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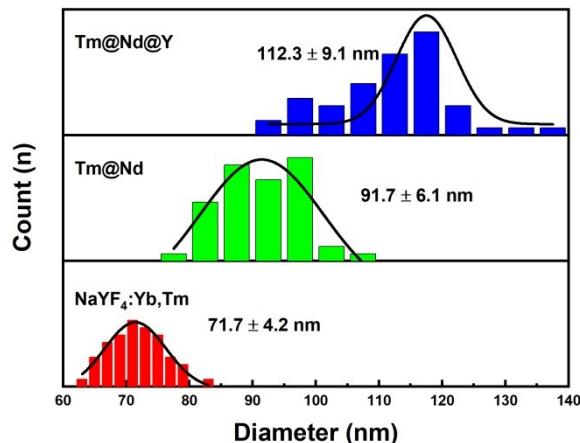
## Supporting Information

### Near-Infrared Light-driven Composite for Smart and Robust Adhesion based on Dynamic Photochemistry

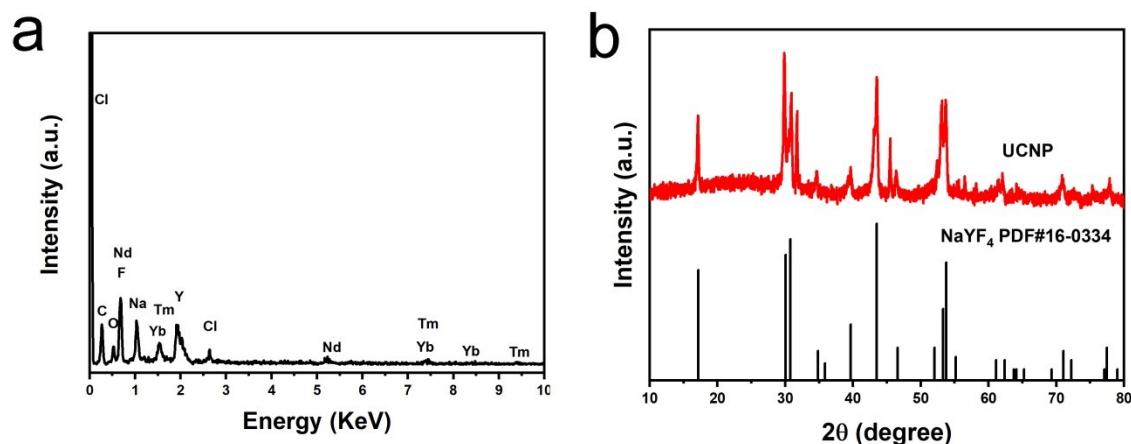
Yuxian Su,<sup>a,b</sup> Tianfu Song,\*<sup>c</sup> Li Liu,\*<sup>a</sup> Shipeng Wen\*<sup>a</sup>

#### Corresponding Authors

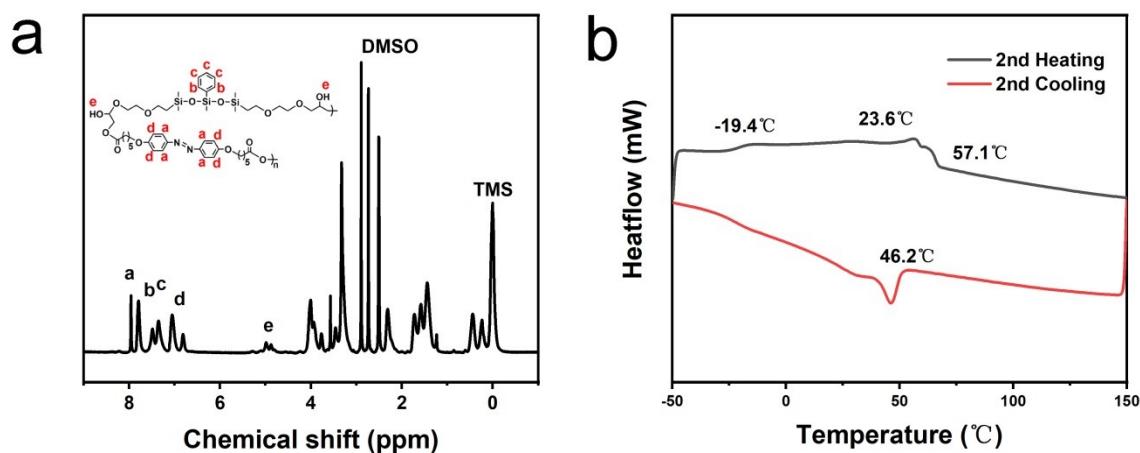
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**Figure S1.** size distributions of NaYF<sub>4</sub>:Yb,Tm, Tm@Nd and Tm@Nd@Y.



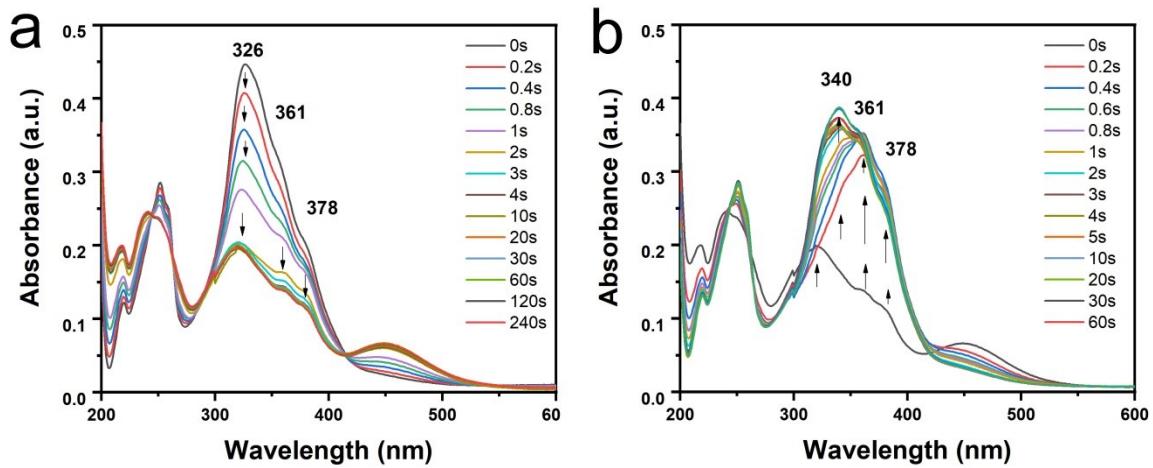
**Figure S2.** (a) EDS image and (b) XRD pattern of  $\text{NaYF}_4:\text{Yb,Tm}@\text{NaYF}_4:\text{Yb,Nd}@\text{NaYF}_4$ .



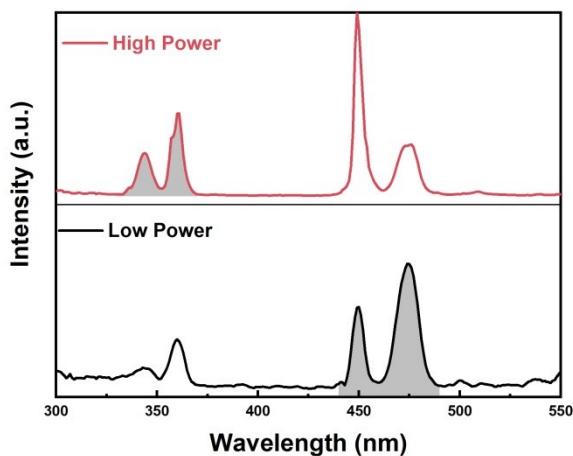
**Figure S3.** NMR hydrogen spectrum (a) and DSC curves (b) of azobenzene polymer with siloxane main chain.

**Table S1.** GPC data of PAzo.

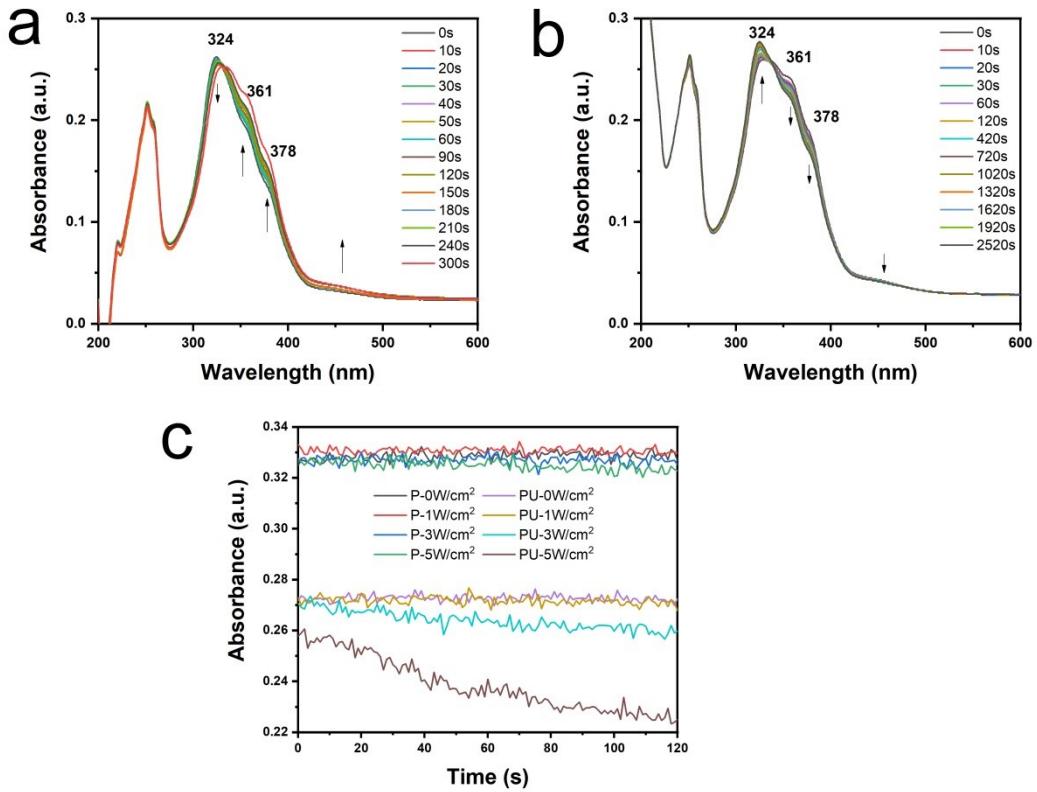
Sample	$M_n$	$M_w$	PDI
PAzo	$1.5 \times 10^4$	$3.3 \times 10^4$	2.19



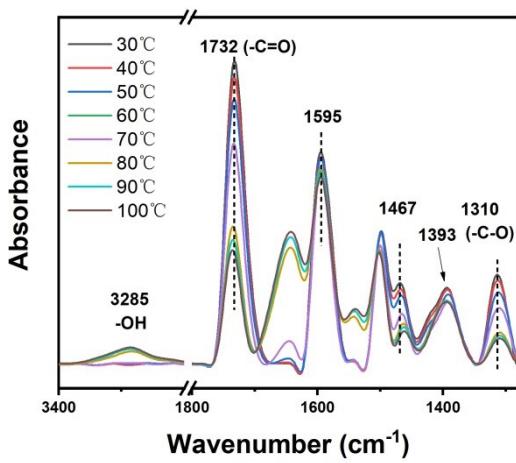
**Figure S4.** Absorption spectra of PAzo/5%UCNP composites under (a) 365 nm and (b) 460 nm irradiation at a power density of  $100 \text{ mW cm}^{-2}$ .



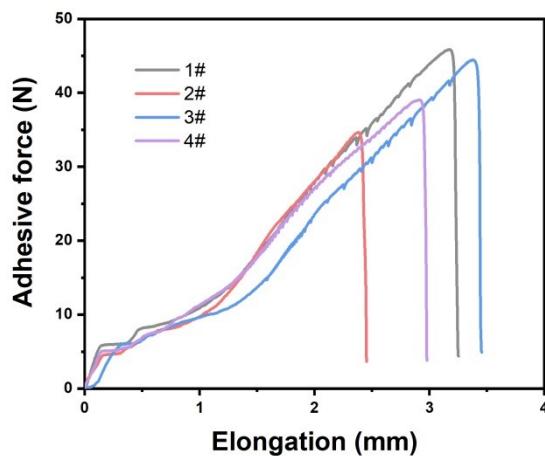
**Figure S5.** luminescence spectra of UCNPs at power densities of  $5 \text{ W cm}^{-2}$  and  $0.5 \text{ W cm}^{-2}$  under  $808 \text{ nm}$  irradiation.



**Figure S6.** Photoisomerization of PAzo/5%UCNP under 808 nm irradiation at power densities of (a) 5 W cm<sup>-2</sup> and (b) 0.5 W cm<sup>-2</sup>. (c) Time-resolved absorption spectra of PAzo (P) and PAzo/5%UCNP (PU) at 328 nm under 808 nm irradiation (5 W cm<sup>-2</sup>).



**Figure S7.** FTIR spectra of PAzo/5%UCNP at different temperatures.



**Figure S8.** adhesion-elongation curves of different composite films.

Note: PAzo (1#), PAzo/1%UCNP (2#), PAzo/5%UCNP (3#), and PAzo/20%UCNP (4#)

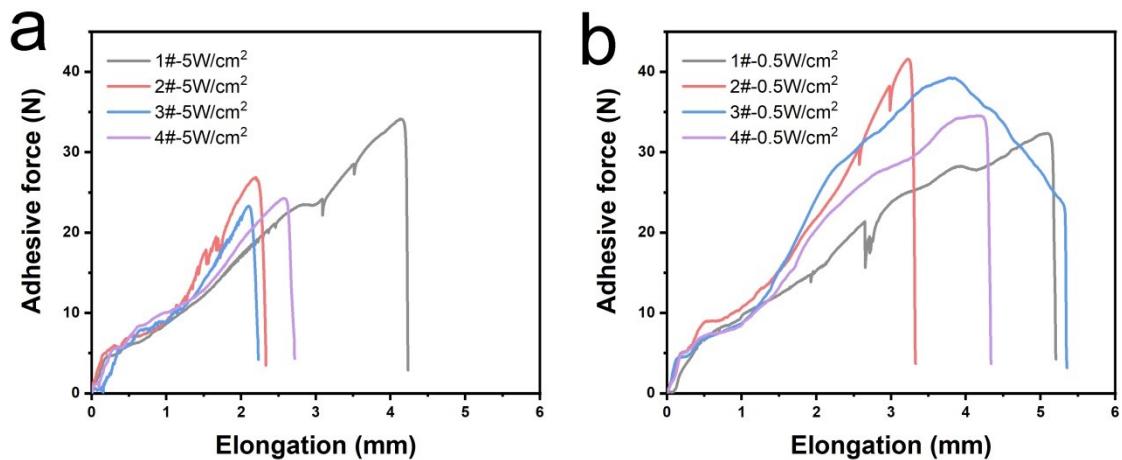
**Table S2.** Parameters of the adhesion-elongation curves.

Film	Maximum adhesion ( $F_m$ , N)	Tensile shear strength (MPa)	Thickness (mm)	Overlapping area (mm <sup>2</sup> )
1#	$44.8 \pm 1.5$	$0.64 \pm 0.07$	$0.091 \pm 0.01$	$71.1 \pm 10.5$
2#	$33.1 \pm 1.5$	$0.66 \pm 0.03$	$0.117 \pm 0.01$	$50.3 \pm 0.0$
3#	$41.6 \pm 2.6$	$0.81 \pm 0.03$	$0.120 \pm 0.02$	$51.5 \pm 1.3$
4#	$37.8 \pm 1.9$	$0.71 \pm 0.03$	$0.107 \pm 0.02$	$53.5 \pm 0.0$

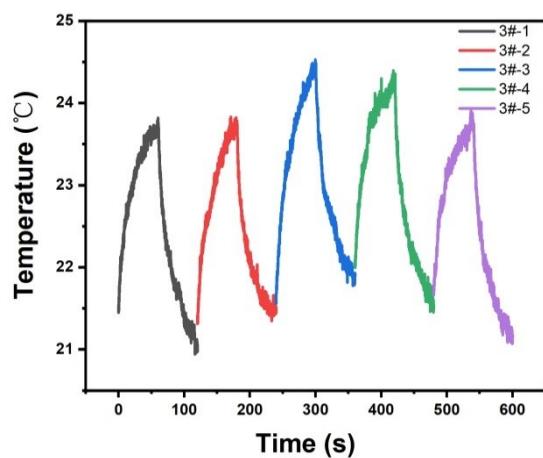
**Table S3.** Parameters of the force-elongation curves of composite films under 808 nm irradiation.

Condition	Film	Maximum adhesion ( $F_m$ , N)	Tensile shear strength (M Pa)	Thickness (mm )	Overlapping area (mm <sup>2</sup> )
$5 \text{ W cm}^{-2}$	1#	$34.9 \pm 1.1$	$0.44 \pm 0.01$	$0.100 \pm 0.00$	$78.5 \pm 0.0$
$5 \text{ W cm}^{-2}$	2#	$24.9 \pm 1.9$	$0.44 \pm 0.02$	$0.167 \pm 0.02$	$56.9 \pm 6.9$

$5 \text{ W cm}^{-2}$	3#	$22.8 \pm 0.8$	$0.38 \pm 0.02$	$0.160 \pm 0.03$	$60.2 \pm 4.9$
$5 \text{ W cm}^{-2}$	4#	$25.3 \pm 1.5$	$0.40 \pm 0.02$	$0.116 \pm 0.00$	$63.6 \pm 0.0$
$0.5 \text{ W cm}^{-2}$	1#	$30.8 \pm 2.2$	$0.39 \pm 0.03$	$0.104 \pm 0.02$	$78.5 \pm 0.0$
$0.5 \text{ W cm}^{-2}$	2#	$38.9 \pm 6.8$	$0.61 \pm 0.05$	$0.158 \pm 0.03$	$64.1 \pm 14.1$
$0.5 \text{ W cm}^{-2}$	3#	$34.7 \pm 6.5$	$0.49 \pm 0.02$	$0.113 \pm 0.04$	$71.1 \pm 10.5$
$0.5 \text{ W cm}^{-2}$	4#	$35.8 \pm 1.8$	$0.54 \pm 0.07$	$0.126 \pm 0.00$	$67.2 \pm 5.1$



**Figure S9.** Adhesion-elongation curves of different composite films under 808 nm irradiation at power densities of (a)  $5 \text{ W cm}^{-2}$  and (b)  $0.5 \text{ W cm}^{-2}$ .



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**Figure S10.** Surface temperature change of PAzo/5%UCNP during 5 cycles under 808 nm irradiation at a power density of  $5 \text{ W cm}^{-2}$ .