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Electronic Supplementary Information for "Formation of ZnO Hexagonal Micro-Pyramids: A Successful Control of the Exposed Polar Surfaces with the Assistance of an Ionic Liquid"

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1. Experimental Detail.

Synthesis of ZnO hexagonal micro-pyramids. In a typical process, the mixture of oleic acid (OA) and ethylenediamine (or trioctylamine, or methylamine) was used as the solvent, the growth environment. 6.6 ml OA and 4.5 ml ethylenediamine (or 30 ml trioctylamine, or 3.2 ml 30% aqueous methylamine solution) were mixed to get an yellow liquid at room temperature, and then 1.46 g anhydrous zinc acetate (Zn (CO_2CH_3)₂) was added. The resulting mixture was rapidly heated to 286°C in 10-15min, and a transparent solution was obtained. Then the solution was kept at 286°C for 1h for the thermo-decomposition of zinc acetate to ZnO. After reaction, the precipitates were collected, washed several times with hexane and ethanol. The final product was dispersed in ethanol for further characterization.

Characterization of Materials. The crystal phase was determined by powder X-ray diffraction (XRD, Panalytical X-Pert diffractometer with copper K α radiation). The morphology, and size of the ZnO hexagonal micro-pyramids were characterized by scanning electron microscopy (SEM, LEO1530). The structures of the ZnO hexagonal micro-pyramids were examined using a FEI Tecnai-F30 FEG instrument with an acceleration voltage of 300 kV. Samples for the electron microscope were prepared by ultrasonic dispersion of the as-prepared product with ethanol. Then, the suspension was dropped onto a conventional carbon-coated copper grid and dried in air before the analysis. Photoluminescence (PL) of the as-prepared products was measured at room temperature with a Hitachi luminescence spectrometer (F-4500) using a Xenon discharge lamp as the excitation light source and the excited wavelength was 325 nm.

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2. ZnO hexagonal micro-pyramid prepared from various mixture of oleic acid and amines



Figure S1. SEM images of ZnO products from the solvent of (A) the mixture of oleic acid and ethylenediamine, (b) the mixture of oleic acid and trioctylamine, and (C) the mixture of oleic acid and methylamine. The results show that the ZnO hexagonal micro-pyramid can be prepared from various mixture of oleic acid and amines.

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3. SEM images of some incomplete ZnO hexagonal micro-pyramids.

Figure S2. (A) SEM image, (B) and (D) HRTEM image and SAED pattern, (C) Schematic model of truncated ZnO micro-pyramids coexisted in the products. The HRTEM image and the SAED pattern in Figure 2(B) show that the truncated micro-pyramid has the same structure as normal ZnO hexagonal micro-pyramid. These truncated ZnO micro-pyramids should be the incomplete ZnO micro-pyramids during the growth. However, the top surface is rather rough, while the bottom surface is relatively flat and smooth. The top surface of the truncated micro-pyramid is (0001) plane with Zn polarity (see Figure 2 (C)), and has been thought to be more active than the oxygenterminated (000 $\overline{1}$) plane, which may also result in the rough surface due to its growth in a very fast rate. As a result, (0001) surface with Zn polarity becomes smaller and smaller during the growth process, and finally vanishes and forms an ideal pyramid. Figure 2D shows a top view TEM image of a truncated micro-pyramid and its corresponding SAED pattern. The SAED pattern can be indexed as diffractions from [0001] zone axis. The indices of diffraction spots imply that side surfaces should be {10 $\overline{1}$ } surfaces.