nature reviews materials
https://doi.org/10.1038/s41578-024-00720-y

**Supplementary information**

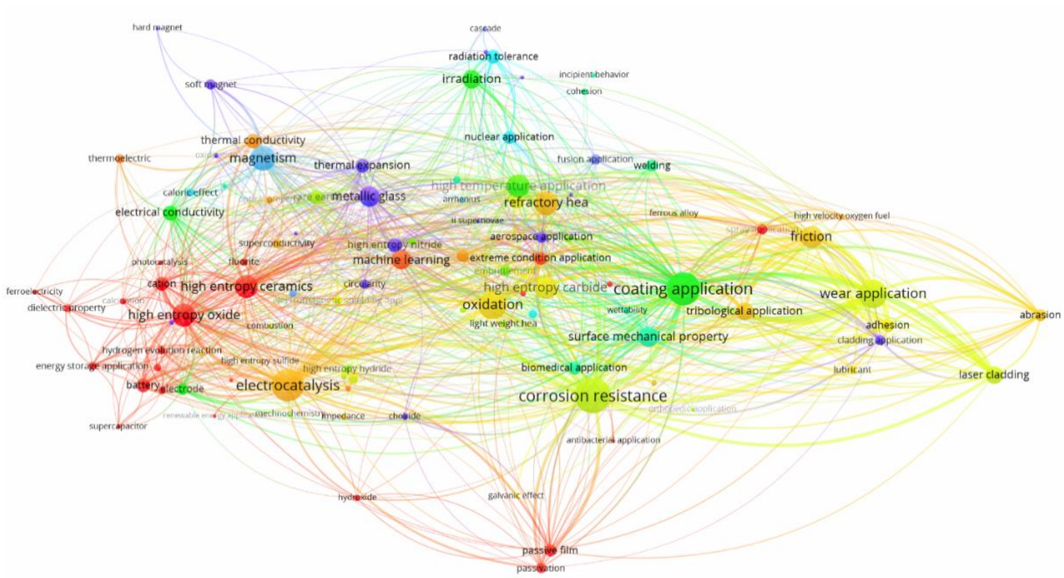
# Multifunctional high-entropy materials

**In the format provided by the authors and unedited**

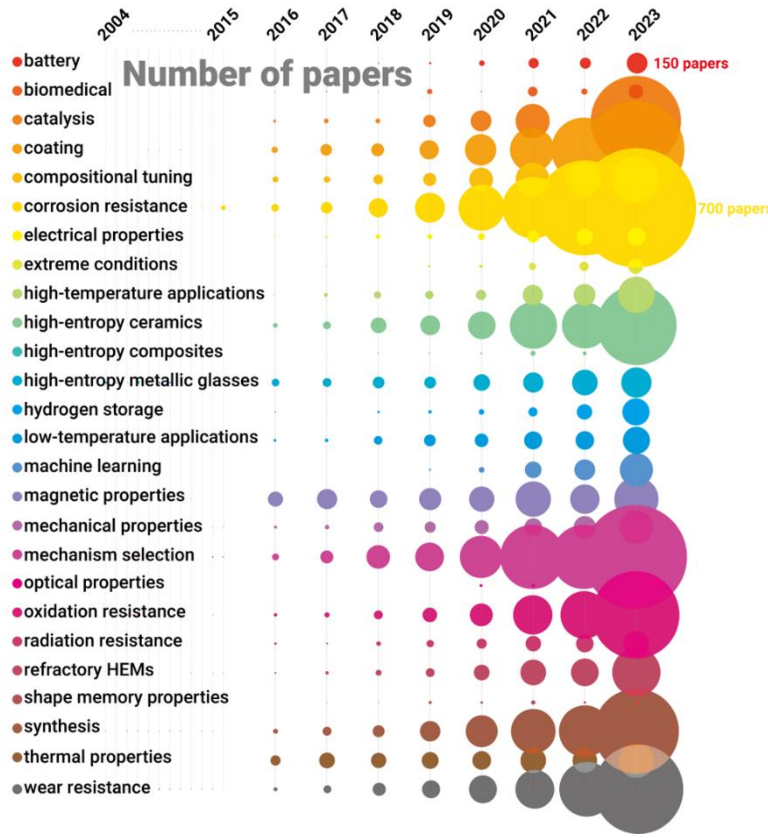
**Supplementary Section 1: Text mining method**

To identify the existing and emergent research interest on HEMs, we collected 13347 abstracts of publications on Web of Science with keywords “high entropy”, “compositionally complex”, and “multicomponent alloy”. The corresponding more than three million words were processed via natural language processing using the VOSviewer software to select key words with more than two occurrences. After removing common phrases and merging those with essentially the same physical meaning (including various abbreviations), the resulting taxonomy were then classified into “processing”, “characterization”, “material system”, “modelling”, “application”, and “microstructure” categories. Focusing on the applications, we obtained 105 keywords with their mapping shown in Figure S1, whereas 25 keywords are further analysed and refined based on the years and highlighted in Figure 2 in the main text.

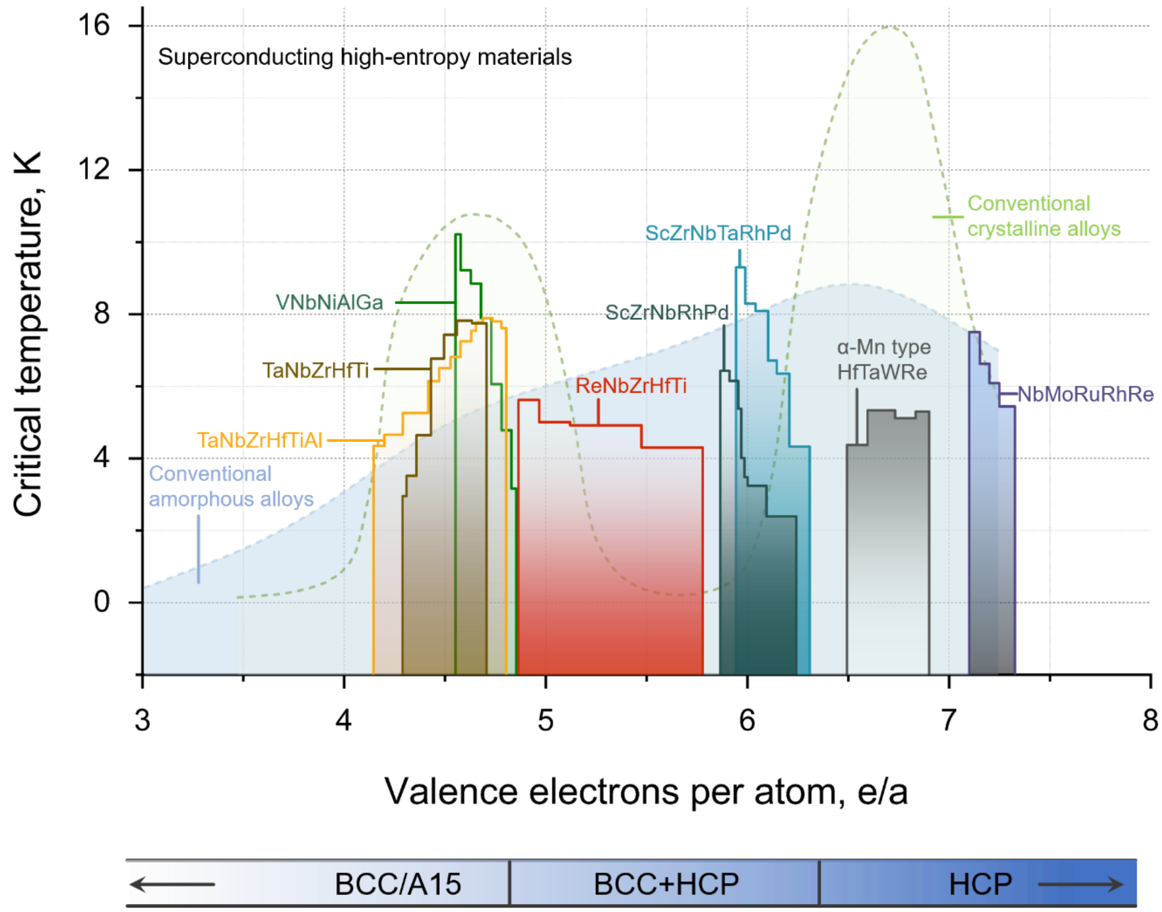
(a)




**(b)**




**Supplementary Figure 1.** (a) Overview of the research interests in the exploration of functional HEMs including promising property connections among different functional characteristics of HEMs, revealing avenues for multi-functional material design. (b) Number of publications on the respective functional features of HEMs.




**Supplementary Figure 2.** Critical temperature as a function of valence-electron count per atom e/a for the reported HEMs and compared with conventional crystalline alloys and amorphous material. Figure adapted with permission from ref 1, American Physical Society.

**Supplementary Table 1.** Comparison of state-of-art maximum *ZT* values of conventional low-entropy thermoelectric materials and entropy-engineered high/medium-entropy materials by different based materials. The temperature, composition and published year are also provided. HEM, MEM and LEM stands for high-entropy material, medium-entropy material and low-entropy material, respectively.

| Base material | Entropy | Composition | *ZT* max | Temperature (K) | Year | Reference |
|---|---|---|---|---|---|---|
| PbTe- based | HEM | $Pb_{0.975}Na_{0.025}Se_{0.5}S_{0.25}Te_{0.25}$ | 2.0 | 900 | 2021 | 2 |
| | | $Pb_{0.9}Na_{0.04}Mn_{0.06}Te_{0.9}S_{0.05}Se_{0.05}$ | 2.1 | 825 | 2024 | 3 |
| | LEM | $Pb_{0.97}Na_{0.03}Te$-(2%MgTe-0.75%GeTe) | 2.8 | 850 | 2024 | 4 |
| | | $FeNb_{0.88}Hf_{0.12}Sb$ | 1.5 | 1200 | 2015 | 5 |
| PbSe-based | HEM | $Pb_{0.89}Sb_{0.012}Sn_{0.1}Se_{0.5}Te_{0.25}S_{0.25}$ | 1.8 | 900 | 2021 | 6 |
| | LEM | $Pb_{0.98}Na_{0.02}Se$-(2.05% $AgInSe_2$) | 1.9 | 873 | 2022 | 7 |
| GeTe-based | MEM | $Ge_{0.63}Mn_{0.15}Pb_{0.1}Sb_{0.06}Cd_{0.06}Te$. | 2.1 | 873 | 2021 | 8 |

| | | | | | | |
|---|---|---|---|---|---|---|
| | LEM | $Ge_{0.9}Sb_{0.1}Te$-2%CdSe | 2.3 | 673 | 2022 | 9 |
| SnTe-based | HEM | $Sn_{0.25}Pb_{0.25}Mn_{0.25}Ge_{0.25}Te$ | 1.5 | 823 | 2021 | 10 |
| | LEM | $(Sn_{0.96}Cd_{0.04}Te_{0.99}I_{0.01})_{0.94}(AgCuTe)_{0.06}$ | 1.7 | 833 | 2022 | 11 |
| $Bi_2Te_3$-based | MEM | $BiAg_{0.05}Sb_{0.95}Te_2Se$ | 1.7 | 450 | 2024 | 12 |
| | LEM | $(Bi_{0.4}Sb_{1.6})_{0.998}Zn_{0.004}Te_{2.995}I_{0.005}$ | 1.5 | 348 | 2022 | 13 |
| $Cu_2Se$-based | HEM | $Cu_{1.87}Ag_{0.13}(In_{0.06}Sn_{0.94})Se_2S$ | 1.5 | 873 | 2022 | 14 |
| | LEM | PbTe-(4%SrTe-2%Na) | 2.2 | 915 | 2012 | 15 |
| $Cu_2Te$-based | HEM | $Cu_2Te_{0.6}S_{0.2}Se_{0.2}$ | 1.4 | 1000 | 2022 | 16 |
| | LEM | $Cu_{1.15}In_{2.29}Te_4$ | 1.0 | 825 | 2019 | 17 |
| SnSe-based | HEM | $(SnSe)_{0.6}(AgSbTe_2)_{0.4}$ | 1.2 | 723 | 2022 | 18 |
| | LEM | $Na_{0.03}Sn_{0.965}Se$ | 3.1 | 783 | 2021 | 19 |
| NbFeSb-based half-Heusler | HEMs | $Nb_{0.8}M_{0.2}FeSb$ | 0.8 | 873 | 2019 | 20 |
| | LEM | $Cu_{1.99}Se$-(0.35%$AgSbF_6$) | 3.0 | 1050 | 2024 | 21 |
| TiNiSn-based half-Heusler | HEM | $Ti_{0.57}Zr_{0.4}Al_{0.02}Ta_{0.01}NiSn_{0.98}Sb_{0.02}$ | 1.4 | 870 | 2023 | 22 |
| | LEM | $Ti_{0.3}Zr_{0.35}Hf_{0.35}Ni_{1.005}Sn$ | 1.0 | 773 | 2019 | 23 |

**Supplementary Table 2.** Composition, cost, thermal expansion and mechanical properties of commercial Invar alloys[24] and Invar HEMs as comparison[25–27]. The price is calculated based on https://tradingeconomics.com/commodity/iron-ore.

| Alloy type | Cost (EUR/T) | Thermal expansion coefficient | Ultimate tensile strength (MPa) |
|---|---|---|---|
| $Fe_{64}Ni_{36}$ Invar | 5997.9 | ~ $1.2 \times 10^{-6}$/K | ~ 600 |
| $Fe_{63}Ni_{32}Co_5$ Super Invar | 6679.2 | ~ 0 | ~ 380 |
| $Fe_{36.5}Co_{54.5}Cr_9$ Stainless Invar | 14899.1 | ~ $-1.2 \times 10^{-6}$/K | / |
| $Fe_{54}Ni_{29}Co_{17}$ Kovar | 9396.3 | ~ $5 \times 10^{-6}$/K | ~ 517 |
| $Fe_{41.8}Ni_{9.4}Co_{40.9}Cr_8$ HEA Invar | 12778.0 | ~ $1.4 \times 10^{-6}$/K | ~ 650 |

References


1. Sun, L. & Cava, R. J. High-entropy alloy superconductors: Status, opportunities, and challenges. *Phys Rev Mater* **3**, (2019).
2. Jiang, B. *et al.* Entropy engineering promotes thermoelectric performance in p-type chalcogenides. *Nat Commun* **12**, 3234 (2021).
3. Zhang, M. *et al.* Improved Thermoelectric Performance of p-Type PbTe by Entropy Engineering and Temperature-Dependent Precipitates. *ACS Appl Mater Interfaces* **16**, 907–914 (2024).
4. Jia, B. *et al.* Pseudo-nanostructure and trapped-hole release induce high thermoelectric performance in PbTe. *Science (1979)* **384**, 81–86 (2024).
5. Fu, C. *et al.* Realizing high figure of merit in heavy-band p-type half-Heusler thermoelectric materials. *Nat Commun* **6**, 8144 (2015).
6. Jiang, B. *et al.* High-entropy-stabilized chalcogenides with high thermoelectric performance. *Science (1979)* **371**, 830–834 (2021).
7. Zhu, Y. *et al.* Multiple valence bands convergence and strong phonon scattering lead to high

thermoelectric performance in p-type PbSe. *Nat Commun* **13**, 4179 (2022).
8. Zhi, S. *et al.* Medium Entropy-Enabled High Performance Cubic GeTe Thermoelectrics. *Advanced Science* **8**, 2100220 (2021).
9. Jin, Y. *et al.* Outstanding CdSe with Multiple Functions Leads to High Performance of GeTe Thermoelectrics. *Adv Energy Mater* **12**, 2103779 (2022).
10. Wang, X. *et al.* Enhanced Thermoelectric Performance in High Entropy Alloys Sn0.25Pb0.25Mn0.25Ge0.25Te. *ACS Appl Mater Interfaces* **13**, 18638–18647 (2021).
11. Wu, G. *et al.* Strengthened phonon scattering and band convergence synergistically realize the high-performance SnTe thermoelectric. *J Mater Chem A Mater* **11**, 649–656 (2023).
12. Ahmad, A. *et al.* Largely enhanced thermoelectric performance in p-type Bi2Te3-based materials through entropy engineering. *Energy Environ Sci* **17**, 695–703 (2024).
13. Zhuang, H.-L. *et al.* High ZT in p-type thermoelectric (Bi,Sb)2Te3 with built-in nanopores. *Energy Environ Sci* **15**, 2039–2048 (2022).
14. Zhang, W. *et al.* Phase-engineered high-entropy metastable FCC Cu2−yAgy(InxSn1−x)Se2S nanomaterials with high thermoelectric performance. *Chem Sci* **13**, 10461–10471 (2022).
15. Biswas, K. *et al.* High-performance bulk thermoelectrics with all-scale hierarchical architectures. *Nature* **489**, 414–418 (2012).
16. Zhang, Z. *et al.* Entropy engineering induced exceptional thermoelectric and mechanical performances in Cu2-yAgyTe1-2xSxSex. *Acta Mater* **224**, 117512 (2022).
17. Li, M. *et al.* Realizing high thermoelectric performance in Cu2Te alloyed Cu1.15In2.29Te4. *J Mater Chem A Mater* **7**, 2360–2367 (2019).
18. Wang, Y. *et al.* Enhanced thermoelectric perfromance in cubic form of SnSe stabilized through enformatingly alloying AgSbTe2. *Acta Mater* **227**, 117681 (2022).
19. Zhou, C. *et al.* Polycrystalline SnSe with a thermoelectric figure of merit greater than the single crystal. *Nat Mater* **20**, 1378–1384 (2021).
20. Yan, J. *et al.* Suppression of the lattice thermal conductivity in NbFeSb-based half-Heusler thermoelectric materials through high entropy effects. *Scr Mater* **157**, 129–134 (2018).
21. Hu, H. *et al.* Highly stabilized and efficient thermoelectric copper selenide. *Nat Mater* **23**, 527–534 (2024).
22. Zhang, X. *et al.* Ultralow lattice thermal conductivity and improved thermoelectric performance in a Hf-free half-Heusler compound modulated by entropy engineering. *J Mater Chem A Mater* **11**, 8150–8161 (2023).
23. Appel, O., Zaharoni, T., Breuer, G., Beeri, O. & Gelbstein, Y. Thermoelectric properties of Ti0.3Zr0.35Hf0.35Ni1.005Sn half-Heusler alloy. *J Appl Phys* **126**, 085110 (2019).
24. Bozorth, R. M. *Ferromagnetism*. (1993).
25. Zheng, S., Sokoluk, M., Yao, G., de Rosa, I. & Li, X. Fe–Ni Invar alloy reinforced by WC nanoparticles with high strength and low thermal expansion. *SN Appl Sci* **1**, 172 (2019).
26. Rao, Z. *et al.* Invar effects in FeNiCo medium entropy alloys: From an Invar treasure map to alloy design. *Intermetallics (Barking)* **111**, 106520 (2019).
27. Rao, Z. *et al.* Machine learning–enabled high-entropy alloy discovery. *Science (1979)* **378**, 78–85 (2022).