EDITORIAL | NOVEMBER 11 2024

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Special Collection: Era of Entropy: Synthesis, Structure, Properties, and Applications of High-Entropy Materials

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Appl. Phys. Lett. 125, 200401 (2024)
https://doi.org/10.1063/5.0245693

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Cite as: Appl. Phys. Lett. **125**, 200401 (2024); doi: 10.1063/5.0245693
Submitted: 28 October 2024 · Accepted: 30 October 2024 ·
Published Online: 11 November 2024

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Note: This paper is part of the APL Special Collection on Era of Entropy: Synthesis, Structure, Properties, and Applications of High Entropy Materials.
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https://doi.org/10.1063/5.0245693

The field of high-entropy materials (HEMs) has emerged as a dynamic area of scientific exploration, driven by the exceptional properties arising from their compositional complexity. Encompassing both high-entropy alloys (HEAs) and high-entropy ceramics (HECs), these materials have garnered significant attention across diverse research domains. From investigations into phase evolution and mechanical characteristics to studies of ionic, electronic, and magnetic behaviors, HEMs demonstrate remarkable potential for a wide array of applications. These range from catalysis and tribology to energy storage and superconductivity. Fundamental research has shed light on crucial phenomena such as configurational entropy, lattice distortion, and sluggish diffusion. These discoveries are paving the way for materials design strategies that enable new functional tunability and resistance to application-specific harsh environments. This burgeoning field promises to revolutionize material design and performance across numerous technological sectors.

This special collection between *Applied Physics Letters* and the *Journal of Applied Physics* provides a timely overview of the latest research in this area. It highlights the growing interest in understanding the impact of high compositional complexity on conventional structure–process–property–performance relationships in HEMs.

High-entropy materials (HEMs) are a class of advanced materials characterized by multicomponent compositions, typically involving five or more principal elements in near-equiatomic proportions. This high compositional complexity creates a high configurational entropy, which favors the formation of single-phase solid solutions over intermetallic, interoxidics, or ordered structures. The unique properties of HEMs stem from several key characteristics. The high-entropy effect stabilizes simple solid solution phases, which can result in novel crystal structures. Severe lattice distortions caused by the presence of various atomic sizes affect both mechanical and thermal properties. Sluggish diffusion further enhances creep resistance, which is valuable for high-temperature applications. For further background on high-entropy materials and entropy stabilization, please refer to the review articles by Miracle and Senkov[1] and McCormack and Navrotsky.[2]

Foundational work in the field includes the pioneering studies of Yeh *et al.*[3] and Cantor *et al.*,[4] who introduced the concept of high-entropy alloys (HEAs). HEAs are metallic systems containing several elements in roughly equal proportions. Notable examples include CoCrFeMnNi and AlCoCrFeNi, which exhibit exceptional mechanical properties such as high strength, excellent fracture toughness, and thermal stability. HEAs are known for their combination of high strength and ductility, making them attractive for structural applications. Rost *et al.*[5] expanded the concept to high-entropy oxides,[6] which further led to expansion into HECs, including carbides,[7] nitrides,[8] diborides,[9] and chalcogenides.[10] HECs, particularly oxides and carbides, are increasingly investigated for not only functionality[11–13] but also applications in extreme environments, where their superior thermal stability and hardness make them ideal for aerospace and defense applications.[14]

Current HEM research often focuses on developing new compositions tailored for specific applications, such as superconducting, catalytic, and magnetic properties. Computational modeling, machine learning, and advanced synthesis techniques like spark plasma sintering and additive manufacturing are being actively explored to create microstructures with enhanced properties. The versatility and robustness of HEMs have opened the door to their use in next-generation technologies.

This collection spans a broad range topics related to phase stability, kinetics, mechanical properties, and functional properties in both HEAs and HECs, totaling 56 articles between *Applied Physics Letters*[12,15–49] and *Journal of Applied Physics*.[50–69]

Many works in this collection focused on synthesis of novel HEMs. Ninomiya *et al.*[40] successfully fabricated polycrystalline submicrometer-sized high-entropy alloy (HEA) particles through laser ablation, highlighting the potential for rapid mass manufacturing for catalysis and tribology applications. Adabasi *et al.*[38] explored high-entropy sulfide thin films, showing increased corrosion resistance and high-temperature performance compared to MoS_2. MOCVD-synthesized CoNiFeCuV/C nanoparticles demonstrated promising dielectric and microwave absorption properties, suggesting their potential for mitigating electromagnetic pollution.[46] HEA nanoparticles for electromagnetic wave absorption were synthesized using electrical wire explosion, showing tunable properties through wire energy deposition and cooling rates.[45] Webb *et al.*[27] studied the high-temperature stability of entropy-stabilized rock salt oxide $(MgCoNiCuZn)_{0.2}O$, finding that Cu and Zn are progressively lost above 1300 °C, leading to multiphase solidification. Li *et al.*[28] investigated spinel high-entropy oxides (HEOs) for gas sensing, showing a strong NO_2 response supported by density functional theory. Guo *et al.*[48] examined grain boundary segregation in complex oxides, finding that cation reducibility affects segregation, leading to Pd and Cu accumulation in oxygen-vacancy-rich regions. The stability of high-entropy oxides (HEOs) under varying synthesis conditions was studied, showing phase competition and decomposition in both rock salt and spinel HEOs, with transformations to metastable phases at high pressures.[22] Finally, the high-entropy oxide $Y_{0.2}La_{0.2}Ce_{0.2}Pr_{0.2}Sm_{0.2}O_2$-$\delta$ demonstrated different crystal structures depending on synthesis methods, with bulk ceramics forming lower-symmetry bixbyite phases and epitaxial thin films forming higher-symmetry fluorite phases, suggesting that synthesis kinetics influence local atomic configurations and material properties.[12]

Mechanical properties of HEMs continue to hold importance for the area of structural applications. Xu *et al.*[39] studied the influence of Al content on bcc lightweight refractory high-entropy alloys, finding that reducing Al enhances plasticity by removing the brittle Al_3Zr_5 phase and promoting dislocation networks at grain boundaries. Rosenkranz *et al.*[65] observed that twinning drives plasticity in hexagonal medium-entropy alloys (MEA), challenging assumptions about high-entropy effects, while Norman *et al.*[36] showed that dislocation formation in polycrystalline rock salt high-entropy oxides varies with crystallographic orientation, impacting mechanical stability. Qiao *et al.*[66] reported that CoCrNi-based alloys exhibit enhanced energy storage capacity from cold work, attributed to fewer immobile dislocations, and Hao *et al.*[39] explored temperature-dependent serrated flow in Cantor alloys, contributing to a predictive model for deformation. Liu *et al.*[70] investigated dynamic strain aging in CoNiV medium-entropy alloys, linking the Portevin-Le Chatelier effect to solute atoms and dislocation pinning. Chang *et al.*[19] examined $CrCoNiSi_{0.3}$ under dynamic tension, noting an increase in yield strength with strain rate and constructing a constitutive model for strain. NbMoCrTiAl alloys demonstrated increased shear stress at grain boundaries due to intermetallic precipitates.[46] Irradiation studies on high-entropy transition metal diborides revealed an initial hardness increase followed by a decrease with continued irradiation, except for $(Hf_{1/3}, Ta_{1/3}, Ti_{1/3})B_2$, which exhibited a steady increase in hardness.[57]

Compositional complexity in high-entropy materials can create novel magnetic, topological, and electronic phases, explaining that the variance in spin, charge, lattice, and orbital interactions can push these materials beyond known phase diagrams, leading to novel phases with potential for quantum technologies.[24] Woodgate and Stauntun, through DFT and MD studies, highlighted the competition between chemical phase ordering and segregation in Ti-doped NbMoTaW and VNbMoTaW HEAs, offering insights into the complex phase behavior of these alloys.[53] Zhao *et al.*[42] investigated strain-driven tuning of magnetic properties in epitaxial high-entropy manganite thin films, observing unique variations in the lattice parameter and a significant reduction in Curie temperature under large epitaxial strain. Strauss *et al.*[60] reviewed inorganic and hybrid high-entropy materials, focusing on their potential for electrochemical energy storage, and highlighted research directions in anode, cathode, and electrolyte components. Jangid *et al.*[43] synthesized and characterized the equiatomic bcc HEA ScVTiHfNb, revealing weakly coupled superconductivity at 4.17 K, with promising low-temperature mechanical properties for superconducting applications. Miruszewski *et al.*[62] reported on the structure and electronic properties of $(Dy_{1-x}Ca_x)(Zr_{0.2}Hf_{0.2}Sn_{0.2}Ti_{0.2}Ge_{0.2})O_7$ pyrochlore oxides, finding low electrical conductivity and a high activation energy of conduction. Tuning lattice spacing through cation ratioing in olivine lithium metal phosphates demonstrated enhanced charge–discharge capacities, laying the groundwork for improved high-voltage batteries.[32]

Rajkowski *et al.*[63] developed a high-throughput approach combining diffusion-multiple experiments with a kinetic model to study interdiffusion in medium- and high-entropy alloys, specifically in quaternary CrFeCoNi alloys. This method efficiently estimates diffusivities, aligning well with existing literature and aiding the rational design of alloys for diffusion-critical applications. Additionally, $(HfZrTiTaNb)B_2$ demonstrated superior oxidation resistance compared to (HfZrTiTaNb)C when tested between 1500 and 1800 °C in oxygen/argon mixtures, attributed to the formation of a dense oxide layer in the borides vs a porous oxide layer in the carbides. At ∼1800 °C, both systems exhibited reduced consumption, likely due to the formation of dense (Hf, Zr, Ti) oxides.[50]

Density functional theory calculations have shown that the Cauchy–Born rule, typically valid in conventional alloys without defects, does not hold in multi-principal element alloys.[49] This violation is due to atomic disorder, which leads to inhomogeneous deformations in these materials. Zhao *et al.*[42] applied machine learning methods to develop a model for alloy ordering based on electronegativity descriptors, using a dataset of 4000 alloy components that included both conventional and high-entropy alloys. The model accurately predicted low-symmetry (disordered) and high-symmetry (intermetallic) structures. Protim Hazarika *et al.*[69] used molecular dynamics simulations to investigate equiatomic and non-equiatomic

HfNbTaTi high-entropy alloys, showing that high strains influence amorphization and deformation. Their findings suggest that alloy strength can be controlled by tuning stoichiometry, as demonstrated by composition-dependent dislocation densities and strain hardening.

High-entropy materials (HEMs), encompassing both high-entropy alloys (HEAs) and high-entropy ceramics (HECs), represent a rapidly growing field of research due to their unique properties stemming from compositional complexity. This collection covers research, from phase evolution and mechanical properties to ionic, electronic, and magnetic behaviors, highlighting the potential of these materials in applications in fields from catalysis and tribology to energy storage and superconductivity. Key studies have advanced our understanding of the effects of configurational entropy, lattice distortion, and sluggish diffusion, contributing to the design of materials that can withstand extreme environments while maintaining superior mechanical and thermal stability.

The continued investigation of HEMs is essential for the development of next-generation technologies, particularly in industries that require materials capable of performing under harsh conditions. The contributions in this collection not only shed light on fundamental aspects of HEM behavior but also pave the way for future innovation through computational modeling, machine learning, and advanced manufacturing techniques. As this field evolves, the insights gained from this research will play a pivotal role in expanding the scope and functionality of these materials.

We believe this special collection will be a valuable resource for researchers and practitioners alike and hope it is well received by the materials science community. By fostering further exploration of high-entropy materials, we aim to contribute to the advancement of the field and the development of new, high-performance materials for a wide range of applications.

The guest editors of this joint special topic want to express their deepest gratitude to all authors who contributed to this collection. A special thank you to the editorial teams at AIP for their organization and guidance during this process. It has been an absolute pleasure serving as guest editors, thank you all.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Christina M. Rost: Writing – original draft (lead); Writing – review & editing (lead). **Alessandro R. Mazza:** Writing – original draft (supporting); Writing – review & editing (supporting). **Scott James McCormack:** Writing – original draft (supporting); Writing – review & editing (supporting). **Katharine Page:** Writing – original draft (supporting). **Abhishek Sarkar:** Writing – original draft (supporting). **T. Zac Ward:** Writing – original draft (supporting); Writing – review & editing (supporting).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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