

Electronic Supplementary Information

## **In Situ Synthesis and Photoresponsive Rupture of Organosilica Nanocapsules**

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### **Experimental Section**

**Materials.** TCPTES was purchased from Gelest. Ammonia water (28%) and RB were purchased from Aldrich. Ibuprofen was purchased from Tokyo Chemical Industry. All reagents were used as received.

**Synthesis of TC-OS NCs.** TCPTES was mixed with ammonia water and the solution was heated at 100°C for a set time (0–24 h). The amount of ammonia water added was 1–15 equivalents relative to TCPTES. The synthesis conditions are summarized in Table 1. After reaction, the NCs were collected by centrifugation (20000×g). The NCs were then redispersed in ethanol and collected by centrifugation (20000×g) to eliminate residual TCPTES in the NCs. This wash sequence was repeated 3 times.

**Synthesis of RB-Encapsulating TC-OS NCs.** TCPTES (50 mM) and RB (0.35 mM) were mixed with ammonia water (1 equiv. relative to TCPTES) and the solution was heated at 100°C for a set time (12 h). These conditions were the same as those used for sample A. RB was miscible in TCPTES and therefore RB-containing TCPTES droplets were formed in the reaction solution. The hydrolysis and condensation of the RB-containing TCPTES droplets yielded RB-encapsulating TC-OS NCs as in the case when using plain TCPTES droplets. The NCs were collected by centrifugation (20000×g).

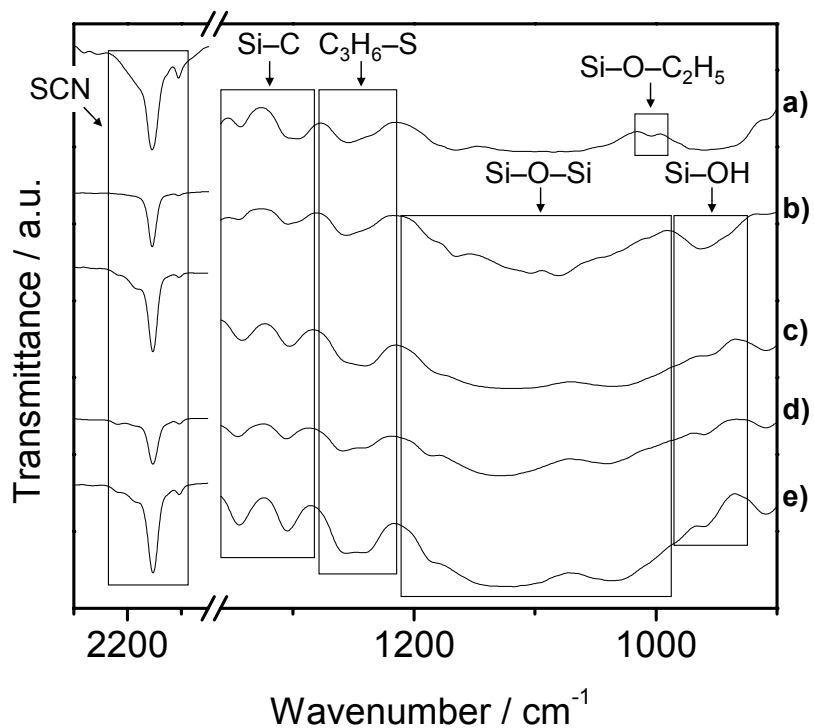
**Synthesis of Ibuprofen-Encapsulating TC-OS NCs.** Ibuprofen-encapsulating TC-OS NCs was synthesized in a similar way to the synthesis procedure for RB-encapsulating TC-OS NCs. TCPTES (50 mM) and ibuprofen (0.35 mM) were mixed with ammonia water (1 equiv.

relative to TCPTES) and the solution was heated at 100°C for a set time (12 h). The products were collected by centrifugation (20000×g). This wash sequence was repeated 3 times

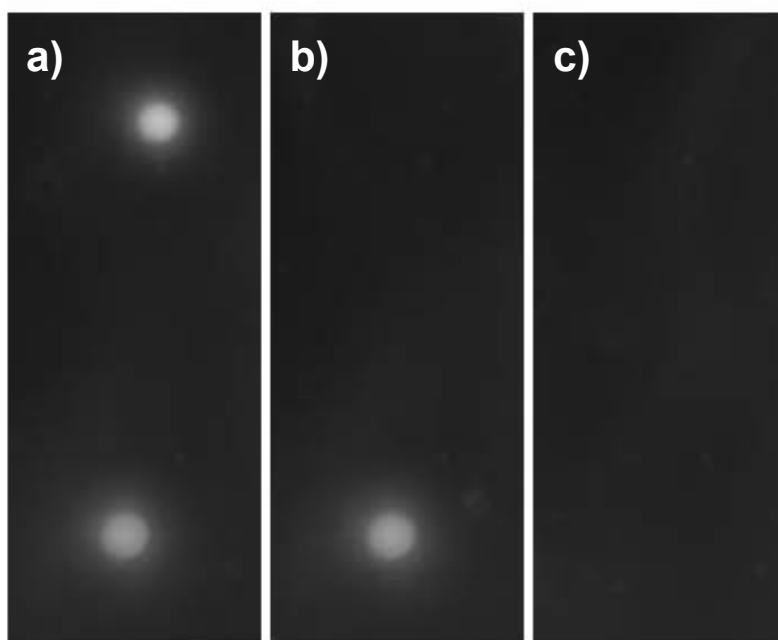
**Characterization.** TEM images were obtained on a H-7650 electron microscope (Hitachi, Tokyo, Japan). FTIR spectra were obtained using a FTIR-6200 instrument (JASCO, Tokyo, Japan).

**Fluorescence Microscope Observation of RB-Encapsulating TC-OS NCs.** RB-encapsulating TC-OS NCs were dispersed in water. The aqueous suspension containing RB-encapsulating TC-OS NCs were dropped onto a glass slide and then the optical responses of the NCs were observed by fluorescent microscopy. A TE2000 fluorescent microscope (Nikon, Kanagawa, Japan) was used, equipped with a 100 W mercury lamp as a light source and CCD camera (Rolera-XR Mono Fast 1394 Cooled, Qimaging, Burnaby, BC, Canada) with Image-Pro Plus software (MediaCybernetics, Silver Spring, MD).

**Release of Ibuprofen from TC-OS NCs.** Ibuprofen-encapsulating TC-OS NCs were dispersed in water and then the aqueous suspension was exposed to light with a 100 W mercury lamp for 1–15 min to rupture the NCs. After light irradiation for a given time, the NCs were separated from the suspension by centrifugation (20000×g). The supernatant liquid included ibuprofen released from the NCs and the amounts of ibuprofen in the supernatant were estimated based on the absorbance at 223 nm measured by UV-Vis spectrophotometer (NanoDrop 1000, Thermo Scientific, MA). This measurement was performed 3 times. We designated the absorbance of the solution containing ibuprofen-encapsulating TC-OS NCs before light irradiation as the amount corresponding to 100% release.



**Fig. S1** FTIR spectra of a) TCPTES, b) sample A, c) sample B, d) sample C and e) sample D.



**Fig. S2** Temporal change in fluorescence images of RB-encapsulating ultrathin TC-OS NCs under light irradiation; a) in 0 s, b) in 2 s and c) in 14 s.