Reply to the Comment on Spontaneously Ordered Sol–Gel Composites with Submicrometer Periodicity

Inorganic/organic composites with different forms (powders, films, and monoliths) have been obtained using different silicone surfactants as the templates under a variety of reaction conditions. It has been demonstrated that almost all of the silicone surfactant molecules enter into SiO_2 matrix when using silicone surfactants as the template for the preparation of the ZSU-L materials. Therefore, one should want to know what structure ZSU-L has in nature. A large number of experiments in our laboratory indicated that ZSU-L has lamellar structure, as confirmed by some evidence including the TEM images of an ultrathin section of the ZSU-L.[1−7] In my previous reply,[8] additional clear evidence was supplied to Su's comments. Here some new proof is presented for identifying the structure of the ZSU-L materials.

Recently, we have been preparing metal-doped ZSU-L materials. Figure 1 shows a typical transmission electron microscopy (TEM) image of an ultrathin section of Pt-doped ZSU-L products (SiO_2). It is clearly shown that Pt particles are arranged along surfactant bilayers, and patterned Pt nanoparticles are formed. Lattice fringes can be clearly seen in the high-resolution transmission electron microscopy (HRTEM) image (inset in Figure 1). It is impossible for ultramicrotoming to induce the formation of aligned Pt particles along the striped lines (surfactant layers). This result and my previous study support the suggestion that ZSU-L materials have macrolamellar structure. The detailed results about metal-doped ZSU-L materials will be published elsewhere and can be argued.

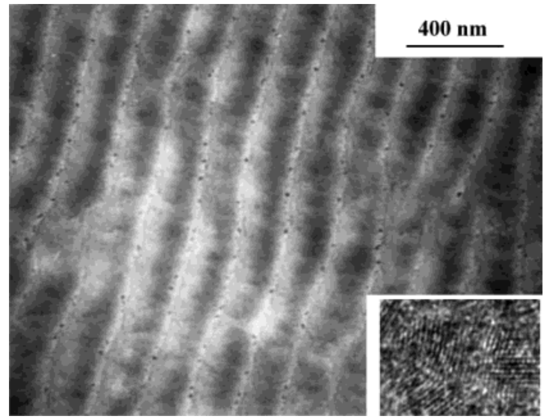

Figure 1. Representative TEM image of an ultrathin section of Pt-doped ZSU-L (SiO_2). Inset is HRTEM image of Pt particles.

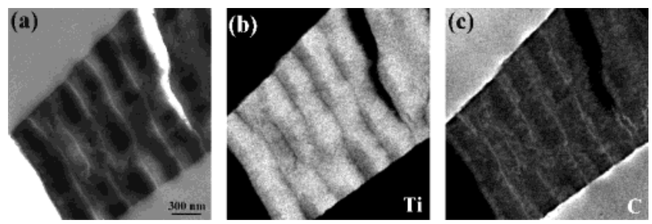

Figure 2. (a) TEM image of an ultrathin section of TiO_2/silicone surfactant composites (ZSU-L); (b) elemental Ti mapping of the image shown in (a); and (c) elemental C mapping of the image shown in (a).

To study whether silicone surfactant molecules were located between walls and walls, energy-filtering elemental mapping using high-resolution electron energy loss spectroscopy (HREELS) analysis was performed on an ultrathin section of the ZSU-L materials (TiO_2). Figure 2 depicts the bright field image (a) together with the elemental Ti (b) and C (c) mapping of the same region. The mapping clearly indicates that silicone surfactant bilayers were sandwiched by titania walls to form ordered macrolamellar structure. The ZSU-L products are constructed of silicone surfactant layers being sandwiched by thick titania walls that are arranged parallel to each othe, which indicates that macrolamellar structured TiO_2 was templated from silicone surfactant. This result supplies additional proof that the ZSU-L materials have macrolamellar structure, which would be not produced by ultramicrotoming.

Although it would not be conclusive, the present results indicate that it is most likely that ZSU-L inherently has macrolamellar structure. My present understanding of these materials is still limited, and a thorough investigation is under way to supply more data for identifying the structure of these materials.

An-Wu Xu

School of Chemistry and Chemical Engineering, Zhongshan University, Guangzhou 510275, China

Received February 26, 2003

CM031045C

* Address correspondence to Dr. Xu via fax (86-20-84111088) or e-mail (cedc17@zsu.edu.cn).

(1) Xu, A. W.; Cai, Y. P.; Zhang, H. X.; Zhang, L. Z.; Yu, J. C. *Angew. Chem., Int. Ed.* **2002**, *41*, 3844. Xu, A. W.; Cai, Y. P.; Zhang, H. X.; Zhang, L. Z.; Yu, J. C. *Angew. Chem.* **2002**, *114*, 4000.
(2) Xu, A. W.; Cai, Y. P.; Zhang, L. Z.; Yu, J. C. *Adv. Mater.* **2002**, *14*, 1064.
(3) Xu, A. W.; Yu, J. C.; Cai, Y. P.; Zhang, H. X.; Zhang, L. Z. *Chem. Commun.* **2002**, 1614.
(4) Xu, A. W. *Chem. Mater.* **2002**, *14*, 3625.
(5) Xu, A. W. *Chem. Lett.* **2002**, 982.
(6) Xu, A. W. *J. Phys. Chem. B* **2002**, *106*, 13161.
(7) Xu, A. W. *Chem. Lett.* **2002**, 878.
(8) Xu, A. W. *Angew. Chem., Int. Ed.* **2003**, *42*, 1574.

10.1021/cm031045c CCC: $27.50
Published on Web 12/16/2003