Supporting Information

[Optical](http://cn.bing.com/dict/clientsearch?mkt=zh-CN&setLang=zh&form=BDVEHC&ClientVer=BDDTV3.4.0.4311&q=%E5%85%89%E6%B3%A2%E9%95%BF) [Wavelength](http://cn.bing.com/dict/clientsearch?mkt=zh-CN&setLang=zh&form=BDVEHC&ClientVer=BDDTV3.4.0.4311&q=%E5%85%89%E6%B3%A2%E9%95%BF) Selective Actuation of Dye Doped Liquid Crystalline

Elastomers by Quasi-Daylight

Figure S1. The NMR measurement plot, and the expounding of NMR peaks of the mesogen RM257.

S-Ⅰ: The dissection of the visible absorber dyes

In our this work, three used visible absorber dyes (ABS407, ABS594 and ABS694) were purchased from the company named Luxottica Exciton. The company refused to disclose the chemical formulation for protecting the trade secret of these dyes. Form the information of the company official website (https://exciton.luxottica.com), we can know that the ABS407 and ABS594 are the kind of Coordination Complex Dyes and the ABS694 is a solvent dye. We dissected the used dyes through studying relevant literatures.

The absorbing band of ABS694 is located in near-infrared region. The most widely

near-infrared dye is type of cyanine dyes, which mainly contain with the cyanine chromophores and squaraine chromophores.

We found the similar absorbing curves in the reference[S1], as shown in following Figure S2. In this reference paper, the spectral maximum of cyanine(ethane salts), as shown in compound **3** of Figure S2, shifted from 600 to 690 nm and its absorption intensity was significantly improved in the presence of cardiolipin. The absorption wavelength ranges of this cyanine(ethane salts) can be tuned by a number of vinylene moieties^[S2] in a conjugated system.

Figure S2. Left: The neutral merocyanine **1** and its derivatives **2-6** (γ-aryl substituted methine salts). Right: The similar curves to the ABS694. Reprinted with permission from ref [S1]

The following Figure S3 in the reference[S3] also shows the similar absorbing curves. In this reference paper, the researchers reported the strategy of changing the main absorbing bands by constituting the coordinative bonds into cyanine dyes. Some other researchers reported the strategy of changing the absorbing properties by substituting Nmeso into cyanine dyes [S4].

Figure S3. UV-vis absorption spectra of constituting coordinative bonds into cyanine dyes. Reprinted with permission from ref [S3]

Furthermore, the squarine and its derivatives also belong to the members of cyanine dyes. The squarine and its derivatives attracted our attention by its special rigid D-π-A-π-D conjugation structures resulting in strong absorption from red to the near-infrared region. As shown in following similar absorbing curves of Figure S4 which was reported in the reference[S5], a squarine derivative showed its chemical formulation and the strong absorption in near-infrared region, and the absorption intensity can be the tuned through the reaction between squarine derivative and cysteine.

Figure S4. The absorption of this squarine derivative. (The strong absorption is this squarine derivative, the lower absorption is the absorption after the reaction between squarine derivative and cysteine.) Reprinted with permission from ref [S5]

Based on above discussion, we can speculate the ABS694 is as much as possible the member of cyanine dyes.

The ABS407 and ABS594 are the coordination complex dyes, whose absorbing bands are located in the visible region. The reference [S6] reported the developed coordination complexes that transition metal(II) complexes with hydrazone ligands containing acetoacetanilide and isoxazole, the chemical formulations and UV-Vis absorption were shown in following Figure S5. From the right figure of Figure S5, we found that the absorption curve of Ni(Ⅱ) is similar to that of the ABS407, as the arrow pointed curve.

Figure S5. The coordination complex formation(left) and the UV-Vis absorption(right). Reprinted with permission from ref [S6]

The reference [S7] reported the copper complex with thiazole azo complexes, whose major absorptions are located in the range of about 500nm to 600nm, as shown in following Figure S6. We found that the absorption characteristic of L -Cu(NO₃)₂ is similar to that of the ABS594, as pointed by the arrow.

Figure S6. The UV-Vis absorption and the chemical formulation of copper complex with thiazole azo complexes. (L: ligand) Reprinted with permission from ref [S7]

So we can speculate that the ABS407 and ABS594 might be based on the azo derivative coordination complexes. The absorption properties of azo derivative coordination complexes can be tuned and improved by changing the coordinative metal and the substitute groups^[S8, S9] or constituting different conjugation effect^[S10,S11] on azo molecule.

S-Ⅱ: The data curves about the photo driven reversible change of actuating forces by using isostrain mode.

Figure S7. The contractile actuation stress changes versus time, in isostrain mode, of the RM/PEG(100:0)-LCE and dye doped RM/PEG(100:0)-LCEs upon the irradiations of filtered light-1 (a), filtered light-2 (b) and filtered light-3 respective. The mark ''×'' at the curves mark the switching off time of light stimulus.

Figure S8. The contractile actuation stress changes versus time, in isostrain mode, of the RM/PEG(90:0)-LCE and dye-doped RM/PEG(90:0)-LCEs upon the irradiations of filtered light-1 (a), filtered light-2 (b) and filtered light-3 respective. The mark "x" at the curves mark the switching off time of light stimulus.

Figure S9. The contractile actuation stress changes versus time, in isostrain mode, of the RM/PEG(80:20)-LCE and dye doped RM/PEG(80:20)-LCEs upon the irradiations of filtered light-1 (a), filtered light-2 (b) and filtered light-3 respective. The mark $\cdot x$ " at the curves mark the switching off time of light stimulus.

S-Ⅲ: The highese temperatures on dye doped LCE materials in response to the irradiations of filtered lights

We measured the temperatures on dye doped LCE materials in response to the irradiations of filtered lights. We found that the highest temperatures on material surfaces increased under higher emitting intensity of the light source. By filtered light-1 irradiating, the highese temperatures on ABS407 doped thiol-acrylate MC-LCEs with different mole ratio of RM257 to PEG250 were basicall equal to each other under the same emitting intensity of the light source. By filtered light-2 irradiating, the highese temperatures on ABS594 doped thiol-acrylate MC-LCEs with different mole ratio of RM257 to PEG250 were basicall equal to each other under the same emitting intensity of the light source. By

filtered light-3 irradiating, the highese temperatures on ABS694 doped thiol-acrylate MC-LCEs with different mole ratio of RM257 to PEG250 were basicall equal to each other under the same emitting intensity of the light source. In addition, under the same emitting intensity of the light source, for the three situations which were ABS407 doped thiolacrylate MC-LCEs irradiated by filtered light-1, the ABS594 doped thiol-acrylate MC-LCEs irradiated by filtered light-2, and the ABS694 doped thiol-acrylate MC-LCEs irradiated by filtered light-3, the arrived highest temperatures on materials were mutually approximate. The highese emitting intensity of our quasi-daylight light source was 3.5×10^5 lux. Figure S10 shows the highest temperatures of dye doped thiol-acrylate MC-LCEs irradiated by the filtered lights under different emitting intensity of light source.

Figure S10. The highest temperatures of dye doped thiol-acrylate MC-LCEs irradiated by the filtered lights under different emitting intensity of light source.

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