**Electronic Supplementary information** 

## Crystalline architectures of $C_{84}$ with tunable morphology and linearly polarized red emission

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*Figure S1.* HPLC profiles of the reaction mixture of  $C_{84}$  isomers with cyclopentadiene.



*Figure S2.* HPLC profiles of the isolated isomers: (a)  $D_{2d}$ -C<sub>84</sub> and (b)  $D_2$ -C<sub>84</sub>.



*Figure S3.* UV-vis spectra of isolated isomers: (a)  $D_{2d}$ -C<sub>84</sub> and (b)  $D_2$ -C<sub>84</sub>.



*Figure S4.* (a,b) SEM images of *r*-HPs of  $D_{2d}$ -C<sub>84</sub> prepared under different conditions. Note: As seen in Figure S4, the size of *r*-HPs is increasing with the decreasing of C<sub>84</sub> concentration, while their aspect ratio remains constant.



*Figure S5.* (a,b) SEM images of *r*-HPs of  $D_{2d}$ -C<sub>84</sub> prepared under different conditions. Note: As seen in Figure S5, the size and aspect ratio of *r*-HPs are decreasing with the increasing of relative volume of IPA.



*Figure S6.* SEM images of *c*-HP of  $D_{2d}$ -C<sub>84</sub> prepared under optimal conditions ( $D_{2d}$ -C<sub>84</sub> (0.2 mg mL<sup>-1</sup>)/MST:IPA = 1:1).



**Figure S7.** SEM images of *r*-HPs of  $D_2$ -C<sub>84</sub> prepared under optimal conditions ( $D_2$ -C<sub>84</sub> (0.2 mg mL<sup>-1</sup>)/MST:IPA = 1:1).



 $D_2$ - $C_{84}$  (0.2 mg mL<sup>-1</sup>)/MST:IPA = 1:1

 $D_2$ - $C_{84}$  (0.2 mg mL<sup>-1</sup>)/MST:IPA = 1:1

*Figure S8.* (a and b,c,d) SEM images of *c*-HPs of  $D_{2d}$ -C<sub>84</sub> prepared under different conditions.



*Figure S9.* Absolute PL quantum yields measured for *r*-HPs and *c*-HPs of  $D_{2d}$ -C<sub>84</sub> (both dispersed in cyclohexane solution at room temperature).

	Table S1.	Fitting res	sults based	l on the b	viexponential	equation of	f
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Device	$\tau_1(ns)$	$A_{l}(\%)$	$\tau_2(ns)$	$A_2(\%)$	$ au_{avg}(ns)^a$	$R^2$			
r-HP	4.38	10.74	0.46	89.26	2.56	0.992			
c-HP	3.85	6.57	0.44	93.43	1.74	0.993			
$ au_{avg} = rac{\sum A_i { au_i}^2}{\sum A_i { au_i}}$									

 $I(t) = A_1 \exp(\frac{-t}{\tau_1}) + A_2 \exp(\frac{-t}{\tau_2})$  for the transient PL spectra of the *r*-HP and *c*-HP.