Supporting Information for 'Electrochemical Stability of biodegradable Zn-Cu alloys through machine-learning accelerated high-throughput discovery'

Kun Luo,[ab] Zhaorui Liu,[ab] Rui Yu, [ab] Tengfei Xu, [ab] Dominik Legut,[cd] Xing. Yin [*e] and Ruifeng Zhang[*ab]

[a]*School of Materials Science and Engineering, Beihang University, Beijing 100191, P. R. China*

[b]*Center for Integrated Computational Engineering (International Research Institute for Multidisciplinary Science) and Key Laboratory of High-Temperature Structural Materials & Coatings Technology (Ministry of Industry and Information Technology), Beihang University, Beijing 100191, P. R. China*

[c]*IT4Innovations, VSB-Technical University of Ostrava, 17. listopadu 2172/15, CZ-70800 Ostrava, Czech Republic*

[d]*Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, 121 16 Prague 2, Czech Republic*

[e]*National Key Laboratory of Nuclear Reactor Technology, Nuclear Power Institute of China, Chengdu 610041, China*

**Corresponding author. Tel: 86-10-8231-5007 E-mail: zrf@buaa.edu.cn (Ruifeng Zhang) and yinxingwt@163.com (Xing Yin)*

Table S1. Calculated lattice constants, formation energies, and volume per atom of partial results of the high-throughput of Zn-Cu alloys from the Inorganic Crystal Structure Database (ICSD) prototype library[1].

	Lattice constants/ Å			Atom number	Formation energy/(eV/atom)	Volume per atom/Å^3	Space group
	a	b	c				
Cu_3Zn	3.685	3.685	14.727	16	-0.0693	12.497	I4/mmm(#139)
	5.218	5.218	12.759	24	-0.0678	12.534	$P6_3$/mmc(#194)
	5.204	5.204	21.293	40	-0.0677	12.487	$P6_3$/mmc(#194)
	5.180	5.187	14.892	32	-0.0677	12.505	Cmca(#64)
	3.699	3.699	25.587	28	-0.0676	12.506	P4/mmm(#123)
	7.388	7.361	7.373	32	-0.0674	12.530	Fmmm(#69)
	3.685	3.685	3.685	4	-0.0673	12.512	$Pm\bar{3}m$(#221)
	5.211	5.211	5.211	8	-0.0670	12.514	$P6_3$/mmc(#194)
	5.210	5.210	4.251	8	-0.0670	12.491	$P6_3$/m(#176)
	3.623	7.566	8.536	16	-0.0664	13.234	$P2_1$/c(#14)
CuZn	5.931	5.931	5.931	16	-0.0910	13.038	$Pm\bar{3}m$(#221)
	5.933	5.933	5.933	16	-0.0909	13.055	$I2_13$(#199)
	5.903	6.052	2.928	8	-0.0906	13.076	Cmmm(#65)
	5.935	5.935	5.935	16	-0.0905	13.064	$I2_13$(#199)
	5.479	8.600	4.440	16	-0.0903	13.075	Cm(#8)
	4.136	4.136	3.052	4	-0.0897	13.052	P4/nmm(#129)
	4.222	5.149	5.146	8	-0.0896	13.076	C2/m(#12)
	5.934	5.934	5.939	16	-0.0896	13.069	I4/mcm(#140)
	5.578	8.751	4.285	16	-0.0894	13.071	Cmca(#64)
	3.868	3.868	3.498	4	-0.0893	13.085	P4/nmm(#129)
Cu_5Zn_8	8.883	8.883	8.883	52	-0.1033	13.482	$I\bar{4}3m$(#217)
	17.101	2.620	11.727	26	0.0105	15.123	C2/m(#12)
	5.364	5.364	22.309	39	0.0779	14.256	$R\bar{3}m$(#166)
$CuZn_3$	5.476	5.476	4.269	8	-0.0713	13.856	$P6_3$/mmc(#194)
	5.448	5.448	19.414	36	-0.0695	13.863	$R\bar{3}m$(#166)
	4.749	5.443	8.552	16	-0.0690	13.818	$P2_1$/c(#14)
	5.447	4.274	4.736	8	-0.0684	13.832	Pmmn(#59)
	4.321	5.394	9.489	16	-0.0673	13.822	Pnma(#62)
	6.959	4.740	6.910	16	-0.0661	13.873	$P2_1$/c(#14)
	6.906	4.741	6.956	16	-0.0660	13.857	$P2_1$/c(#14)
	4.250	2.777	4.706	4	-0.0587	13.885	Pmm2(#25)
	5.457	5.457	21.459	40	-0.0549	13.835	$P6_3$/mmc(#194)

	7.207	7.207	7.630	24	-0.0472	14.3	$P\bar{3}c1$(#165)
$CuZn_4$	4.782	4.285	13.679	20	-0.0564	14.016	Pnma(#62)
	11.826	4.773	5.127	20	-0.0509	14.056	C2/c(#15)
	4.310	5.835	11.624	20	-0.0438	14.62	$P2_12_12_1$(#19)
	5.191	16.643	5.068	30	-0.0348	14.324	Cm(#8)
	7.199	5.403	5.581	10	-0.0208	14.764	C2/m(#12)
	4.389	12.379	11.552	40	-0.0199	15.333	Pbca(#61)
	6.080	6.080	3.804	10	-0.0173	14.063	I4/m(#87)
	9.893	4.058	8.049	20	-0.0079	14.874	$P2_1/c$(#14)
	4.148	13.178	5.237	20	0.0049	14.316	Cmcm(#63)
	6.463	6.463	6.768	20	0.0096	14.134	$P4_12_12$(#92)
$CuZn_5$	4.784	4.784	8.581	12	-0.0390	14.174	$P6_322$(#182)
	12.600	80832	6.606	48	-0.0361	14.732	$P2_1/c$(#14)
	2.879	9.894	12.399	24	-0.0150	14.715	Cmcm(#63)
	5.108	8.450	8.368	24	-0.0094	14.460	C2/m(#12)
	12.060	4.292	7.290	24	-0.0090	15.127	C2/m(#12)
	6.773	10.661	11.133	48	-0.0079	15.151	$P2_1/c$(#14)
	6.150	6.150	9.795	24	-0.0002	15.435	I4/mcm(#140)
	6.821	15.182	6.801	48	0.0236	14.674	Pnma(#62)
	7.661	9.539	4.896	24	0.0303	14.908	Cmcm(#63)
	12.787	12.787	4.589	48	0.0397	15.631	$I\bar{4}2d$(#122)

Table S2. The setup of hyper-parameters used in sklearn for XGBoost. The explanation of each hyper-parameter can be found in Ref [2].

Parameter	Value
num_round (n_estimators)	300
seed	0
eta (learning_rate)	0.1
colsampel_bytree	0.7
subsample	0.90
max_depth	5
min_child_weight	1
lambda	1
gamma	0.05
objective	reg: squarederror
verbostity	1

Fig. S1 Comparison of DFT-calculated formation energy and predicted formation energy with models created using the (a) SOAP, (b) MBTR, (c) Ewald, (d) Sine matrix, (e) ACSF. (f) Average performance of models constructed using five descriptors after five-fold cross-validation.

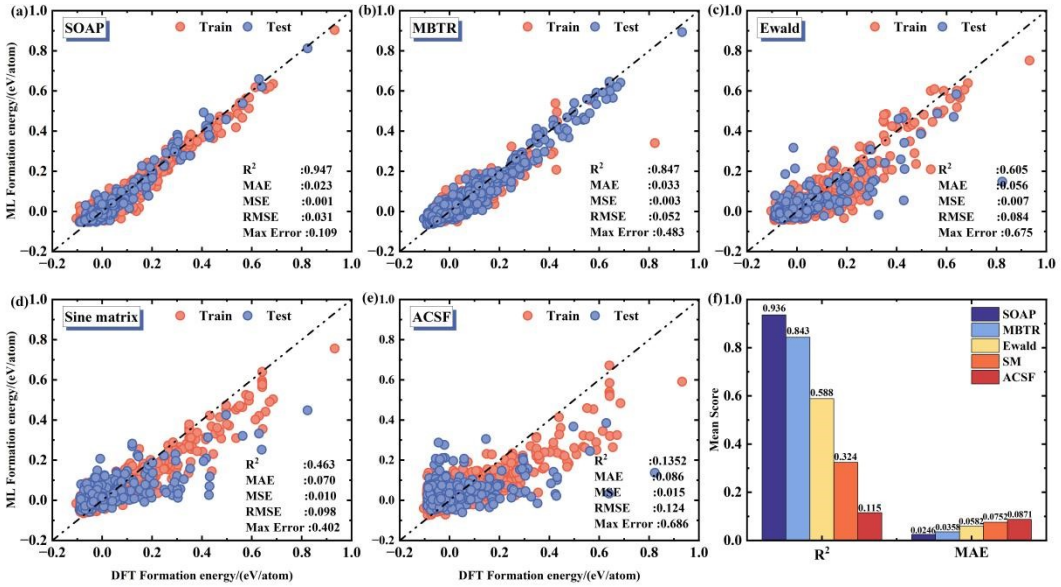


Fig. S2 Phonon dispersion curves result for (a) Cu_3Zn, (b) $CuZn_3$.

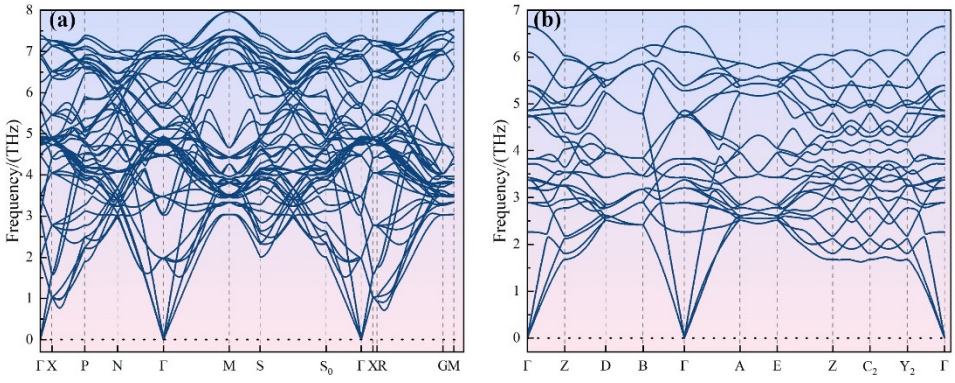


Fig. S3 AIMD simulation curves result and the structural changes for (a) Cu_3Zn, (b) $CuZn_3$.

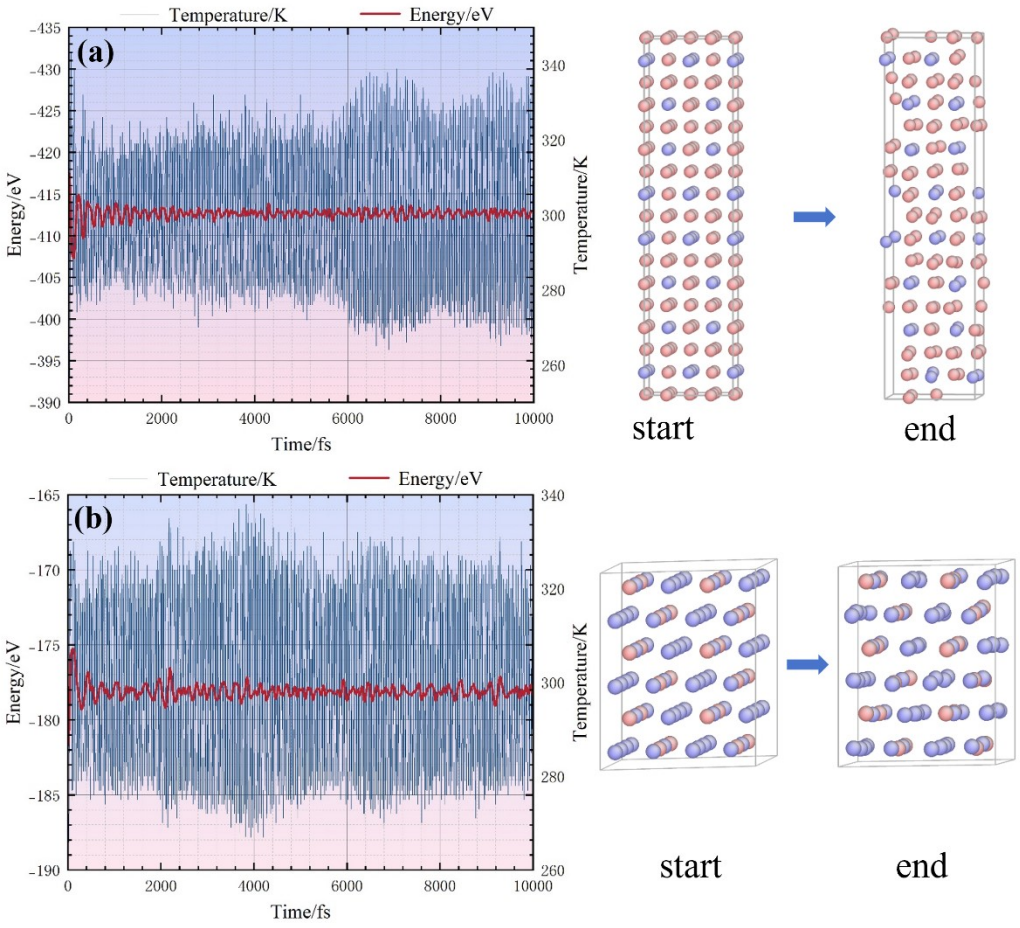


Fig. S4 R^2 variation curve and histograms of MAE, MSE performance of the model on the training and validation sets under five-fold-cross-validation. The model we used is the fifth fold.

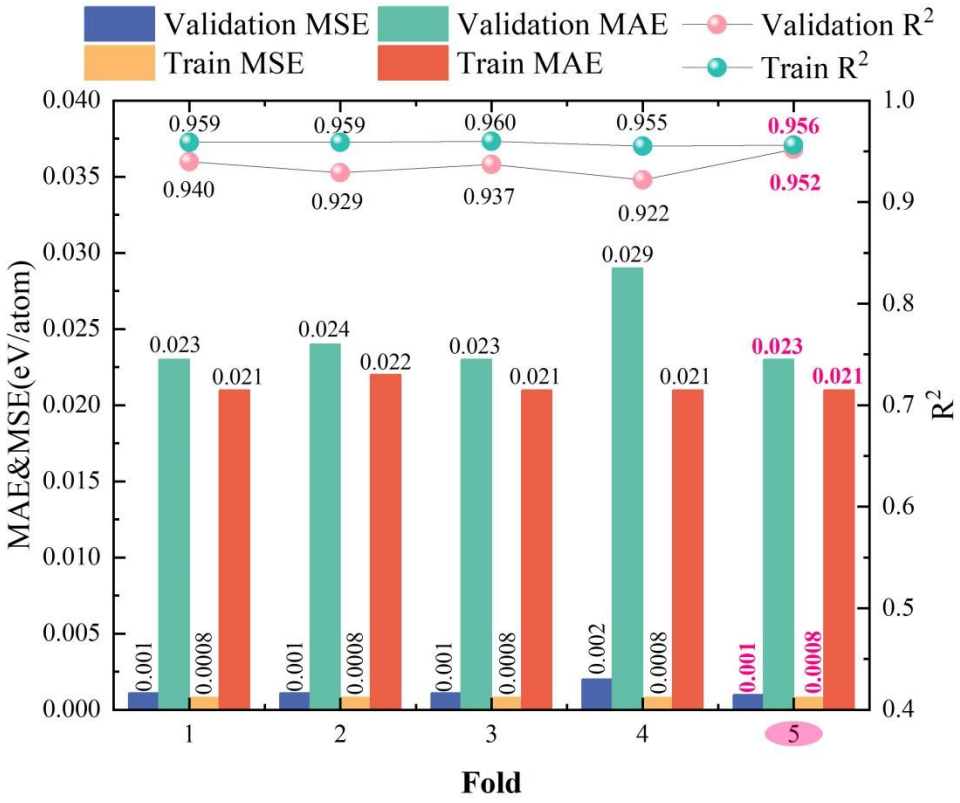


Fig. S5 The structural changes for $CuZn_5$ from the start (a) to the end (b) during performing AIMD simulation

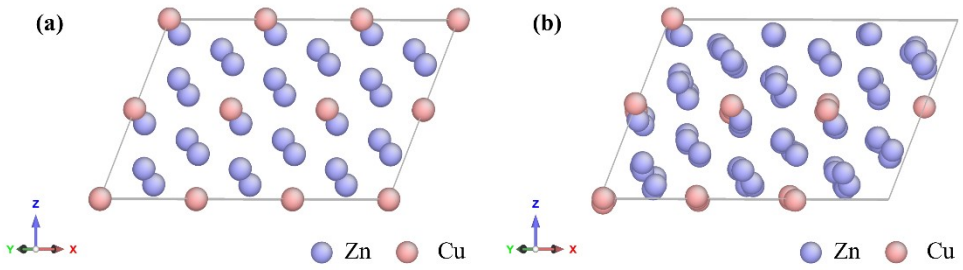


Fig. S6 Pourbaix diagrams in a pure aqueous solution at 298 K， including (a) Cu: Zn = 3:1(b) Cu: Zn = 1:1, (c) Cu: Zn = 5:8, (d) Cu: Zn=1:3，Pourbaix diagrams with chlorine in the solution at 298 K， including (e) Cu: Zn = 3:1(f) Cu: Zn = 1:1, (g) Cu: Zn = 5:8, (h) Cu: Zn=1:3.

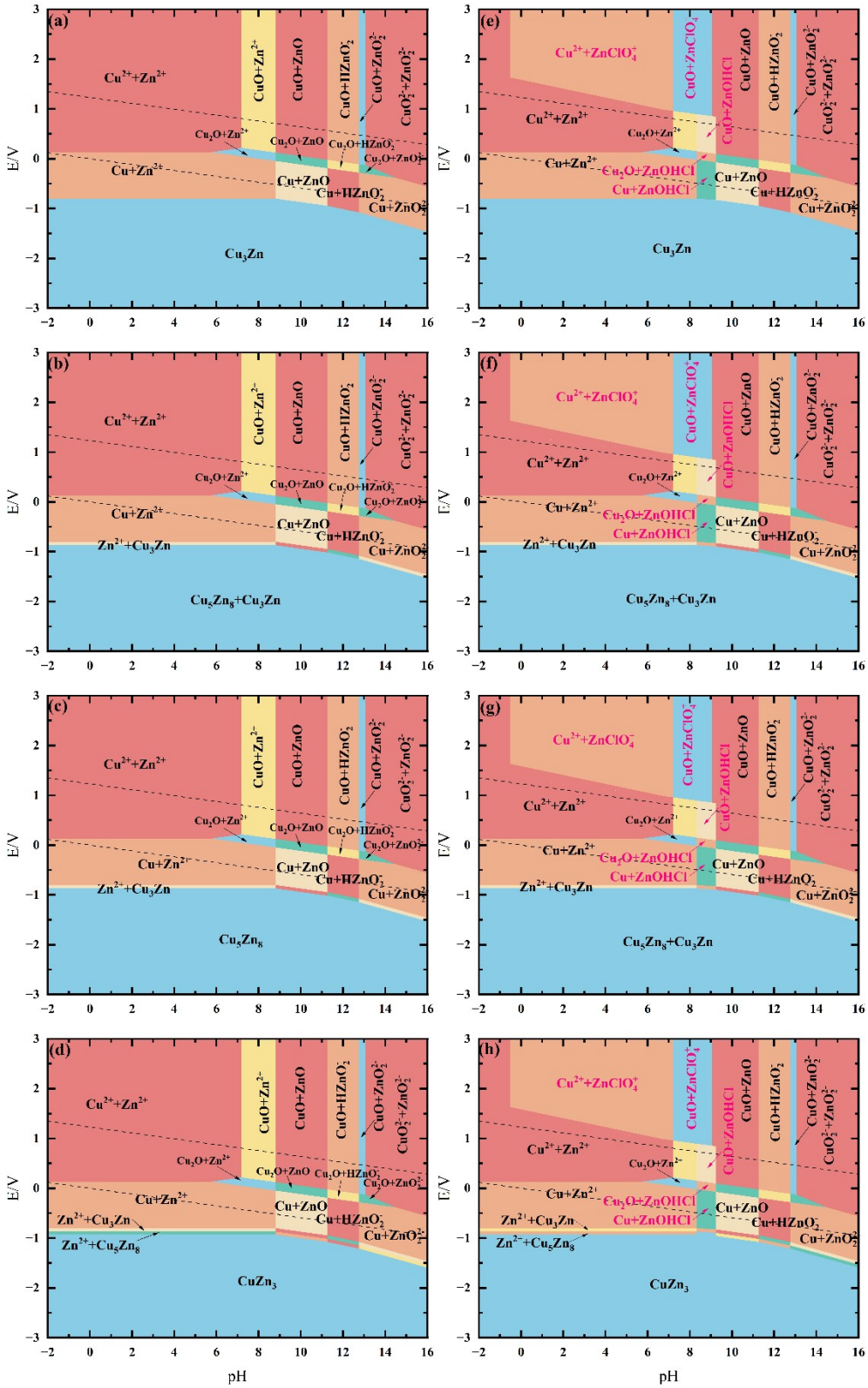


Fig. S7 Pourbaix diagrams of Cu: Zn=1:10(Zn-rich alloys) in a pure aqueous solution at 298 K takes into account both cases (a) without $CuZn_5$ and (b)with $CuZn_5$.

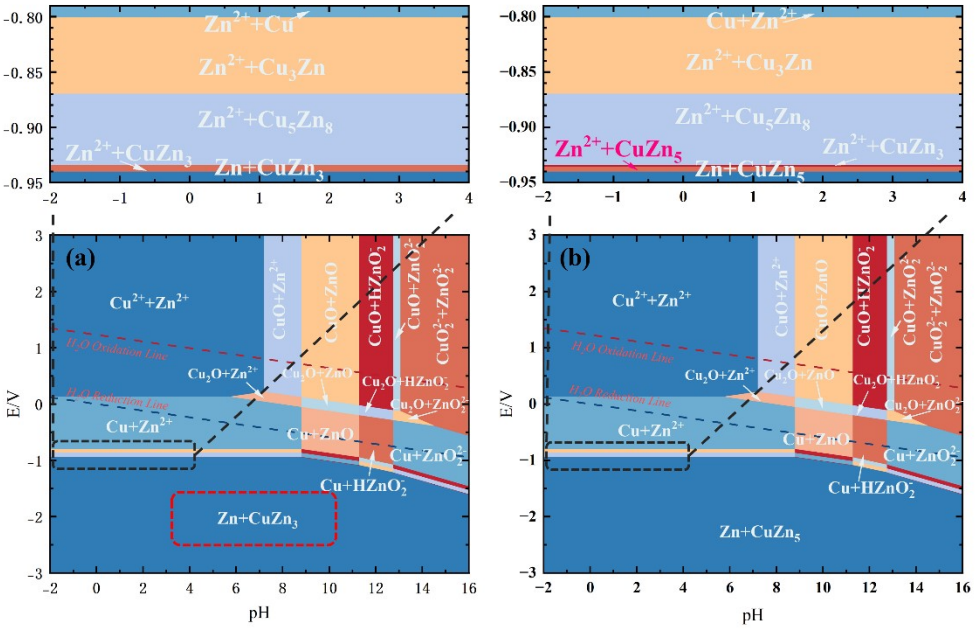


Fig. S8 Pourbaix diagrams of chlorine.

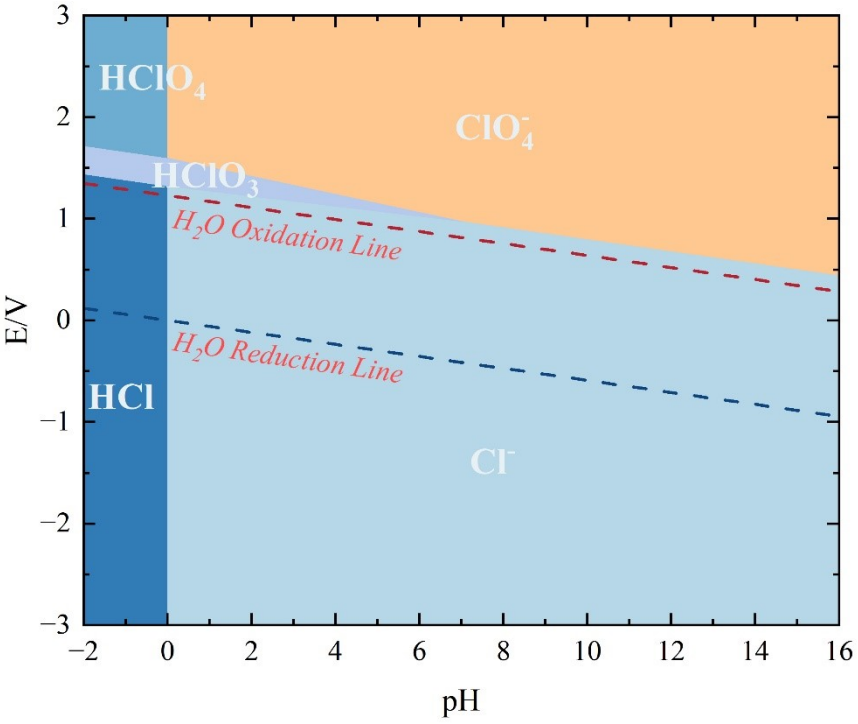
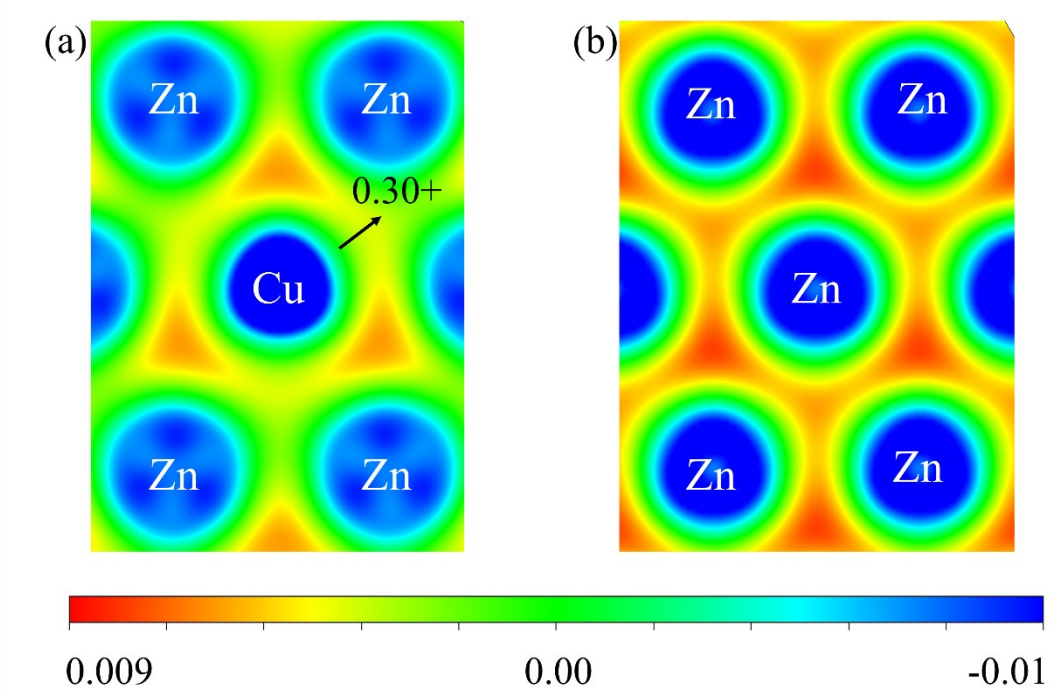


Fig. S9 Valence charge density differences (VCDD) and electron charges (marked with one-way arrows) of $CuZn_5$(a) 2D (1 1 1) and pure Zn (b) 2D (0 0 1), respectively.



References

[1] A. Belsky, M. Hellenbrandt, V.L. Karen, P. Luksch, New developments in the Inorganic Crystal Structure Database (ICSD): accessibility in support of materials research and design, Acta Crystallographica Section B: Structural Science, 58 (2002) 364-369.
[2] T. Chen, C. Guestrin, Xgboost: A scalable tree boosting system, in: Proceedings of the 22nd acm sigkdd international conference on knowledge discovery and data mining, 2016, pp. 785-794.