Supplementary Information

In situ TEM observations of growth mechanisms of PbO

nanoparticles from Sm-doped PMN-PT matrix

Shuang Zhang^{a,†}, Xue Tian^{a,†}, Ying Zheng, Yongcheng Zhang^{a,*}, Wanneng Ye^{a,*}

^a College of Physics, State Key Laboratory of Bio-fibers and Eco-textiles, Qingdao University, Qingdao 266071, China

* Corresponding authors. E-mail addresses: qdzhyc@163.com, ywn@qdu.edu.cn.

[†] Shuang Zhang and Xue Tian contributed equally to this work.

Negligible heating effect of electron beam

The maximum temperature rise T of sample caused by the electron beam is calculated,¹

$$\Delta T = \frac{I}{\pi k e} \left(\frac{\Delta E}{t} \right) \ln \frac{b}{r \theta}$$

(S1)

where *I* is the beam current, *k* is the thermal conductivity, *e* is the electron charge, ΔE is the total energy loss per electron in a sample of thickness t,² *b* is the carbon film radius, r_0 is the beam radius. In our experimental conditions, the accelerate voltage of the TEM is 200 keV, the electron beam current density is 60 pA/cm², I = 9.6 nA, b = 1.5 mm, $r_0 = 400$ nm, k = 2.683 W/m/K.³ The maximum temperature rise is calculated to be 3 K, which is consistent with previous reports.^{2,4,5} Thus, the temperature rise caused by electron beam irradiation in this work can be ignored.

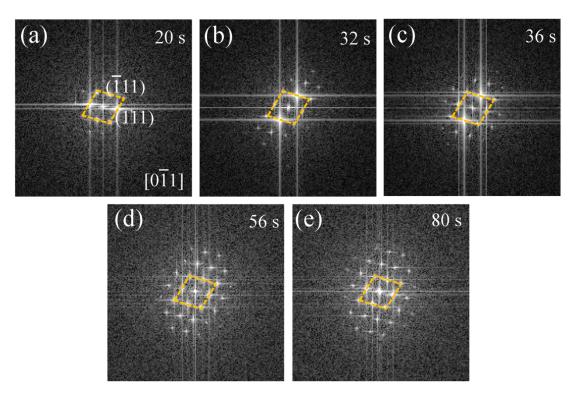


Fig. S1. The FFT patterns correspond to the HRTEM images of newly formed PbO nanoparticle in Fig. 1(b)–(f).

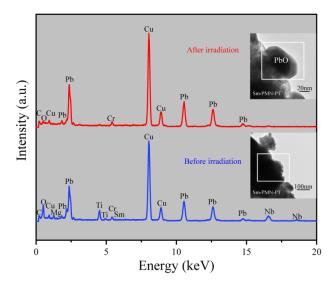


Fig. S2. EDX spectra obtained from Sm-PMN-PT particle (blue line) before and (red line) after electron beam irradiation. The inset images show the observed area.

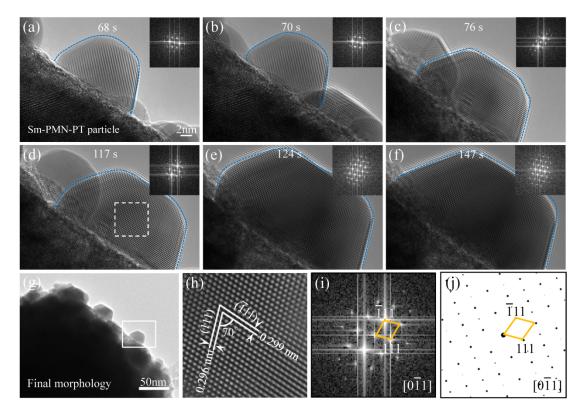


Fig. S3. The growth of nanoparticle (marked by blue dot line) separates from Sm-PMN-PT particle with 5 %mol excess PbO. (a)–(f) The growth process of nanoparticle and the FFT patterns in the upper right corner correspond to HRTEM images, respectively. (g) The final morphology of the focused area. (h) The enlarged area and (i) the corresponding FFT pattern obtained from the area marked white dot line in (d). (j) The simulated pattern along $[0\bar{1}1]$.

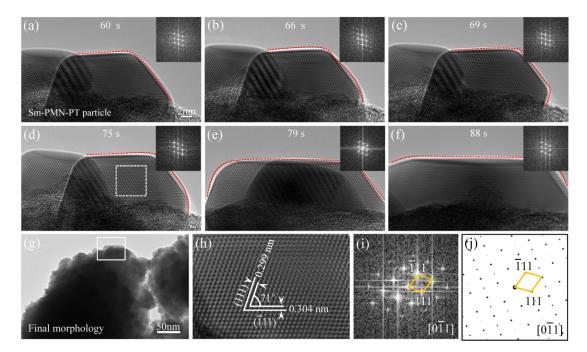


Fig. S4. The growth of nanoparticle (marked by red dot line) separates from Sm-PMN-0.30PT particle with 15 %mol excess PbO. (a)–(f) The growth process of nanoparticle and the FFT patterns in the upper right corner correspond to HRTEM images, respectively. (g) The final morphology of the focused area. (h) The enlarged area and (i) the corresponding FFT pattern obtained from the area marked with white line in (d). (j) The simulated pattern along $[0\bar{1}1]$.

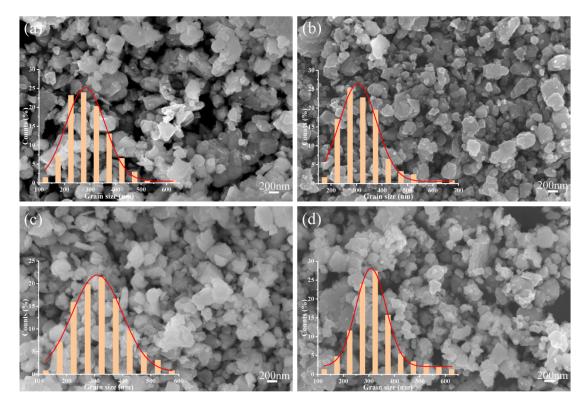


Fig. S5. (a)–(d) SEM images and the statistical distribution of grain sizes of Sm-PMN-PT powder samples with normal stoichiometry, 5 mol%, 10 mol% and 15 mol% excess PbO, respectively.

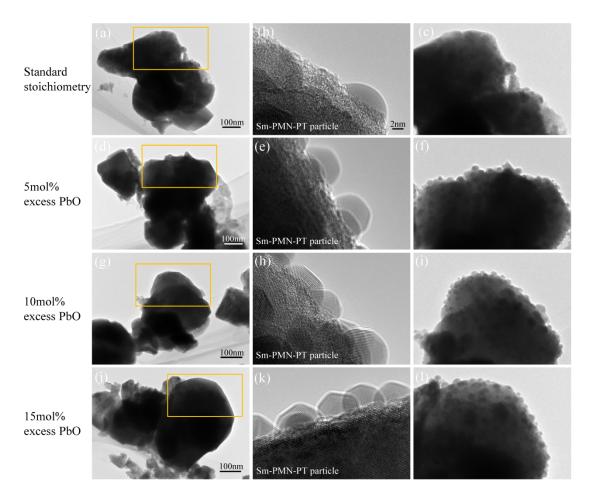


Fig. S6. The morphologies of Sm-PMN-PT powder samples with normal stoichiometry, 5 mol%, 10 mol% and 15 mol% excess PbO before and after electron beam irradiation.

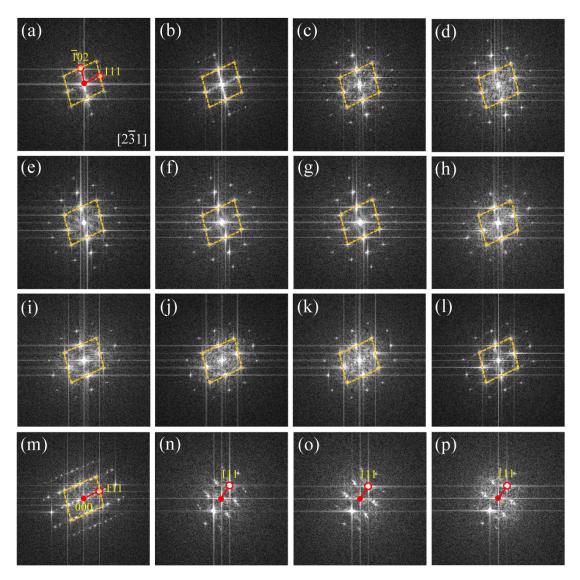


Fig. S7. The FFT patterns correspond to PbO nanoparticle 1 in Fig. 5.

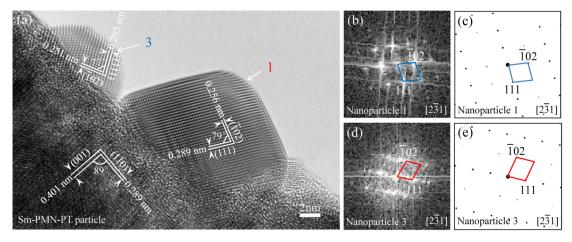


Fig. S8. (a) The indexed crystal planes of Sm-PMN-PT matrix, PbO nanoparticles 1 and 3. (b) and (d) corresponding FFT patterns of nanoparticles 1 and 3 in (a). (c) and (e) simulated patterns along $[2\overline{3}1]$ based on orthorhombic structured PbO.

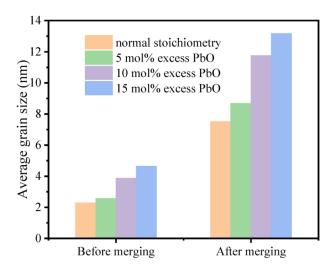


Fig. S9. The average sizes of PbO nanoparticles formed from four Sm-PMN-PT samples with normal stoichiometry, 5 mol%, 10 mol% and 15 mol% excess PbO before and after merging.

Supplementary Movie S1 shows PbO nanodroplets are separating from Sm-PMN-PT particle and these nanodroplets are constantly fluctuating in the initial stage of electron beam irradiation.

Supplementary Movie S2 shows a PbO nanoparticle and a small PbO nanodroplet rapidly merge into a large PbO nanoparticle.

References

- I. Jenčič, M. W. Bench, I. M. Robertson and M. A. Kirk, J. Appl. Phys., 1995, 78, 974–982.
- 2 W. Qin, T. Nagase and Y. Umakoshi, Acta Mater., 2009, 57, 1300–1307.
- 3 M. M. Ammar, M. M. Halawa, N. A. Ghoneim, A. F. Abbas and H. A. El Batal, J. Am. Ceram. Soc., 1982, 65, c174–c175.
- 4 S. M. Liu, A. Ishii, S. B. Mi, S. Ogata, J. Li and W. Z. Han, *Small*, 2022, 18, 2105881.
- 5 T. Yokota, M. Murayama and J. M. Howe, *Phys. Rev. Lett.*, 2003, **91**, 265504.