

## Supplementary Information (SI)

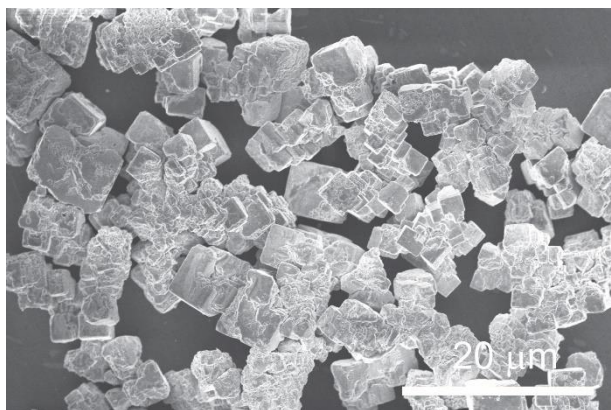
### Morphology control of $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$ crystallites under alkaline hydrothermal conditions

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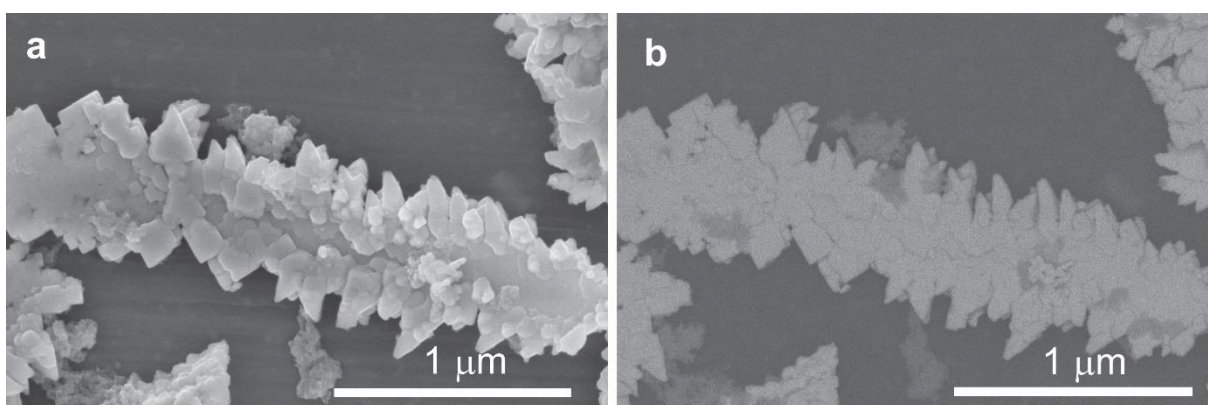
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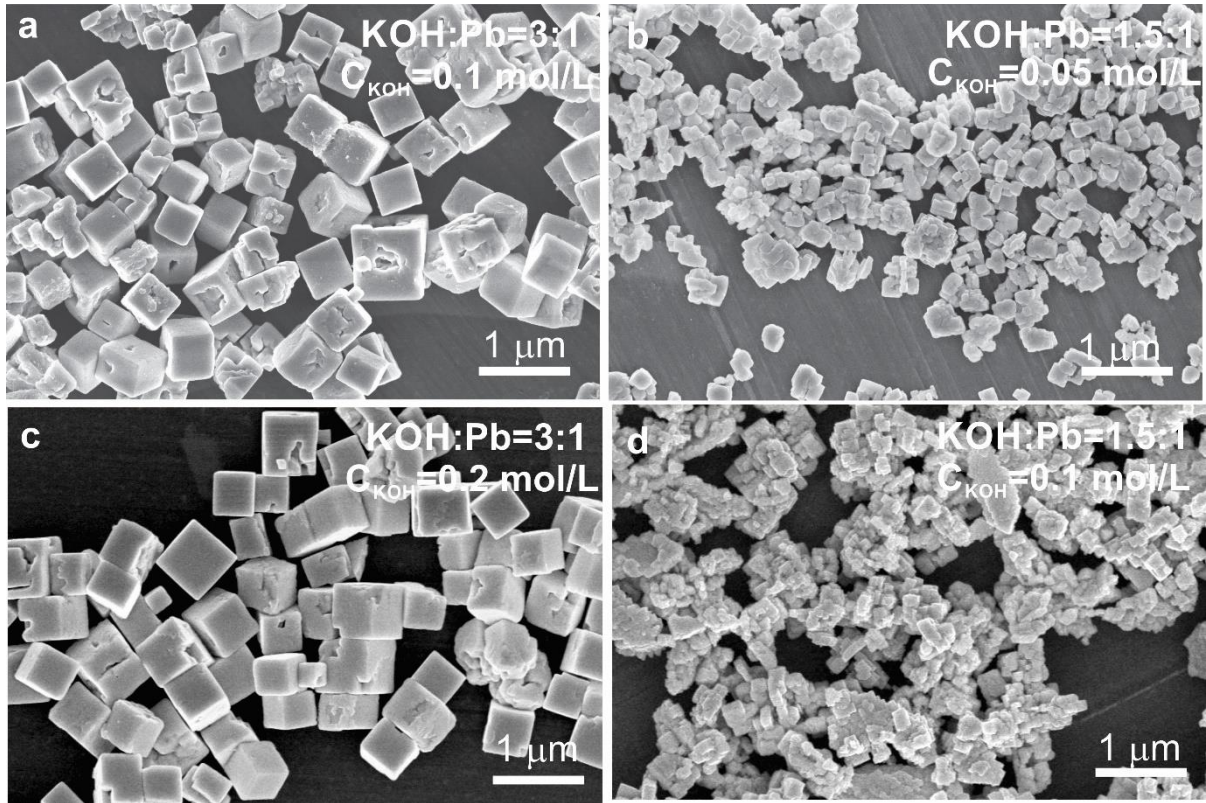
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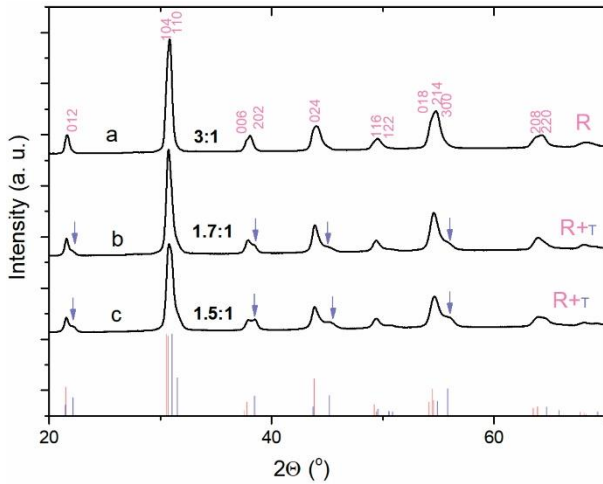
**Figure S1:** SEM micrograph (SE image) of the PZT06 reaction product obtained through one-step precipitation process and hydrothermal reaction at 235°C (12h) at KOH:Pb=20:1.



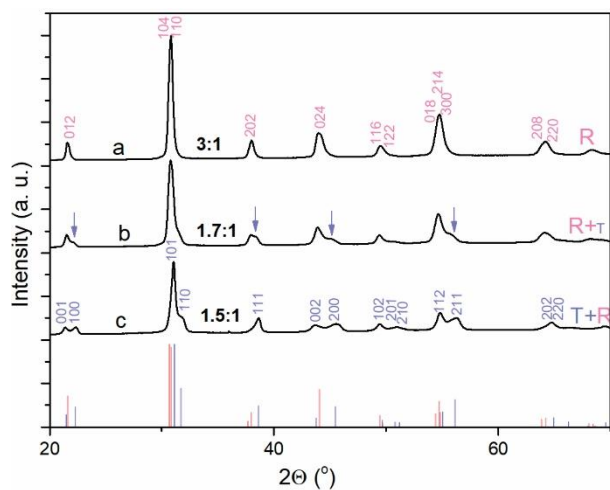
**Figure S2:** SEM micrographs (a, b) of the PZT06 reaction product obtained through one-step precipitation process and hydrothermal reaction at 235°C (12h) at KOH:Pb=3:1: (a)  $\Rightarrow$  SE image, b $\Rightarrow$ BSE image.



**Figure S3:** SEM micrographs of the PZT06 reaction product obtained through two-step precipitation process and hydrothermal reaction at 235°C (12h) (a, b) using the concentrations of Pb-, Zr- and Ti-precursors as shown in Table 1 ( $C_{\text{TALH}}=0.0112$  mol/L,  $C_{\text{ZrOCl}_2}=0.0168$  mol/L,  $C_{\text{Pb}(\text{CH}_3\text{COO})_2}=0.035$  mol/L) and (c, d) two-times larger concentrations of Pb-, Zr- and Ti-precursors ( $C_{\text{TALH}}=0.0224$  mol/L,  $C_{\text{ZrOCl}_2}=0.0336$  mol/L,  $C_{\text{Pb}(\text{CH}_3\text{COO})_2}=0.07$  mol/L). The concentrations of KOH was doubled in c, resulting in KOH:Pb=3:1, but in d remained the same as for normal Pb- Zr-, Ti-concentration experiment (0.1 mol/l), giving KOH:Pb=1.5:1.



**Figure S4:** Powder XRD pattern of the PZT07 reaction product obtained through two-step precipitation process and hydrothermal reaction at 235°C (12 h) and at (a) KOH:Pb=3:1, (b) KOH:Pb=1.7:1, (c) KOH:Pb=1.5:1. The main diffraction lines of rhombohedral (R) and tetragonal (T) phase are indexed according to ICDD pdf reference code 04-016-9524 (S.G.: R3c) and ICDD pdf reference code 01-070-4262 (S.G.: P4mm), respectively. The prevailing phase is denoted with larger letter. The blue arrows indicate the occurrence of some characteristic diffractions of the tetragonal phase.



**Figure S5:** Powder XRD pattern of the PZT08 reaction product obtained through two-step precipitation process and hydrothermal reaction at 235°C (12 h) and at (a) OH:Pb=3:1, (b) KOH:Pb=1.7:1, (c) KOH:Pb=1.5:1. The main diffraction lines of rhombohedral (R) and tetragonal (T) phase are indexed according to ICDD pdf reference code 01-078-4666 (S.G.: R3c) and ICDD pdf reference code 04-017-5715 (S.G.: P4mm), respectively. The prevailing phase is denoted with larger letter. The blue arrows indicate the occurrence of some characteristic diffractions of the tetragonal phase.