



Nanosheet synthesis using liquid crystals as templates

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Abstract

We have synthesized unprecedented amorphous aluminosilicate nanosheets by the Two-dimensional Reactor in Amphiphilic Phase (TRAP) method, which we proposed as a new nanosheet synthesis method using liquid crystals. Four types of zeolite nanosheets could be converted from a single type of amorphous aluminosilicate nanosheet by the Dry Gel Conversion (DGC) method using Structure-Directing Agent (SDA). Cracking tests of low-density polyethylene (LDPE) catalyzed by one of the zeolite nanosheets, CHA-type zeolite nanosheets, showed that the ratio of lower olefins was higher than that of conventional CHA-type zeolite crystals.

Background & Results

Nanostructured materials show shape-derived properties. Nanosheets are so thin that they are highly dispersible and exhibit nanosize effects, and they are easy to handle thanks to their width, which is more than 100 times greater than their thickness. Nanosheet synthesis by top-down exfoliation, a method commonly used in the past, cannot synthesize nanosheets of non-exfoliated materials. Therefore, bottom-up methods by anisotropic growth using various interfaces as templates have been investigated. A synthesis method that can be scaled up for practical use and that allows control of nanosheet size at low cost is required. The authors have recently developed a nanosheet synthesis method using liquid crystals as templates (TRAP method).

Here, we proposed a new method to synthesize amorphous aluminosilicate nanosheets using the TRAP method and to convert them into various types of zeolite nanosheets by DGC. Four types of zeolite nanosheets were obtained from the same amorphous aluminosilicate.

First, amorphous aluminosilicate nanosheets were synthesized using hydrophilic TRAPs in the HL phase of decane solutions of ionic amphiphilic molecules. The raw materials were tetraethyl orthosilicate and aluminum isopropoxide. The resulting powder was an amorphous aluminosilicate nanosheet with a thickness of 1.96 ± 0.67 nm and a width of 395 ± 137 nm.

The amorphous aluminosilicate nanosheets were then immersed in an aqueous solution of SDA and dried to form a gel. Crystallization in a steam at 160°C , we obtained four types of zeolite nanosheets (PHI, CHA, SOD, and MFI types) by changing the structure-defining agent.

Furthermore, we confirmed that the CHA-type zeolite nanosheets work as the catalyst of cracking of low-density polyethylene (LDPE). The ratio of lower olefins was better than conventional CHA-type zeolite crystals.

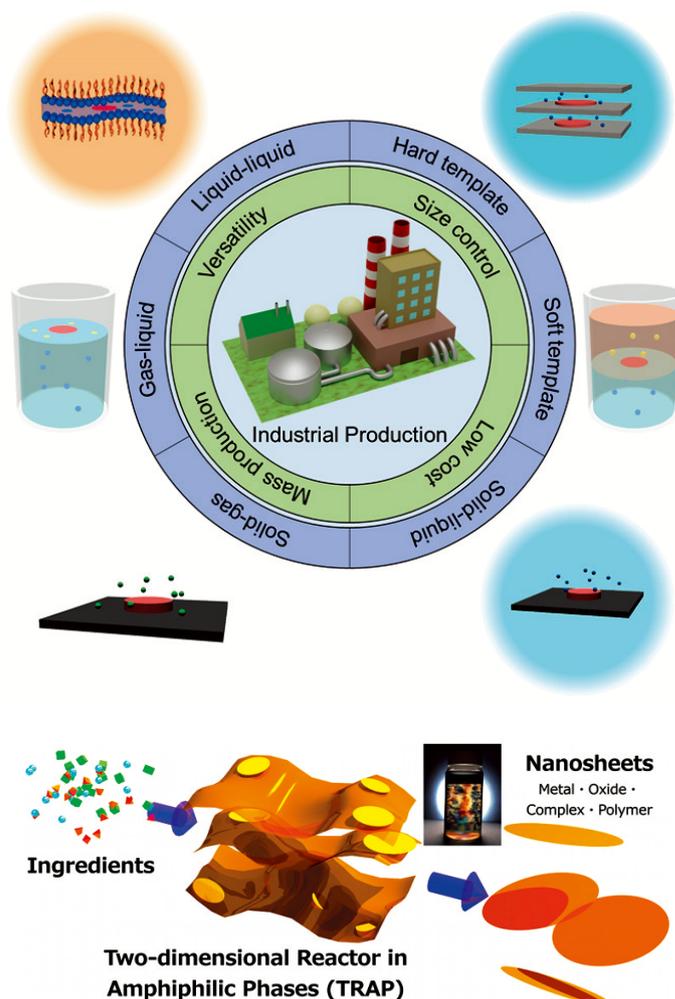
Significance of the research and Future perspective

The methodology of this study is applicable to many types of zeolites. Due to their thickness and large external surface area, zeolite nanosheets have great potential for various applications such

as methanol-olefin catalysts, polymer degradation, and separation membranes.

The MFI and CHA types in this study are commonly used as catalysts; MFI-type zeolite nanosheets have been reported in another nanosheet synthesis methods, but this is the first report on the CHA type.

This method has another advantage; it is easy to scale up this method due to the use of self-assembly as templates and the low cost of the amphiphilic molecules.

**Patent** JP7290230, JP6978783Sasaki, Koki; Uchida, Yoshiaki; Nishiyama, Norikazu. Bottom-up synthesis of nanosheets at various interfaces. *ChemPlusChem*. 2023, 88 (10), e202300255. doi: 10.1002/cplu.202300255**Treatise** Sasaki, Koki; Uchida, Yoshiaki; Nishiyama, Norikazu et al. Amorphous aluminosilicate nanosheets as universal precursors for the synthesis of diverse zeolite nanosheets for polymer-cracking reactions. *Angewandte Chemie International Edition*. 2022, 61 (46), e202213773. doi: 10.1002/anie.202213773
Uchida, Yoshiaki; Nishizawa, Takuma; Nishiyama, Norikazu et al. Nanosheet formation in hyperswollen lyotropic lamellar phases. *Journal of the American Chemical Society*. 2016, 138 (4), 1103-1105. doi: 10.1021/jacs.5b11256**URL** <http://www.cheng.es.osaka-u.ac.jp/nishiyamalabo/english/research/359/3141.html>**Keyword** aluminosilicate, lamellar phase, nanosheets, zeolite, catalysts