

## Fabrication of helical hybrid silica bundles

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Solvents	LL-12	LL-10	LL-9	LL-8	LL-7	LL-6
Water	I	I	I	I	I	I
Hexane	I	I	I	I	I	I
Cyclohexane	I	I	I	I	I	I
Ethyl acetate	I	I	I	I	I	I
Acetone	I	I	I	I	I	I
Acetonitrile	I	I	I	I	I	I
Methanol	P	O-gel(100)→P	P	Tl-gel(60)	P	S(50)
Ethanol	Tl-gel(40)	Tl-gel(30)→P	G(50)→P	Tl-gel(30)	P	PG(50)
1-Propanol	Tl-gel(40)	G-gel(60)→P	G(50)→P	Tl-gel(30)	P	PG(50)
1-Butanol	Tl-gel(40)	Tl-gel(50)→P	Tl-gel(45)	Tl-gel(30)	PG(50)	PG(50)
Cyclohexanone	Tl-gel(8)	Tl-gel(10)	Tl-gel(10)	Tl-gel(15)	Tl-gel(20)	Tl-gel(10)
THF	LG(50)	Tl-gel(10)	PG(20*)	Tl-gel(10)	PG(20*)	PG(20*)
1,4-Dioxane	Tl-gel(10)	Tl-gel(10)	O-gel(10)	Tl-gel(15)	Tl-gel(20)	Tl-gel(20)
Toluene	T-gel(5)	T-gel(5)	T-gel(30)	T-gel(10)	T-gel(15)	LG(25*)
Chlorobenzene	LG(T 50)	T-gel(5)	Tl-gel(20)	T-gel(5)	Tl-gel(30)	T-gel(30)
Nitrobenzene	Tl-gel(25)	Tl-gel(10)	Tl-gel(20)	Tl-gel(20)	Tl-gel(35)	Tl-gel(20)
DMF	O-gel(15)	Tl-gel(30)	PG(50)	Tl-gel(30)	PG(50)	Tl-gel(40)
DMSO	Tl-gel(40)	T-gel(40)	PG(50)	T-gel(40)	PG(50)	S(50)
Chloroform	T-gel(15)	T-gel(20)	VS(50)	T-gel(30)	S(50)	T-gel(30)

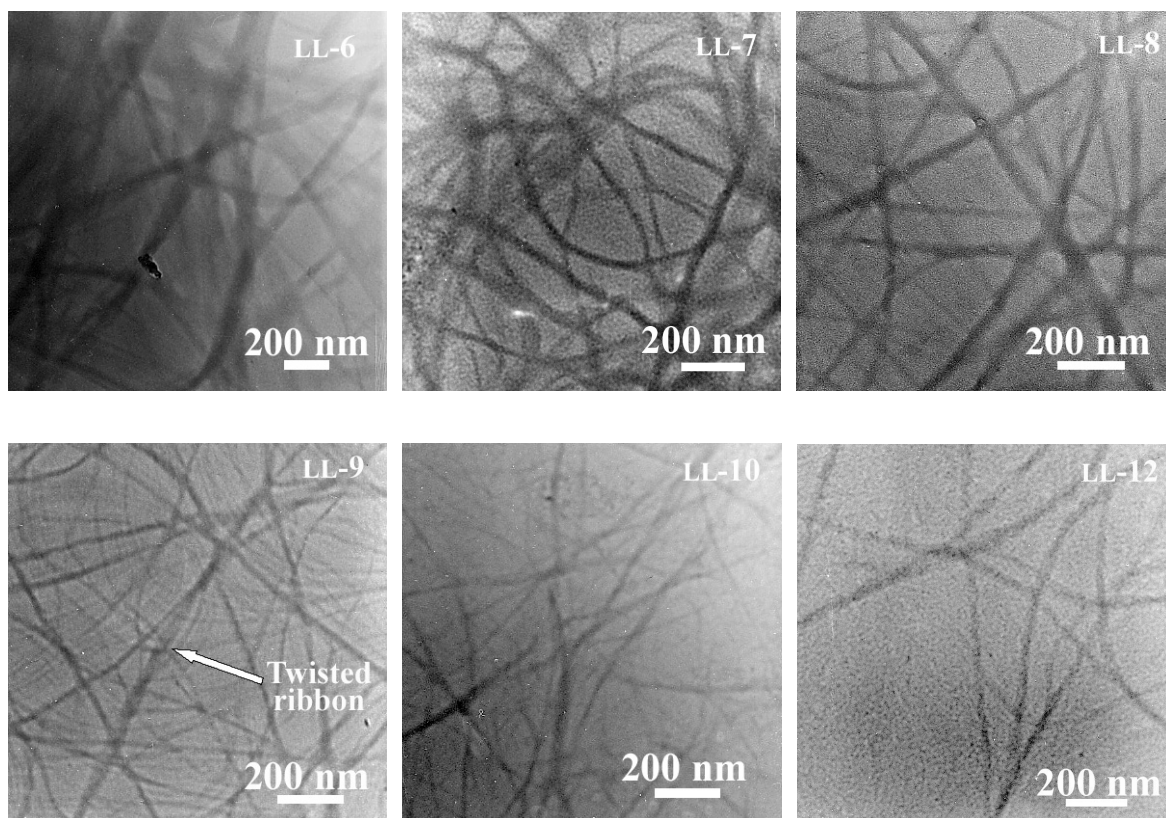
**Table S1** Gelation properties and MGCs (mg/mL) of the silsesquioxanes.

T-gel; Transparent gel, Tl-gel; Translucent gel, O-gel; Opaque gel

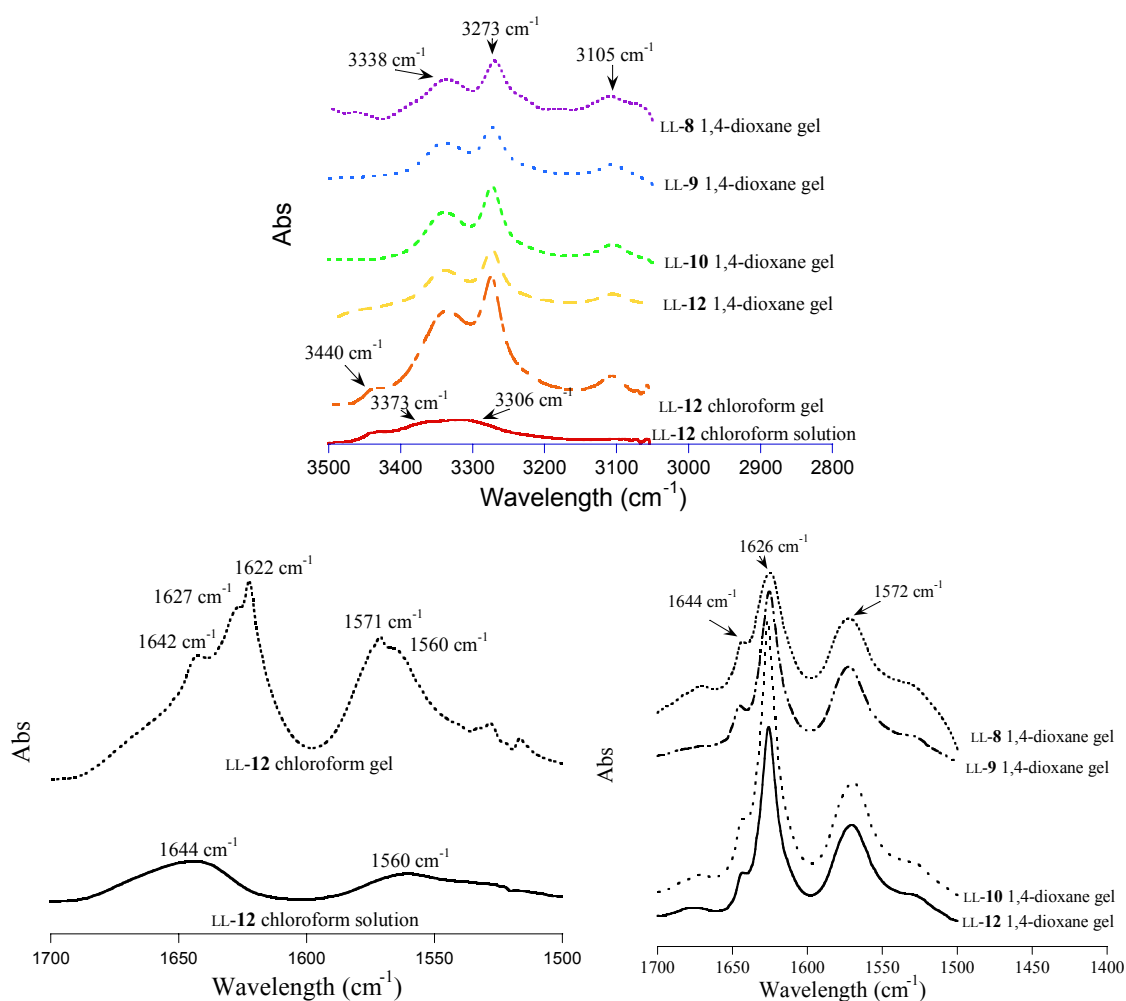
S; Soluble, I; Insoluble, P; Precipitated, LG; Loose gel, PG; Partial gel, VS; Viscous solution

(\*); Almost insoluble at the concentration above values.

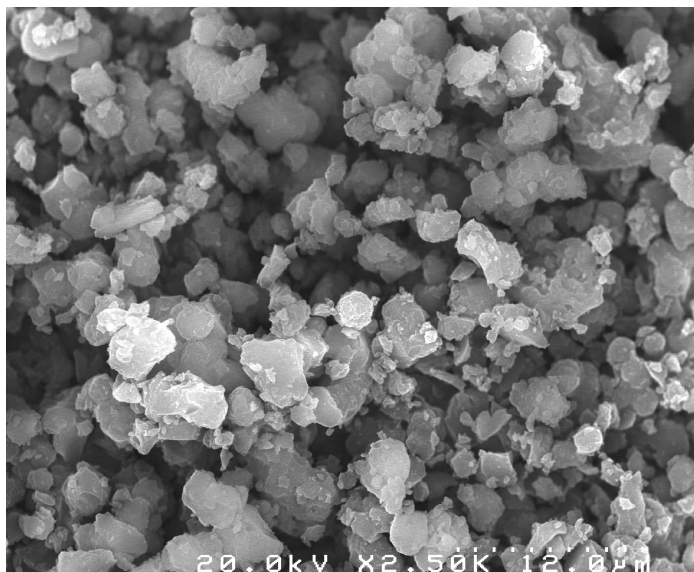
→; Overnight.



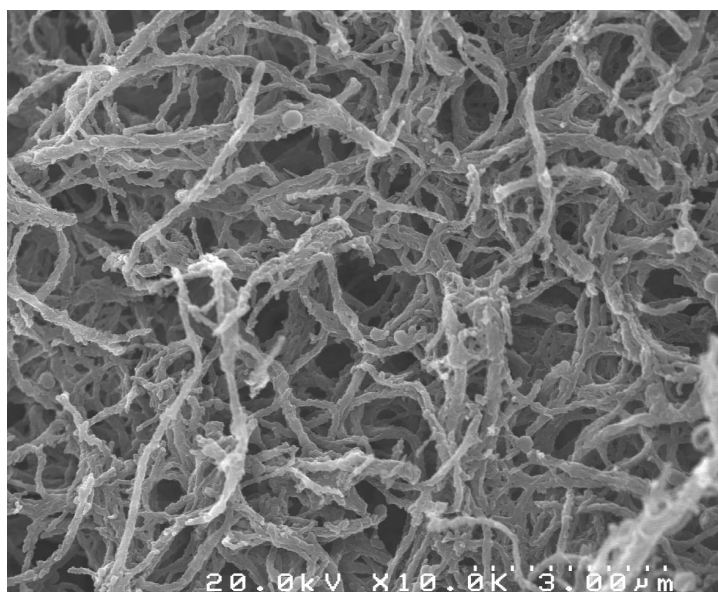
**Figure S1.** TEM images of the xerogel of the silisequioxanes prepared in dioxane (1 or 2 mg/mL).



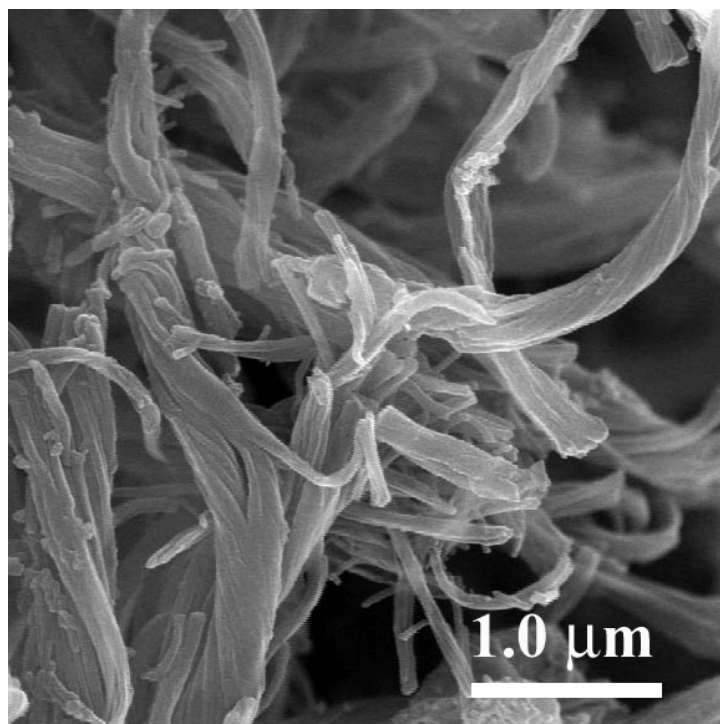
**Figure S2.** FT-IR spectra of the silsesquioxane in chloroform solution, chloroform gel and 1,4-dioxane gels (Concentration: LL-12 chloroform solution: 5.0 mg/mL; LL-12 chloroform gel: 20 mg/mL; 1,4-dioxane gels: 5.0 mg/mL).



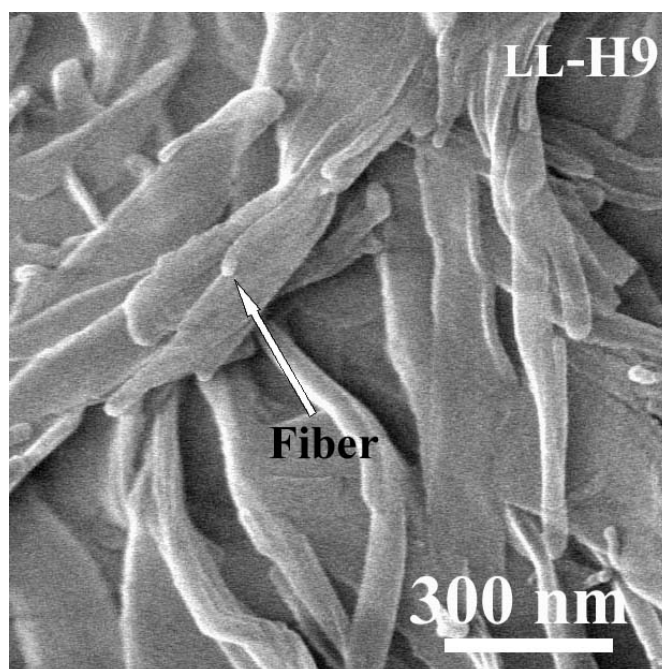
**Fig. S3.** FESEM image of LL-H6 (preparation condition: 20 mg of LL-6, 1.0 mL of 1,4-dioxane, and 1.0 mL of 4.0 M HCl aq.).



**Fig. S4.** FESEM image of LL-H7 (preparation condition: 20 mg of LL-7, 1.0 mL of 1,4-dioxane, and 1.0 mL of 4.0 M HCl aq.).



**Fig. S5.** FESEM image of LL-H12 by **Method 3** (Preparation condition: 100 mg of LL-12, 1.0 mL of 1,4-dioxane, and 1.0 mL of 1.0 M HaOH aq.).



**Fig. S6.** FESEM image of LL-H9 by **Method 4** (Preparation condition: 20 mg of LL-9, 1.0 mL of 1,4-dioxane, and 1.0 mL of 1.0 M HaOH aq.).

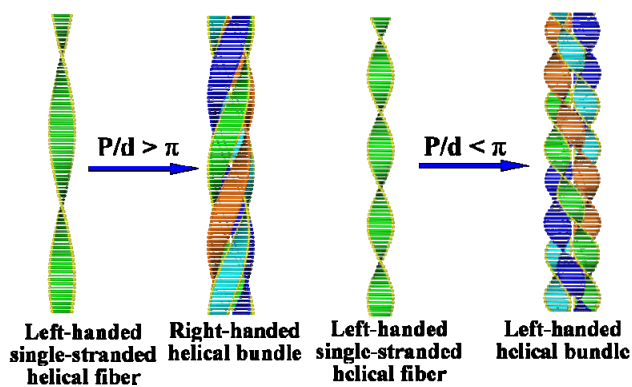


Fig. S7. Plausible model for formation of helical bundles.

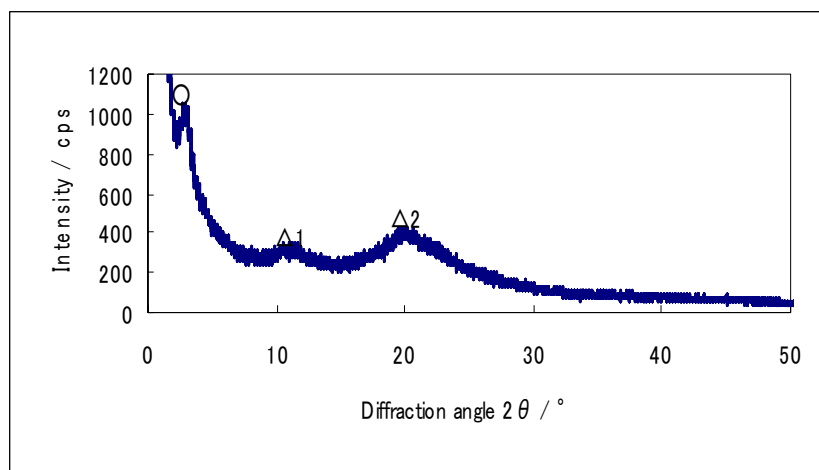
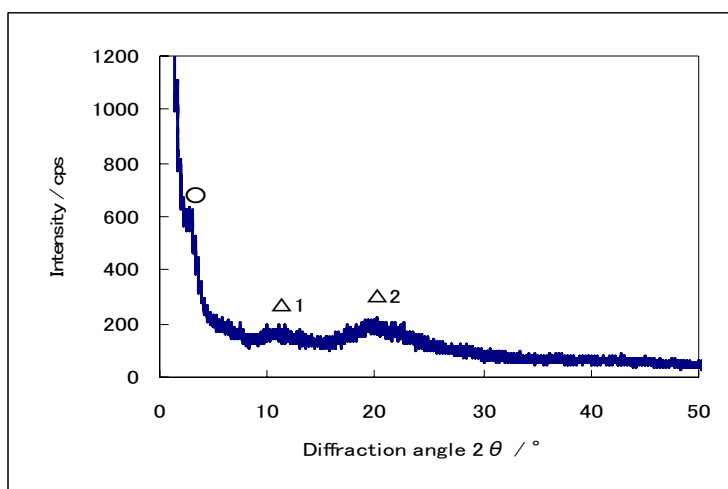
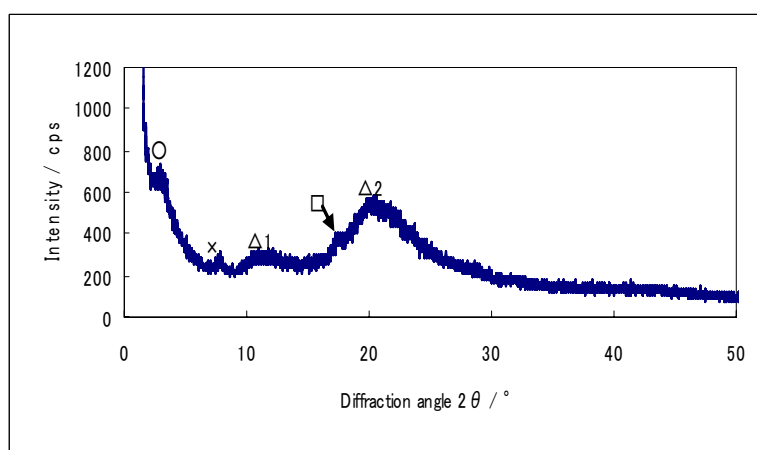


Fig. S8. Wide angle X-ray diffraction graphs of hybrid silica. (Acidic catalysis condition: 50 mg of LL-12, 1.0 mL of 1,4-dioxane, and 1.0 mL of 4.0 M HCl.)



**Fig. S9.** Wide angle X-ray diffraction graphs of hybrid silica. (Basic condition: 25 mg of LL-12, 1.0 mL of 1,4-dioxane, and 1.0 mL of 1.0 M NaOH)



**Fig. S10.** Wide angle X-ray diffraction graphs of hybrid silica. (TBAF condition: 100 mg of LL-12, 1.0 mL of dioxane, and 1.0 mL of 0.05 M TBAF)

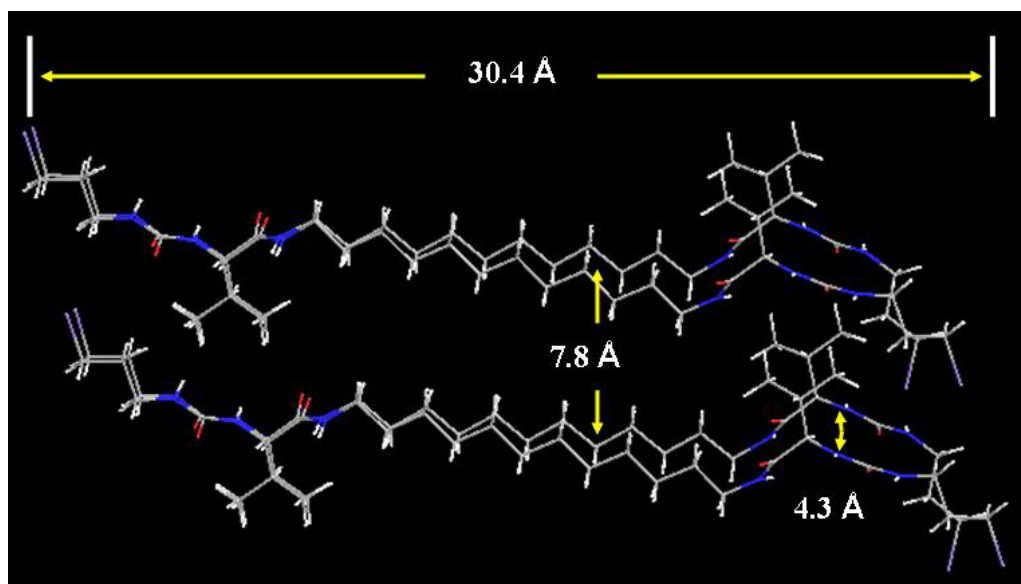


Fig. S11. Structure model of polysilsesquioxane LL-H12.



Fig. S12. Shape of a water droplet on the surface of DD-H12 thin film.