## SUPPORTING INFORMATION

## Slow Kinetic Evolution of Nanohelices Based on Gemini Surfactant Self-Assemblies with Various Enantiomeric Excess; chiral segregation towards racemic mixture.

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**Figure S1:** TEM images of twisted ribbons (a) and helical ribbons (b). Schemes to indicate the width and pitch in twisted ribbon (c) and helical ribbon (d).



**Figure S2:** TEM images of 16-2-16 tartrate with ee=0.5 (a, b, c) and 0.7 (d, e, f) at different aging times (4h, 1d, 4d). The images (g, h) are SEM images of 16-2-16 tartrate with ee=0.33 after sol-gel transcription to form silica helices around the organic ones. With SEM, the handedness can be observed without negative staining.





**Figure S3:** Cryo-TEM images of twisted ribbons and helical ribbons formed by 16-2-16 tartrates with ee=0.9 aging 2h (a) and aging 5h (b), ee=1 aging 2h (c) and aging 5h (d). In the bottom image, we show how the pitches and widths are measured from the Cryo-TEM Images.





**Table S1:** Pitch length and width of twisted ribbons and helical ribbons formed by 16-2-16 tartrate with high ee values (ee>0.8). The statistics were done with The statistics were done from a number of TEM images from different samples of the same composition, different TEM grids of one sample, and different grid squares, in order to avoid the sampling problems as it is well known that TEM images may not be representative if we only take one or two images to do the statistics. For each sample, around 100 TEM images were taken in which there are in the average 5-30 helices were observed.. Just to show how it is done, we show below how typically those values were obtained from TEM images. For each value, around 100 helices were counted.

ee	Aging time	Morphology	Pitch/nm	Width/nm	Pitch/Width
0.9	2h	Twisted ribbons	$108.23 \pm 14.23$	$11.94 \pm 1.52$	9.1
	5h	Twisted ribbons	$109.75 \pm 7.01$	$19.23 \pm 2.46$	5.7
		Helical ribbons	$77.531 \pm 5.01$	$26.93 \pm 2.62$	2.9
1	2h	Twisted ribbons	$108.32 \pm 7.61$	$13.53 \pm 2.48$	8.0
	5h	Twisted ribbons	$108.43 \pm 5.31$	$20.40 \pm 3.42$	5.4
		Helical ribbons	$79.39 \pm 7.63$	$27.92 \pm 5.50$	2.8
	5d	Tubes	$54.10 \pm 9.84$	$47.73 \pm 6.30$	1.3

**Figure S4:** CD spectra of 16-2-16 L- and D- tartrate (1 mM) with aging times of 0 min and 1h (a). CD spectra of 16-2-16 L- tartrate for 1 mM (b) and 10 mM (c) with various aging times at 20 °C. CD values at 206 nm, as a function of time, for two concentrations (d).



**Figure S5:** CD spectra of 16-2-16 tartrate with ee = 0.25 (L-tartrate > D-tartrate, red lines) and ee = -0.25 (L-tartrate < D-tartrate, black lines) aging at 0 min (a), 1h (b) and 2d (c).



**Figure S6:** a) Gemini dications are intrinsically achiral but their headgroups N<sup>+</sup>-C-C-N<sup>+</sup> adopt two mirror-image conformations, Z or  $\Sigma$  through the formation of strong ion pair with dianion tartrate. b) When ee = 0.5, with mixture of 16-2-16 L-tartrate and 16-2-16 D-tartrate 1:3, first, they form separately right handed and left handed small helices formed by double bilayer. With time, small helices with opposite handedness merge to form larger and thicker helices with larger helicities. The images were developed from our previous paper in ref (17)



**Figure S7:** X-ray scattering patterns of ee = 0.25 Gemini tartrate in 50 mM aged with various duration. Blue arrow shows the disappearance of the peak at  $q = 1.7 \text{ Å}^{-1}$ 

