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Supplementary Information for

Interpretable and uncertainty-informed machine learning to accelerate the design and discovery of lead-free piezoceramics with large piezoelectric constant

Heng Hu[a], Bin Wang[a], Didi Zhang[a], Kang Yan[a,*], Tao Tan[b,*], Dawei Wu[a,*]

[a] State Key Laboratory of Mechanics and Control of Mechanical Structures, College of Aerospace Engineering, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

[b] Faculty of Applied Sciences, Macao Polytechnic University, Macao, 999078, China

*Corresponding authors

E-mail addresses: yankang@nuaa.edu.cn (K. Yan), taotan@mpu.edu.mo (T. Tan), dwu@nuaa.edu.cn (D. Wu).

Table S1 Initial database.

K (%)	Na (%)	Li (%)	Ba (%)	Ca (%)	$Bi_{0.5}K_{0.5}$ (%)	$Bi_{0.5}Na_{0.5}$ (%)	$Bi_{0.5}Li_{0.5}$ (%)	Nb (%)	Sb (%)	Zr (%)	d_{33} (pC/N)
35.64	53.46	9.90	0.00	0.00	1.00	0.00	0.00	94.05	4.95	1.00	170
34.92	52.38	9.70	0.00	2.00	1.00	0.00	0.00	92.15	4.85	3.00	204
34.56	51.84	9.60	0.00	3.00	1.00	0.00	0.00	91.20	4.80	4.00	230
34.38	51.57	9.55	0.00	3.50	1.00	0.00	0.00	90.73	4.78	4.50	209
34.20	51.30	9.50	0.00	4.00	1.00	0.00	0.00	90.25	4.75	5.00	189
33.84	50.76	9.40	0.00	5.00	1.00	0.00	0.00	89.30	4.70	6.00	174
34.92	52.38	9.70	2.00	0.00	1.00	0.00	0.00	92.15	4.85	3.00	225
34.56	51.84	9.60	3.00	0.00	1.00	0.00	0.00	91.20	4.80	4.00	201
34.38	51.57	9.55	3.50	0.00	1.00	0.00	0.00	90.73	4.78	4.50	168
34.20	51.30	9.50	4.00	0.00	1.00	0.00	0.00	90.25	4.75	5.00	135
45.54	49.50	3.96	0.00	0.00	1.00	0.00	0.00	93.06	5.94	1.00	283
44.62	48.50	3.88	0.00	2.00	1.00	0.00	0.00	91.18	5.82	3.00	372
44.16	48.00	3.84	0.00	3.00	1.00	0.00	0.00	90.24	5.76	4.00	395
43.70	47.50	3.80	0.00	4.00	1.00	0.00	0.00	89.30	5.70	5.00	313
43.24	47.00	3.76	0.00	5.00	1.00	0.00	0.00	88.36	5.64	6.00	130
45.60	49.40	0.00	0.00	1.00	0.00	4.00	0.00	95.00	0.00	5.00	310[1]
45.60	49.40	0.00	0.00	1.00	0.00	4.00	0.00	94.05	0.95	5.00	330
45.60	49.40	0.00	0.00	1.00	0.00	4.00	0.00	93.10	1.90	5.00	380
45.60	49.40	0.00	0.00	1.00	0.00	4.00	0.00	92.15	2.85	5.00	470
45.60	49.40	0.00	0.00	1.00	0.00	4.00	0.00	91.20	3.80	5.00	180
52.00	48.00	0.00	0.00	0.00	0.00	0.00	0.00	95.00	5.00	0.00	80[2]
51.48	47.52	0.00	0.00	1.00	0.00	0.00	0.00	94.05	4.95	1.00	180
50.96	47.04	0.00	0.00	2.00	0.00	0.00	0.00	93.10	4.90	2.00	237
50.44	46.56	0.00	0.00	3.00	0.00	0.00	0.00	92.15	4.85	3.00	162
49.40	45.60	0.00	0.00	5.00	0.00	0.00	0.00	90.25	4.75	5.00	62
47.50	47.50	5.00	0.00	0.00	0.00	0.00	0.00	95.00	5.00	0.00	205[3]
47.26	47.26	4.98	0.00	0.50	0.00	0.00	0.00	94.53	4.98	0.50	270
47.03	47.03	4.95	0.00	1.00	0.00	0.00	0.00	94.05	4.95	1.00	190
46.55	46.55	4.90	0.00	2.00	0.00	0.00	0.00	93.10	4.90	2.00	185
46.08	46.08	4.85	0.00	3.00	0.00	0.00	0.00	92.15	4.85	3.00	120
45.60	45.60	4.80	0.00	4.00	0.00	0.00	0.00	91.20	4.80	4.00	110
45.13	45.13	4.75	0.00	5.00	0.00	0.00	0.00	90.25	4.75	5.00	100
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	96.03	0.97	3.00	87[4]
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	94.09	2.91	3.00	163
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	92.15	4.85	3.00	217
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	91.18	5.82	3.00	267

44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	90.21	6.79	3.00	225
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	89.24	7.76	3.00	220
44.23	47.92	4.85	0.00	1.50	0.00	1.50	0.00	87.30	9.70	3.00	180
48.00	52.00	0.00	0.00	0.00	0.00	0.00	0.00	96.00	4.00	0.00	155[5]
47.52	51.48	0.00	0.00	0.00	0.00	1.00	0.00	95.04	3.96	1.00	200
47.04	50.96	0.00	0.00	0.00	0.00	2.00	0.00	94.08	3.92	2.00	260
46.56	50.44	0.00	0.00	0.00	0.00	3.00	0.00	93.12	3.88	3.00	380
46.32	50.18	0.00	0.00	0.00	0.00	3.50	0.00	92.64	3.86	3.50	410
46.08	49.92	0.00	0.00	0.00	0.00	4.00	0.00	92.16	3.84	4.00	440
45.84	49.66	0.00	0.00	0.00	0.00	4.50	0.00	91.68	3.82	4.50	280
45.60	49.40	0.00	0.00	0.00	0.00	5.00	0.00	91.20	3.80	5.00	90
48.00	48.00	0.00	0.00	0.00	0.00	4.00	0.00	92.16	3.84	4.00	435[6]
48.00	48.00	0.00	0.00	0.00	0.00	3.60	0.40	92.16	3.84	4.00	425
48.00	48.00	0.00	0.00	0.00	0.00	3.20	0.80	92.16	3.84	4.00	410
48.00	48.00	0.00	0.00	0.00	0.00	2.40	1.60	92.16	3.84	4.00	360
48.00	48.00	0.00	0.00	0.00	0.00	1.60	2.40	92.16	3.84	4.00	330
48.00	48.00	0.00	0.00	0.00	0.00	0.80	3.20	92.16	3.84	4.00	325
48.00	48.00	0.00	0.00	0.00	0.00	0.00	4.00	92.16	3.84	4.00	300
47.52	51.48	0.00	0.00	0.00	0.00	1.00	0.00	95.04	3.96	1.00	182[7]
47.04	50.96	0.00	0.00	0.00	0.00	2.00	0.00	94.08	3.92	2.00	220
46.56	50.44	0.00	0.00	0.00	0.00	3.00	0.00	93.12	3.88	3.00	312
46.08	49.92	0.00	0.00	0.00	0.00	4.00	0.00	92.16	3.84	4.00	512
45.60	49.40	0.00	0.00	0.00	0.00	5.00	0.00	91.20	3.80	5.00	295
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	96.00	0.00	4.00	174[8]
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	92.16	3.84	4.00	223
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	91.20	4.80	4.00	320
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	90.24	5.76	4.00	370
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	89.28	6.72	4.00	425
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	88.32	7.68	4.00	320
45.60	45.60	4.80	4.00	0.00	0.00	0.00	0.00	87.36	8.64	4.00	159

The first 15 data are derived from experiment records and the rest are derived from published literature.

Table S2 Defined material features based on the properties of perovskite A-site and B-site elements (atoms or ions).

No.	Feature	Description
1	t	Tolerance factor calculated by Shannon's ionic radii
2	μ	Octahedral factor calculated by Shannon's ionic radii
3	*EN-A*	Electronegativity (absolute scale) difference between A, B-site cations and anions[9]

4	*EN-P*	Electronegativity (Pauling scale) difference between A, B-site cations and anions[10]
5	*EN-MB*	Electronegativity (Matyonov-Batsanov) difference between A, B-site cations and anions[11]
6	*EI*	First energy ionization difference between A, B-site cations and anions[12]
7	*EA*	Electron affinity difference between A, B-site cations and anions[12]
8	*CR*	Pseudopotential core radii difference between A, B-site cations and anions[13]
9	*V*	Atomic volume fraction of the A, B-site elements[14]
10	*RDCE*	Valence electron distance (Schubert) fraction of the A, B-site elements[15]
11	*RDVE*	Core electron distance (Schubert) fraction of the A, B-site elements[15]
12	*PE*	Period fraction of the A, B-site elements
13	*W*	Relative atomic mass fraction of the A, B-site elements[12]
14	*EFF-S*	Nuclear effective charge (Slater) fraction of the A, B-site elements[15]
15	*EFF-C*	Nuclear effective charge (Clementi) fraction of the A, B-site elements[15]
16	*AN*	Atomic number fraction of the A, B-site elements
17	*R*	Shannon's (1976) ionic radii fraction of the A, B-site elements[16]
18	*AO*	Ideal bond distances fraction of the A, B-site elements[17]
19	*D*	Ionic displacement fraction of the A, B-site elements[18]
20	*CVW*	Crystallographic van der Waals radii fraction of the A, B-site elements[19]
21	*EVW*	Equilibrium van der Waals radii fraction of the A, B-site elements[19]
22	*P*	Polarizability fraction of the A, B-site elements[12]
23	*AR*	Atomic radius fraction of the A, B-site elements[20]
24	*GR*	Group fraction of the A, B-site elements
25	*RCOV*	Covalent radii fraction of the A, B-site elements[15]
26	*z/rk*	“Nominal charge/ Shannon’s ionic radii” fraction of the A, B-site elements
27	*Vec/Z*	“Valence electron number / nominal charge” fraction of the A, B-site elements
28	T_{R-O}	Dependence of the rhombohedral to orthorhombic ferroelectric transition temperature on the doping element[21]
29	T_{O-T}	Dependence of the tetragonal to orthorhombic ferroelectric transition temperature on the doping element[21]

30	T_C	Dependence of the cubic to tetragonal ferroelectric transition temperature on the doping element[21]
31	*K/Na*	Ratio of the A-site K to Na cation

Table S3 Effects of dopants on phase transition temperatures.[22]

Substitutes	K^+	Na^+	Li^+	Ba^{2+}	Ca^{2+}	Bi^{3+}	Nb^{5+}	Sb^{5+}	Zr^{4+}
$\boldsymbol{T_{R\_O}}$	0	0	0	0	0	+3	0	+2	+3
$\boldsymbol{T_{O\_T}}$	0	0	-3	-2	-1	-3	0	-2	-3
$\boldsymbol{T_C}$	0	0	+2	-2	-1	-2	0	-3	-2

Formulation of material features

The tolerance factor *t* and octahedral factor μ are key factors to the stability of perovskite. They are defined as

$$t = \left(R_A + R_O\right) / \sqrt{2}\left(R_B + R_O\right) \tag{1}$$

$$\mu = R_B / R_O \tag{2}$$

, where R_A and R_B are the mole averaged ionic radius of the A-site and B-site ions, and R_o is the ionic radius of the oxygen.

As mentioned in the literature,[23] electronegativity, first energy ionization, electron affinity, and pseudopotential core radii are factors mainly used to compare the difference between cations and anions. So, the feature No.3-8 are defined as

$$X_{AB-O} = \left(\left|X_A - X_O\right| + \left|X_B - X_O\right|\right) / 2 \tag{3}$$

$$X_A = f_K X^K + f_{Na} X^{Na} + f_{Li} X^{Li} + f_{Ba} X^{Ba} + f_{Ca} X^{Ca} + f_{Bi} X^{Bi} \tag{3.1}$$

$$X_B = f_{Nb}X^{Nb} + f_{Sb}X^{Sb} + f_{Zr}X^{Zr} \tag{3.2}$$

$$X_O = f_O X^O \tag{3.3}$$

, where f is the mole fraction, and X corresponds to the properties of each element.

Features No.9-27 were defined by the ratios of those properties for A-site and B-site, which are calculated using the following equation:

$$X_{A/B} = X_A / X_B \tag{4}$$

, in which X_A and X_B are previously defined in Eq. (3.1) and Eq. (3.2), respectively.

Chemical modification can effectively improve the piezoelectric performance of KNN materials by shifting the rhombohedral–orthorhombic (R–O), orthorhombic–tetragonal (O–T), and rhombohedral–tetragonal (R–T) phase transition temperatures near room temperature. Previous studies have reported the contributions of the individual elements to phase transition temperature. For example, Ba^{2+}, Ca^{2+}, Bi^{3+}, Sb^{5+}, and Zr^{4+} decreased O-T phase transition temperature. In addition, Sb^{5+} and Zr^{4+} could also increase the R-O phase transition temperature.[21,22] In this work, the effect of a doped element is assigned positive if it increases the transition temperature and negative if it reduces the transition temperature. The absolute value for each element depends on how much it affects the transition temperature. Value 0 is assigned to K, Na, Nb, and to the elements that have almost no effect on the transition temperature. The detailed information is given in Table S3. The features No.28-30 were calculated by

$$X_{A+B} = X_A + X_B \tag{5}$$

The feature of the potassium-sodium ratio is defined as

$$K / Na = f_K / f_{Na} \tag{6}$$

Table S4 Groups of features with strong correlations. The first feature in each group is retained while the rest are removed.

Group number	Features with strong correlation ($r > 0.95$)	Retained feature
1	*t*, *R*, *AO*, *CVW*, *RCOV*, *EVW*	*t*
2	*CR*, *V*, *RDVE*	*CR*
3	*W*, *EFFC*, *AN*	*W*
4	*D*, *Vec/Z*	*D*
5	*P*, *AR*	*P*

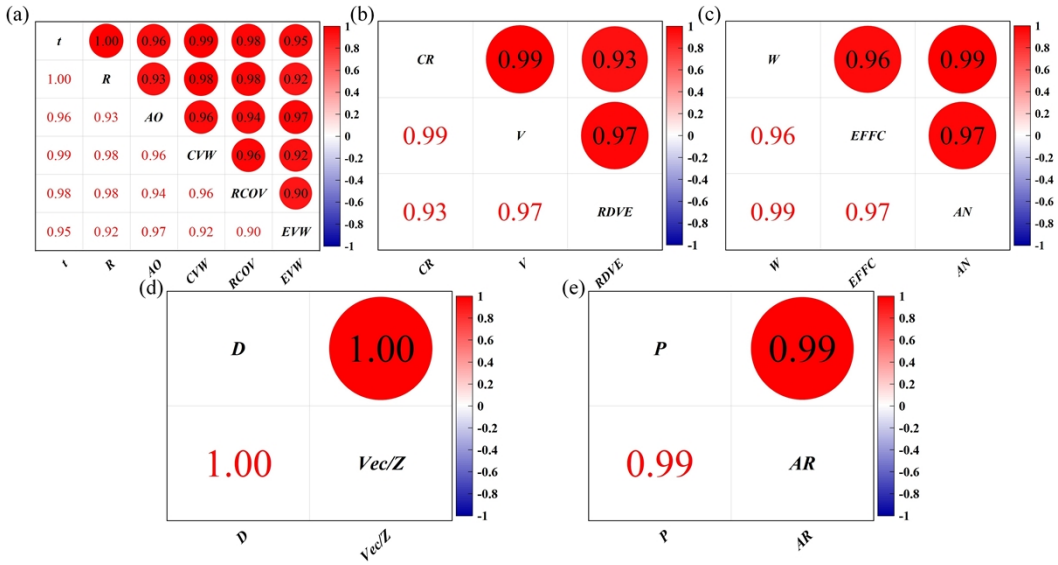

Figure S1 Heatmaps describing the Pearson correlation of features with strong correlations ($r > 0.95$). (a)-(b) represent the features in group 1-5, respectively.

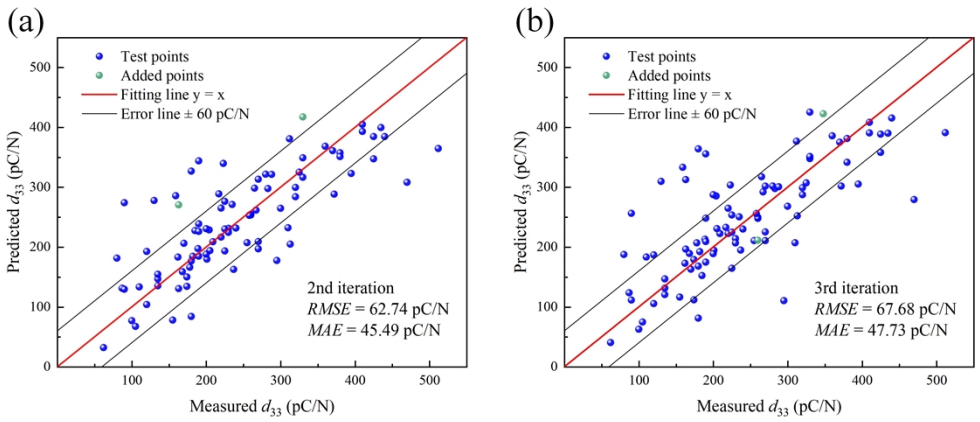

Figure S2 Global performance of ANN models for the second and third experimental iterations.

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