## Supporting information

# Solvothermal Reaction and Piezoelectric Response of Oriented $\mathrm{KNbO}_{3}$ Polycrystal 

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Figure S1. XRD patterns of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/isopropylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h ,
respectively.


Figure S2. XRD patterns of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/propylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S3. XRD patterns of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethyacetate mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S4. SEM images of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/isopropylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S5. SEM images of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/propylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S6. SEM images of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethyacetate mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S7. EDS images of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethylnediamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$ and (c) $12: 15$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively, (d), (e) and (f) are the element maps of (a), (b) and (c).


Figure S8. (a) and (e) TEM images of cuboid KN particles. (b-d) and (f-g) TEM images are the enlarged views of (a) and (e) images, respectively. The KN particles obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethylnediamine mixed solvent with volume ratios of (a) $0: 27$ and (b) $2: 25$ at $230^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S9. (A-a-b and B-a-b) HRSEM images, (A-c and B-c) TEM images, (A-d,h,j and B-d,h,j) HRTEM images, and (A-e-g,i,k and B-e-g,i,k) Fast Fourier Transform
(FFT) patterns of sliced cuboid KN particles obtained via the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethylnediamine mixed solvent with volume ratios of (A) $0: 27$ and (B) $2: 25$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively. The (A-d,h,j and B-d,h,j) HRTEM images are derived from the black dotted bordered rectangle in the (A-c and B-c) TEM images, respectively. The (A-e-g,i,k and B-e-g,i,k) FFT patterns are derived from the red square in the (A-d,h,j and B-d,h,j) HRTEM images, respectively.


Figure S10. (a) TEM images and (b) SAED patterns of the samples obtained by solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethylnediamine (12:15) at $230^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S 11 . AFM topography images of cuboid $\mathrm{K}_{2} \mathrm{Nb}_{2} \mathrm{O}_{6} \cdot \mathrm{nH}_{2} \mathrm{O}$ polycrystals.


Figure S12. Raman spectra of the specimens obtained using the solvothermal treatment of 3 mL of PHN solution in 27 mL of the water/isopropylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S13. Raman spectra of the specimens obtained using the solvothermal treatment of 3 mL of PHN solution in 27 mL of the water/propylamine mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S14. Raman spectra of the specimens obtained using the solvothermal treatments of 3 mL of PHN solution in 27 mL of the water/ethyacetate mixed solvent with volume ratios of (a) $0: 27$, (b) $2: 25$, (c) $12: 15$, (d) $17: 10$ at $230{ }^{\circ} \mathrm{C}$ for 12 h , respectively.


Figure S 15 . TG profiles of $\mathrm{K}_{2} \mathrm{Nb}_{2} \mathrm{O}_{6} \cdot \mathrm{nH}_{2} \mathrm{O}$ specimen.


Figure S 16 . TG profiles of $\mathrm{K}_{4} \mathrm{Nb}_{6} \mathrm{O}_{17} \cdot \mathrm{nH}_{2} \mathrm{O}$ specimen.

In this study, values of physical properties for the mentioned solvents are derived from reference [S1], and summarized in Table S1.

Table S1 Values of physical properties for the mentioned solvents ${ }^{[51]}$

| Molecular formulat | $\mathrm{p} K_{l}$ | $\mathrm{p} K_{2}$ | Viscosity | $\begin{gathered} D^{\mathrm{a}} \\ (\mathrm{C} \cdot \mathrm{~m}) \end{gathered}$ | Surface tension |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  | (mN • $\mathrm{s} \cdot \mathrm{m}-1)$ |  | a/ (dyn/cm) | b/ [dyn/(cm $\left.\left.\cdot{ }^{\circ} \mathrm{C}\right)\right]$ |
| $\mathrm{H}_{2} \mathrm{~N} \leadsto \mathrm{NH}_{2}$ | 6.85 (+2) | $9.92(+1)$ | 1.54 | 1.99 | 44.77 | 0.1398 |
| $\sim \mathrm{NH}_{2}$ | $10.568(+1)$ | - | 0.353 | 1.26 | 24.86 | 0.1243 |
|  | 10.64 (+1) | - | 0.36 | - | 19.91 | 0.09719 |
|  | - | - | 0.455 | 1.81 | 26.29 | 0.1161 |
|  | - | - | 1.078 | 1.69 | 24.05 | 0.0832 |

${ }^{\square} D$ : dipole moment
(S1) LANGE’ S HANDBOOK OF CHEMISTRY. James G. Speight, Ph.D. Library of Congress Catalog Card Number 84-643191, ISSN 0748-4585, 1998.

