Designation of Intra-layer and Intercalated High Entropy Quasi-2D Compounds
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Abstract: Here, we designed two promising schemes to realize the high-entropy structure in a series of quasi-2D compounds, transition metal dichalcogenides (TMDCs). In the intra-layer high-entropy plan, (HEM)X2 compounds with high-entropy structure in the MX2 slabs were obtained, here HEM means high-entropy metals. On the other hand, in the intercalation plan, we intercalated HEM into the inter-layer vacancies in MX2 to form (HEM)X2 compounds, x in the range of 0.2~1, in which HEM is mainly composed of 3d transition metal elements, such as FeCoCrNiMn. Meanwhile, millimetre-sized single crystals were obtained by chemical vapor transport method. For physical properties, superconductivity and magnetic ordering were detected in (HEM)X2 and (HEM)mMX2, respectively. Our work provides a new way to design low dimensional high-entropy compounds, especially, the intercalation method which can realize a high-entropy two-dimensional magnetic triangular lattice. The high-entropy TMDCs found by us possess great potentials to find new physics in low-dimensional high-entropy structures and future applications.

Keywords: high-entropy, TMDCs, quasi-2D, low dimensional materials, superconductor, magnetic order

Introduction

With the flourish of two-dimensional started with the discovery of graphene[1–4], quasi-two-dimensional compounds[5,6] which can be easily exfoliated to single or few-layered have been paid lots of attention, especially transition metal dichalcogenides (TMDCs). TMDCs are a big family of compounds with a chemical formula MX2, in which M=transition metals (Ti, V, Nb, Ta, Mo, Hf, and so on), X=chalcogen elements (S, Se, Te). TMDCs have not only versatile chemical compositions but also various polymorphs[7], such as 1T, 2H, 3R, and so on. Various famous materials with exotic physical properties (extreme magnetoresistance[8], superconductivity, type-II Weyl fermions[9–11], quantum spin Hall effect[12], and so on) and excellent application prospects (electro- or photo- catalysis, photo-electronics[13][14], energy storage, pollution reduction[15] and so on) were found in TMDCs. These compounds show excellent structural compatibility which can be doped in both M and X sites[16]. Meanwhile, transition metal metals,
alkali metal metals, alkali earth metal atoms, rare-earth metal atoms, organic molecules can be intercalated into the vacancies between two X-atoms layers from two MX₂ slabs to form intercalated compounds[17–19] such as Cu₂TiSe₂[20], (NH₃)₂LiNbSe₂ and so on.

Recently, high-entropy structuralize were found to be an effective way to searching new materials. The idea of high-entropy alloy was proposed by Ye [21,22]. At present, more than 400 kinds of high-entropy alloys were found. Since 2015, first high-entropy oxide (MgCoNiCuZn)O was found by M. Rost et al. [23], more attention was paid to high-entropy compounds. At present, the family of high-entropy compounds have been expanded to borides [24] oxides [23,25,26], carbides[27,28], sulfides[29], silicide[30] and so on. Some of them show excellent properties surpassing those of the constituent materials[31], such as colossal dielectric constant [32], ultra-low thermal conductivity [29,33], enhanced mechanical [34], energy-storage ability [35] and so on. On the other hand, how the high-entropy effect on superconductivity [36–42] and magnetic order [25,26,28,43–45] are quite concerned in condensed matter physics. Although so many high-entropy compounds have been found, the high-entropy compounds in low-dimensional are still rare.

Considering the versatile chemical composition and structure compatibility, TMDCs can be an excellent platform to realize the high-entropy two-dimension (2D) or quasi-2D material. In this work, we found two feasible ways to realized high-entropy structure in TMDCs. One way is the high-entropy structuralizing at the M site, and the other is the intercalation of high-entropy alloy into the intra-layer vacancies in MX₂. Meanwhile, intercalating various 3d transition metal elements may introduce high structural and magnetic entropy simultaneously in the intercalated layer. Taken possible magnetic frustration in triangular lattice into consideration, the magnetism in the 2D magnetic layer will be very interesting and complicated. In this work, various high-entropy TMDCs were found by us. Meanwhile, millimeter-sized single crystals of high-entropy TMDCs were grown by a chemical vapor transport method. Crystal structure, electrical and magnetic properties of the high-entropy quasi-2D compounds were characterized. Surprisingly, superconductivity with a Tc=7.4 K was observed in a Mo-rich high-entropy compound, and magnetic ordering with Currie temperatures in the range of 11 K ~ 30 K was detected. The two high-entropy schemes we proposed can also be applicable for other quasi-2D of quasi-1D systems. More advanced characterizations are called to reveal the new physics and potential applications in high-entropy quasi-2D compounds.

Materials and methods

Preparation of (HEM)S₂ powder: The high-entropy alloys were produced by the arc-melting method. Then we crushed the high-entropy alloys into powders in a glove box as protected by Ar-gas atmosphere. Then high-entropy alloy powder (TiZrNbMoTa, TiVNbMoTa, or TiVHfNbTa) was carefully mixed with the X=S, Se, Te powders. Then load the mixed powder into a silica tube and then sealed the tube under high vacuum. At last, the sample was heated to 1073-1273 K in 300 minutes and keep sintering for more than 2 days.

Preparation of (HEM),MX₂ powder: First, we synthesized the MX₂ powders by solid-state sintering of mixed M element and X element powders. The high-entropy alloys (FeCoCrNiMn, FeCoCrNi, or
FeCoCrNiAl) were carefully grounded with as-prepared MX₂ (TiS₂, TiSe₂, NbS₂) powder, and then sealed the mixed powder into a silica tube under high vacuum.

Single crystal growth: The as-prepared powders by solid-state sintering with 150 mg iodine as transport agent were sealed in a silica tube under high vacuum. The silica tube was heated in a two-zone furnace in which the ingredients were placed at the hot end (1273 K - 1373 K). Keep sintering for 7-15 days, and the high-entropy crystals were obtained at the cool end. The temperature gradient was set as 100-150 K.

Characterization: X-ray diffractions were applied on a D8 Advance (Bruker Germany). Elemental compositions were characterized by the energy-dispersive X-ray spectrum (EDS, Oxford) at Field-emission scanning electron microscopy (FESEM, FEI, NANOSEM-450). Transport properties and magnetism were characterized by a Physical Properties Measurements System 16 T (Quantum Design, U. S.).

Results

1 T and 2 H phases are the most commonly seen polymorphs in TMDCs. Here we take 1T phase as an example to illustrate the two schemes to realize high-entropy structures in TMDCs. In the single MX₂ slab, one layer of M atoms was sandwiched by two X-atom layers, and one M atoms are octahedrally coordinated with six X-atoms. MX₂ slabs are mainly van-der-Waals connected with each other; as a result, they are easy to be exfoliated. As shown in Figure 1, if we substitute M elements with HEM, we will get (HEM)X₂ which owns a high-entropy structure inside the MX₂ slabs, as shown in Figure 1(c). In the intra-layer case, HEM composed of IVB-, VB-, and VIB- group elements and some precious metals are preferred which can form MX₂ compounds independently. Here we use equiatomic high-entropy alloys (HEA) as ingredients such as TiZrNbMoTa, TiVNbMoTa, and TiHfNbTaV.

In the other high-entropy scheme, we take advantage of the vacancies between MX₂ slabs. In 1T phase, it has large octahedral vacancies formed by six X-atoms between two X layers from different slabs. As previous works reported, V, Fe, Co, Cr, Ni, Mn can be intercalated into the inter-layer vacancies, form M'MX₂, M’=3d transition metals. The introduction of 3d elements will bring rich magnetic properties, such as ferromagnetism and spin-glass. Here we think about an extreme situation. When several kinds of 3d metals were intercalated into the inter-layer vacancies simultaneously, they may all contribute to the magnetism and have different magnetic moments. Consequently, it may form a 2D layer with both high structural and magnetic entropy. At present, even co-intercalation of two kinds of 3d elements in TMDCs has never been reported. Therefore we designed an intercalated high-entropy structure (HEM)MX₂, in which the HEM means the 3d transition metals. Here we use HEA to react with MX₂ at high temperatures, and surprisingly five kinds of 3d elements (FeCoCrNiMn) were successfully intercalated simultaneously. We also tried to use mechanically mixed powder of five elements as ingredients, while the resulting products are multi-phased.

Chemical vapor transport (CVT) method is an effective method to grow the single crystals of MX₂ compounds. We found CVT is also effective for the single-crystal growth of (HEM)X₂ and (HEM)MX₂. Iodine
was used as transport agent in this work. As shown in Figure 2, the photos of several single crystals were presented. Most of the as-grown single crystals are hexagonal or triangular thin plates in millimeter-size.

It should be noted that the formulas are the nominal composition in this article if not specified. Figure 3 shows the chemical composition analysis of (HEM)2X and (HEM)MX2 single crystals. Mapping scan of each element in (Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)0.333TiS2 were given in Figure 3(a). No obvious inhomogeneous were observed, and the as-determined composition is (Fe0.20Co0.21Cr0.21Ni0.19Mn0.19)0.333TiS2 which is almost equiatomic. When x=0.5, the as-determined chemical composition is (Fe0.15Co0.15Cr0.20Ni0.10Mn0.10)TiS2, in which the atom ratio of Fe Co Cr Ni are almost equal, while as-determined content of Mn is ~30 % less than the nominal composition. Similar cases was also observed in other (HEM)MX2 such as the (Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)0.5NbS2 with a chemical composition of (Fe0.14Co0.10Cr0.21Ni0.084Mn0.067)NbS2, as shown in Figure 3(d).

Both equiatomic and non-equiatomic (HEM)2X were found. For (Ti0.2V0.2Hf0.2Nb0.2Ta0.2)Se2, the composition of HEM are almost equiatomic, as (Ti0.2V0.21Nb0.19Mo0.18Ta0.22)Se2. And a mapping EDX analysis of a single crystal with in chemical composition of (Ti0.38Zr0.08Nb0.22Mo0.10Ta0.26)Se2 was shown in Figure 3(b). Meanwhile, other non-equiatomic (HEM)X2 compounds, such as a Mo-rich (HEM)X2 compounds: (Mo0.76Nb0.15Ta0.05)Se2, and (Mo0.65Nb0.22Ta0.10V0.03)Se2 were found. All (HEM)MX2 and (HEM)X2 compounds we obtained and related properties of them were listed in Table S1. And more information about the EDX results was presented in Supporting Information.

Using X-ray diffractions, we studied the crystal structure evolution with the intercalated content x. The XRD patterns of (HEM)2TiS2 and (HEM)4TiSe2 were shown in Figure 4. As-prepared TiS2 and TiSe2 are both 1 T phase (space group: P-3m1) well in line with PDF#15-0853 and PDF#30-1383, respectively. In the case of low intercalated content x=0.2, (Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)0.2TiS2, Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)0.5TiSe2, (Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)0.5TiSe2, the intercalation will remain the crystal structure of matrix. Here we focus on the case in (Fe0.2Co0.2Cr0.2Ni0.2Mn0.2)TiSe2. As shown in Figure 4(g) and (e), all peaks can be well indexed by the space group P-3m1, the (002) peak move to higher angles compared with TiSe2. When x=0.5, it cannot be well indexed by P-3m1 but reduced into a space group P321 with no obvious change in the size of lattice. When x content was increased above 0.6, the lattice distortion will be strong enough to make the (HEM)4TiSe2 change into orthorhombic with an expanded lattice of 1a × √3b × 2c compared with the samples with low x content; similar phenomenon was also reported in other works about the 3d transition metal intercalation such as Fe3TiSe2[46–48]. Lattice parameters of samples with x in the range of 0.2–0.6 were refined by Jade, as shown in Figure 4(i). With the increasing of intercalation content (x<0.5), the lattice parameter a increased and c decreased. The decreased c value should be assigned to the enhanced static-force connection between MX2 slabs.

Transport properties and magnetism of several (HEM)X2 single crystals were characterized. And surprisingly we found superconductivity in a Mo-rich (HEM)Se2 compound in a chemical composition (Mo0.76Nb0.15Ta0.09)Se2. The residual resistance ratio RRR=\( R_{300K}/R_{7.5K} \approx 7.2 \), which indicates the high
crystallinity of as-grown single crystal. As shown in Figure. 5(a) and (b), under zero field, the onset critical transition temperature is about 7.4 K which is even slightly higher than the value 7.2 K in NbSe$_2$. The superconducting transition finished at 7.06 K. As shown in Figure 4(c), the upper critical field $H_{c2}$ is about 2750 Oe. It should be noted that MoSe$_2$ is a semiconductor with an indirect bandgap of 1.08 eV[49]. To our best knowledge, no superconductor has ever been reported in Mo-site doped MoSe$_2$. In contrast, no superconductivity was observed in the other Mo-rich (HEM)Se$_2$ single crystal (Mo$_{0.66}$Nb$_{0.22}$Ta$_{0.10}$V$_{0.03}$)Se$_2$. It shows metallic behavior when $T>17$ K. However, below 17 K, an Anderson-localized R-T behavior was observed, which might be caused by the high-entropy structure.

The magnetism of (HEM)$_x$TiS$_2$ and (HEM)$_x$TiSe$_2$ with $x=0.2$ were shown in Figure 6. As shown in Figure 6 (a), (c), and (e), obvious divergence was observed in the zero-field cooling (ZFC) and field cooling (FC) curves under 1000 Oe. As shown in Figure 6 (g), a remanent magnetization moment was observed in the magnetization curve at 2 K of (Fe$_{0.2}$Co$_{0.2}$Cr$_{0.2}$Ni$_{0.2}$Al$_{0.2}$)$_{0.2}$TiSe$_2$. It suggests the ferromagnetism in (HEM)$_{0.2}$TiX$_2$. The Currie temperatures were determined by minimum of $dM/dT$ as shown in Figure 6 (b), (d), and (f). The magnetism of (HEM)$_x$TiX$_2$ with higher $x$ content were given in Supporting Information. And the highest measured Currie temperature is up to 30 K.

Discussion and Conclusion

In conclusion, we proposed two effective ways to realize the 2D high-entropy structure in TMDCs, which can also be applied in other quasi-2D or quasi-1D compounds. Various (HEM)X$_2$ with intra-layer high-entropy structure and (HEM)$_x$MX$_2$ with a high-entropy layer between MX$_2$ slabs were obtained here. Meanwhile, both samples with equiatomic and non-equatomic compositions of HEM were found. Using high-entropy alloys as ingredients will be helpful to avoid the formation of multi-phases, especially for (HEM)$_x$MX$_2$. Various large-size high-entropy single crystals were grown by the CVT method. The layered structure remained with the introduction of a high-entropy structure so that the as-grown single crystals can be easily exfoliated, which provides the opportunities to realized monolayered of few-layered high-entropy materials. We also reveal that high-entropy structuralizing could be a new way to manipulate the superconductivity or novel magnetic order in TMDCs. Superconductivity with a $T_c$ up to 7.4 K was observed in a Mo-rich (HEM)X$_2$. Magnetic ordering in the high-entropy magnetic layer with various 3d transition metal elements with Curie temperatures in the range of 11 K ~ 30 K were detected in (HEM)$_x$MX$_2$. Meanwhile, the structure evolution of $x$ content reveals the possible superlattice formed in the 2D high-entropy atom layer. Our work will evoke more enthusiasm about exploring high-entropy compounds and the high-entropy effects in low-dimensional systems.

Figures and Figure Captions
Figure 1. Two schemes to realize high-entropy structures in MX$_2$. (a) Crystal structure of MX$_2$ viewed from the $\alpha$ axis, here we take 1T phase as an example. (b) High-entropy metal elements. (c) Crystal structure of (HEM)X$_2$. (d) Crystal structure of (HEM)$_3$MX$_2$.

Figure 2. The photo images of several single crystals of (HEM)$_2$X$_2$, and (HEM)$_x$TiSe$_2$. 
Figure 3. Energy dispersion X-ray spectroscopy analysis of several (HEM)X₂ and (HEM)MX₂ compounds. Mapping analysis of (a) (Fe₀.2Co₀.2Cr₀.2Ni₀.2Mn₀.2)₀.3TiS₂ and (b) (Ti₀.2Nb₀.2Mo₀.2Ta₀.2V₀.2)S₂; Point-scan analysis of (c) (Ti₀.2V₀.2Nb₀.2Ta₀.2Hf₀.2)S₂, (d) (Fe₀.2Co₀.2Cr₀.2Ni₀.2Mn₀.2)₀.5NbS₂.
Figure 4. The X-ray diffraction analysis of (HEM)$_x$MX$_2$. The red stars indicate the featured peaks as caused by lattice distortions.

Fig. 5 Transport properties. (a) Temperature dependent resistance of Mo$_{0.76}$Nb$_{0.15}$Ta$_{0.09}$Se$_2$. (b) Field-dependent $R$-$T$ in the range of 1-10 K of Mo$_{0.76}$Nb$_{0.15}$Ta$_{0.09}$Se$_2$. (c) Field dependent $H_{c2}$. (d) $R$-$T$ of Mo$_{0.65}$Nb$_{0.22}$Ta$_{0.10}$V$_{0.03}$Se$_2$. 
Figure 6. The magnetic properties of (HEM)$_{x}$TiX$_2$. (a), (c), and (e) are ZFC and FC curves measured at 1000 Oe of (Fe$_{0.2}$Co$_{0.2}$Cr$_{0.2}$Ni$_{0.2}$Al$_{0.2}$)$_{0.2}$TiSe$_2$, (Fe$_{0.2}$Co$_{0.2}$Cr$_{0.2}$Ni$_{0.2}$Al$_{0.2}$)$_{0.2}$TiS$_2$, and (Fe$_{0.2}$Co$_{0.2}$Cr$_{0.2}$Ni$_{0.2}$Mn$_{0.2}$)$_{0.2}$TiS$_2$, respectively. (b), (d), (f) are the corresponding $dM/dT$ analysis. (g) Field-dependent magnetization of (Fe$_{0.2}$Co$_{0.2}$Cr$_{0.2}$Ni$_{0.2}$Al$_{0.2}$)$_{0.2}$TiSe$_2$ in 0-7 Tesla.

Conflict of interest
The authors declare no interest conflicts in this work.

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References
[1] Geim AK, Novoselov KS. The rise of graphene. Nat Mater 2007;6. https://doi.org/10.1038/nmat1849.

[2] Novoselov KS, Geim AK, Morozov S V, Jiang D, Zhang Y, Dubonos S V., et al. Electric Field Effect in Atomically Thin Carbon Films Supplementary. Science (80-) 2004;5. https://doi.org/10.1126/science.aab1343.

[3] Novoselov KS, Jiang D, Schedin F, Booth TJ, Khotkevich V V., Morozov S V., et al. Two-
dimensional atomic crystals. Proc Natl Acad Sci U S A 2005;102. https://doi.org/10.1073/pnas.0502848102.

[4] Novoselov KS, Geim AK, Morozov S V., Jiang D, Katsnelson MI, Grigorieva I V., et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 2005;438. https://doi.org/10.1038/nature04233.

[5] Chhowalla M, Shin HS, Eda G, Li LJ, Loh KP, Zhang H. The chemistry of two-dimensional layered transition metal dichalcogenide nanosheets. Nat Chem 2013;5. https://doi.org/10.1038/nchem.1589.

[6] Wilson JA, Di Salvo FJ, Mahajan S. Charge-density waves and superlattices in the metallic layered transition metal dichalcogenides. Adv Phys 1975;24. https://doi.org/10.1080/00018737500101391.

[7] Wilson JA, Yoffe AD. The transition metal dichalcogenides discussion and interpretation of the observed optical, electrical and structural properties. Adv Phys 1969;18. https://doi.org/10.1080/00018736900101307.

[8] Ali MN, Xiong J, Flynn S, Tao J, Gibson QD, Schoop LM, et al. Large, non-saturating magnetoresistance in WTe2. Nature 2014. https://doi.org/10.1038/nature13763.

[9] Tamai A, Wu QS, Cucchi I, Bruno FY, Ricco S, Kim TK, et al. Fermi arcs and their topological character in the candidate type-II Weyl semimetal MoTe2. Phys Rev X 2016;6. https://doi.org/10.1103/PhysRevX.6.031021.

[10] Li P, Wen Y, He X, Zhang Q, Xia C, Yu ZM, et al. Evidence for topological type-II Weyl semimetal WTe2. Nat Commun 2017;8. https://doi.org/10.1038/s41467-017-02237-1.

[11] Soluyanov AA, Gresch D, Wang Z, Wu Q, Troyer M, Dai X, et al. Type-II Weyl semimetals. Nature 2015;527:495–8. https://doi.org/10.1038/nature15768.

[12] Ye M, Zhang D, Yap YK. Recent advances in electronic and optoelectronic devices based on two-dimensional transition metal dichalcogenides. Electron 2017;6. https://doi.org/10.3390/electronics6020043.

[13] Wang QH, Kalantar-Zadeh K, Kis A, Coleman JN, Strano MS. Electronics and optoelectronics of two-dimensional transition metal dichalcogenides. Nat Nanotechnol 2012;7. https://doi.org/10.1038/nnano.2012.193.

[14] Zhang X, Teng SY, Loy ACM, How BS, Leong WD, Tao X. Transition metal dichalcogenides for the application of pollution reduction: A review. Nanomaterials 2020;10. https://doi.org/10.3390/nano10061012.

[15] Tedstone AA, Lewis DJ, O'Brien P. Synthesis, Properties, and Applications of Transition Metal-Doped Layered Transition Metal Dichalcogenides. Chem Mater 2016;28. https://doi.org/10.1021/acs.chemmater.6b00430.

[16] Titov AN, Bryntse I, Titova SG, Toporova N V. layer compounds 2001:293–300.

[17] Dresselhaus MS. Intercalation In Layered Materials. vol. 12. 1987. https://doi.org/10.1557/S0883769400068093.

[18] Jacobson AJ, Nazar LF. Intercalation Chemistry. 2006. https://doi.org/10.1002/0470862106.ia098.

[19] Morosan E, Zandbergen HW, Dennis BS, Bos JWG, Onose Y, Klimczuk T, et al. Superconductivity in Cu x TiSe 2. Nat Phys 2006;2:544–50. https://doi.org/10.1038/nphys360.

[20] Rost CM, Sachet E, Borman T, Moballegh A, Dickey EC, Hou D, et al. Entropy-stabilized oxides. Nat Commun 2015;6. https://doi.org/10.1038/ncomms9485.

[21] Gild J, Zhang Y, Harrington T, Jiang S, Hu T, Quinn MC, et al. High-Entropy Metal Diborides: A New Class of High-Entropy Materials and a New Type of Ultrahigh Temperature Ceramics. Sci Rep 2016;6:2.11. https://doi.org/10.1038/srep37946.

[22] Witte R, Sarkar A, Kruk R, Eggert B, Brand RA, Wende H, et al. High-entropy oxides: An emerging
prospect for magnetic rare-earth transition metal perovskites. Phys Rev Mater 2019;3:1–9.
https://doi.org/10.1103/PhysRevMaterials.3.034406.

[26] Mao A, Xiang HZ, Zhang ZG, Kuramoto K, Zhang H, Jia Y. A new class of spinel high-entropy oxides with controllable magnetic properties. J Magn Magn Mater 2020;497:165884.
https://doi.org/10.1016/j.jmmm.2019.165884.

[27] Zhou J, Zhang J, Zhang F, Niu B, Lei L, Wang W. High-entropy carbide: A novel class of multicomponent ceramics. Ceram Int 2018;44:22014–8.
https://doi.org/10.1016/j.ceramint.2018.08.100.

[28] Li Y, Lu J, Li M, Chang K, Zha X, Zhang Y, et al. Multielemental single–atom-thick A layers in nanolaminated V2(Sn, A) C (A = Fe, Co, Ni, Mn) for tailoring magnetic properties. Proc Natl Acad Sci U S A 2020;117:820–5. https://doi.org/10.1073/pnas.1916256117.

[29] Zhang RZ, Gucci F, Zhu H, Chen K, Reece MJ. Data-Driven Design of Ecofriendly Thermoelectric High-Entropy Sulfides. Inorg Chem 2018;57:13027–33.
https://doi.org/10.1021/acs.inorgchem.8b02379.

[30] Gild J, Braun J, Kaufmann K, Marin E, Harrington T, Hopkins P, et al. A high-entropy silicide: (Mo0.2Nb0.2Ta0.2Ti0.2)Si2. J Mater 2019;5:337–43.
https://doi.org/10.1016/j.jmat.2019.03.002.

[31] Oses C, Toher C, Curtarolo S. High-entropy ceramics. Nat Rev Mater 2020.
https://doi.org/10.1038/s41578-019-0170-8.

[32] Bérandan D, Franger S, Dragoe D, Meena AK, Dragoe N. Colossal dielectric constant in high entropy oxides. Phys Status Solidi - Rapid Res Lett 2016;10:328–33.
https://doi.org/10.1002/pssr.201600043.

[33] Yan X, Constantin L, Lu Y, Silvain JF, Nastasi M, Cui B. (Hf0.2Zr0.2Ta0.2Nb0.2Ti0.2)O high-entropy ceramics with low thermal conductivity. J Am Ceram Soc 2018;101:4486–91.
https://doi.org/10.1111/jace.15779.

[34] Yeh JW. Recent progress in high-entropy alloys. Ann Chim Sci Des Mater 2006.
https://doi.org/10.3166/acsm.31.633-648.

[35] Sarkar A, Velasco L, Wang D, Wang Q, Talasila G, de Biasi L, et al. High entropy oxides for reversible energy storage. Nat Commun 2018;9:5. https://doi.org/10.1038/s41467-018-05774-5.

[36] Von Rohr FO, Cava RJ. Isoelectronic substitutions and aluminium alloying in the Ta-Nb-Hf-Zr-Ti high-entropy alloy superconductor. Phys Rev Mater 2018;2:1–15.
https://doi.org/10.1103/PhysRevMaterials.2.034801.

[37] Guo J, Wang H, Von Rohr F, Wang Z, Cai S, Zhou Y, et al. Robust zero resistance in a superconducting high-entropy alloy at pressures up to 190 GPa. Proc Natl Acad Sci U S A 2017;114:13144–7.
https://doi.org/10.1073/pnas.1716981114.

[38] Koželj P, Vrtnik S, Jelen A, Jazbec S, Jagličič Z, Maiti S, et al. Discovery of a superconducting high-entropy alloy. Phys Rev Lett 2014;113:1–5.
https://doi.org/10.1103/PhysRevLett.113.107001.

[39] Stolze K, Cevallos FA, Kong T, Cava RJ. High-entropy alloy superconductors on an α-Mn lattice. J Mater Chem C 2018;6:10441–9.
https://doi.org/10.1039/c8tc03337d.

[40] Ye YF, Wang Q, Lu J, Liu CT, Yang Y. High-entropy alloy: challenges and prospects. Mater Today 2016.
https://doi.org/10.1016/j.mattod.2015.11.026.

[41] Sogabe R, Goto Y, Mizuguchi Y. Superconductivity in REO0.5F0.5BiS2 with high-entropy-alloy-type blocking layers. Appl Phys Express 2018;11. https://doi.org/10.7567/APEX.11.053102.

[42] Sun L, Cava RJ. High-entropy alloy superconductors: Status, opportunities, and challenges. Phys Rev Mater 2019;3:1–33.
https://doi.org/10.1103/PhysRevMaterials.3.095301.

[43] Vinnik DA, Trofimov EA, Zhvulin VE, Zaitseva O V., Gudkova SA, Starikov AY, et al. High-entropy oxide phases with magnetoplumbite structure. Ceram Int 2019;45:12942–8.
https://doi.org/10.1016/j.ceramint.2019.03.221.

[44] Mao A, Xiang HZ, Zhang ZG, Kuramoto K, Zhang H, Jia Y. A new class of spinel high-entropy oxides with Key Laboratory of Metallurgical Emission Reduction & Resources Recycling. J Magn Magn Mater 2019:165884.
https://doi.org/10.1016/j.jmmm.2019.165884.

[45] Mao A, Xiang HZ, Zhang ZG, Kuramoto K, Yu H, Ran S. Solution combustion synthesis and magnetic property of rock-salt (Co 0.2 Cu 0.2 Mg 0.2 Ni 0.2 Zn 0.2 )O high-entropy oxide nanocrystalline powder. J Magn Magn Mater 2019;484:245–52.
https://doi.org/10.1016/j.jmmm.2019.04.023.

[46] Tazuke Y, Miyashita T, Nakano H, Sasaki R. Magnetic properties of MxTiSe2 (M=Mn, Fe, Co).
Phys Status Solidi Curr Top Solid State Phys 2006;3:2787–90. https://doi.org/10.1002/pssc.200669523.

[47] Lyding JW, Ratajack MT, Kannewurf CR, Goodman WH, Ibers JA, Marsh RE. Structure, electrical transport, and optical properties of a new ordered iron intercalated dichalcogenide, Fe0.34TiSe2. J Phys Chem Solids 1982;43. https://doi.org/10.1016/0022-3697(82)90049-X.

[48] Shintomi M, Tazuke Y, Takahashi H. Structural and magnetic properties of FexTiSe2 intercalation compounds. Mol Cryst Liq Cryst Sci Technol Sect A Mol Cryst Liq Cryst 2000;341:27–32. https://doi.org/10.1080/10587250008026112.

[49] Kam KK, Parkinson BA. Detailed photocurrent spectroscopy of the semiconducting group VIB transition metal dichalcogenides. J Phys Chem 1982;86:463–7. https://doi.org/10.1021/j100393a010.