Supporting Information


Sara Inoue and Shinobu Fujihara*

Department of Applied Chemistry, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan

E-mail: shinobu@applc.keio.ac.jp

One may wonder whether the 2D-structured LBZB (layered basic zinc benzoate) film could be obtained exclusively by our method utilizing the continuous transport of benzoic acid through the immiscible liquid–liquid interface. For a comparative study, we tried to prepare LBZB in a single-phase system where all the components (zinc nitrate hexahydrate, urea, and benzoic acid) were dissolved in water.

The concentrations of benzoic acid, Zn$^{2+}$, and urea in the aqueous solution were adjusted to 0.02, 0.1, and 1.0 mol/dm$^3$, respectively, corresponding to [B] : [Z] : [U] = 0.2 : 1 : 10 in one liquid phase. It should be noticed that the concentration of benzoic acid was limited as low as 0.02 mol/dm$^3$ in this case, because of its relatively low solubility in water. The solution was maintained at 80 °C for 24 h, similarly to the experiment for the biphasic xylene–water system.

It was found that a solid phase (designated sample E) was formed on the internal surface of the container by the heterogeneous nucleation. Figure S1 shows an XRD pattern and TG-DTA curves of the sample E. No diffraction peak is seen in the lower 2θ range (2θ < 10 °), indicating that LBZB is not
formed in the single-phase medium. Instead, all of diffraction peaks can be indexed as zinc carbonate hydroxide, Zn$_5$(CO$_3$)$_2$(OH)$_6$ (ICDD 19-1458). In the TG-DTA curves, first two weight losses with endothermic peaks at 118 and 153 °C are assigned to the departure of water and the dehydration of the zinc hydroxide layers, respectively. A gradual weight loss occurring at temperatures between 150 and 240 °C is similar to the pyrolysis behavior of layered basic zinc carbonate (LBZC), which undergoes the release of CO$_3^{2-}$ in the temperature range of 180 – 260 °C.\textsuperscript{7,13} However, the last weight loss between 350 and 400 °C, accompanied by an exothermic peak at 370 °C, cannot be explained with the Zn$_5$(CO$_3$)$_2$(OH)$_6$ composition. According to the FT-IR analysis, we noticed the presence of the benzoate anion in the sample E, possibly resulting from the chemical adsorption. As observed in the TG-DTA of LBZB (see Figure 4), the last weight loss is ascribed to the decomposition and oxidation of the benzoate group.

From the XRD, FT-IR, and TG-DTA measurements, the sample E obtained in the single, aqueous medium was basically the layered hydroxide zinc compound intercalating the carbonate ions into the zinc hydroxide layers, in spite of the presence of the benzoate anion. This result indicates that LBZB is not formed in the aqueous phase unless benzoic acid is continuously delivered from the organic phase under the distribution law.

![Figure S1.](image.png)

**Figure S1.** (a) An XRD pattern and (b) TG-DTA curves of the sample E synthesized in the single liquid phase.