First-Principles Calculations of the Structural, Electronic, Optical, and Mechanical Properties of 21 Pyrophosphate Crystals

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1. Optical properties

We use OLCAO method to calculate the optical properties of crystalline materials based on the theory of inter-band optical absorptions within the random phase approximation. According to this approach, the optical conductivity is given by:

$$\sigma_1(\hbar w) = \frac{2\Pi e\hbar^2}{3m^2\omega\Omega}\sum_{n,l}|\langle n|\vec{p}|l\rangle^2 f_l[1-f_n]\delta(E_n - E_l - \hbar\omega) \quad (1)$$

f_l is the Fermi-Dirac function of the occupied band state ι and δ function makes sure that the energy is conserved in the transition process between occupied state and unoccupied state with energy Eι and En respectively. Ω is the volume of the unit cell.

Assuming that there is no intra-band transitions for the materials with an energy gap in their band structure, so that the optical properties can be described by the complex dielectric function which is related to the $\sigma_1(\hbar w)$ by:

$$\varepsilon_2(w) = 4\pi\frac{\sigma_1(w)}{w} \quad (2)$$

By transforming the summation over the unit cell in the equation (3) to an integral over the BZ in **k** space, $\varepsilon_2(w)$ can be given by:

$$\varepsilon_2(\hbar\omega) = \frac{e^2}{\pi m\omega^2}\int dk^3 \sum_{n,l}\left|\langle\Psi_n(\vec{k},\vec{r})\right|-i\hbar\vec{\nabla}\left|\Psi_l(\vec{k},\vec{r})\rangle\right|^2 f_l(\vec{k})[1 - f_n(\vec{k})]\delta(E_n(\vec{k}) - E_l(\vec{k}) - \hbar\omega) \quad (3)$$

The real part of the dielectric function can be derived from ε2 by using Kramers-Kronig relation:

$$\varepsilon_1(\hbar\omega) = 1 + \frac{2}{\pi}P\int_0^\infty \frac{s\varepsilon_2(\hbar\omega)}{s^2-\omega^2}\,ds \quad (4)$$

In OLCAO method, the momentum matrices in the Eqs (2) and (4) for the optical transitions can be easily calculated as sums of two –center integrals between GTOs[1].

2. Mechanical properties

Ab initio calculation method within DFT for the mechanical properties is very powerful method these days. In this section, and to calculate the mechanical properties, we followed the method of the Hongzhi Yao et al. work[2]. In this work, a scheme has been applied to calculate the elastic constants of crystals. Starting with the simple Hook's famous law that can relate the stress components σ_i with the strain components ε_j by relation:

$$\sigma_i = \sum_{j=1}^{6} C_{ij}\, \varepsilon_j \tag{5}$$

C_{ij} is the elastic constants of the crystals. From knowing the elastic constants, we calculate other mechanical properties like compliance tensor S_{ij}, Young's modulus (E), Bulk modulus (K), Shear modulus (G), and Poisson's ratio (η). Here, the Voigt – Reuss –Hill (VRH)[3,4] approximation has been used to derive the above mechanical properties. According to this approximation, the upper and lower bounds for the structural parameters, like bulk and shear modulus are respectively given by:

$$K_V = \frac{(C_{11}+C_{22}+C_{33})}{9} + \frac{2(C_{12}+C_{13}+C_{23})}{9} \tag{6}$$

$$K_R = \frac{1}{(S_{11}+S_{22}+S_{33})+2(S_{12}+S_{13}+S_{23})} \tag{7}$$

$$G_V = \frac{(C_{11}+C_{22}+C_{33}-C_{12}-C_{13}-C_{23})}{15} + \frac{(C_{44}+C_{55}+C_{66})}{5} \tag{8}$$

$$G_R = \frac{15}{4(S_{11}+S_{22}+S_{33})-4(S_{12}+S_{13}+S_{23})+3(S_{44}+S_{55}+S_{66})} \tag{9}$$

So, the average values of the mechanical properties are given by:

$$K = \frac{(K_V+K_R)}{2} \tag{10}$$

$$G = \frac{(G_V+G_R)}{2} \tag{11}$$

$$E = \frac{9KG}{(3K+G)} \tag{12}$$

$$\eta = \frac{(3K-2G)}{2(3K+G)} \tag{13}$$

3. The Supplementary Tables

Table S1. Young's modulus (E), bulk modulus (K), shear modulus (G), Poisson's ratio (η), Pugh's ratio (G/K), and Vicker's hardness(H_V) for the 21 crystals.

#	Crystal	E(GPa)	K(GPa)	G(GPa)	η	G/K	H_V(GPa)
1	K_2Zn-1	28.712	21.999	11.194	0.283	0.509	2.361
2	K_2Zn-2	41.432	28.241	16.500	0.256	0.584	3.632
3	K_2Cu-3	32.303	26.329	12.467	0.296	0.474	2.349
4	K_2Cu-4	42.251	31.018	16.595	0.273	0.535	3.301
5	K_2Ni-5	20.853	16.749	8.067	0.293	0.482	1.759
6	K_2Ni-6	37.787	30.673	14.593	0.295	0.476	2.639
7	K_2Co-7	37.497	25.825	14.903	0.258	0.577	3.334
8	K_2Mg-8	42.945	27.731	17.290	0.242	0.623	4.041
9	Rb_2Zn-9	38.921	27.716	15.372	0.266	0.555	3.260
10	Rb_2Mg-10	40.606	27.239	16.222	0.252	0.596	3.673
11	Rb_2Co-11	37.848	28.985	14.757	0.283	0.509	2.871
12	$(NH_4)_2Zn$-12	49.931	34.213	19.865	0.257	0.581	4.118
13	$(NH_4)_2Mg$-13	46.434	33.579	18.288	0.269	0.545	3.612
14	$(NH_4)_2Ni$-14	44.468	38.973	16.975	0.310	0.436	2.658
15	$(NH_4)_2Co$-15	45.794	37.910	17.631	0.299	0.465	2.938
16	$(NH_4)_2Mn$-16	47.698	35.165	18.721	0.274	0.532	3.572
17	Tl_2Zn-17	37.208	26.668	14.678	0.268	0.550	3.123
18	Tl_2Ni-18	33.009	27.107	12.725	0.297	0.469	2.355
19	Tl_2Mg-19	37.723	26.564	14.930	0.263	0.562	3.240
20	Tl_2Mn-20	40.013	27.919	15.864	0.261	0.568	3.423
21	Tl_2Co-21	36.767	27.260	14.416	0.275	0.529	2.950

Table S2. The calculated elastic constants C_{ij} (GPa) for the 21 crystals.

#	Crystal	C_{11}	C_{12}	C_{13}	C_{21}	C_{22}	C_{23}	C_{31}	C_{32}	C_{33}	C_{44}	C_{55}	C_{66}
1	K_2Zn-1	27.25	11.57	17.63	11.57	38.22	10.98	17.63	10.97	64.11	10.48	10.23	8.71
2	K_2Zn-2	77.63	8.92	5.02	8.92	60.02	20.78	5.02	20.78	61.86	16.05	10.42	12.10
3	K_2Cu-3	44.29	14.10	27.81	14.10	63.34	8.89	27.81	8.89	38.34	5.67	21.41	14.65
4	K_2Cu-4	94.55	6.73	6.75	6.73	61.88	21.90	6.75	21.90	64.28	14.73	10.17	9.55
5	K_2Ni-5	36.94	10.64	16.31	10.64	31.92	11.56	16.31	11.56	28.00	7.84	7.15	8.17
6	K_2Ni-6	98.74	2.88	12.45	2.88	55.23	21.68	12.45	21.68	67.31	13.45	8.34	6.65
7	K_2Co-7	69.07	13.84	8.69	13.84	50.12	21.03	8.69	21.03	52.49	14.68	14.46	14.26
8	K_2Mg-8	77.01	9.06	4.66	9.06	59.20	19.48	4.66	19.48	61.66	17.74	10.71	13.99
9	Rb_2Zn-9	77.62	7.40	7.63	7.40	55.60	19.74	7.63	19.74	59.36	14.19	9.88	10.69
10	Rb_2Mg-10	75.76	8.01	6.93	8.01	56.44	18.59	6.93	18.59	58.64	16.28	10.50	11.78
11	Rb_2Co-11	85.87	8.85	8.14	8.85	54.96	19.92	8.14	19.92	59.52	16.53	8.67	7.94
12	$(NH_4)_2Zn$-12	92.55	15.09	9.08	15.09	67.30	20.72	9.08	20.72	67.18	17.03	13.15	17.54
13	$(NH_4)_2Mg$-13	98.64	9.19	10.49	9.16	64.04	22.95	10.49	22.95	72.56	17.34	12.41	11.20
14	$(NH_4)_2Ni$-14	107.7	13.59	19.70	13.59	64.34	22.72	19.70	22.72	79.55	12.78	10.65	9.56
15	$(NH_4)_2Co$-15	112.1	13.68	17.54	13.68	65.27	22.26	17.54	22.26	70.66	13.36	12.43	10.27
16	$(NH_4)_2Mn$-16	105.6	13.09	11.51	13.09	63.56	24.21	11.51	24.21	70.34	18.20	12.40	12.51
17	Tl_2Zn-17	73.80	7.57	7.62	7.57	56.59	20.00	7.62	20.00	59.24	12.78	10.60	9.90
18	Tl_2Ni-18	79.66	8.99	8.83	8.99	56.12	19.95	8.83	19.95	60.88	12.93	8.20	6.54
19	Tl_2Mg-19	69.38	8.78	8.41	8.78	60.09	19.57	8.41	19.57	56.91	16.68	10.52	9.61
20	Tl_2Mn-20	81.01	7.28	8.44	7.28	55.47	20.59	8.44	20.59	61.28	15.53	11.75	9.98
21	Tl_2Co-21	71.57	7.82	9.48	7.82	57.52	20.39	9.48	20.39	60.85	16.21	9.44	8.09

Table S3. The calculated elastic constants C_{ij} (GPa) for the crystals 2, 4, 6, and the crystals from 7 to 21.

#	Crystal	C_{14}	C_{15}	C_{16}	C_{24}	C_{25}	C_{26}	C_{34}	C_{35}	C_{36}	C_{41}	C_{42}	C_{43}
2	K_2Zn-2	0.550	1.546	6.645	5.696	1.292	4.759	5.892	6.985	1.973	0.550	5.696	5.892
4	K_2Cu-4	-1.335	-3.959	6.998	3.931	-0.035	2.128	1.987	1.589	2.227	-1.335	3.931	1.987
6	K_2Ni-6	-4.120	-5.780	4.561	0.866	0.499	-0.465	-1.983	-1.825	1.705	-4.110	0.866	-1.983
7	K_2Co-7	6.064	2.106	7.559	-3.402	1.820	9.264	-3.350	13.601	3.078	6.064	-3.403	-3.350
8	K_2Mg-8	1.150	2.922	7.597	5.294	1.292	4.937	4.882	7.515	1.962	1.150	5.294	4.882
9	Rb_2Zn-9	-0.311	0.174	5.856	5.658	1.696	2.871	4.806	6.229	2.703	-0.311	5.658	4.806
10	Rb_2Mg-10	0.277	1.873	6.766	5.426	1.414	3.365	3.430	6.787	2.665	0.277	5.426	3.430
11	Rb_2Co-11	1.767	-2.140	3.554	3.558	1.480	2.605	3.130	4.991	1.619	1.767	3.558	3.130
12	$(NH_4)_2Zn$-12	-3.754	-3.681	1.661	4.574	-2.231	2.183	9.928	1.488	1.647	-3.754	4.574	9.928
13	$(NH_4)_2Mg$-13	0.664	0.233	5.678	6.198	0.814	6.701	4.589	6.545	2.757	0.664	6.198	4.589
14	$(NH_4)_2Ni$-14	-0.587	-2.088	-0.607	0.798	1.722	-2.803	0.886	1.063	0.422	-0.587	0.798	0.886
15	$(NH_4)_2Co$-15	-0.992	-2.398	4.369	1.494	1.324	2.610	0.445	2.733	2.130	-0.992	1.494	0.445
16	$(NH_4)_2Mn$-16	2.073	0.448	8.053	3.936	1.663	7.186	3.380	5.966	3.636	2.073	3.936	3.380
17	Tl_2Zn-17	0.127	0.131	8.503	5.427	1.865	5.705	5.196	7.217	2.820	0.126	5.427	5.196
18	Tl_2Ni-18	1.250	-2.633	4.583	5.406	1.455	6.105	5.676	6.356	1.747	1.250	5.406	5.676
19	Tl_2Mg-19	0.616	1.696	9.970	5.380	1.593	5.161	3.821	7.588	2.162	0.616	5.380	3.821
20	Tl_2Mn-20	0.105	2.772	9.592	3.513	1.753	3.957	2.668	7.877	2.968	0.105	3.513	2.668
21	Tl_2Co-21	0.894	-0.866	5.797	4.335	1.586	6.103	3.350	6.700	2.342	0.894	4.335	3.360

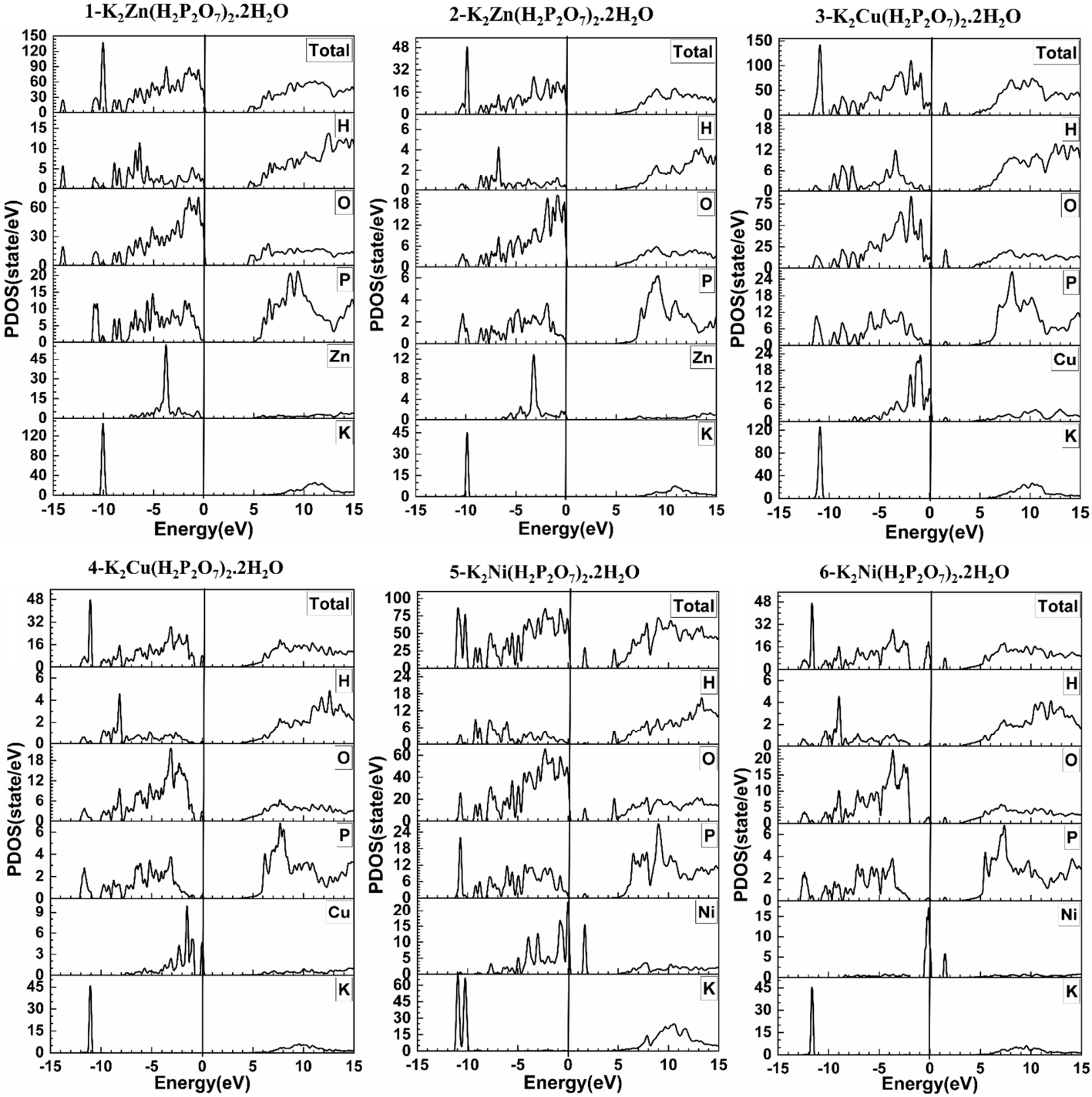
Table S4. The calculated elastic constants C_{ij} (GPa) for the crystals 2, 4, 6, and the crystals from 7 to 21.

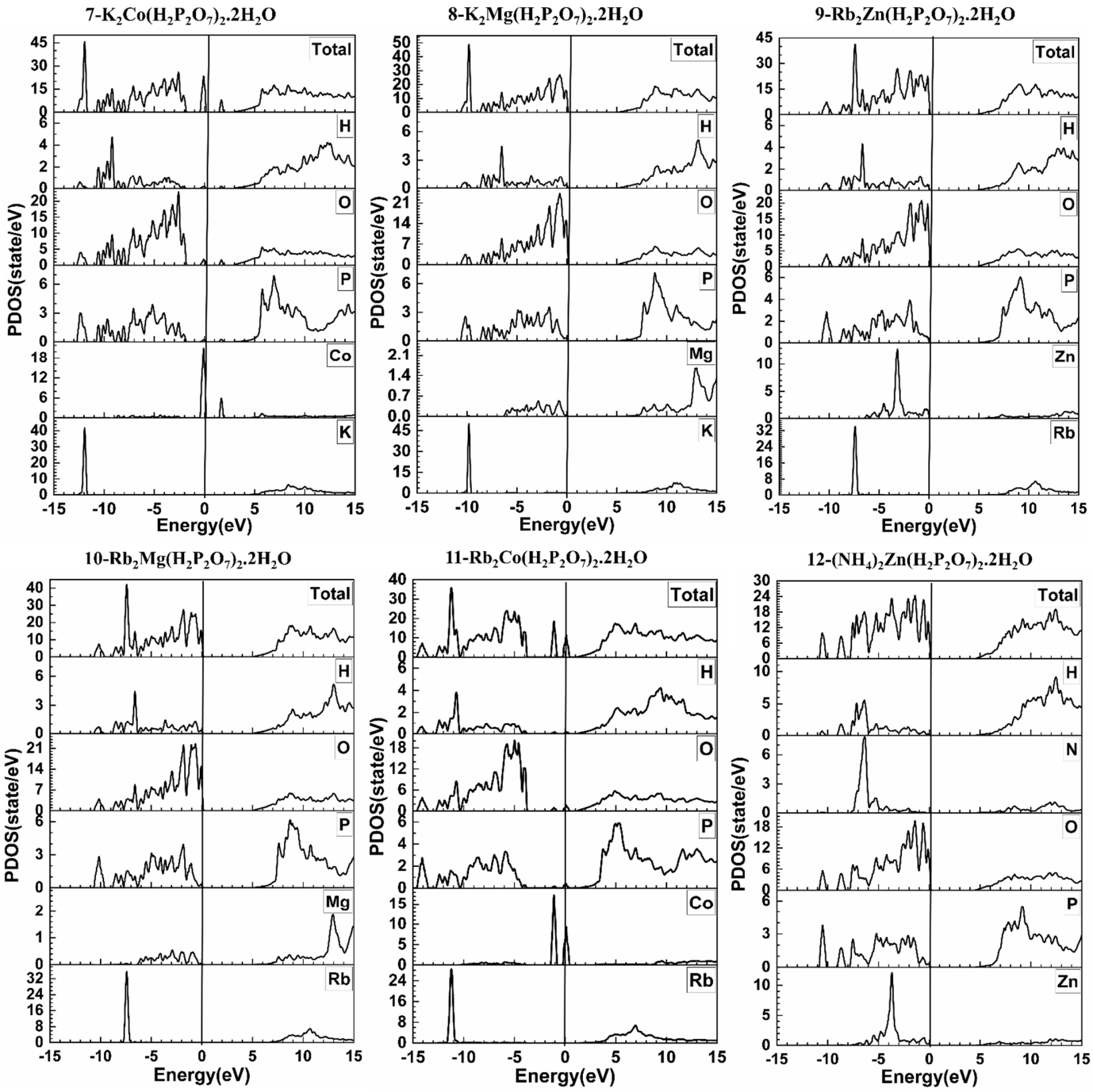
#	Crystal	C_{45}	C_{46}	C_{51}	C_{52}	C_{53}	C_{54}	C_{56}	C_{61}	C_{62}	C_{63}	C_{64}	C_{65}
2	K_2Zn-2	0.650	2.270	1.546	1.292	6.985	0.650	1.913	6.645	4.759	1.973	2.270	1.913
4	K_2Cu-4	-0.920	0.321	-3.960	-0.035	1.588	-0.920	0.426	6.998	2.128	2.227	0.321	0.426
6	K_2Ni-6	-1.349	-0.263	-5.780	0.499	-1.825	-1.349	0.604	4.561	-0.465	1.705	-0.263	0.604
7	K_2Co-7	2.950	1.247	2.106	1.820	13.601	2.950	1.445	7.559	9.264	3.078	1.247	1.445
8	K_2Mg-8	1.140	2.649	2.922	1.292	7.515	1.140	2.522	7.597	4.937	1.962	2.649	2.522
9	Rb_2Zn-9	1.044	2.498	0.174	1.696	6.229	1.044	1.291	5.856	2.871	2.703	2.498	1.291
10	Rb_2Mg-10	1.082	2.533	1.873	1.414	6.787	1.082	1.734	6.766	3.365	2.665	2.533	1.732
11	Rb_2Co-11	1.618	3.052	-2.140	1.480	4.991	1.618	-0.476	3.554	2.605	1.619	3.052	-0.476
12	$(NH_4)_2Zn$-12	-0.383	-0.690	-3.681	-2.231	1.488	-0.383	-1.722	1.661	2.183	1.647	-0.690	-1.722
13	$(NH_4)_2Mg$-13	0.411	2.853	0.233	0.814	6.545	0.411	1.475	5.678	6.701	2.757	2.853	1.475
14	$(NH_4)_2Ni$-14	-1.099	0.464	-2.088	1.721	1.063	-1.099	0.272	-0.607	-2.803	0.421	0.464	0.272
15	$(NH_4)_2Co$-15	-0.346	1.761	-2.398	1.324	2.733	-0.346	0.868	4.369	2.610	2.130	1.761	0.868
16	$(NH_4)_2Mn$-16	1.126	2.903	0.448	1.663	5.966	1.126	2.076	8.053	7.188	3.636	2.903	2.076
17	Tl_2Zn-17	0.775	2.423	0.131	1.865	7.217	0.775	1.232	8.503	5.705	2.820	2.423	1.232
18	Tl_2Ni-18	0.989	2.390	-2.633	1.455	6.356	0.989	-2.108	4.583	6.105	1.747	2.390	-2.108
19	Tl_2Mg-19	1.283	2.772	1.696	1.593	7.588	1.283	1.629	9.970	5.161	2.162	2.772	1.629
20	Tl_2Mn-20	1.112	2.245	2.772	1.753	7.877	1.112	1.784	9.592	3.957	2.968	2.245	1.784
21	Tl_2Co-21	0.948	2.122	-0.866	1.586	6.700	0.948	-0.989	5.797	6.103	2.342	2.122	-0.989

Table S5. The calculated bulk and shear modulus' Voigt and Reuss estimates (K_V, K_R, G_V, G_R), the universal anisotropic index (A^U), and the percentage of anisotropy in the compression and shear for the 21 crystals (A_{comp}, A_{shear}).

#	Crystal	K_V (GPa)	K_R (GPa)	G_V (GPa)	G_R (GPa)	A^U	A_{comp}	A_{shear}
1	K_2Zn-1	23.325	20.674	11.843	10.545	0.744	0.060	0.058
2	K_2Zn-2	29.885	26.909	18.702	14.299	1.650	0.052	0.133
3	K_2Cu-3	27.509	26.247	14.690	10.245	2.218	0.024	0.178
4	K_2Cu-4	32.385	30.555	19.245	13.946	1.960	0.029	0.160
5	K_2Ni-5	19.320	14.412	8.520	7.614	0.936	0.146	0.056
6	K_2Ni-6	32.812	30.615	17.973	11.213	3.086	0.035	0.232
7	K_2Co-7	28.754	22.980	17.221	12.586	2.090	0.112	0.155
8	K_2Mg-8	29.363	26.335	19.467	15.114	1.555	0.054	0.126
9	Rb_2Zn-9	29.125	26.720	17.473	13.271	1.673	0.043	0.137
10	Rb_2Mg-10	28.654	26.080	18.197	14.247	1.485	0.047	0.122
11	Rb_2Co-11	30.463	28.768	17.524	11.991	2.366	0.029	0.188
12	$(NH_4)_2Zn$-12	35.202	33.479	21.687	18.043	1.062	0.025	0.092
13	$(NH_4)_2Mg$-13	35.609	32.099	21.029	15.546	1.873	0.052	0.150
14	$(NH_4)_2Ni$-14	40.402	38.827	19.636	14.313	1.900	0.020	0.157
15	$(NH_4)_2Co$-15	39.440	37.271	20.179	15.083	1.748	0.028	0.145
16	$(NH_4)_2Mn$-16	37.456	33.262	21.336	16.106	1.750	0.059	0.140
17	Tl_2Zn-17	28.890	24.715	16.951	12.405	2.001	0.078	0.155
18	Tl_2Ni-18	30.243	25.590	16.127	9.322	3.832	0.083	0.267
19	Tl_2Mg-19	28.878	24.539	17.337	12.524	2.098	0.081	0.161
20	Tl_2Mn-20	30.043	26.081	18.215	13.513	1.892	0.071	0.148
21	Tl_2Co-21	29.479	25.508	16.898	11.935	2.235	0.072	0.172

4. The Supplementary Figures

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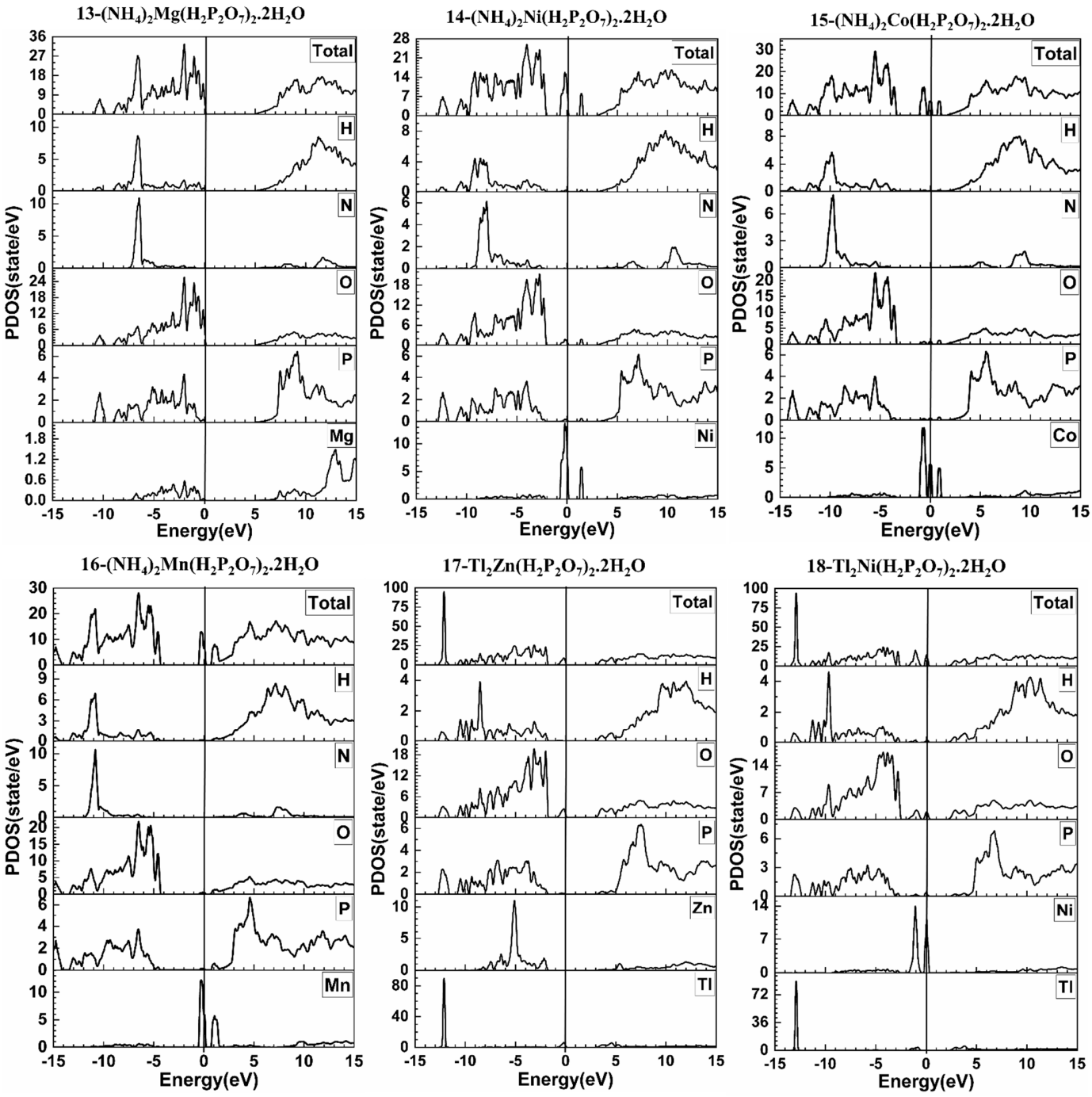
8-K2Mg(H2P2O7)2.2H2O
9-Rb2Zn(H2P2O7)2.2H2O
10-Rb2Mg(H2P2O7)2.2H2O
11-Rb2Co(H2P2O7)2.2H2O
12-(NH4)2Zn(H2P2O7)2.2H2O
Total
H
O
P
Co
K
Mg
Rb
Zn
N
PDOS(state/eV)
Energy(eV)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15-(NH4)2Co(H2P2O7)2.2H2O
16-(NH4)2Mn(H2P2O7)2.2H2O
17-Tl2Zn(H2P2O7)2.2H2O
18-Tl2Ni(H2P2O7)2.2H2O
Total
H
N
O
P
Mg
Ni
Co
Mn
Zn
Tl
PDOS(state/eV)
Energy(eV)

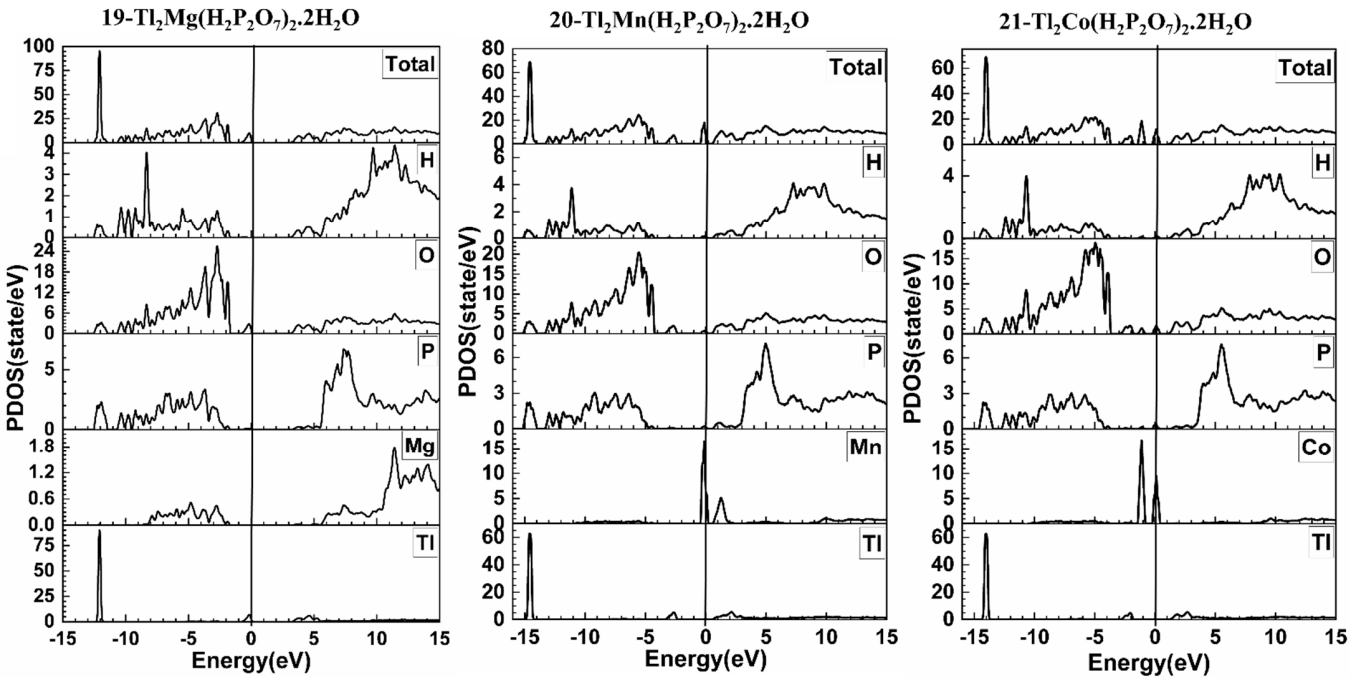
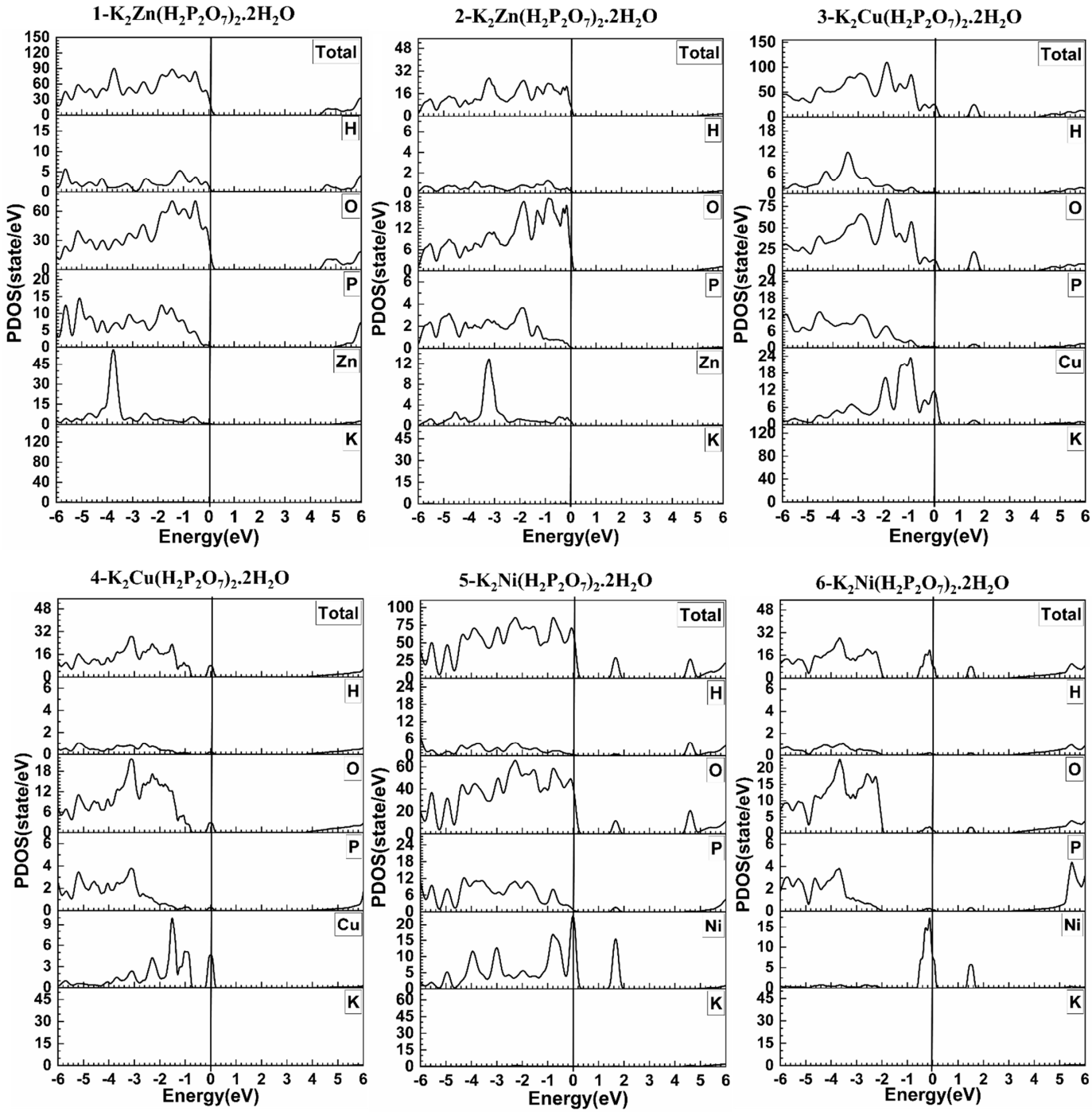


Fig.S1 Calculated partial density of states (PDOS) for the 21 crystals.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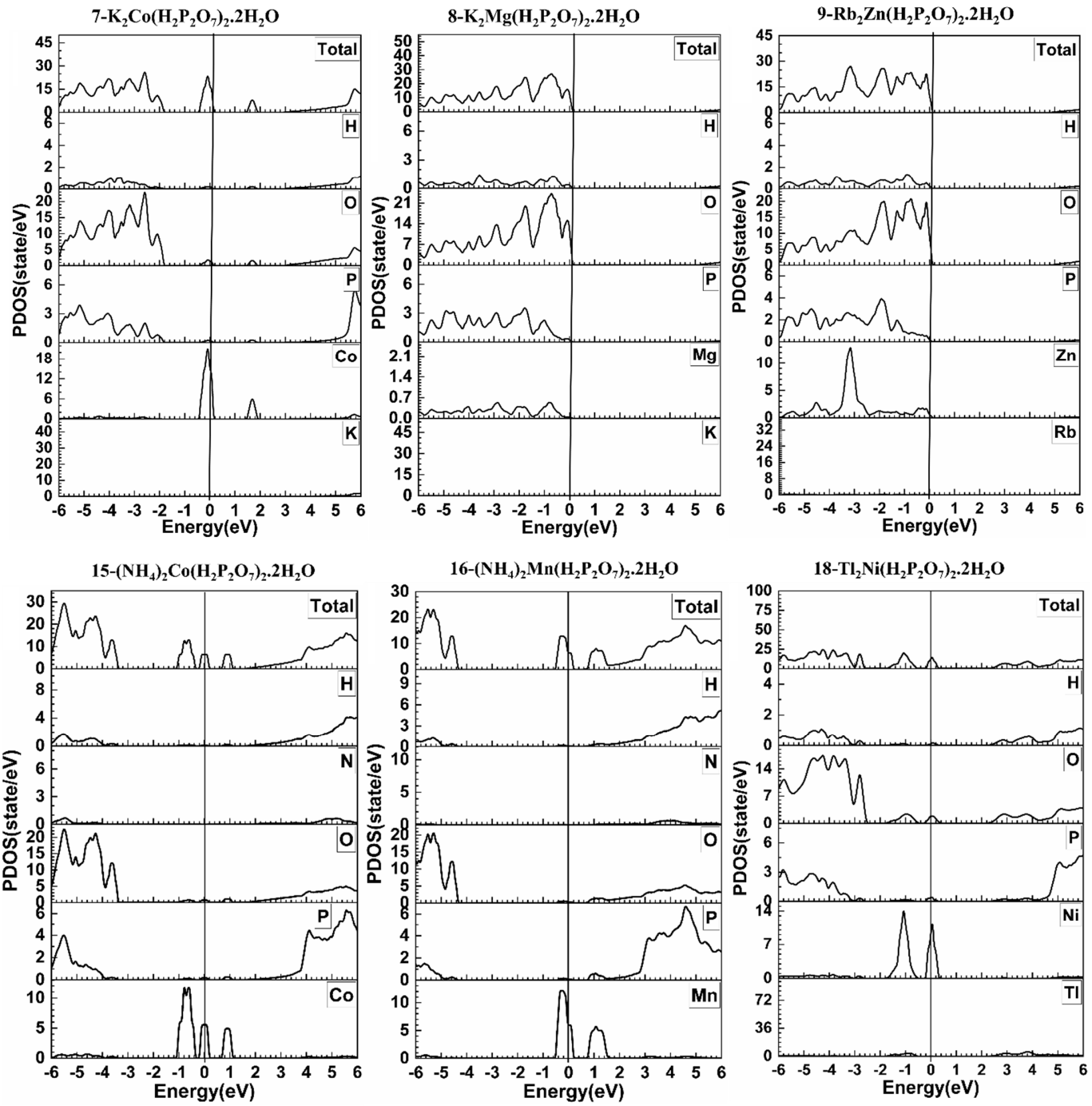
2-K2Zn(H2P2O7)2.2H2O
3-K2Cu(H2P2O7)2.2H2O
4-K2Cu(H2P2O7)2.2H2O
5-K2Ni(H2P2O7)2.2H2O
6-K2Ni(H2P2O7)2.2H2O
Total
H
O
P
Zn
Cu
Ni
K
PDOS(state/eV)
Energy(eV)


7-K2Co(H2P2O7)2.2H2O
8-K2Mg(H2P2O7)2.2H2O
9-Rb2Zn(H2P2O7)2.2H2O
15-(NH4)2Co(H2P2O7)2.2H2O
16-(NH4)2Mn(H2P2O7)2.2H2O
18-Tl2Ni(H2P2O7)2.2H2O
Total
H
O
P
Co
K
Mg
Zn
Rb
N
Mn
Ni
Tl
PDOS(state/eV)
Energy(eV)

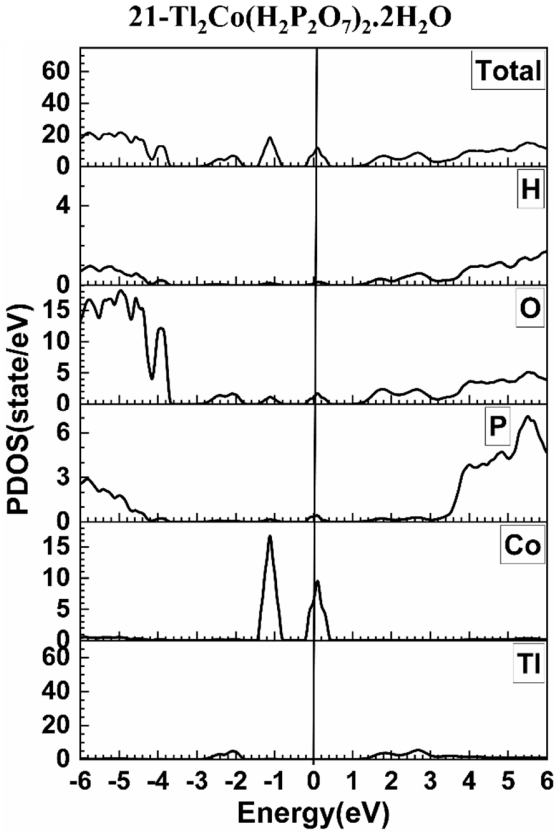
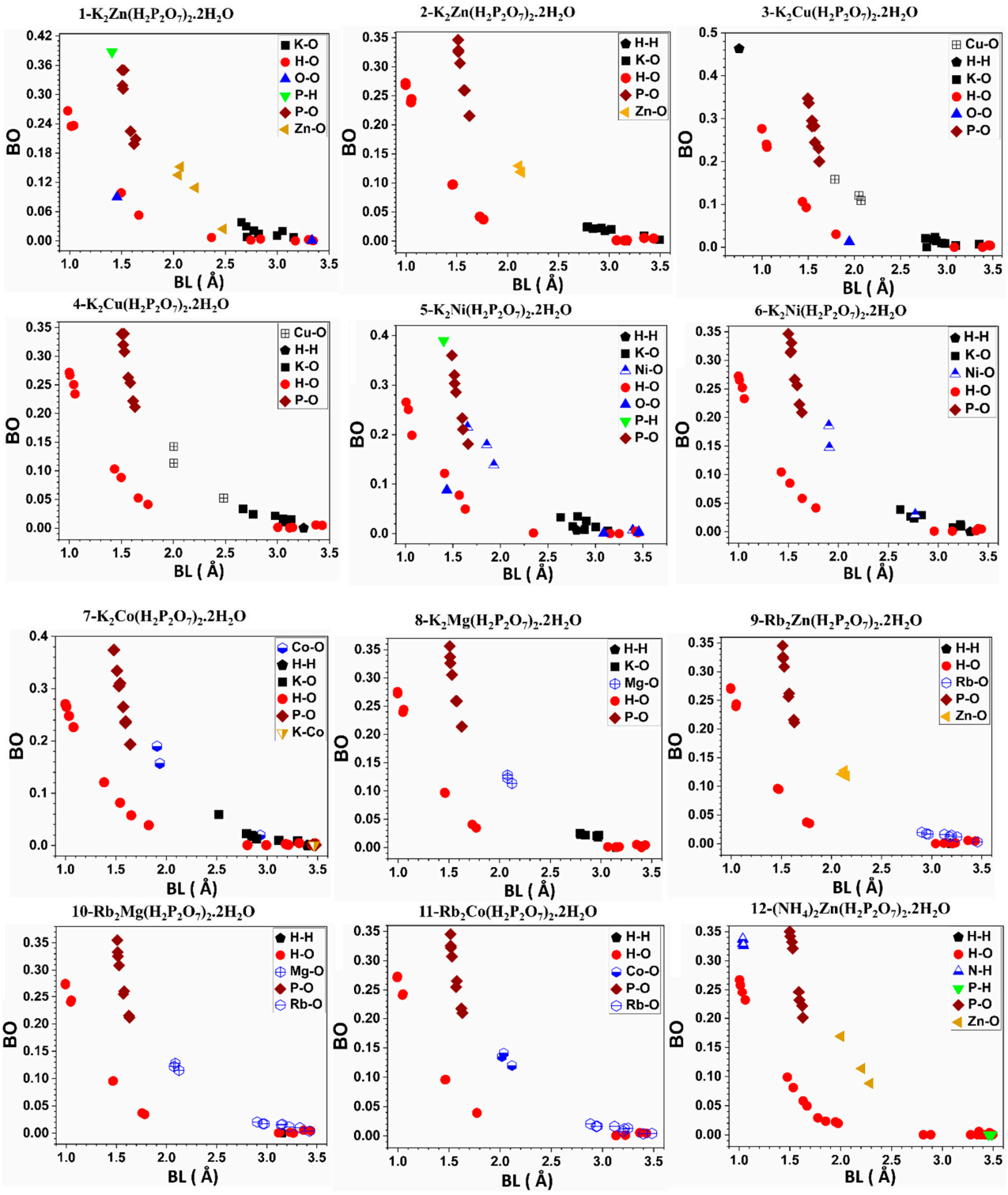


Fig.S2 Calculated TDOS and PDOS of crystals from 1 to 9, and crystals: 15, 16, 18, and 21.


1-K2Zn(H2P2O7)2.2H2O
K-O
H-O
O-O
P-H
P-O
Zn-O
2-K2Zn(H2P2O7)2.2H2O
H-H
K-O
H-O
P-O
Zn-O
3-K2Cu(H2P2O7)2.2H2O
Cu-O
H-H
K-O
H-O
O-O
P-O
4-K2Cu(H2P2O7)2.2H2O
Cu-O
H-H
K-O
H-O
P-O
5-K2Ni(H2P2O7)2.2H2O
H-H
K-O
Ni-O
H-O
O-O
P-H
P-O
6-K2Ni(H2P2O7)2.2H2O
H-H
K-O
Ni-O
H-O
P-O
7-K2Co(H2P2O7)2.2H2O
Co-O
H-H
K-O
H-O
P-O
K-Co
8-K2Mg(H2P2O7)2.2H2O
H-H
K-O
Mg-O
H-O
P-O
9-Rb2Zn(H2P2O7)2.2H2O
H-H
H-O
Rb-O
P-O
Zn-O
10-Rb2Mg(H2P2O7)2.2H2O
H-H
H-O
Mg-O
P-O
Rb-O
11-Rb2Co(H2P2O7)2.2H2O
H-H
H-O
Co-O
P-O
Rb-O
12-(NH4)2Zn(H2P2O7)2.2H2O
H-H
H-O
N-H
P-H
P-O
Zn-O
BO
BL (Å)

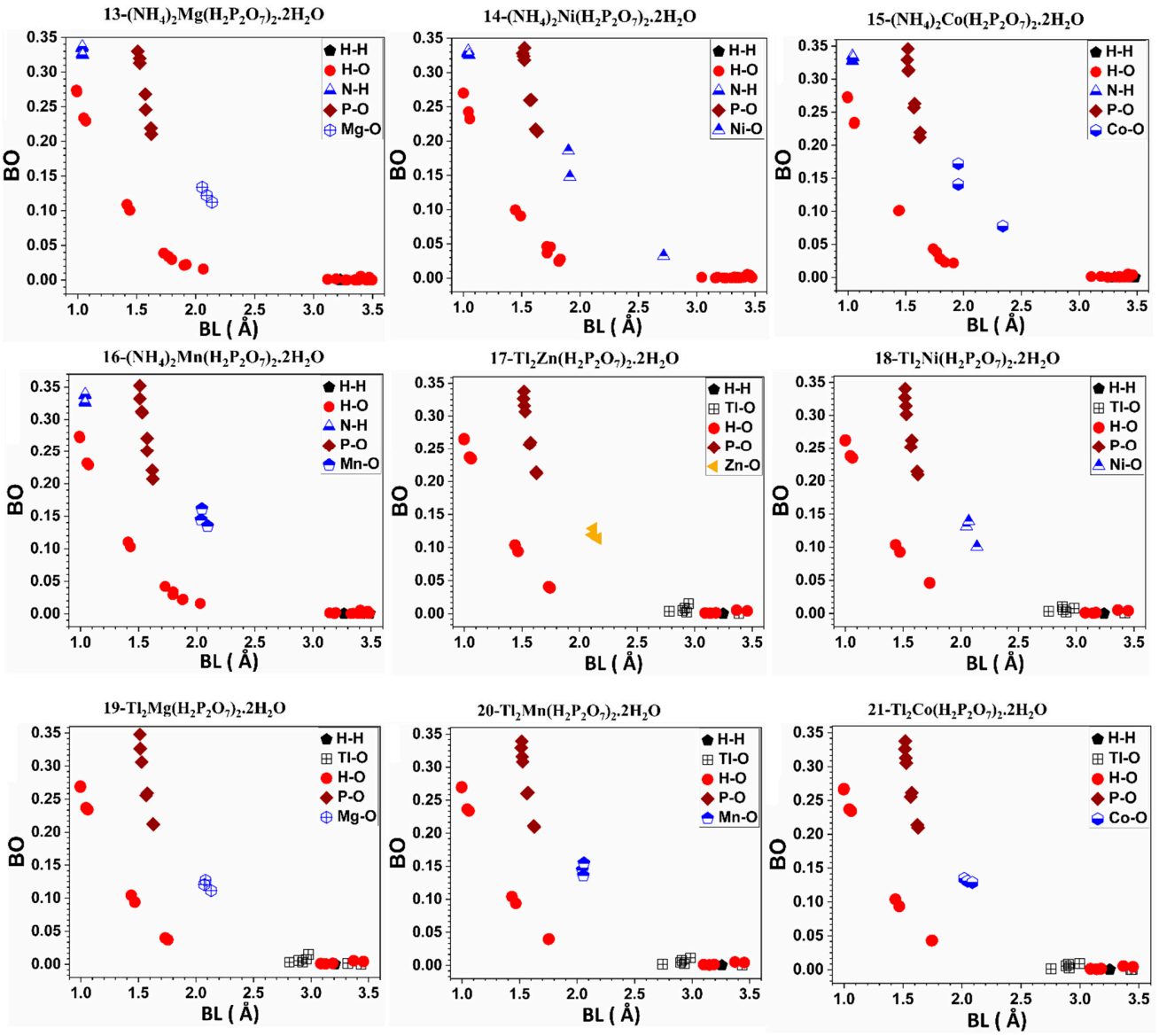


Fig.S3 Calculated BO vs BL for the 21 crystals.

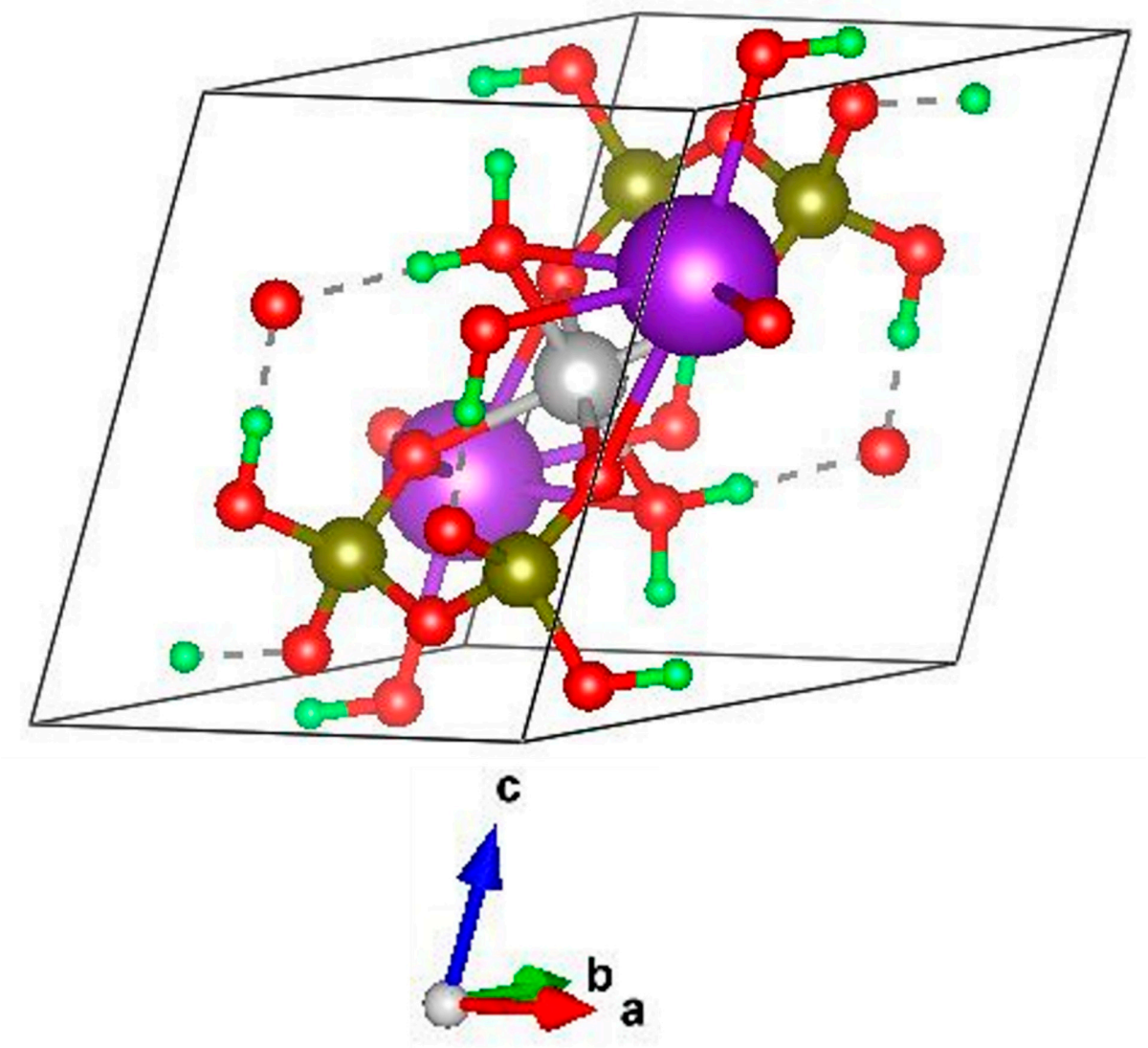
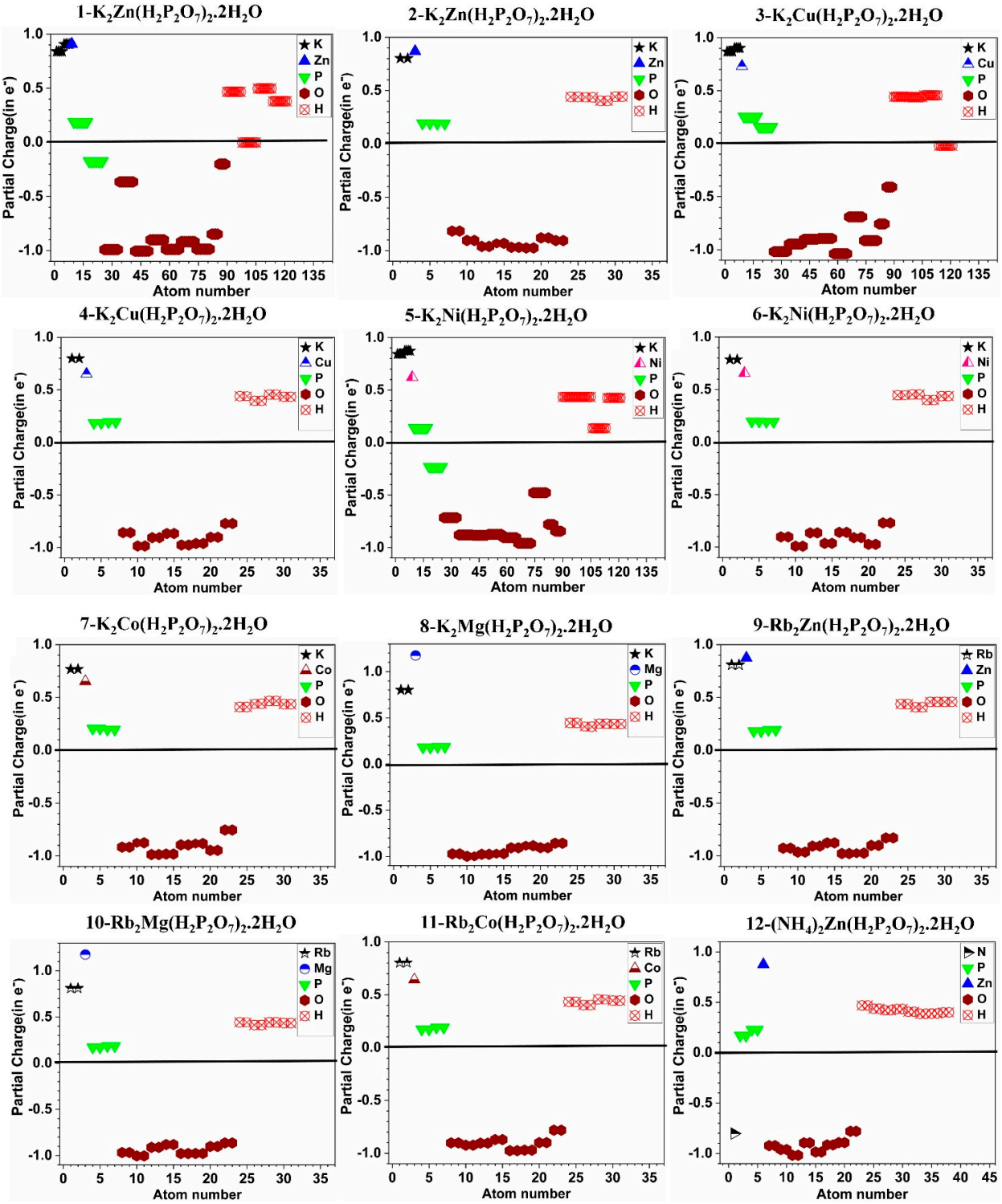


Fig.S4. Crystal structure of crystals 2 in ball and stick format. Crystal 2 with triclinic phase.


1-K2Zn(H2P2O7)2.2H2O
2-K2Zn(H2P2O7)2.2H2O
3-K2Cu(H2P2O7)2.2H2O
4-K2Cu(H2P2O7)2.2H2O
5-K2Ni(H2P2O7)2.2H2O
6-K2Ni(H2P2O7)2.2H2O
7-K2Co(H2P2O7)2.2H2O
8-K2Mg(H2P2O7)2.2H2O
9-Rb2Zn(H2P2O7)2.2H2O
10-Rb2Mg(H2P2O7)2.2H2O
11-Rb2Co(H2P2O7)2.2H2O
12-(NH4)2Zn(H2P2O7)2.2H2O
Partial Charge(in e-)
Atom number
K
Zn
Cu
Ni
Co
Mg
Rb
N
P
O
H

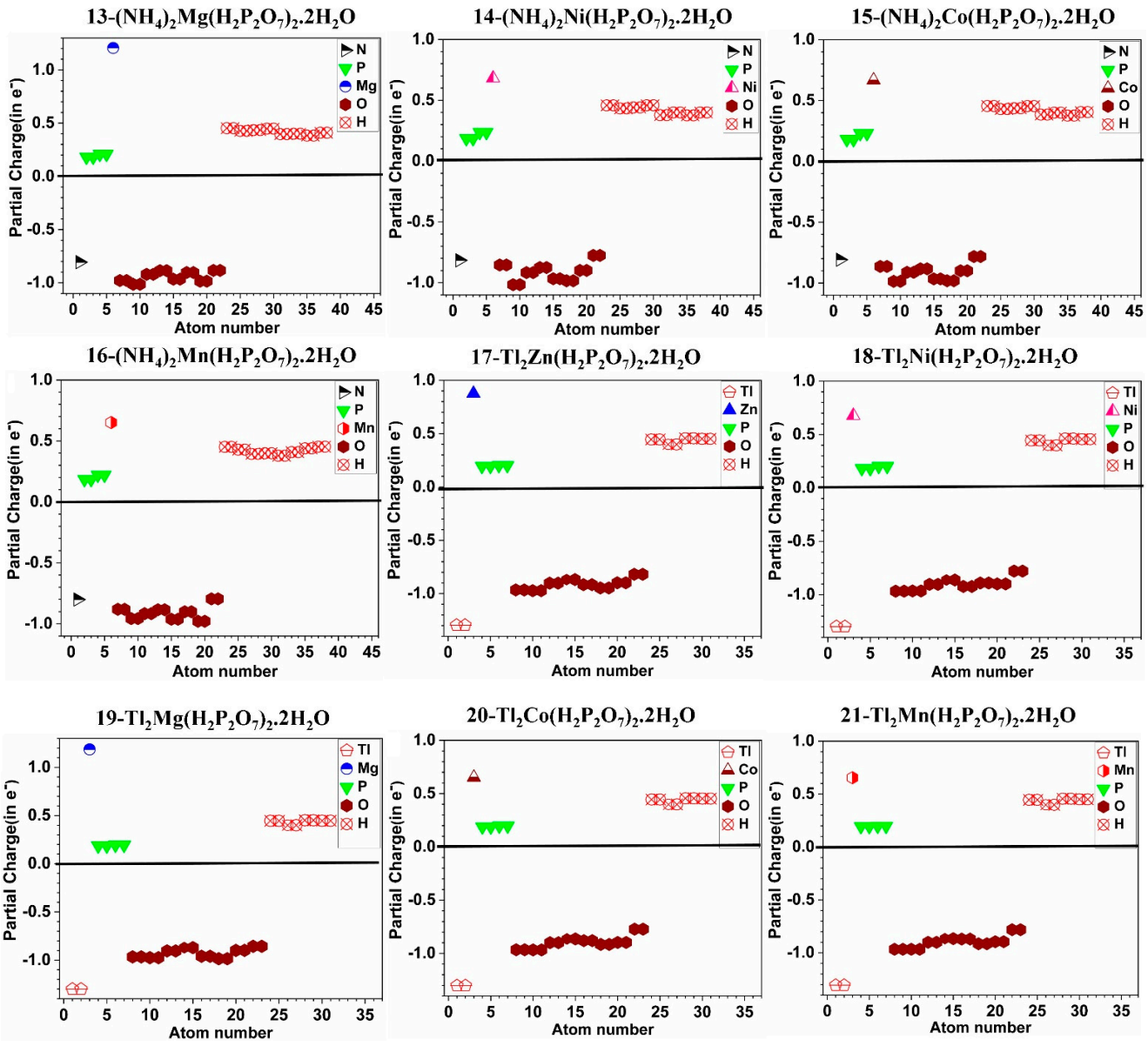


Fig.S5. Calculated partial charge distribution in the 21 pyrophosphate crystals.

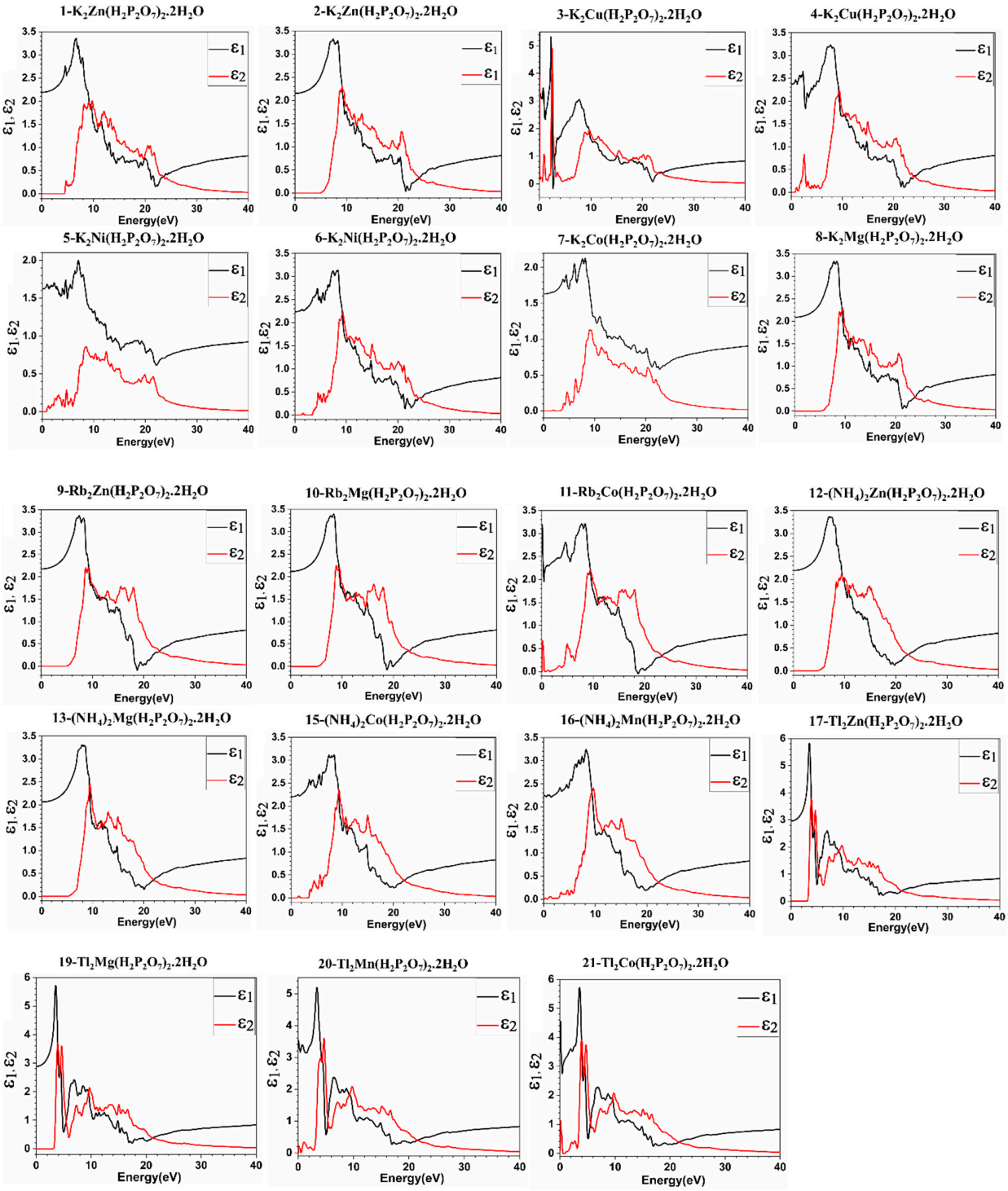


Fig.S6. Calculated optical dielectric functions for the 21 crystals.

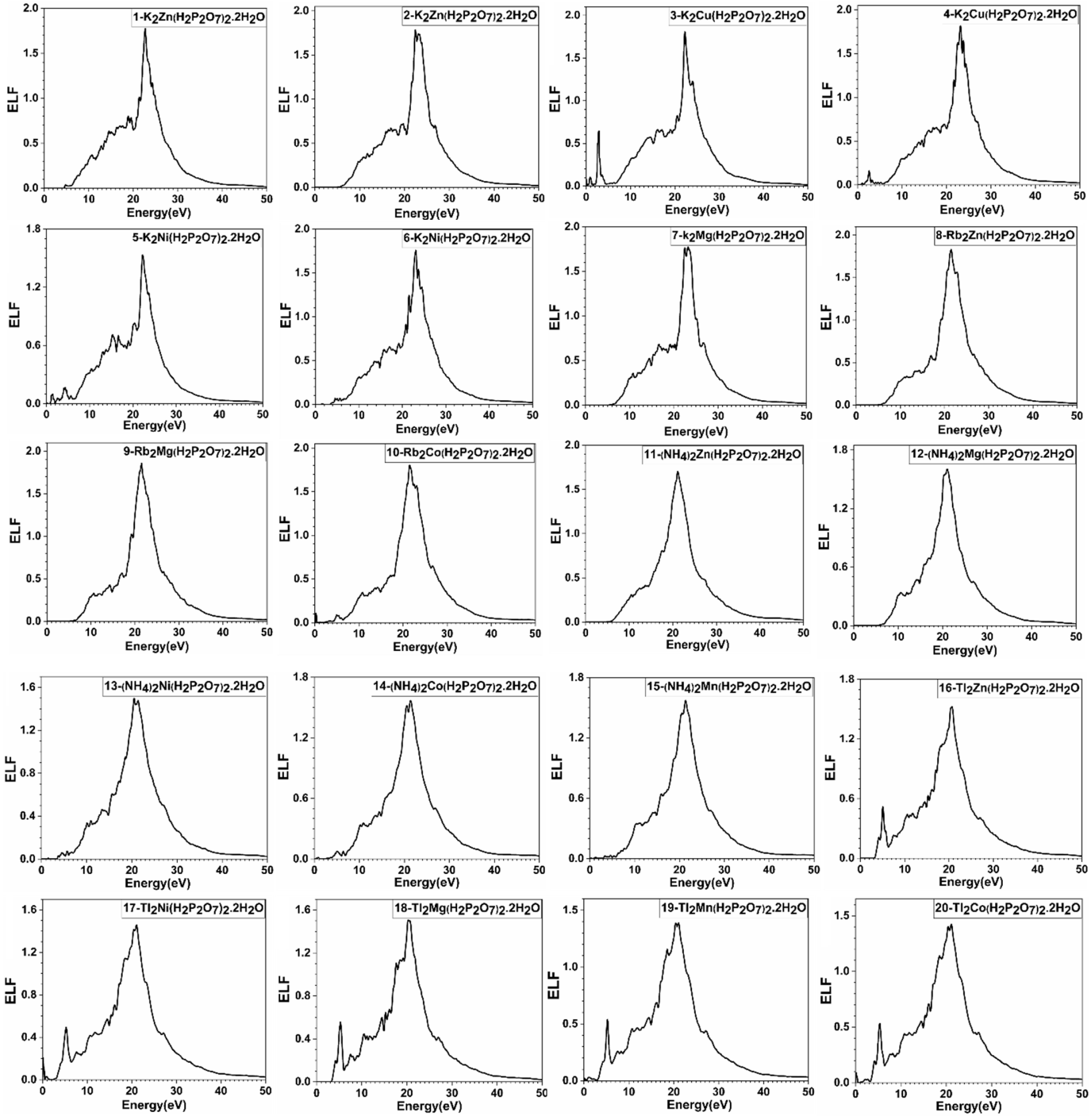
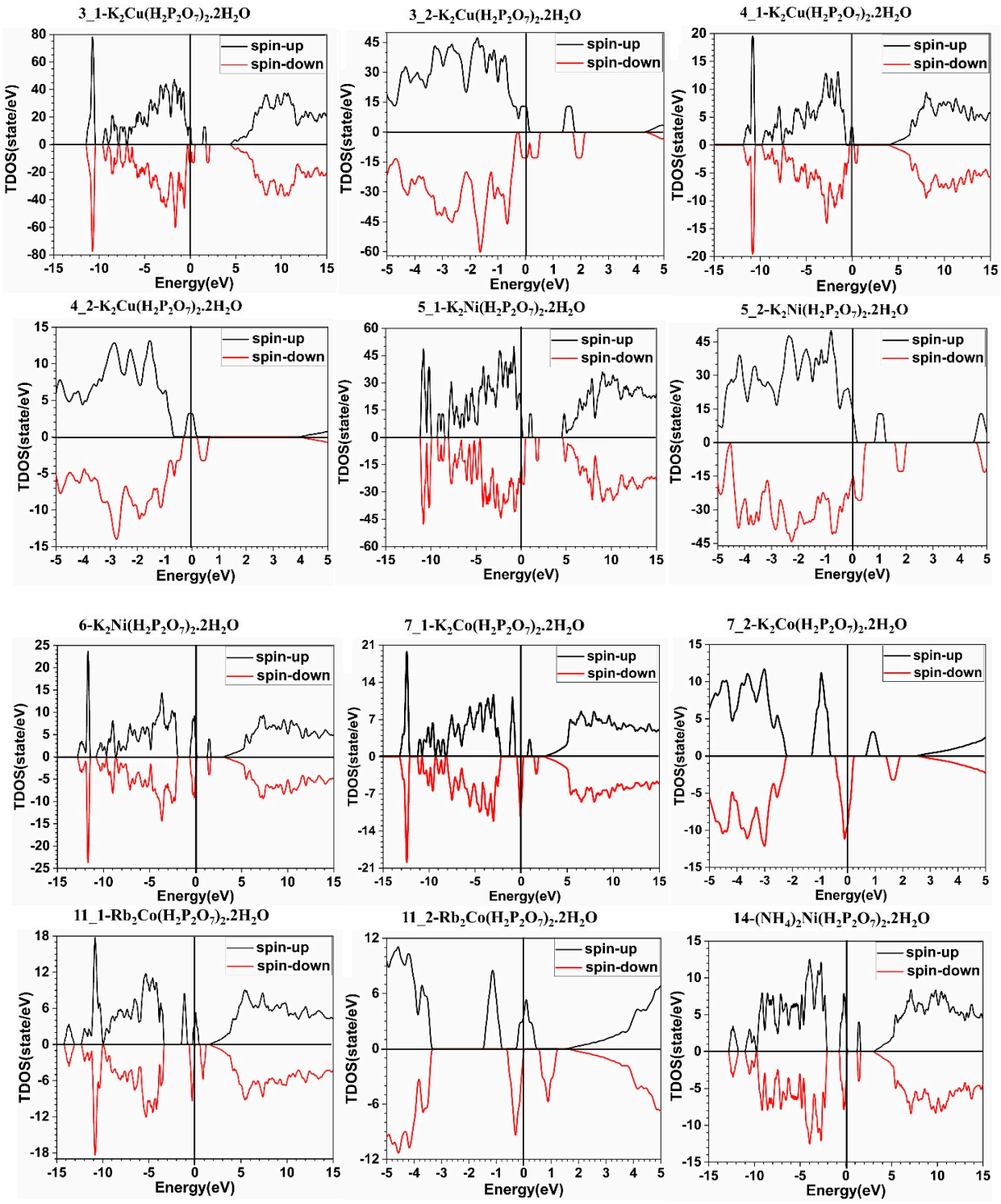


Fig.S7 Calculated energy lose function (ELF) for the 21 crystals.


3_1-K2Cu(H2P2O7)2.2H2O
3_2-K2Cu(H2P2O7)2.2H2O
4_1-K2Cu(H2P2O7)2.2H2O
4_2-K2Cu(H2P2O7)2.2H2O
5_1-K2Ni(H2P2O7)2.2H2O
5_2-K2Ni(H2P2O7)2.2H2O
6-K2Ni(H2P2O7)2.2H2O
7_1-K2Co(H2P2O7)2.2H2O
7_2-K2Co(H2P2O7)2.2H2O
11_1-Rb2Co(H2P2O7)2.2H2O
11_2-Rb2Co(H2P2O7)2.2H2O
14-(NH4)2Ni(H2P2O7)2.2H2O
spin-up
spin-down
TDOS(state/eV)
Energy(eV)

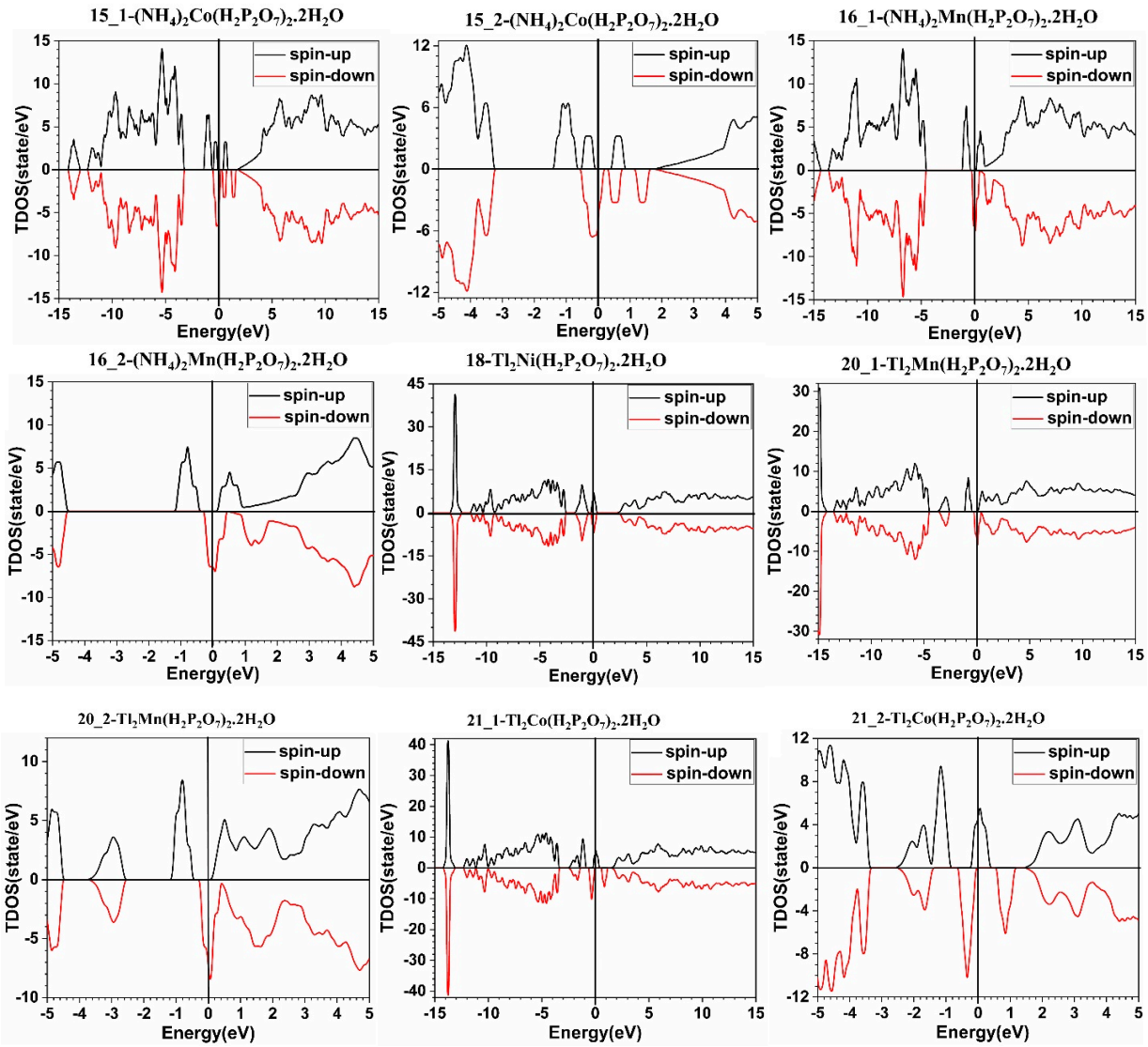


Fig.S8. Calculated TDOS with spin-polarized effect for the crystals 3, 4, 5, 6, 11, 14, 15, 16, 18, 20, and 21. The abbreviation #_**2** means that this figure belong to the same # crystal but in different range of energy. For example, 3_1 and 3_2 figures represent the same crystal.

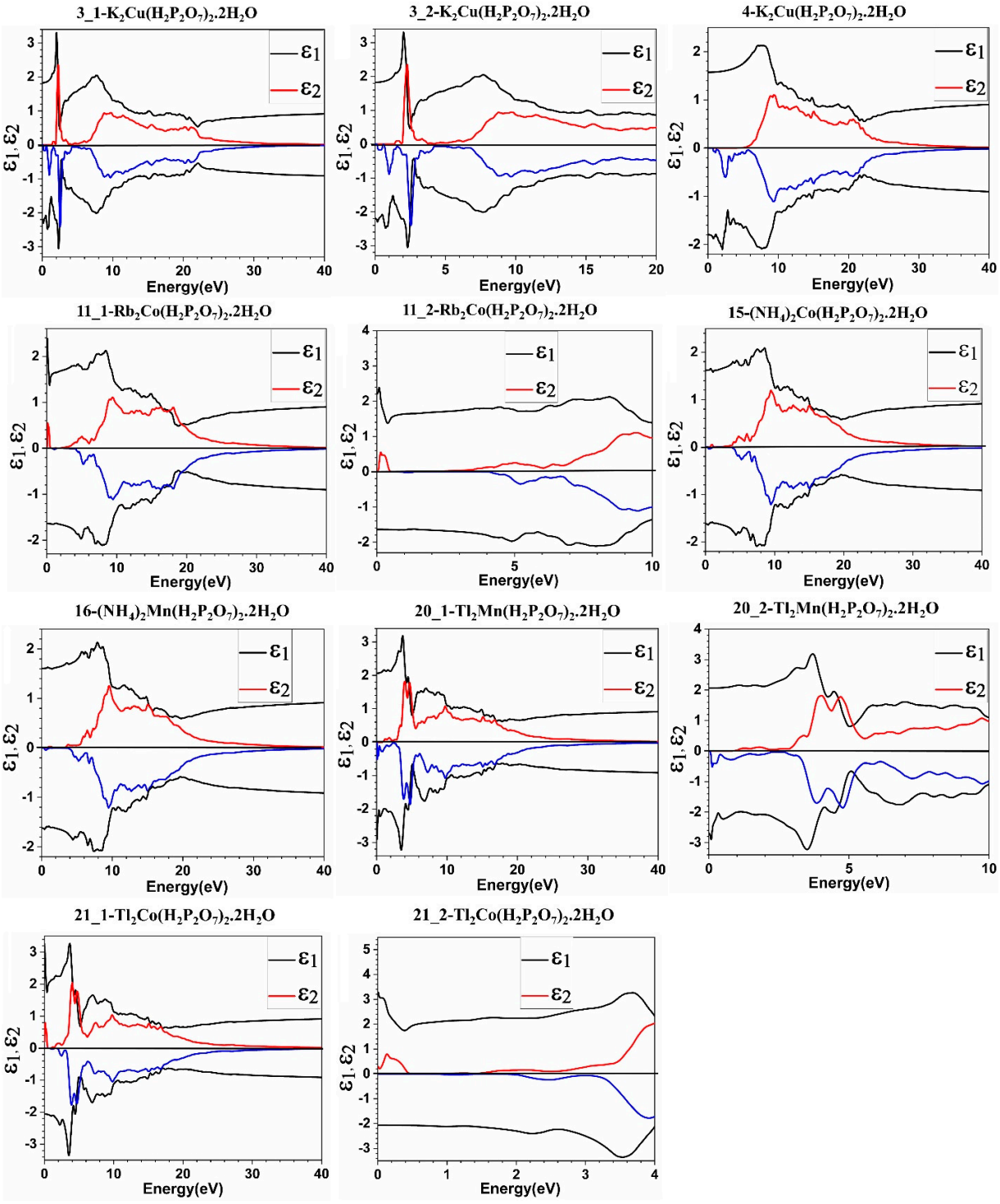


Fig.S9 Calculated spin –up and spin-down refractive indices of crystals: 3, 4, 11, 15, 16, 20, and 21. The abbreviation **#_2** means that this figure belong to the same # crystal but in different range of energy. For example, 3_1 and 3_2 figures represent the same crystal.

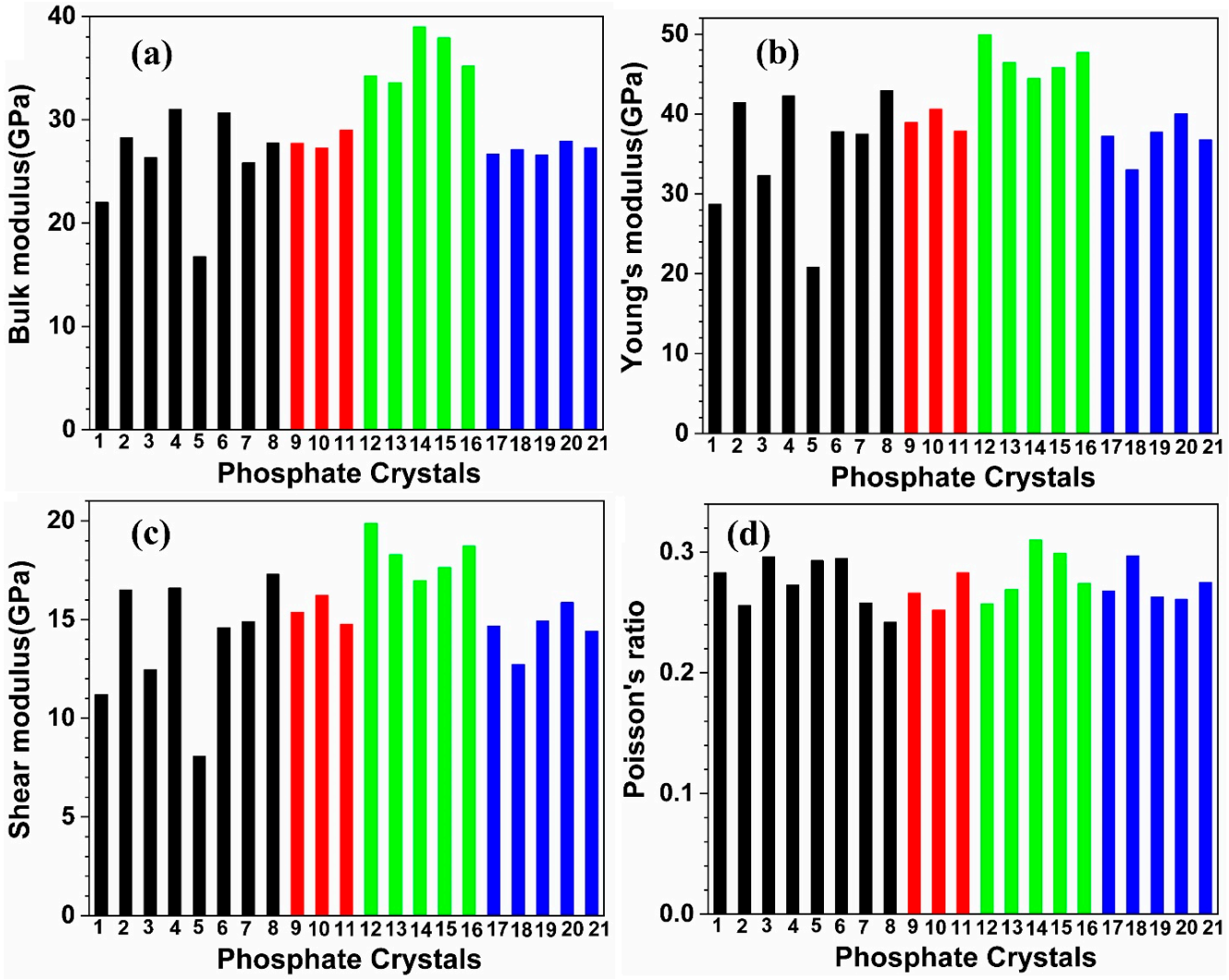


Fig.S10 (a–d) Distribution of bulk modulus (**a**), Young's modulus(**b**), shear modulus(**c**), and Poisson's ratio (**d**) for the 21 crystals. The black color is for the crystals from 1 to 8, red color is for crystals from 9 to 11, green color is for crystals from 12 to 16, and blue color is for crystals from 17 to 21.

5. References

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[3] Reuss, A. Berechnung der Fließgrenze von Mischkristallen auf Grund der Plastizitätsbedingung für Einkristalle .. **1929**, *9*, 49–58, https://doi.org/10.1002/zamm.19290090104.

[4] Hill, R. The Elastic Behaviour of a Crystalline Aggregate. *Proc. Phys. Soc. Sect. A* **1952**, *65*, 349, doi:10.1088/0370-1298/65/5/307.