**Supporting Information**

**Synergically enhanced piezocatalysis performance of eco-friendly** **(K0.52Na0.48)NbO3 through ferroelectric polarization and defects**

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**Fig. S1. (a) Parameters obtained from Rietveld refinement, SEM images of (K0.52Na0.48)Nb1 – *x*(FeCo)*x*O3 ceramics for (b) *x* = 0, (c) *x* = 0.01, (d) *x* = 0.02 and the pulverized powders for (e) *x* = 0, (f) *x* = 0.01, (g) *x* = 0.02.**

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**Fig. S2. XPS spectra of (a-d) Fe 2p and (e-h) Co 2p for the samples with *x* = 0, 0.01, 0.015, and 0.02.**

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**Fig. S3. (a) Polarization–electric–field (*P*–*E*) loops and (b) current–electric–field (*J*–*E*) curves of KNNFC0.02 under an applied electric field of 36 kV and 100 Hz at room temperature.**

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**Fig. S4. Temperature dependence of dielectric permittivity (*ɛ*′) and loss (tan *δ*) at various frequency in the temperature range of 10–800 K for (a, e) KNNFC0, (b, f) KNNFC0.01, (c, g) KNNFC0.015, and (d, h) KNNFC0.02. (i, j) Ac conductivity of KNNFC0.02 at 1 MHz in logarithmic scale as a function of inverse temperature in the temperature range of 10–800 K.**

Fig. S4(a–h) gives the KNNFC*x* piezocatalyst dielectric constant (*ɛ*') and loss (tan*δ*) as a function of temperature over a temperature range of 10–800 K, and a frequency range of 1000 Hz to 1 MHz. It can be seen that the ferroelectric rhombohedral-orthorhombic (R–O), ferroelectric orthorhombic to tetragonal (O–T) and ferroelectric tetragonal to paraelectric cubic (T–C) phase transitions are observed in all the KNNFC*x* systems, occurring at 100–150, 450–500, and 650–700 K, respectively.

With the FeCo concentration increase to *x* = 0.020 (Fig. S4(d) and (h)), the dielectric relaxation III disappears, and a new dielectric relaxation IV occurs in the temperature range of 10–100 K. The ln*σ*–1000/*T* curve (Fig. S4(i–j)) corresponding to the dielectric relaxation regions I, II, and IV (Fig. S4(d) and (h)) is fitted. The activation energy of K/Na ions vacancies and oxygen vacancies (I and II, Fig. S4(i)) increase. Compared to other FeCo doped samples, the *x* = 0.020 sample did not exhibit a very large concentration of defects (K/Na ions vacancies and oxygen vacancies), indicating a potential impurity phase in this sample. Further, the activation energy obtained by fitting the ln*σ*–1000/*T* curve of the temperature region of dielectric relaxation IV is only about 0.003 eV, which is usually generated by the impurity excited state conduction [1]. This conclusion is consistent with the unexpectedly high leakage current in Fig. S3, the abnormally large 2*P*r and *E*c in Fig. 2(e), and the inferior piezocatalytic performance shown in Fig. 3(d–f).

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**Fig. S5. (a) Relative concentration ratios *C*/*C*0 of RhB versus degradation time under different experimental conditions and color change of the RhB dye solution with time under the ultrasonic power of (b) 100 W and (c) 150 W.**

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**Fig. S6. Mott–Schottky plots of (a) KNNFC0.01 and (b) KNNFC0.015 at selective frequencies.**

**References**

[1] T. Inushima, T. Matsushita, S. Ohya, H. Shiomi, Hopping conduction via the excited states of boron in p-type diamond, *Diam. Relat. Mater.*, 9(2000), p. 1066-1070.