Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

SUPPORTING INFORMATION

Prediction of Dielectric Constants of ABO_3-Type Perovskites Using Machine Learning and First-Principle Calculations

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Model (ε_E)	R^2	RMSE	MAE
Light Gradient Boosting Machine	0.84	0.12	0.05
Extreme Gradient Boosting	0.81	0.13	0.06
Random Forest	0.81	0.13	0.06
Gradient Boosting Regressor	0.79	0.14	0.07
CGCNN	0.74	0.14	0.07
Ridge Regression	0.73	0.16	0.08
Bayesian Ridge	0.72	0.16	0.08
Orthogonal Matching Pursuit	0.70	0.16	0.09
Support Vector Machine	0.59	0.19	0.11
K Neighbors Regressor	0.57	0.20	0.11
Decision Tree	0.56	0.20	0.09
Huber Regressor	0.54	0.21	0.13
Elastic Net	0.48	0.22	0.13
AdaBoost Regressor	0.48	0.22	0.17
Lasso Regression	0.45	0.22	0.14
Passive Aggressive Regressor	0.09	0.28	0.20
Lasso Least Angle Regression	0.00	0.30	0.22
Linear Regression	-0.13	0.26	0.09

Model (ε_i)	R^2	RMSE	MAE
Light Gradient Boosting Machine	0.67	0.26	0.16
Random Forest	0.62	0.28	0.18
Extreme Gradient Boosting	0.62	0.28	0.18
CGCNN	0.59	0.29	0.19
Gradient Boosting Regressor	0.58	0.30	0.19
Ridge Regression	0.49	0.33	0.22
Bayesian Ridge	0.47	0.33	0.22
Linear Regression	0.46	0.34	0.22
Orthogonal Matching Pursuit	0.44	0.34	0.24
K Neighbors Regressor	0.32	0.38	0.26
Support Vector Machine	0.30	0.38	0.26
Huber Regressor	0.29	0.39	0.27
Elastic Net	0.25	0.40	0.27
Lasso Regression	0.22	0.41	0.28
AdaBoost Regressor	0.19	0.41	0.32
Decision Tree	0.18	0.41	0.26
Lasso Least Angle Regression	0.00	0.46	0.32
Passive Aggressive Regressor	-1.87	0.73	0.58

Table S1. R^2, RMSE, and mean absolute error scores for the 18 predictive ML models for the electronic (left, ε_E) and ionic (right, ε_i) contributions.

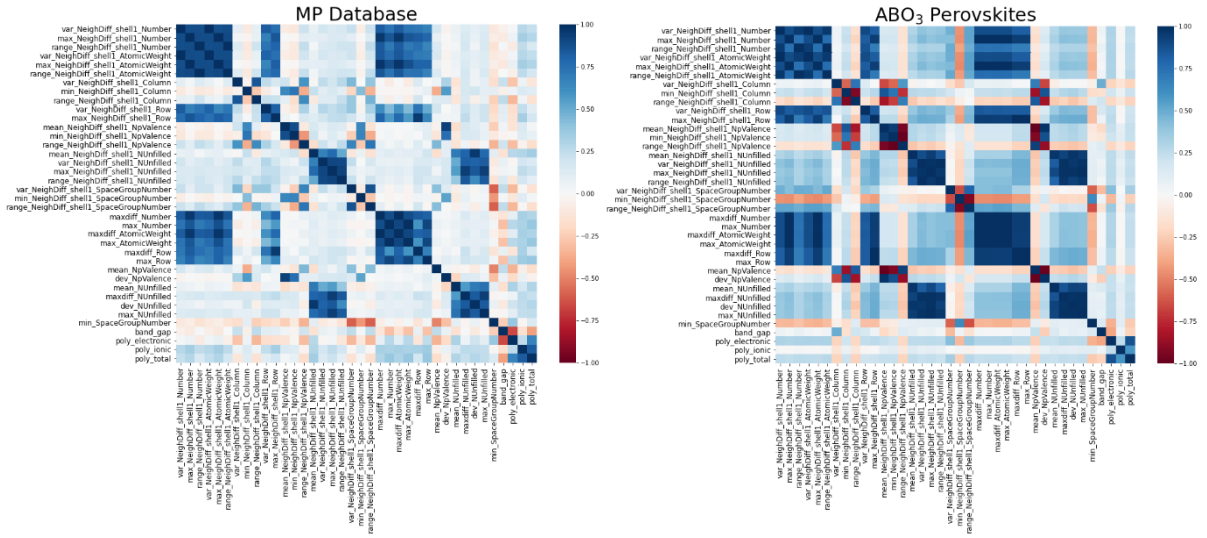

Figure S1. Correlation matrix of the dielectric constant (ε_P) with the (a) MP database and (b) ABO_3 perovskites.

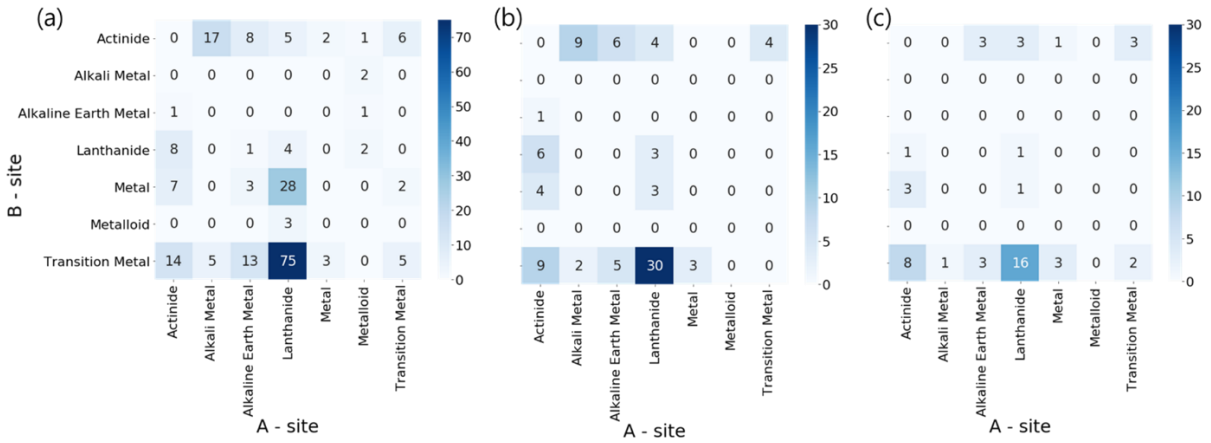

Figure S2. (a) A- and B-site element groups of the 216 calculated ABO_3 perovskites, (b) 89 perovskites with values over the median of the dielectric constants, and (c) 49 structures with values over the median values for both electronic and ionic contributions.

Materials group	Properties	Algorithms	Reference
ABO_3 perovskites	Thermodynamic stability	ET and KRR	[1]
	Stable, metastable	GBDT	[2]
	Formability, cubic structure stability	RF and GBR	[3]
	Formation energy, bandgap	GBR	[4]
	Curie temperature	SVM	[5]
	Maximum magnetic entropy change	GPR	[6]
	Ionic conductivity	SVM	[7]
	Specific surface area (SSA)	SVM	[8]
ABX_3 perovskites	Stability	GBR and CNN	[9]
	Bandgap	ACE	[10]
	Stability	ERT	[11]
	Formability and stability	SVM	[12]
	Formability and interfacial properties	SVM	[13]
	Phase stability	XGBoost	[14]
	Bandgap	RF	[15]
	Dielectric breakdown strength	KRR, RF and LASSO	[16]
	Stability, bandgap and spontaneous polarization	GBC and GBR	[17]

Table S2. Previous studies for predicting materials properties of ABO_3 and ABX_3 perovskites. [18]

Materials group	Total dielectric tensor	Electronic contribution	Ionic contribution	Reference
Ternary oxides	○	○	○	[19]
Inorganic compounds	○	○	○	[20]
Binary non-oxides, ternary fluorides	○	○	○	[21]
Inorganic materials	○	-	-	[22]
Metal oxides	○	○	○	[23]

Table S3. Previous database for dielectric constants using DFPT calculations

REFERENCES

[1] W. Li, R. Jacobs, and D. Morgan, "Predicting the thermodynamic stability of perovskite oxides using machine learning models," *Comput. Mater. Sci.*, vol. 150, no. January, pp. 454–463, 2018, doi: 10.1016/j.commatsci.2018.04.033.

[2] H. Liu *et al.*, "Screening stable and metastable ABO3 perovskites using machine learning and the materials project," *Comput. Mater. Sci.*, vol. 177, no. January, p. 109614, 2020, doi: 10.1016/j.commatsci.2020.109614.

[3] P. V Balachandran, A. A. Emery, J. E. Gubernatis, T. Lookman, C. Wolverton, and A. Zunger, "Predictions of new ABO3 perovskite compounds by combining machine learning and density functional theory," *Phys. Rev. Mater.*, vol. 2, no. 4, p. 43802, Apr. 2018, doi: 10.1103/PhysRevMaterials.2.043802.

[4] L. Chen, "A progressive learning method for predicting the band gap of ABO 3 perovskites using an instrumental," pp. 3127–3136, 2020, doi: 10.1039/c9tc06632b.

[5] X. Zhai, M. Chen, and W. Lu, "Accelerated search for perovskite materials with higher

Curie temperature based on the machine learning methods," *Comput. Mater. Sci.*, vol. 151, no. April, pp. 41–48, 2018, doi: 10.1016/j.commatsci.2018.04.031.

[6] Y. Zhang and X. Xu, "Machine learning the magnetocaloric effect in manganites from lattice parameters," *Appl. Phys. A Mater. Sci. Process.*, vol. 126, no. 5, pp. 1–12, 2020, doi: 10.1007/s00339-020-03503-8.

[7] L. Xu, L. Wencong, P. Chunrong, S. Qiang, and G. Jin, "Two semi-empirical approaches for the prediction of oxide ionic conductivities in ABO3 perovskites," *Comput. Mater. Sci.*, vol. 46, no. 4, pp. 860–868, 2009, doi: 10.1016/j.commatsci.2009.04.047.

[8] L. Shi, D. Chang, X. Ji, and W. Lu, "Using Data Mining to Search for Perovskite Materials with Higher Specific Surface Area," *J. Chem. Inf. Model.*, vol. 58, no. 12, pp. 2420–2427, 2018, doi: 10.1021/acs.jcim.8b00436.

[9] X. Li *et al.*, "Computational screening of new perovskite materials using transfer learning and deep learning," *Appl. Sci.*, vol. 9, no. 24, 2019, doi: 10.3390/app9245510.

[10] V. Gladkikh, D. Y. Kim, A. Hajibabaei, A. Jana, C. W. Myung, and K. S. Kim, "Machine Learning for Predicting the Band Gaps of ABX3 Perovskites from Elemental Properties," *J. Phys. Chem. C*, vol. 124, no. 16, pp. 8905–8918, 2020, doi: 10.1021/acs.jpcc.9b11768.

[11] J. Schmidt, J. Shi, P. Borlido, L. Chen, S. Botti, and M. A. L. Marques, "Predicting the Thermodynamic Stability of Solids Combining Density Functional Theory and Machine Learning," *Chem. Mater.*, vol. 29, no. 12, pp. 5090–5103, Jun. 2017, doi: 10.1021/acs.chemmater.7b00156.

[12] G. Pilania, P. V. Balachandran, C. Kim, and T. Lookman, "Finding new perovskite halides via machine learning," *Front. Mater.*, vol. 3, no. April, pp. 1–7, 2016, doi: 10.3389/fmats.2016.00019.

[13] D. Jain, S. Chaube, P. Khullar, S. Goverapet Srinivasan, and B. Rai, "Bulk and surface DFT investigations of inorganic halide perovskites screened using machine learning and materials property databases," *Phys. Chem. Chem. Phys.*, vol. 21, no. 35, pp. 19423–19436, 2019, doi: 10.1039/c9cp03240a.

[14] H. Park *et al.*, "Learn-and-Match Molecular Cations for Perovskites," *J. Phys. Chem. A*, vol. 123, no. 33, pp. 7323–7334, 2019, doi: 10.1021/acs.jpca.9b06208.

[15] K. Takahashi, L. Takahashi, I. Miyazato, and Y. Tanaka, "Searching for Hidden Perovskite Materials for Photovoltaic Systems by Combining Data Science and First Principle Calculations," *ACS Photonics*, vol. 5, no. 3, pp. 771–775, Mar. 2018, doi: 10.1021/acsphotonics.7b01479.

[16] C. Kim, G. Pilania, and R. Ramprasad, "Machine Learning Assisted Predictions of Intrinsic Dielectric Breakdown Strength of ABX3 Perovskites," *J. Phys. Chem. C*, vol. 120, no. 27, pp. 14575–14580, Jul. 2016, doi: 10.1021/acs.jpcc.6b05068.

[17] S. Lu, Q. Zhou, L. Ma, Y. Guo, and J. Wang, "Rapid Discovery of Ferroelectric Photovoltaic Perovskites and Material Descriptors via Machine Learning," *Small Methods*, vol. 3, no. 11, pp. 1–10, 2019, doi: 10.1002/smtd.201900360.

[18] Q. Tao, P. Xu, M. Li, and W. Lu, "Machine learning for perovskite materials design and discovery," *npj Comput. Mater.*, vol. 7, no. 1, pp. 1–18, 2021, doi: 10.1038/s41524-021-00495-8.

[19] I. Petousis *et al.*, "Data Descriptor: High-throughput screening of inorganic compounds for the discovery of novel dielectric and optical materials," *Sci. Data*, vol. 4, pp. 1–12, 2017, doi: 10.1038/sdata.2016.134.

[20] I. Petousis *et al.*, "Benchmarking density functional perturbation theory to enable high-throughput screening of materials for dielectric constant and refractive index," *Phys. Rev. B*, vol. 93, no. 11, pp. 1–8, 2016, doi: 10.1103/PhysRevB.93.115151.

[21] M. Lee, Y. Youn, K. Yim, and S. Han, "High-throughput ab initio calculations on dielectric constant and band gap of non-oxide dielectrics," *Sci. Rep.*, vol. 8, no. 1, pp. 1–8, 2018, doi: 10.1038/s41598-018-33095-6.

[22] K. Morita, D. W. Davies, K. T. Butler, and A. Walsh, "Modeling the dielectric constants of crystals using machine learning," *J. Chem. Phys.*, vol. 153, no. 2, 2020, doi: 10.1063/5.0013136.

[23] A. Takahashi, Y. Kumagai, J. Miyamoto, Y. Mochizuki, and F. Oba, “Machine learning models for predicting the dielectric constants of oxides based on high-throughput first-principles calculations,” *Phys. Rev. Mater.*, vol. 4, no. 10, 2020, doi: 10.1103/PhysRevMaterials.4.103801.