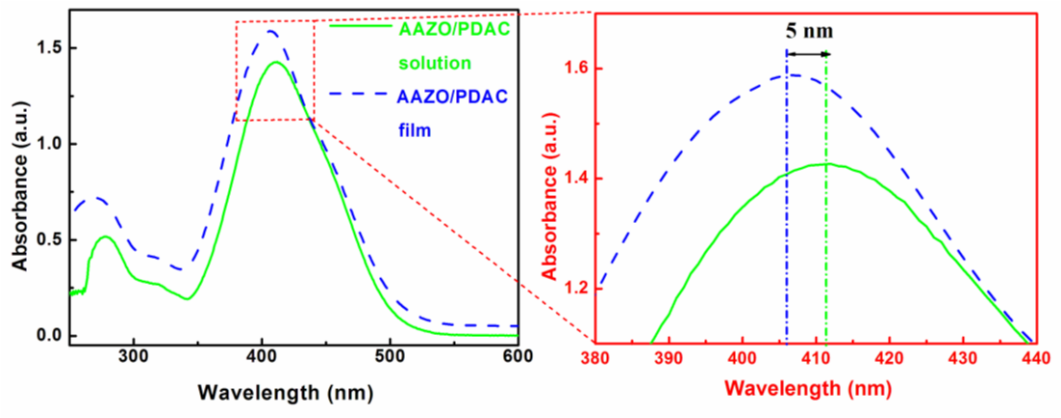


Figure S7. UV-Vis absorption spectra of AAZO/PDAC in DMF/H_2O mixture (50:1 in volume) (green solid line) and the film (blue dash line). The right enlarged spectra corresponds to the selected wavelength (from 380 nm to 440 nm) of the left spectra.

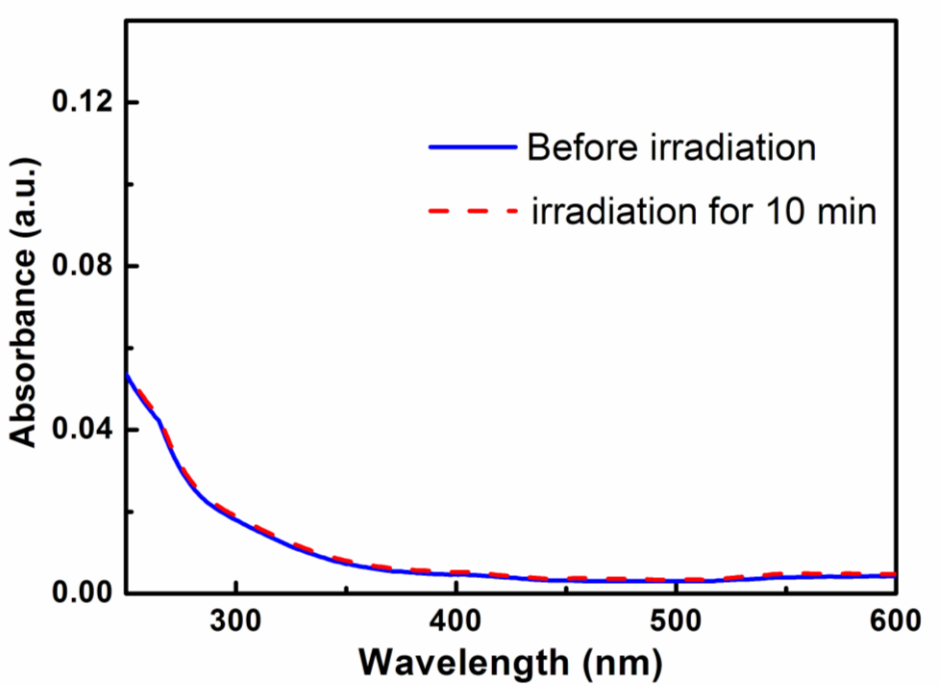


Figure S8. UV-Vis absorption spectra of PDAC solution before (blue solid line) and after 10 min irradiation (red dash line).

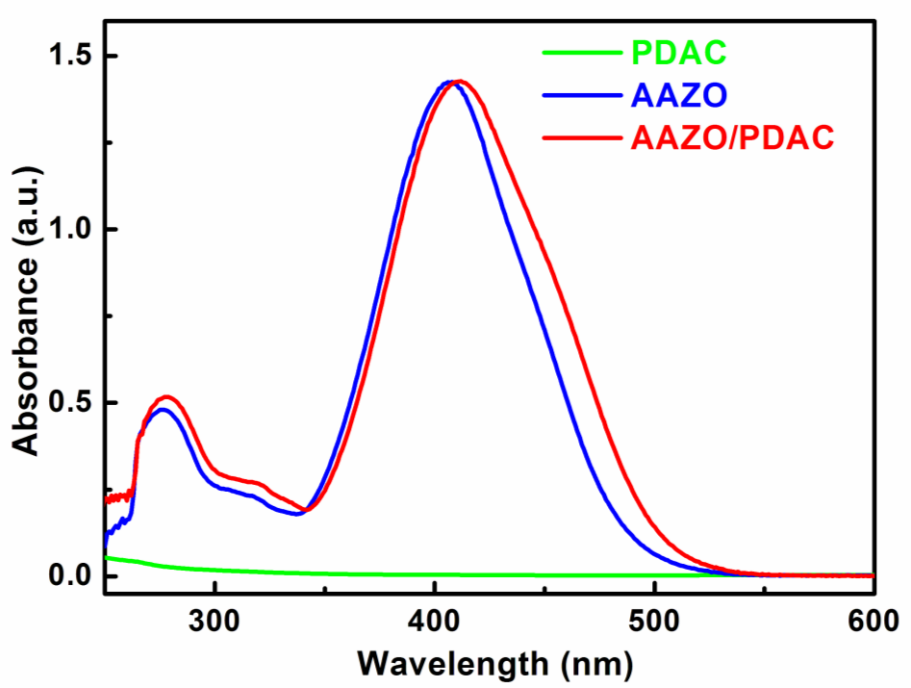


Figure S9. UV-Vis absorption spectra of AAZO, PDAC and AAZO/PDAC.

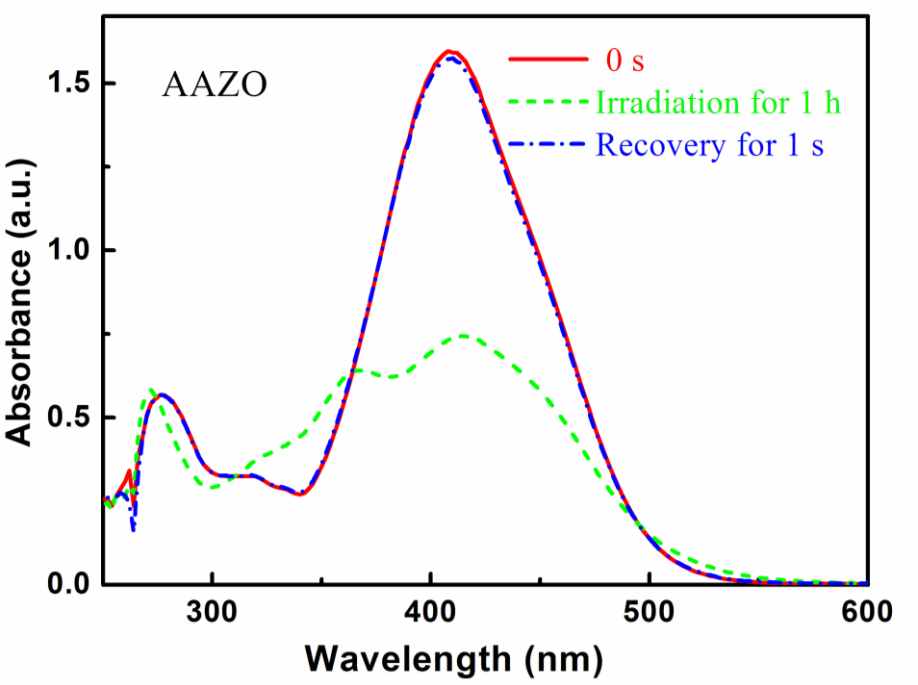


Figure S10. Time-evolved absorption spectra of pure AAZO under the irradiation of UV light at 365 nm for 1 h and after the irradiation in darkness for 1 s.

$$\ln(\frac{A_\infty - A_t}{A_\infty - A_0}) = -k_{rev}t \qquad \textbf{(S1)}$$

where A_0 is the absorption intensity of AZO at a photostationary equilibrium between *trans*- and *cis*-isomers after the irradiation, A_t is the absorption intensity of AZO kept in darkness for a "t" time, A_∞ is the absorption intensity of AZO after the complete *cis*-to-*trans* reversion.

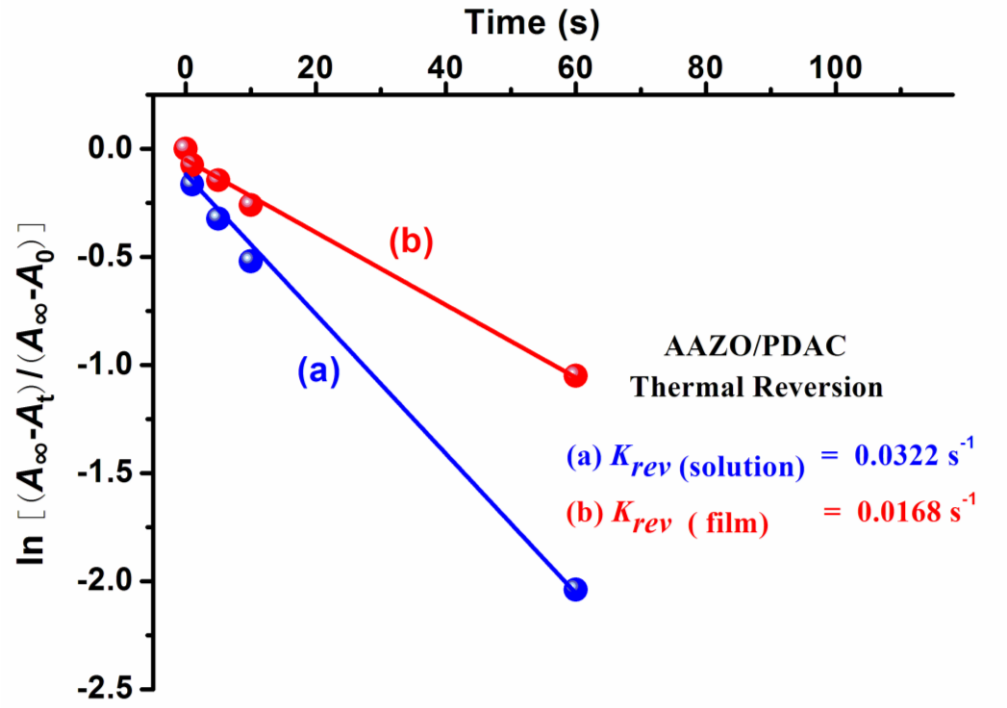


Figure S11. First plots of *cis*-to-*trans* thermal reversion of AAZO/PDAC in DMF/H_2O mixture (50:1 in volume) and the film with different $\boldsymbol{k_{rev}}$ (the inset).

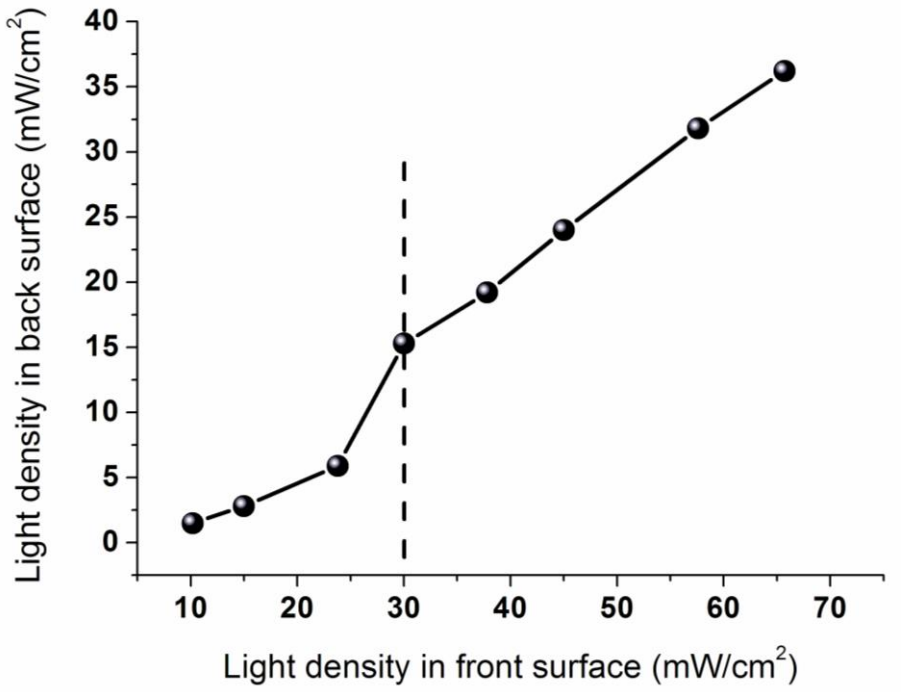


Figure S12. The tested light density in the front/back surface of the AAZO/polymer film with a thickness of 10 μm.

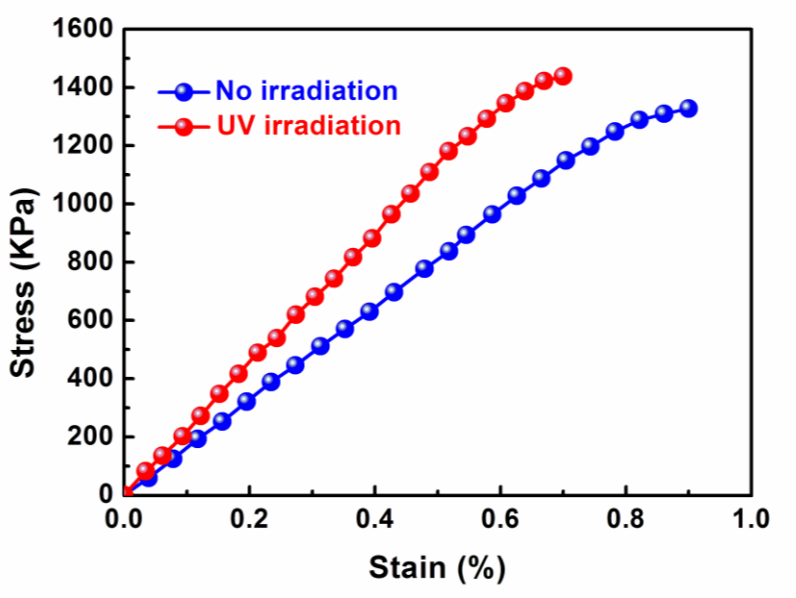


Figure S13. Representative stress-strain responses of AAZO/polymer films in uniaxial tension.