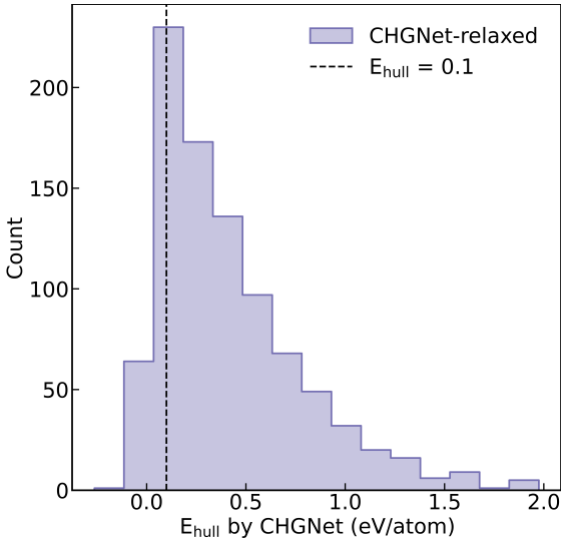
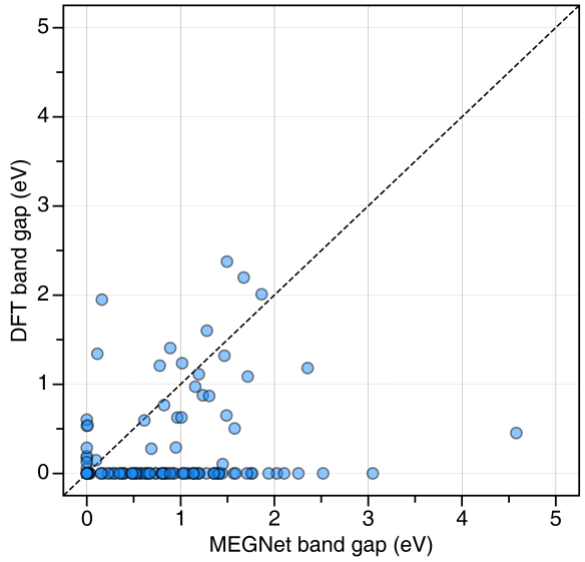
# Supplemental Information




**Supplementary Figure 1 | Distribution of energy above hull ($E_{hull}$) of 917 generated crystals.** The results are given at the CHGNet calculation level.




**Supplementary Figure 2 | Band gap comparison for generated crystal structures.** The DFT band gaps found in **Supplementary Table 1**. **Supplementary Table 2**

**Supplementary Table 1 | Symmetry, band gaps and stability of 35 semiconductors out of 135 inputs**

| ID | Formula | $N_{sites}$ | Space group | $E_{hull}$ (eV/atom)[a] | $E_g^{WyCryst+}$ (eV) | $E_g^{DFT}$ (eV) | Phonon[b] |
|---|---|---|---|---|---|---|---|
| 48 | $Zr_8Mn_4O_{24}$ | 36 | 205 (Pa-3) | **0.03** | 1.83 | 2.20 | Stable |
| 64 | $P_4Ir_4Se_{16}$ | 24 | 198 ($P2_13$) | 0.32 | 0.85 | 0.15 | Unstable |
| 80 | $K_{12}Bi_4Se_{20}$ | 36 | 198 ($P2_13$) | 0.64 | 1.39 | 0.77 | Unstable |
| 84 | $Zr_4Rh_{16}S_{20}$ | 40 | 198 ($P2_13$) | 0.27 | 0.64 | 0.05 | Unstable |
| 102 | $Cs_2V_2I_8$ | 12 | 194 ($P6_3/mmc$) | 0.09 | 0.62 | 0.17 | Unstable |
| 132 | $Cs_2Tl_6Te_{10}$ | 18 | 194 ($P6_3/mmc$) | 0.48 | 0.51 | 0.28 | Unstable |
| 202 | $K_8Mn_2S_6$ | 16 | 186 ($P6_3mc$) | 0.21 | 1.25 | 0.19 | Unstable |
| 203 | $Na_6Mn_2Se_8$ | 16 | 186 ($P6_3mc$) | **0.06** | 1.24 | 0.59 | Stable |
| 239 | $Rb_{12}Hg_4S_{10}$ | 26 | 186 ($P6_3mc$) | **0.10** | 1.89 | 1.41 | Stable |
| 251 | $K_{12}Hg_8S_{10}$ | 30 | 186 ($P6_3mc$) | 0.33 | 1.69 | 0.53 | Unstable |
| 352 | $Pt_3Pb_3F_{24}$ | 30 | 166 (R-3m) | 0.16 | 2.06 | 0.50 | Unstable |
| 378 | $Cs_6Pt_9Se_{21}$ | 36 | 166 (R-3m) | **0.06** | 1.01 | 0.13 | Stable |
| 385 | $Pt_3Pb_6F_{24}$ | 33 | 166 (R-3m) | 0.17 | 2.01 | 2.01 | Unstable |
| 556 | $Cd_6Sn_6Br_{36}$ | 48 | 161 (R3c) | 0.10 | 1.40 | 1.60 | Unstable |
| 566 | $Tl_9Sb_3O_9$ | 21 | 160 (R3m) | 0.17 | 1.63 | 1.24 | Unstable |
| 579 | $Fe_3Bi_9O_{18}$ | 30 | 160 (R3m) | 0.19 | 1.77 | 1.95 | Unstable |
| 584 | $Tl_3As_9S_{15}$ | 27 | 160 (R3m) | 0.10 | 1.29 | 1.34 | Unstable |
| 586 | $Tl_9As_9S_{12}$ | 30 | 160 (R3m) | 0.22 | 1.30 | 0.29 | Stable |
| 597 | $Y_9Ag_3S_{27}$ | 39 | 160 (R3m) | 0.09 | 0.87 | 1.09 | Unstable |
| 612 | $Ge_3Pb_9S_{36}$ | 48 | 160 (R3m) | 0.36 | 1.29 | 0.63 | Unstable |
| 669 | $Hg_6Pb_3F_{18}$ | 27 | 148 (R-3) | 0.18 | 1.20 | 2.38 | Unstable |
| 693 | $Cd_9P_6Se18$ | 33 | 148 (R-3) | 0.20 | 0.95 | 0.88 | Stable |
| 703 | $Cd_{12}Ge_{12}O_{18}$ | 42 | 148 (R-3) | 0.23 | 0.75 | 0.29 | Stable |
| 704 | $Co_9Pt_3F_{36}$ | 48 | 148 (R-3) | 0.20 | 0.97 | 0.60 | Unstable |
| 713 | $Al_6Ge_5S_{11}$ | 22 | 143 (P3) | 0.20 | 2.16 | 0.87 | Stable |
| 786 | $Rb_8V_8Br_{28}$ | 44 | 64 (Cmce) | 0.16 | 0.97 | 0.65 | Unstable |
| 851 | $K_6Au_{14}S_{12}$ | 32 | 55 (Pbam) | 0.14 | 0.97 | 0.97 | Unstable |
| 880 | $Na_4Ag_4S_8$ | 16 | 33 ($Pna2_1$) | 0.10 | 1.36 | 1.11 | Stable |
| 898 | $Cr_4Hg_4Cl_{16}$ | 24 | 19 ($P2_12_12_1$) | 0.08 | 0.73 | 1.21 | Stable |
| 905 | $Al_4Bi_8Br_{36}$ | 48 | 15 (C2/c) | 0.18 | 1.89 | 1.18 | Unstable |
| 925 | $Ba_1Br_3N_2$ | 6 | 10 (P2/m) | 0.21 | 1.08 | 0.45 | Unstable |
| 931 | $Cs_2P_3Se_8$ | 13 | 6 (Pm) | 0.12 | 1.38 | 0.10 | Unstable |
| 934 | $K_{12}Cd_4O_{10}$ | 26 | 5 (C2) | 0.10 | 1.30 | 1.32 | Stable |
| 942 | $K_1Tl_3O_4$ | 8 | 2 (P-1) | 0.09 | 0.47 | 0.54 | Stable |
| 944 | $Na_3In_3S_4$ | 10 | 2 (P-1) | 0.15 | 1.59 | 0.63 | Unstable |

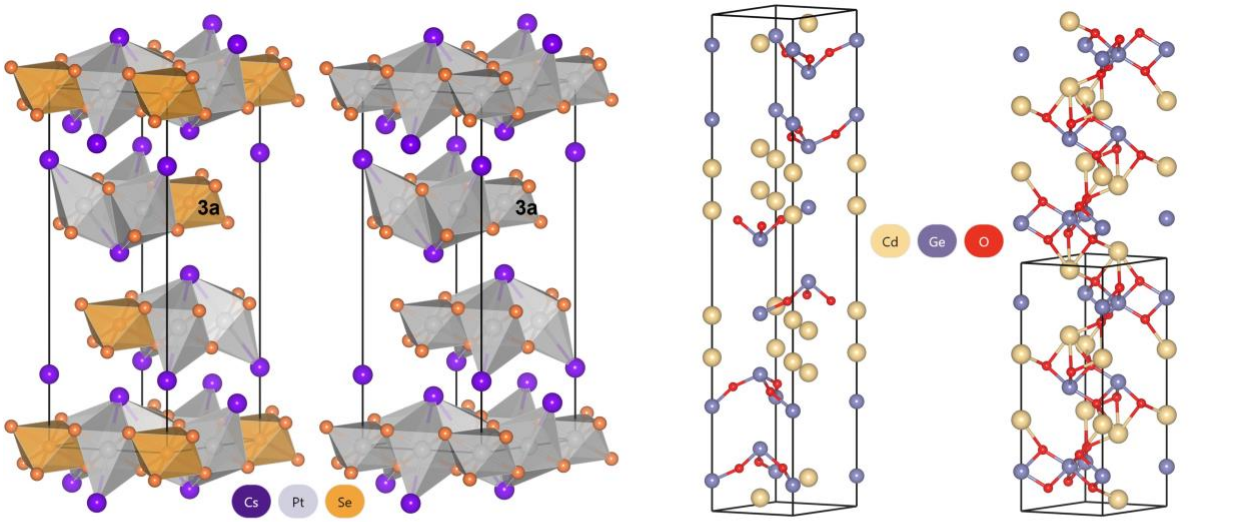
[a] DFT calculated results; Phonon stable semiconductors with $E_{hull}$ <= 0.10 eV/atom are in bold.

[b] Estimated from density functional perturbation theory (DFPT) calculations at the Γ q-point.

**Supplementary Table 2 | Similar structures in the Materials Project to the new 8 semiconductors**

| ID | Space group | Formula | $E_g$ (eV) | MP-ID[a] | Space group | Formula[b] | ICSD ID | $E_g$ (eV)[c] |
|---|---|---|---|---|---|---|---|---|
| 378 | 166 (R-3m) | $Cs_6Pt_9Se_{21}$ | 0.13 | mp-573316 | 166 (R-3m) | $Cs_6Pt_{12}Se_{18}$ | 69440 | 1.05 |
| 703 | 148 (R-3) | $Cd_{12}Ge_{12}O_{18}$ | 0.29 | mp-8275 | 148 (R-3) | $Cd_6Ge_6O_{18}$ | 30971 | 1.41 |
| 586 | 160 (R3m) | $Tl_9As_9S_{12}$ | 0.29 | mp-9791 | 160 (R3m) | $Tl_9As_3S_9$ | 100292 | 1.22 |
| 203 | 186 ($P6_3mc$) | $Na_6Mn_2Se_8$ | 0.59 | mp-14780 | 186 ($P6_3mc$) | $Na_{12}Mn_2Se_8$ | 65449 | 1.13 |
| | | | | mp-10232 | 156 (P3m1) | $NaMnSe_2$ | 50818 | 0 (1.06)[d] |
| | | | | mp-29745 | 15 (C2/c) | $Na_{16}Mn_{16}Se_{24}$ | 50820 | 0 (1.60)[e] |
| 713 | 143 (P3) | $Al_6Ge_5S_{11}$ | 0.87 | — | — | — | — | — |
| 693 | 148 (R-3) | $Cd_9P_6Se_{18}$ | 0.88 | mp-1079559 | 148 (R-3) | $Cd_6P_6Se_{18}$ | 620234 | 1.05 |
| 239 | 186 ($P6_3mc$) | $Rb_{12}Hg_4S_{10}$ | 1.41 | mp-1190842 | 186 ($P6_3mc$) | $Rb_{12}Hg_2S_8$ | 639158 | 1.60 |
| 48 | 205 (Pa-3) | $Zr_8Mn_4O_{24}$ | 2.20 | mp-754513 | 161 (R3c) | $Zr_6Mn_6O_{18}$ | — (0.04)[f] | 2.70 |
| | | | | mp-763464 | 1 (P1) | $Zr_4MnO_9$ | — (0.05)[f] | 2.66 |

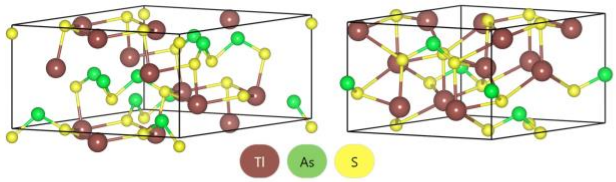
[a] The identifier (ID) in the Materials Project.
[b] The formulae are in the reduced form.
[c] The band gap is as-queried, calculated at the GGA(+*U*)-PBE level.
[d] DFT calculated from ref [1], for which the MP gives zero gaps.
[e] DFT+*U* calculated from ref [2], where another analog with reduced formula of $Na_4Mn_6Se_8$ is presented with C2/m symmetry and band gap of 1.59 eV, and the measured optical band gaps for $Na_{16}Mn_{16}Se_{24}$ and $Na_4Mn_6Se_8$ are 2.03 and 2.04 eV, respectively.
[f] The energy above the convex hull given by the Materials Project website (Accessed date: 2024 Sept 12).



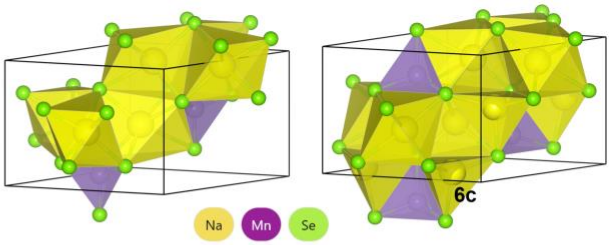

| (378) | mp-573316 |
|---|---|
| R-3m (166) | R-3m (166) |
| $Cs_6Pt_9Se_{21}$ | $Cs_6Pt_{12}Se_{18}$ |
| Cs: 6c (0, 0, 0.21)<br>Pt: 9e (1/2, 0, 0)<br>**Se1: 3a (0, 0, 0)**<br>Se2: 18h (0.49, -0.49, 0.27) | Cs: 6c (0, 0, 0.20)<br>**Pt1: 3a (0, 0, 0)**<br>Pt2: 9e (1/2, 0, 0)<br>Se: 18h (0.49, -0.49, 0.27) |

| (703) | mp-8275 |
|---|---|
| R-3 (148) | R-3 (148) |
| $Cd_{12}Ge_{12}O_{18}$ | $Cd_6Ge_6O_{18}$ |
| Cd1: 6c (0, 0, 0.31)<br>**Cd2: 6c (0, 0, 0.39)**<br>Ge1: 6c (0, 0, 0.06)<br>**Ge2: 6c (0, 0, 0.21)**<br>O: 18f (0.32, 0.27, 0.25) | Cd: 6c (0, 0, 0.37)<br>Ge: 6c (0, 0, 0.16)<br>O: 18f (0.34, 0.06, 0.10) |

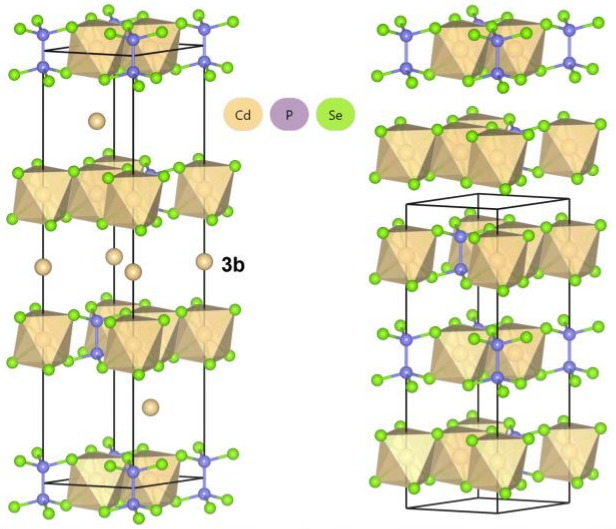
**Supplementary Figure 3 | Structure comparison between WyCryst+-designed structures and those analogs with the same space group from the Materials Project.** Sequentially, the Wyckoff positions involve (1) substitution/replacement, (2) addition, (3) swap and addition, (4) deletion, (5) addition, (6) addition, respectively.



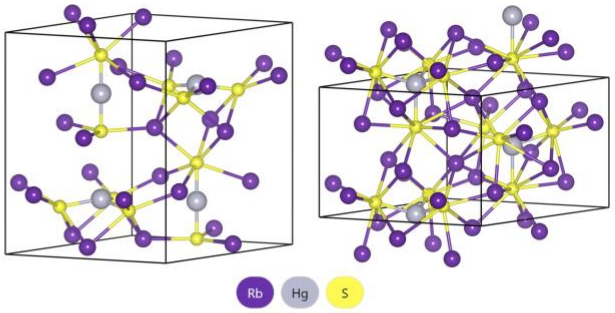

| (586) | mp-9791 |
|---|---|
| R3m (160) | R3m (160) |
| $Tl_9As_9S_{12}$ | $Tl_9As_3S_9$ |
| Tl: 9b (0.19, -0.19, 0.24)<br>**As: 9b (0.91, -0.91, 0.13)**<br>S1: 9b (0.82, -0.82, 0.97)<br>**S2: 3a (0, 0, 0.92)** | Tl: 9b (0.80, -0.80, 0.21)<br>As: 3a (0, 0, 0.44)<br>S: 9b (0.12, -0.12, 0.30) |




| (203) | mp-14780 |
|---|---|
| $P6_3mc$ (186) | $P6_3mc$ (186) |
| $Na_6Mn_2Se_8$ | $Na_{12}Mn_2Se_8$ |
| Na: 6c (0.48, -0.48, 0.95)<br>Mn: 2b (1/3, 2/3, 0.57)<br>Se1: 6c (0.20, 0.20, 0.67)<br>Se2: 2b (1/3, 2/3, 0.24) | Na1: 6c (0.47, -0.47, 0.87)<br>**Na2: 6c (0.15, -0.15, 0.54)**<br>Mn: 2b (1/3, 2/3, 0.25)<br>Se1: 6c (0.19, 0.19, 0.14)<br>Se2: 2b (1/3, 2/3, 0.60) |

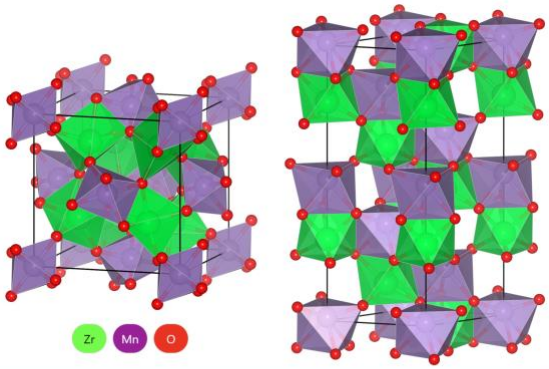



| (693) | mp-1079559 |
|---|---|
| R-3 (148) | R-3 (148) |
| $Cd_9P_6Se_{18}$ | $Cd_6P_6Se_{18}$ |
| Cd1: 6c (0, 0, 0.33)<br>**Cd2: 3b (0, 0, 1/2)**<br>P: 6c (0, 0, 0.04)<br>Se: 18f (0.67, 0.68, 0.06) | Cd: 6c (0, 0, 0.17)<br>P: 6c (0, 0, 0.45)<br>Se: 18f (0.34, 0.99, 0.25) |




| (239) | mp-1190842 |
|---|---|
| $P6_3mc$ (186) | $P6_3mc$ (186) |
| $Rb_{12}Hg_4S_{10}$ | $Rb_{12}Hg_2S_8$ |
| Rb1: 6c (0.48, -0.48, 0.55)<br>Rb2: 6c (0.15, -0.15, 0.29)<br>Hg1: 2b (1/3, 2/3, 0.21)<br>**Hg2: 3b (1/3, 2/3, 0.73)**<br>S1: 2b (1/3, 2/3, 0.04)<br>**S2: 2b (1/3, 2/3, 0.38)**<br>S3: 6c (0.19, -0.19, 0.71) | Rb1: 6c (0.47, -0.47, 0.88)<br>Rb2: 6c (0.15, -0.15, 0.21)<br>Hg: 2b (1/3, 2/3, 0.50)<br>S1: 2b (1/3, 2/3, 0.17)<br>S2: 6c (0.19, -0.19, 0.60) |

**Supplementary Figure 3** (Continued)




| (48) | mp-754513 |
|---|---|
| Pa-3 (205) | R3c (161) |
| $Zr_8Mn_4O_{24}$ | $Zr_6Mn_6O_{18}$ |
| Zr: 8c (0.21, 0.21, 0.21)<br>Mn: 4c (0, 0, 0)<br>O: 24d (0.06, 0.64, 0.31) | Zr: 6a (0, 0, 0.30)<br>Mn: 6a (0, 0, 0.01)<br>O: 18b (0.02, 0.38, 0.06) |

**Supplementary Figure 4 | Structure comparison between the WyCryst+-designed structure and the analog with the different space group from the Materials Project.** The coordination environment of Zr in the two (Zr-O polyhedra) are not the same. The former consists of distorted [$ZrO_6$] trigonal prisms while the latter distorted [$ZrO_6$] octahedra.

## References

1. Ghermoul, N. et al. Ab initio prediction of half-metallicity in the NaMnZ2 (Z = S, Se, Te) ternary layered compounds. Comput. Condens. Matter 33, e00754 (2022).

2. Pak, C. Low Dimensional Electron Correlated Materials. Florida State University (2018); http://purl.flvc.org/fsu/fd/2018_Su_Pak_fsu_0071E_14679.

# Source data

Source data for Figure 4