*Supporting Information for*:

# A Light-Responsive Polyurethane Liquid Crystal Elastomer Actuators with Photochromic and Multiple Shape Memory Effect

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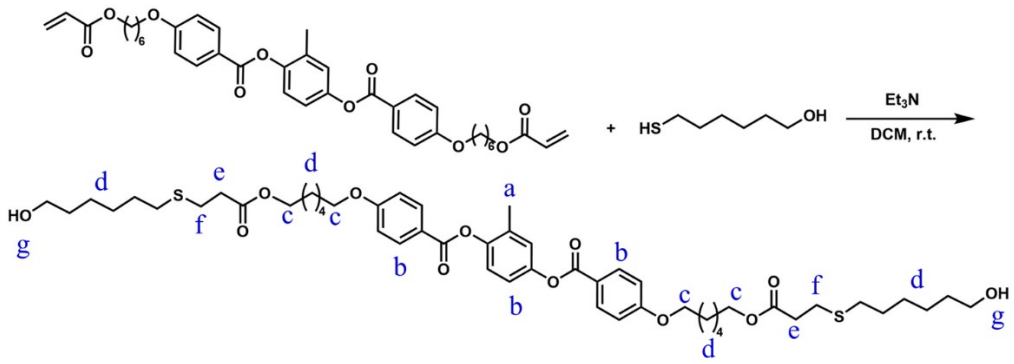
## CONTENTS

## 1. Experimental Procedures

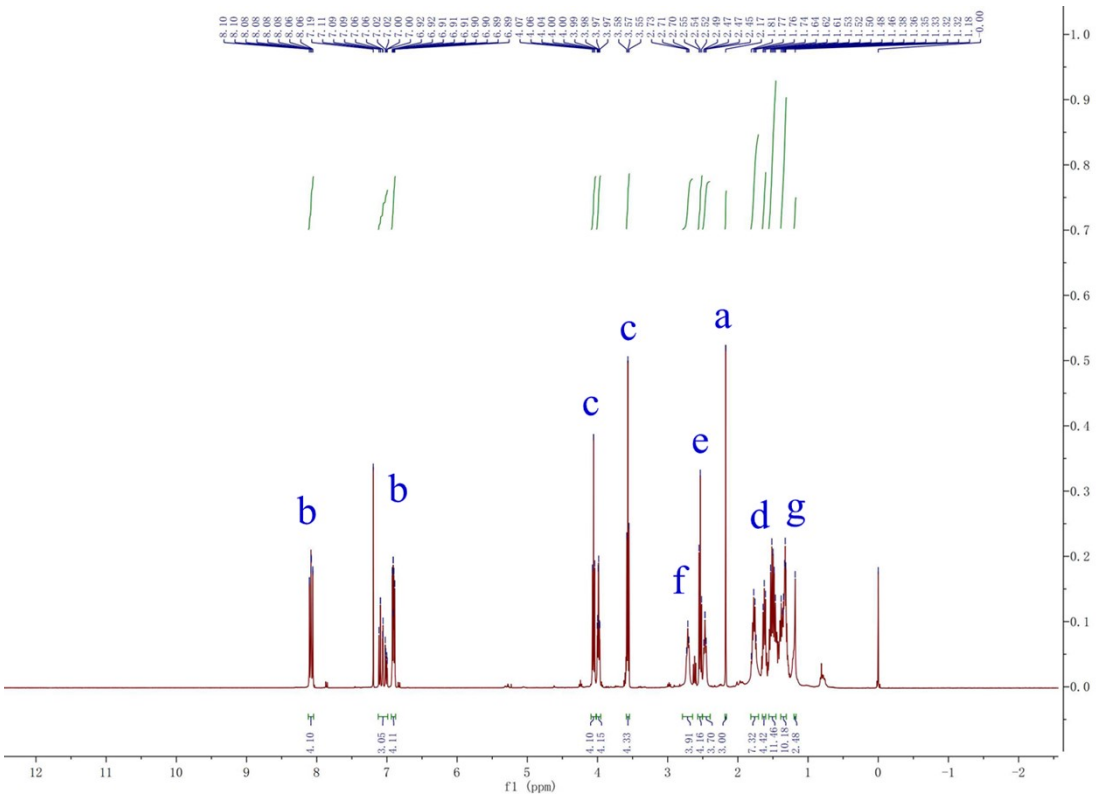
**Preparation of the 2-methyl-1,4-phenylene bis(4-((6-((3-((6-hydroxyhexyl)thio)propanoyl)oxy)hexyl)oxy)benzoate)**

The 2-methyl-1,4-phenylene bis(4-((6-((3-((6-hydroxyhexyl)thio)propanoyl)oxy)hexyl)oxy)benzoate) was synthesized via the thiol-acrylate Michael addition reaction between RM82 and 6-mercapto-1-hexanol (1:2 mol ratio) with TEA as catalyst. A mixture of compound RM82 (2.07 g, 5 mmol) and 6-mercapto-1-hexanol (1.34 g, 10 mmol) were dissolved in 50 mL of dehydrated DCM, and add triethylamine (1.38 mL, 10 mmol) to the above solution. The mixture was stirring at room temperature for 24 h. The reaction mixture was quenched by pouring into $NH_4Cl$ solution. The target product was extracted with ethyl acetate and dried with anhydrous $MgSO_4$. After the solvent was removed, the obtained white solid was purified by silica gel column chromatography to give the 2-methyl-1,4-phenylene bis(4-((6-((3-((6-hydroxyhexyl)thio)propanoyl)oxy)hexyl)oxy)benzoate). The $^1$H-NMR spectrum of the product showed the corresponding proton peaks with well-matched integrals.

$^1$H NMR (400 MHz, Chloroform-*d*) δ 8.12 – 8.05 (m, 4H), 7.12 – 6.99 (m, 3H), 6.94 – 6.88 (m, 4H), 4.06 (t, *J* = 6.6 Hz, 4H), 3.99 (td, *J* = 6.4, 2.1 Hz, 4H), 3.57 (t, *J* = 6.5 Hz, 4H), 2.71 (t, *J* = 6.6 Hz, 4H), 2.54 (t, *J* = 7.3 Hz, 4H), 2.50 – 2.39 (t, 4H), 2.17 (s, 3H), 1.77 (q, *J* = 9.8, 6.4 Hz, 7H), 1.65 – 1.60 (m, 4H), 1.50 (dt, *J* = 13.7, 7.0 Hz, 11H), 1.39 – 1.31 (m, 10H), 1.18 (s, 2H).




**Scheme 1**. Synthesis of the LCE-OH




$^{1}$H NMR spectrum ($CDCl_3$, room temperature, 400MHz)

## 2. Supplementary characterization Figure

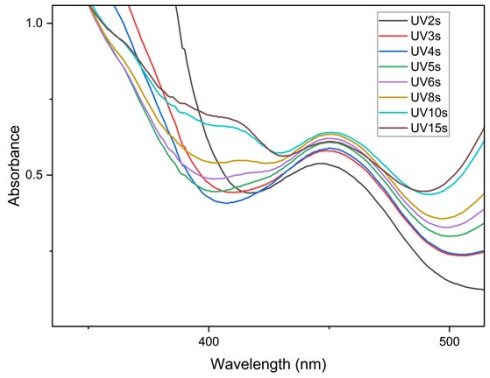



**Figure S1**. Localized magnified UV visible absorption spectra of the mixture of BHHAB and SP in DMF solution in Figure 2c.

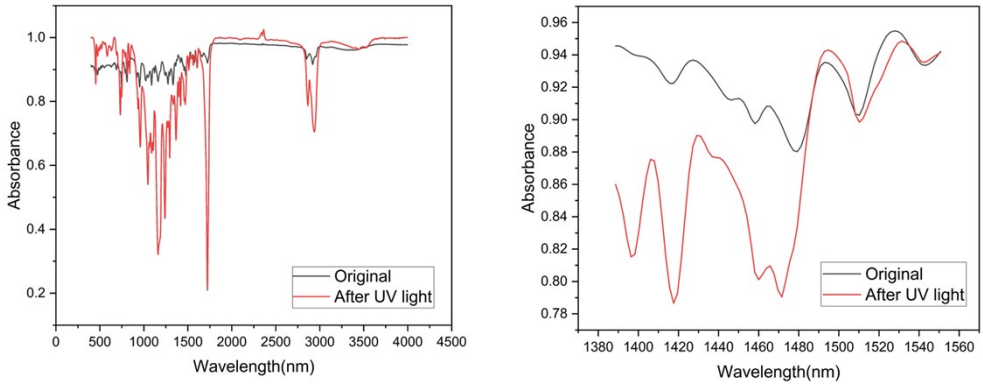



**Figure S2**. IR spectra of the films after UV irradiation.

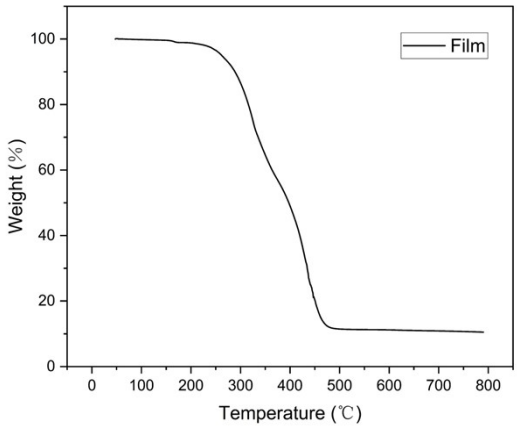



**Figure S3**. Thermogravimetric Analysis (TGA) of the film.

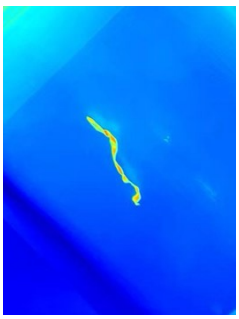


**Figure S4**. Infrared images of the thermal effects on film induced by UV light (100 mW $cm^{-2}$).

## 3. Supporting Movies

**Supplementary Movie 1.** Simultaneous photoinduced helicoid deformation and photochromic effects of film upon irradiation with UV (365 nm, 100 mW $cm^{-2}$) at room temperature.

**Supplementary Movie 2.** Simultaneous photoinduced helicoid deformation and photochromic effects of film one by one upon irradiation with UV (365 nm, 100 mW $cm^{-2}$) at room temperature.