



# Supplementary Materials

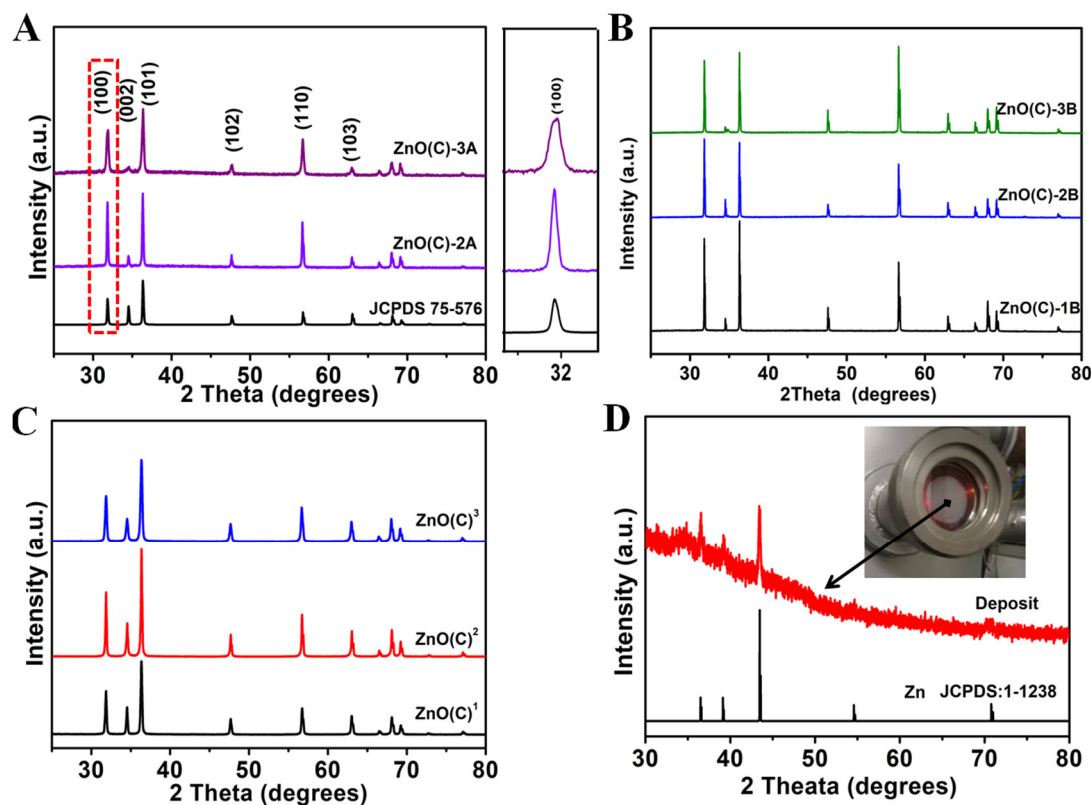
## Fabrication of ZnO Ceramics with Defects by Spark Plasma Sintering Method and Investigations of Their Photoelectrochemical Properties

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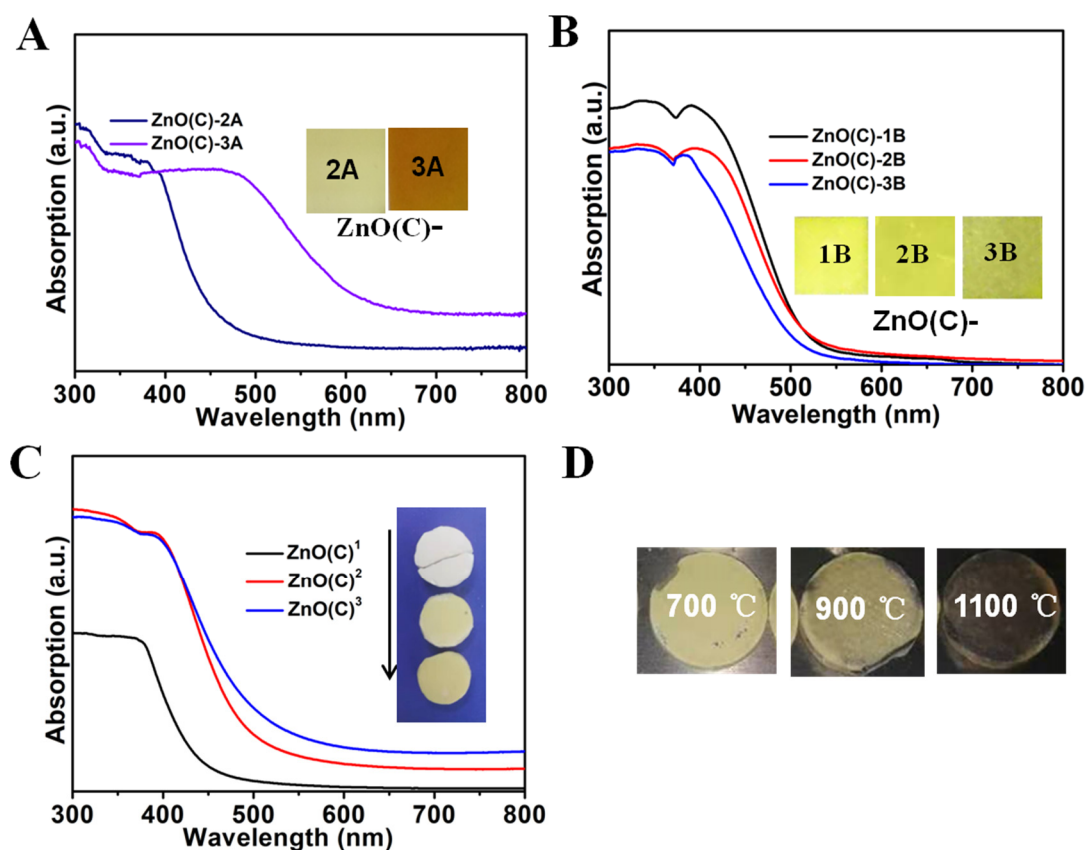
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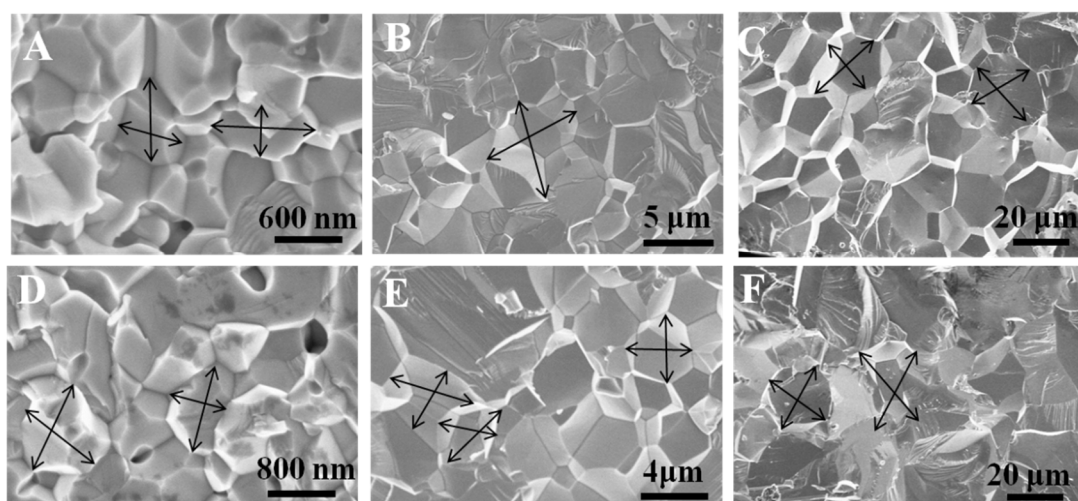
**Figure S1.** (A) XRD patterns of the ZnO(C)-2A (3A) samples; (B) XRD patterns of the ZnO(C)-1B (2B and 3B) samples; (C) XRD patterns of the ZnO(C)<sup>1</sup>, ZnO(C)<sup>2</sup> and ZnO(C)<sup>3</sup> samples; (D) XRD of the deposits on the inner surface of quartz sheet. insert: digital photograph of the observation window of a vacuum furnace with silver-gray zinc deposited on the inner surface of the quartz glass.

**Table S1.** The quality changes of the three samples after 1h treatment in 1100 °C vacuum condition.

	ZnO(C)-1	ZnO(C)-2	ZnO(C)-3
before	39.2 mg	44.7 mg	45.5 mg
after	28.5 mg	35.3 mg	36.1 mg



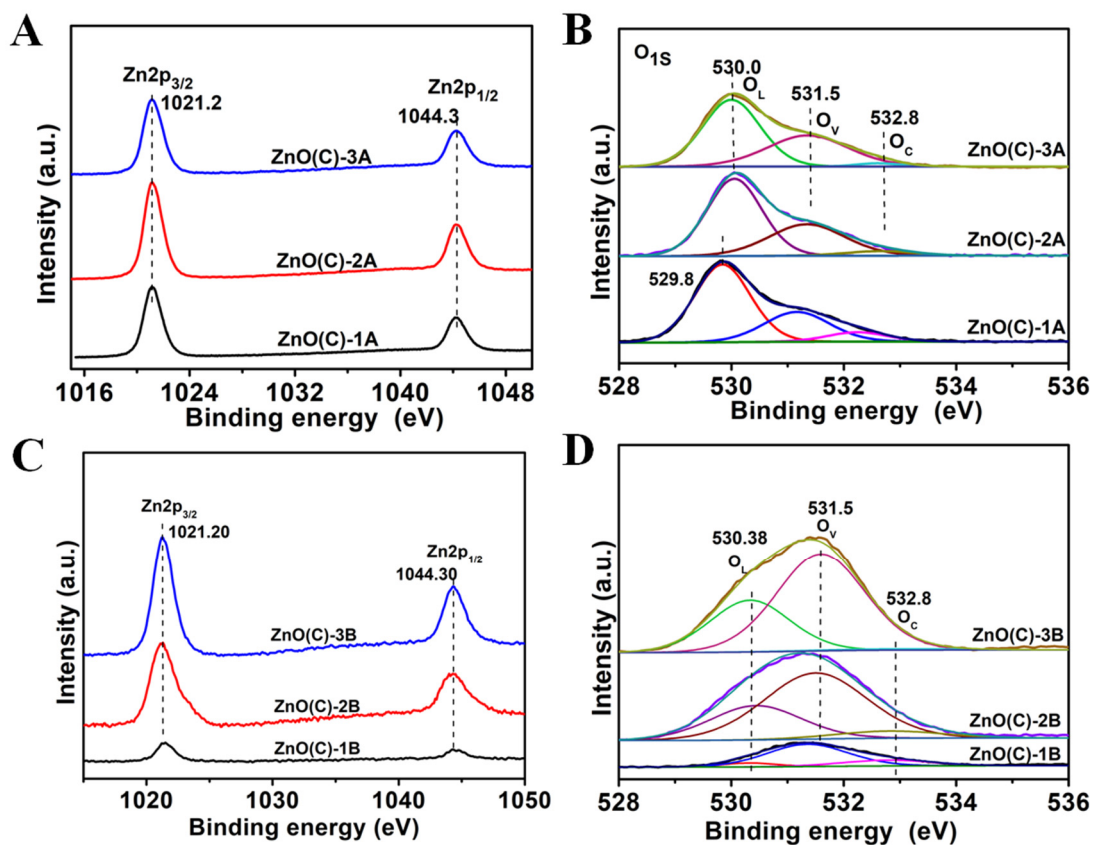
**Figure S2.** (A) UV-Vis DRS of the ZnO(C)-2A (3A) samples, insert: the digital photographs of ZnO(C)-2A (3A); (B) UV-Vis DRS of the ZnO(C)-1B (2B and 3B) samples, insert: the digital photographs of ZnO(C)-1B (2B and 3B); (C) UV-Vis DRS of the ZnO(C)<sup>1</sup>, ZnO(C)<sup>2</sup> and ZnO(C)<sup>3</sup> samples, insert: the digital photographs of the three samples; (D) The digital photographs of three ceramics prepared at different temperatures, with graphite paper around them cleaned with sandpaper.

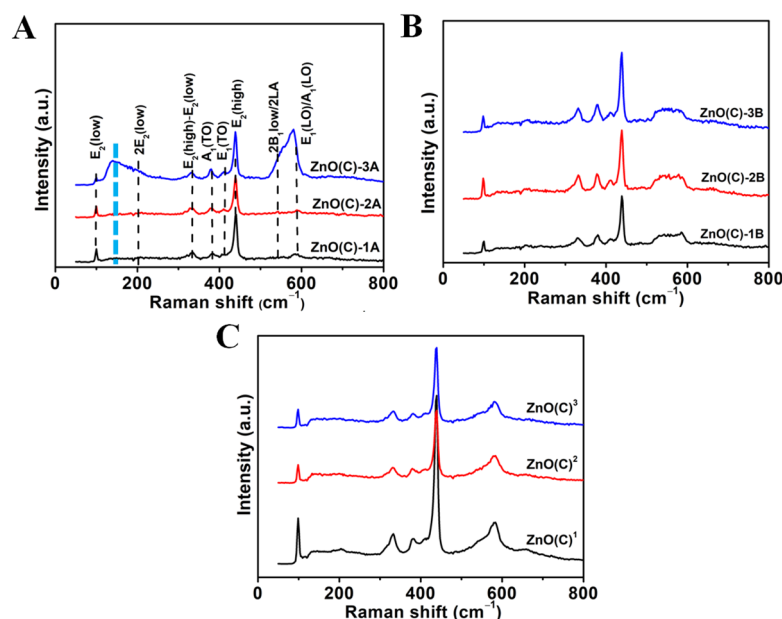


**Figure S3.** SEM images of the ceramics obtained at different temperature and the ceramics annealed at 500 °C for 5 h in ambient, (A) ZnO(C)-1, (B) ZnO(C)-2, (C) ZnO(C)-3, (D) ZnO(C)-1A, (E) ZnO(C)-2A and (F) ZnO(C)-3A. Insert: the “ $\longleftrightarrow$ ” represents the auxiliary line for grain size measurement.

**Table S2.** The density ( $\rho$ ) and relative density of the ZnO powder and ceramics prepared at 700 °C, 900 °C and 1100 °C.

Temperature	700 °C		900 °C		1100 °C	
Density and Relative density	$\rho$ (g/cm <sup>3</sup> )	Relative density (%)	$\rho$ (g/cm <sup>3</sup> )	Relative density (%)	$\rho$ (g/cm <sup>3</sup> )	Relative density (%)
SPS method	5.54	98.1	5.65	99.6	5.70	100
Traditional sintering method	4.90	86.7	5.60	99.1	5.65	99.6

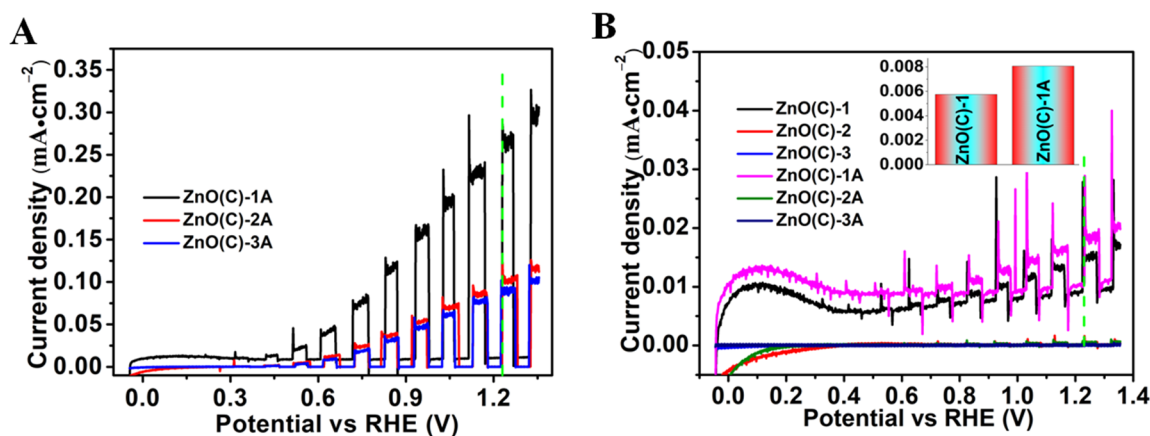
**Figure S4.** XPS patterns of the ZnO powder and ZnO(C)-1A (2A and 3A), (A) Zn 2p peak and (B) O1s peak; XPS patterns of the ZnO(C)-1B (2B and 3B), (C) Zn 2p peak and (D) O1s peak.



**Figure S5.** (A) Raman spectrum of ZnO(C)-1A (2A and 3A) samples; (B) Raman spectrum of ZnO(C)-1B (2B and 3B) samples; (C) Raman spectrum of ZnO(C)<sup>1</sup>, ZnO(C)<sup>2</sup> and ZnO(C)<sup>3</sup> samples.

**Table S3.** The resistivity mobility and density results of ZnO(C)-1, ZnO(C)-2 and ZnO(C)-3.

	ZnO(C)-1	ZnO(C)-2	ZnO(C)-3
Resistivity ( $\Omega\cdot\text{cm}$ )	1187.52	0.1435	0.0561
Mobility ( $\text{cm}^2/\text{Vs}$ )	7.94	26.63	38.94
Density ( $\text{cm}^{-3}$ )	$6.65 \times 10^{14}$	$1.63 \times 10^{18}$	$2.86 \times 10^{18}$



**Figure S6.** (A) Photocurrent density versus applied potential (J-V) curves measured for the ZnO(C)-1A (2A and 3A) photoanodes with a scan rate of 20 mV/s under AM 1.5G illumination or visible light irradiation in 0.1 M NaSO<sub>4</sub> solution (PH = 6.8) and (B) Photocurrent density versus applied potential (J-V) curves measured for the ZnO(C)-1 (2 and 3) and ZnO(C)-1A (2A and 3A) photoanodes with a scan rate of 20 mV/s under visible light irradiation by 300W Xe arc lamp equipped with a UV/IR cut filter ( $\lambda \geq 420$  nm) inset, the photocurrent values at 1.23 V<sub>RHE</sub>.

**Table S4.** Photocurrents ( $I_{1.5}$  and  $I_v$ ) of ZnO(C)-1(1A) under the AM 1.5G illumination and visible light at 1.23 V<sub>RHE</sub>.

	$I_{1.5}$ (mA/cm <sup>2</sup> )	$I_v$ (mA/cm <sup>2</sup> )	$I_v/I_{1.5}$
ZnO(C)-1	0.14	0.005	3.6 %
ZnO(C)-1A	0.27	0.008	2.9 %