

# Supporting Information

*for*

## **Mechano-responsive Calix[4]arene-based Molecular Gels: Agitation Induced Gelation and Hardening**

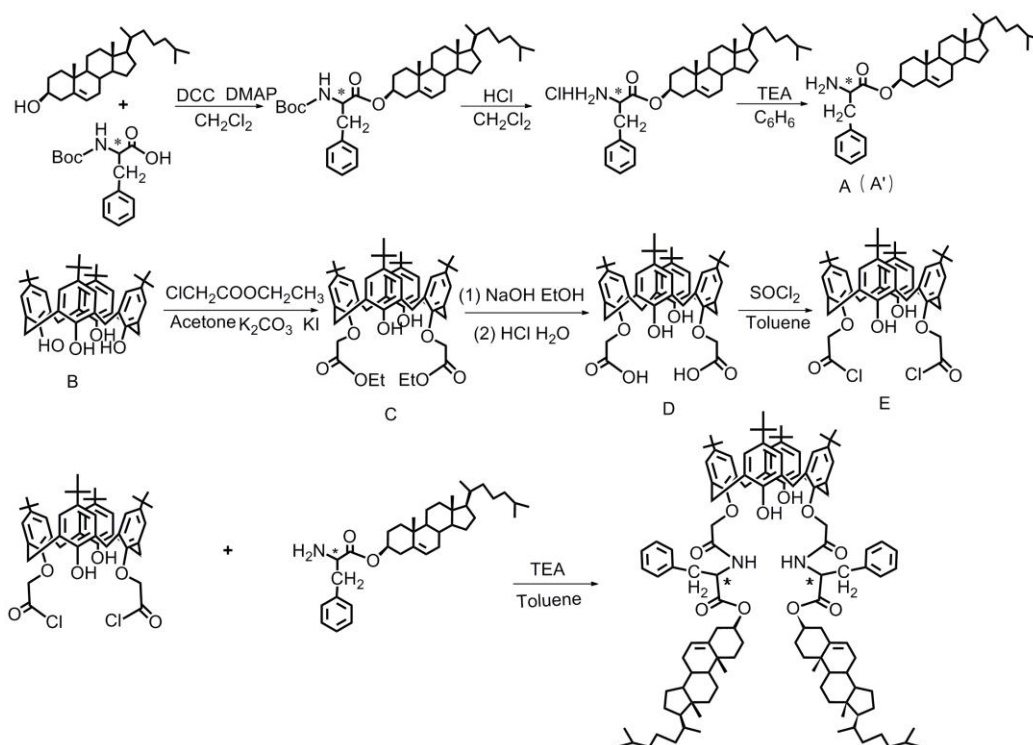
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## 1. Synthetic procedures



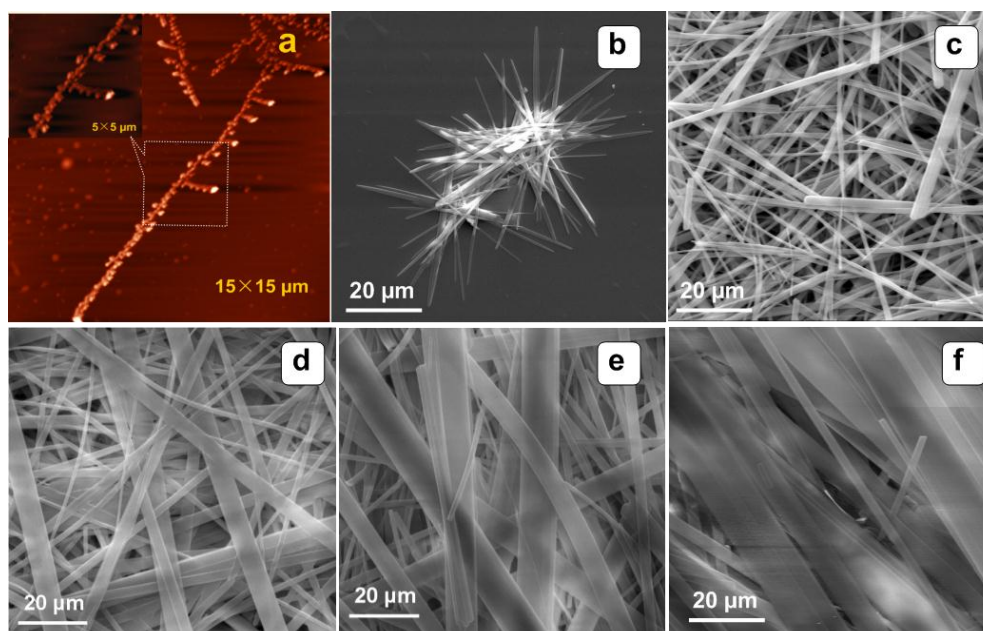
**Scheme S1.** Synthesis routes of the cholesteryl derivatives of calix[4]arene with L- or D-phenylalanine in the linkers (**1** and **2**). With the exception of A, the others including calix[4]arene are the intermediates **B**, **C**, **D** and **E**.

## 2. Gelation behaviors of the two compounds

**Table S1.** Gelation behaviors of **1** and **2** (2.5%, w/v)

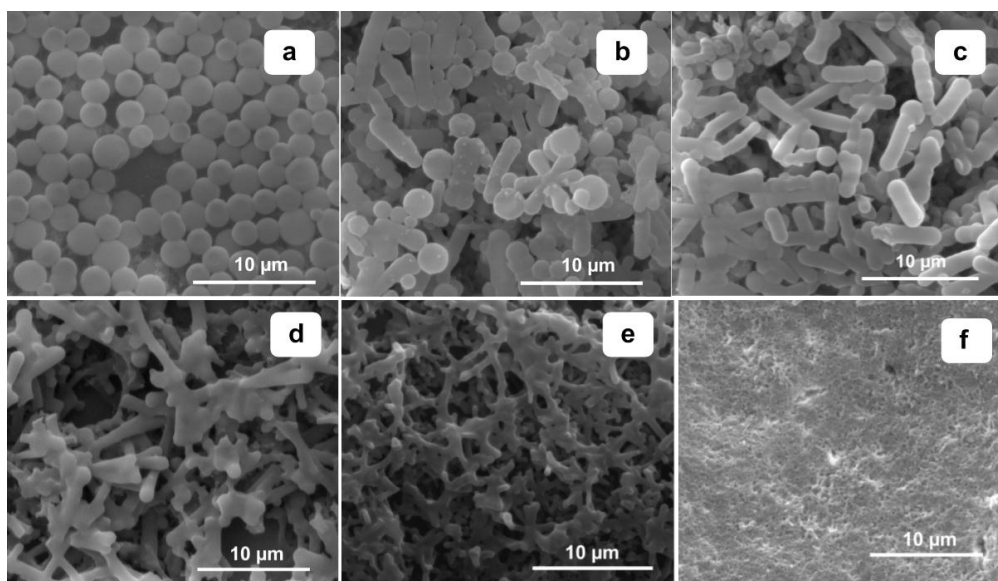
<b>Solvent</b>	<b>1</b>	<b>2</b>	<b>Solvent</b>	<b>1</b>	<b>2</b>
Methanol	I	I	<i>n</i> -Hexane	P	S
Ethanol	I	I	<i>n</i> -Heptane	P	S
<i>n</i> -Propanol	PG	S	<i>n</i> -Octane	P	S
<i>n</i> -Butanol	G	S	<i>n</i> -Nonane	P	S
<i>n</i> -Pentanol	G	S	<i>n</i> -Decane	P	S
<i>n</i> -Hexanol	PG	S	Cyclohexane	S	S
<i>n</i> -Heptanol	S	S	Acetonitrile	I	I
<i>n</i> -Octanol	S	S	Isopropanol	P	G
<i>n</i> -Nonanol	S	S	Kerosene	S	S
<i>n</i> -Decanol	S	S	Ethyl acetate	S	S
Acetone	I	S	DMSO	P	P
Benzene	S	S	DMF	S	S
Toluene	S	S	CHCl <sub>2</sub>	S	S
THF	S	S	CHCl <sub>3</sub>	S	S
<i>n</i> -Pentane	P	I	CCl <sub>4</sub>	S	S

### 3. Concentration dependence of the gel-networks of 1/*n*-pentanol



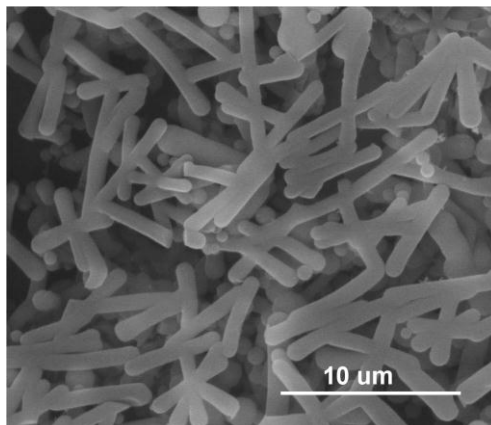
**Figure S2** AFM (a) and SEM (b-f) images of the aggregates of **1** in its *n*-pentanol gel (a, 0.01%; b, 0.1%; c, 0.5%; d, 1%; e, 1.5%; f, 2%, w/v)

#### 4. Sonication time-dependence of the gel-networks of 2/isopropanol



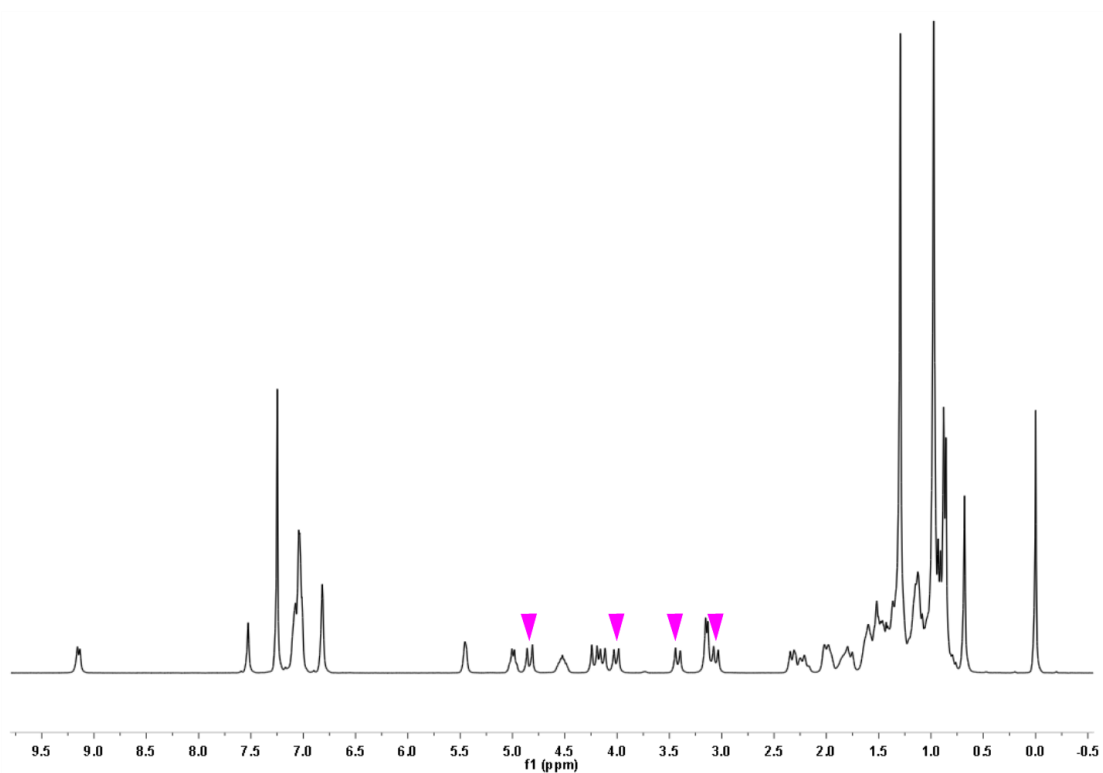
**Figure S3** The SEM images of the xerogels obtained from the 2/isopropanol gels prepared with different sonication time at room temperature ( a, 3 min; b, 6 min; c, 9 min; d, 12 min; e, 15 min; f, 36 min)

#### 4. Gel networks of the gel formed after 30 h after dissolution of **1** in the solvent (2.5%, w/v) via heating



**Figure S4** SEM image of the aggregates in the xerogel of **2** obtained from its isopropanol gel which was prepared by dissolving **2** in isopropanol (2.5%, w/v) via heating. However, it is to be noted that the gel can be only obtained 30 h later of the dissolution. Even so, a very small or light shaking is needed to make the system (a very viscous solution) become a gel.

## 5. $^1\text{H}$ NMR spectra of **1**



**Figure S5**  $^1\text{H}$  NMR spectra of **1** in  $\text{CDCl}_3$