## Supporting information

# Massive hydration-driven swelling of layered perovskite niobate crystals in aqueous solutions of organoammonium bases 

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## S1. Powder XRD data of $\mathrm{KCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10}$ before and after acid treatment.



Fig. S1 XRD patterns of (a) $\mathrm{KCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10}$ and (b) $\mathrm{HCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10} \cdot 1.5 \mathrm{H}_{2} \mathrm{O}$ obtained by the acid treatment of $\mathrm{KCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10}$. All the peaks can be indexed in terms of orthorhombic $[a=0.3875(1)$ $\mathrm{nm}, b=0.7709(3) \mathrm{nm}, c=2.9486(8) \mathrm{nm}]$ and tetragonal $[a=0.3855(1) \mathrm{nm}, c=1.6225(2) \mathrm{nm}]$ unit cell dimensions for $\mathrm{KCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10}$ and $\mathrm{HCa}_{2} \mathrm{Nb}_{3} \mathrm{O}_{10} \cdot 1.5 \mathrm{H}_{2} \mathrm{O}$, respectively, which are compatible with previous reports. ${ }^{1,2}$

S2. Polarized optical microscope image containing non-swollen component of the crystals.


Fig. S2 Polarized optical microscope image for the sample in the TBAOH solution at $\mathrm{TBA}^{+} / \mathrm{H}^{+}$ $=0.1$, highlighting the non-swollen component of the crystals.

S3. Powder XRD data of swollen crystals at a relative humidity of $95 \%$.


Fig. S3 XRD data of the swollen samples in the TMAOH (upper panel) and DMAE (lower panel) solutions at indicated $\mathrm{N}^{+} / \mathrm{H}^{+}$molar ratios. The numerals on the peak correspond to the interlayer distances.

## Reference

1. A. J. Jacobson, J. T. Lewandowski and J. W. Johnson, J. Less-Common Met., 1986, 116, 137-146.
2. M. Dion, M. Ganne and M. Tournoux, Mater. Res. Bull., 1981, 16, 1429-1435.
