

## Supplementary Information

# Formation of Double-Cone-Shaped ZnO Mesocrystals by Addition of Ethylene Glycol to ZnO Dissolved Choline Chloride–Urea Deep Eutectic Solvents and Observation of Their Manners of Growth

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**Table S1.** Experimental conditions for the growth of ZnO crystals in CU-DES:ZnO mixed with EG

Run No.	CU-DES:ZnO ZnO conc. / mass%	CU-DES:ZnO mass / g	EG volume / mL	EG addition rate / mL min <sup>-1</sup>	Holding temp. / °C	Aging time / h
1	3.3	20	80	1.0	150	24
2	7.2	20	80	1.0	150	24
3	16.7	20	80	1.0	150	24
4	16.7	20	80	1.0	150	0
5	16.7	20	80	1.0	150	12
6	16.7	20	80	2.0	150	24
7	16.7	20	80	8.0	150	24
8	16.7	20	80	1.0	120	24
9	16.7	20	80	1.0	90	24

*Detail of calculation of crystallite size by Scherrer's equation*

According to “X-ray Diffraction Procedures: For Polycrystalline and Amorphous Materials 2<sup>nd</sup> ed., (Harold P. Klug, Leroy E. Alexander, JOHN WILEY & SONS, New York, 1967)”, “Elements of X-ray Diffraction 2<sup>nd</sup> ed. (B. D. Cullity, Addison-Wesley, Massachusetts, 1978)”, if we suppose small uniform cubic crystallite, the crystallite size is calculated by Scherrer's equation;

$$L = \frac{0.94\lambda}{\beta \cos\theta}$$

$\theta$ : angle of diffraction peak,  $\lambda$ : wavelength of X-ray,  $L$ : diameter of cubic crystallite,  $\beta$ : full width of half maximum (FWHM) of specific peak.

$\beta$  is calculated by the following equation;

$$\beta^2 = B_M^2 - B_S^2$$

$B_M$ : measured FWHM,  $B_S$ : FWHM of standard sample (crystallite size > 100 nm)

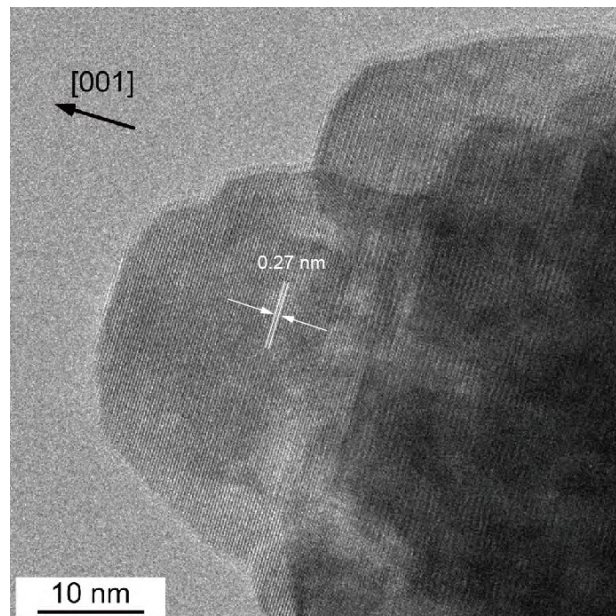
In this study, polycrystalline silicon powder with particle size of 5  $\mu\text{m}$  (KOJUNDO Chemical Laboratory Co., Ltd., 99.9%) was used as a standard sample.  $B_S$  was 0.11°, which was average FWHM of {111}, {220}, and {311}.

The crystallite sizes of the obtained samples are calculated from Scherrer's equation using their XRD peaks. The crystallite sizes and the average particles sizes are summarized in the **Table S2**;

**Table S2.** Crystallite size of the samples calculated from Scherrer's equation and particle dimensions the particles measured from SEM images

Run No.	Crystallite size calculated from each XRD peak / nm			Particle size / $\mu\text{m}$
	{002}	{110}	{112}	Length $\times$ Width
1	29	23	22	$1.05 \times 0.41$
2	45	30	27	$2.42 \times 0.81$
3	45	29	27	$3.54 \times 1.26$
4	27	19	18	n.d.
5	47	25	23	$1.12 \times 0.56$
6	45	34	29	$2.07 \times 0.82$
7	49	32	27	$1.32 \times 0.61$
8	48	27	27	$1.05 \times 0.45$
9	24	19	15	n.d.

The double-cone-shaped and ellipsoidal ZnO mesocrystals were obtained in **Run Nos. 1, 2, 3, 5, 6, 7, and 8.**



**Figure S1.** Bright-field TEM images and SAED patterns of a ZnO particle prepared with CU-DES (ZnO 16.7 mass%) and aging time for 12 h (**Run No. 5**). Arrows in the TEM images indicate a direction along [001]