Multi-transition metal-based HEO cathodes promote cationic and anionic redox activity with a strong increase in energy density and a longer cycle life. The presence of multiple cations stabilized the anionic redox process by inhibiting the reversible oxygen loss and providing a very high level of structural reversibility. Additionally, the entropy stabilization effect in these cathodes suppressed phase transitions and reduced capacity fading under high-voltage cycling conditions.

In SIBs, HEO cathodes such as $Na_3(VFeMnCoNi)O_4$ have exhibited superior cycling stability and rate performance, owing to their structural robustness and tunable redox potentials. These materials benefit from entropy-induced stabilization, which minimizes the volume changes associated with sodium intercalation and enhances electrode durability [61].

1.6.2 High-Entropy Sulfides and Phosphides for Conversion Reactions

HESs and HEPs have been explored as conversion-type electrodes, offering high theoretical capacities through multi-step electrochemical reactions. Unlike conventional intercalation-based electrodes, which rely on reversible ion insertion, conversion electrodes store charge through chemical transformation reactions, often involving the formation and dissolution of intermediate phases.

1.6.2.1 High-Entropy Sulfide Anodes

HESs, such as $(CoCuZnSnFe)S_2$, have demonstrated superior cycling stability due to entropy-stabilized phase formation [66]. The incorporation of multiple cations in the sulfide matrix suppresses structural collapse and ensures long-term electrochemical reversibility. These anodes exhibit minimal volume expansion, reducing mechanical stress and enhancing battery lifespan.

1.6.2.2 High-Entropy Phosphate Cathodes

High-entropy phosphate cathodes offer structural robustness and tunable voltage profiles, making them suitable for LIBs and SIBs. One such example, a NASICON-type $Na_3VFe_{0.5}(TiMnZrCuAl)_{0.5}(PO_4)_3$ cathode, demonstrated a high specific capacity of $158.8\ mA\,h\,g^{-1}$ and retained 95% of its capacity after 170 cycles, making it one of the most stable sodium-ion cathodes reported [10].

HEAs have gained attention as cathode materials due to their exceptional electronic conductivity and phase stability. Unlike traditional cathodes, which often suffer from phase segregation and degradation over extended cycling, HEAs provide a homogeneous reaction environment that supports multi-electron redox processes.

For example, CoCrFeMnNi-based HEAs have demonstrated excellent cycling stability and mechanical robustness, making them attractive for high-power energy storage applications. Additionally, in Li-S batteries, high-entropy metallic alloys have been used as cathode hosts, effectively suppressing the polysulfide shuttle effect and extending cycle life.

High-entropy electrodes represent a new materials design strategy for energy storage applications, offering enhanced electrochemical performance, superior structural stability, and prolonged cycle life. HEOs, HESs, high-entropy phosphates, and HEAs have been successfully employed in LIBs, SIBs, and ASSBs, demonstrating their potential as next-generation energy storage materials. Future advancements in computational modeling, in situ characterization, and scalable synthesis techniques will be critical for the commercialization of high-entropy electrodes. By addressing current challenges and optimizing their properties, these materials are expected to redefine energy storage technologies, paving the way for high-performance and sustainable battery systems.

1.7 High-Entropy Electrolytes and Interface Engineering

Aside from the bulk attributes of high-entropy electrolytes (HEEs), interface engineering is also vital for maximizing battery performance. Engineering stable electrolyte–electrode interfaces is critical in minimizing the interfacial resistance, enhancing ion transfer, and suppressing undesirable side reactions that lower battery durability [68, 69]. The design rules, structural aspects, and electrochemical attributes of the HEEs are covered under this section in addition to the contribution of interface engineering to enhance their incorporation within energy storage devices.

1.7.1 High-Entropy Solid-State Electrolytes

SSEs are an essential constituent of ASSBs because of their enhanced safety, thermal stability, and mechanical robustness. High-entropy solid-state electrolytes (HESEs) extends the above advantages with the inclusion of multiple cationic and anionic species to further enhance structural stability and ionic conductivity.

1.7.1.1 Composition and Structural Characteristics

HESEs typically consist of a combination of oxides, sulfides, halides, or polyanionic frameworks. These materials exhibit disordered lattice structures, where entropy-driven stabilization suppresses phase decomposition and promotes the formation of single-phase solid solutions. Notable examples of HESEs include high-entropy lithium argyrodite (HE-Li_6PS_5X) electrolytes, which incorporate Cl^-, Br^-, and I^- anions to enhance Li-ion mobility, polyanionic sulfide-based HESEs, such as $Li_6PS_5(Cl_{0.33}Br_{0.33}I_{0.33})$, which exhibit an exceptionally high ionic conductivity of $22.7\,mS\,cm^{-1}$ at room temperature and $Li_{3.25}[Si_{0.25}P_{0.75}]S_4$, an entropically stabilized fast-ion conductor that achieves $1.22\,mS\,cm^{-1}$ ionic conductivity, significantly outperforming conventional β-Li_3PS_4.

1.7.1.2 Ionic Transport Mechanisms in HESEs

The presence of multiple cations and anions in HEEs creates a complex energy landscape for ion migration. This compositional disorder due to the presence of multiple ions results in reduced activation energy for ion diffusion, facilitating fast ionic transport, increased number of percolation pathways, that leads efficient ion conduction even in polycrystalline phases and suppression of grain boundary resistance which improves overall electrolyte performance in SSBs.

Computational studies and experimental observations have confirmed that high-entropy-driven site disorder leads to an increase in lithium and sodium diffusion coefficients, making HESEs highly competitive with conventional SSEs.

1.7.1.3 Interfacial Engineering in High-Entropy Solid Electrolytes

One of the primary challenges in HEEs is achieving stable electrode–electrolyte interfaces that minimize interfacial resistance and prevent side reactions. Interface engineering strategies for HESEs include surface coatings (e.g. $LiNbO_3$, $Li_4Ti_5O_{12}$) to stabilize SEIs in lithium-metal batteries and interfacial engineering with high-entropy fluorides (HEFEs), which form stable passivation layers that suppress dendrite formation.

1.7.1.4 Mitigation of Interfacial Decomposition

Interfacial decomposition remains a major concern in HEE systems. Strategies to mitigate these effects is focused on grain boundary modifications to reduce contact resistance between polycrystalline phases in HESEs and pre-lithiation and artificial SEI formation to prevent electrolyte degradation upon cycling.

Advanced characterization techniques, such as in situ XPS and EIS, have been employed to monitor and optimize interface formation in HESE systems. HEEs represent a transformative approach to electrolyte design, offering enhanced ionic conductivity, superior phase stability, and improved interfacial compatibility for next-generation energy storage systems. By harnessing configurational entropy and multicomponent interactions, HESEs, HELEs, and high-entropy polymer electrolytes (HEPEs) exhibit outstanding electrochemical performance. With increasing advancements, the future of energy storage will be revolutionized by the increasing prominence of HEEs, which will facilitate safer, more efficient, and enhanced performance battery technology.

1.7.2 High-Entropy Liquid and Gel Electrolytes

Although SSEs have gained significant attention, liquid and gel-based HEEs are also being developed for applications in hybrid batteries and flexible energy storage devices. These electrolytes take advantage of solute–solvent interactions in multicomponent ionic systems, modifying transport properties and electrochemical stability.

1.7.2.1 Multi-Salt HELEs

HELEs incorporate multiple lithium or sodium salts to optimize solvation dynamics and enhance ionic conductivity. Studies on multi-salt electrolyte formulations containing LiTFSI, LiFSI, and $LiClO_4$ have demonstrated that the disordered solvation environment reduces ion association, promoting free-ion conductivity.

In SIBs, HEEs have been designed using a combination of NaFSI, $NaClO_4$, and NaTFSI, achieving improved electrochemical window stability and increased Na-ion diffusion rates compared to conventional single-salt electrolytes.

1.7.2.2 High-Entropy Polymer Electrolytes

The development of HEPEs has enabled the fabrication of flexible and highly conductive gel electrolytes for next-generation energy storage devices. These materials integrate multiple polymeric matrices and ionic liquid additives to improve mechanical flexibility, ionic transport, and thermal stability. For instance, HEPEs are composed of PEO, PVDF, and PAN matrices, which exhibit enhanced compatibility with lithium and sodium metal anodes and multicomponent ionic gels incorporating LiTFSI, NaFSI, and deep eutectic solvents, achieving stable performance in flexible batteries.

1.8 Advanced Characterization Techniques for HEMs

It is vital to utilize a combination of structural, spectroscopic, and electrochemical techniques to unravel the intrinsic characteristics of HEMs. A discussion in this section focuses on the most critical X-ray, electron microscopy, electrochemical, and in situ/operando techniques used to characterize HEMs and their usage specifically in electrodes, electrolytes, and interfaces for energy storage applications.

1.8.1 Structural and Phase Analysis Techniques

1.8.1.1 X-ray Diffraction (XRD) and Rietveld Refinement

Given that HEMs often form single-phase solid solutions due to entropy stabilization, XRD is instrumental in distinguishing between homogeneous solid solutions and multiphase composites. Rietveld refinement of XRD data enables precise lattice parameter determination, site occupancy analysis, and phase fraction quantification. Similarly, synchrotron XRD provides high-resolution diffraction patterns, allowing for the study of short-range disorder, stacking faults, and lattice distortions in HEMs and pair distribution function (PDF) analysis extends XRD capabilities by providing insights into local atomic arrangements, particularly in materials exhibiting short-range ordering despite long-range crystallinity.

1.8.1.2 Neutron Diffraction and Total Scattering Techniques

Neutron diffraction offers advantages over XRD for HEMs containing low-Z elements (e.g. Li, Na, O, F, S), which have weak X-ray scattering contrast. Time-of-flight neutron diffraction (TOF-ND) enables high-resolution structure determination for complex multi-element HEMs. In the same manner, total scattering and atomic PDF analysis reveal local atomic environments, enabling the study of lattice distortions and cation disorder. Moreover, *in situ* neutron diffraction is particularly useful for tracking phase transformations and defect evolution in HEM electrodes and electrolytes during charge/discharge cycles.

1.8.2 Chemical State and Electronic Structure Characterization

HEMs exhibit complex multi-element interactions, variable oxidation states, and defect-driven electronic properties, necessitating advanced spectroscopic techniques.

1.8.2.1 X-ray Photoelectron Spectroscopy (XPS) and Auger Electron Spectroscopy (AES)

XPS provides elemental composition, oxidation states, and surface chemistry information. Due to the diverse chemistry of HEMs, XPS is crucial for deconvoluting multiple oxidation states in multi-cationic HEMs (e.g. $Fe^{2+/3+/4+}$, $Mn^{2+/3+/4+}$), identifying cation migration and surface reconstructions in battery electrodes and tracking electrochemical degradation in HEMs through post-cycling XPS analysis. In addition, Auger Electron Spectroscopy (AES) is often used alongside XPS to study electronic structure modifications and defect chemistry at electrode–electrolyte interfaces.

1.8.2.2 X-ray Absorption Spectroscopy

XAS is particularly powerful for probing local electronic structures and coordination environments in high-entropy systems. X-ray absorption near-edge structure (XANES) provides oxidation state information, which is critical for understanding redox activity and charge compensation mechanisms in HEM-based electrodes [21]. Similarly, extended X-ray absorption fine structure (EXAFS) analysis permits the exact measurement of cation–anion bonding geometries and coordination distortions, which determine the manner by which structural disorder is influenced by entropy in HEMs.

1.8.3 Electron Microscopy for Microstructural and Compositional Analysis

High-resolution electron microscopy is an essential method used to examine the morphology, grain boundary structure, and atomic-scale defects of HEMs.

1.8.3.1 Scanning Electron Microscopy and Energy-Dispersive X-ray

Spectroscopy Scanning electron microscopy (SEM) enables microstructural characterization for the detection of the grain size, surface morphology, and phase distribution of the HEM and energy-dispersive X-ray spectroscopy (EDS) mapping attests to the uniform distribution of various main elements, one of the main characteristics of well-synthesized HEMs.

1.8.3.2 Transmission Electron Microscopy and Electron Energy Loss Spectroscopy (EELS)

High-resolution transmission electron microscopy (HRTEM) facilitates imaging of lattice structures, dislocation defects, and grain boundary characteristics at the atomic level [70]. Selected area electron diffraction (SAED) enables phase identification of nanostructured HEMs and information from the local bonding state, defect state, and electronic state can be obtained from electron energy loss spectroscopy (EELS) analysis, which is a complementary tool to XAS [23].

1.8.4 In Situ and Operando Characterization Techniques

To understand the electrochemical and structural development of HEMs in real time, in situ and operando methodologies are vital. In situ XRD records phase transitions and lattice parameters in real time during charge–discharge cycling [71]. Operando XAS monitors dynamic changes of oxidation states in battery electrodes. In situ TEM facilitates the observation of the process of lithiation/delithiation-induced defect formation, providing valuable information regarding structural degradation mechanisms in high-entropy electrodes.

The in-depth characterization of HEMs involves a multidimensional approach involving X-ray, neutron, and electron microscopy, spectroscopic, and electrochemical studies. All these methodologies yield useful information about the phase stability, defect chemistry, ion transport, and interface evolution so the rational design of high-performance HEMs for application in energy storage is facilitated. Future studies need to concentrate on combining sophisticated in situ and operando methodologies to further define the basic mechanisms that control electrochemical performance, durability, and degradation in HEMs. All this will open the way for the large-scale application of HEMs for next-generation LIBs, SIBs, and ASSBs.

1.9 Challenges, Prospects, and Commercialization Pathways

The development of HEMs has created a new paradigm in the field of material science, especially in applications for energy storage. The application of configurational entropy helps materials attain increased phase stability, tunable electrochemical performance, and improved ionic conductivity. Though HEMs offer the said attributes, their large-scale usage in commercial battery systems is hindered by a number of scientific as well as technical roadblocks (see Figure 1.5). The complexity factor at the synthesis step, electrochemical stability, financial feasibility, and large-scale production are the main challenges hindering the use of HEMs. The following section discusses the major barriers to the commercialization of HEM, presents prospective research directions for their exploitation, and details how the large-scale production of HEMs can be achieved.

One of the main difficulties in the construction of high-entropy battery material is the complexity and scalability of synthesis. For maintaining phase purity and uniform distribution of the multiple elements, which is essential for consistent electrochemical activity, synthesis of the HEM is usually performed at a high temperature using the solid-state sintering method, the sol–gel process, SPS, and mechanochemical synthesis. All such syntheses are power-hungry and hard to scale. Batch-to-batch differences in composition further complicate their industrial production. Another key challenge is the electrochemical and structural stability of the long-term cycling of the HEMs. Although configurational disorder alleviates phase segregation, a level of gradual compositional ordering is still present in certain high-entropy systems, affecting their capacity retention and overall electrochemical performance. Interfacial stability is also an issue with the high-entropy electrodes and HEEs specifically in ASSBs since such materials can show large interfacial resistance and reactivity, which can eventually cause degradation. Interface engineering strategies involving surface coatings and artificial SEI films will be essential for long-term electrochemical stability. The availability and the cost of raw materials are also vital factors that determine the commercialization of HEMs. The inclusion of costly components like cobalt (Co), nickel (Ni), and vanadium (V) can dramatically raise the cost of production. For further economic feasibility, researchers are investigating the application of earth-abundant and low-cost elements like iron (Fe), manganese (Mn), and titanium (Ti) as replacements. Moreover, the availability of resources and geopolitical considerations for the extraction of rare metals might also affect the future supply chain of HEM.

Despite these limitations, HEMs have tremendous potential for the next wave of energy storage. One of the most promising research avenues is the application of high-throughput computational simulation and artificial intelligence (AI) for material discovery and to speed up the development of advanced high-performance HEMs [72, 73]. Machine learning (ML) techniques can examine large datasets to identify the best combination of elements that maximize electrochemical performance and reduce material expenses. Furthermore, density functional theory (DFT) calculations and molecular dynamics (MD) simulations can gain insight into ion diffusion mechanisms, defect energies, and phase stability, which can be used for rational material design with the aid of high-throughput screening strategies. Another area of focus is the interfacial engineering to enhance the stability of electrolytes and high-entropy electrodes. Thin-film coatings, artificial SEI films, and grain boundary modifications can all be used to lower interfacial resistance and enhance charge transfer. Multi-layer electrode designs coupling high-entropy phases with highly conductive scaffolds could further be beneficial. Another vital area for future studies is the development of low-cost and sustainable materials. To make high-entropy battery materials economically viable, researchers are also looking at the application of naturally prevalent elements. Recycling and upcycling methodologies for spent battery materials can also decrease reliance on critical resources. The application of green process technologies like low-sintering temperature and solvent-free synthesis can also reduce the overall impact of the development of high-entropy materials.

To gain universal industrial applications, scalable and economic manufacturing methods have to be developed for high-entropy compounds. Dry roll-to-roll processing and chemical vapor deposition (CVD) would be used to mass-produce thin-film high-entropy electrodes [74–76]. 3D printing and additive manufacturing also open possibilities for the creation of tailored electrode geometries with tailored porosity and mechanics [77]. Sustainable synthesis strategies like aqueous processing can further enhance cost-effectiveness. Another essential aspect of commercialization is that the HEMs should be integrated well with conventional battery technology. The HEEs and electrodes must be made compatible with conventional LIB and SIB designs with straightforward adoption by battery manufacturers. Hybrid designs with conventional battery material combined with the high-entropy phase can be an intermediate step toward the use of entirely entropy-maximized energy storage devices. For the development of HEMs closer to commercialization, cooperation between government agencies, academic institutions, and industry will be essential. Technology transfer can be accelerated by public–private collaborations, enabling research ideas to go from lab-scale demonstrations to pilot-scale production.

Standardized testing procedures should also be put in place to guarantee that HEM-based battery materials fulfill industry and regulatory standards for durability, safety, and dependability. Governments and funding organizations ought to encourage research projects aimed at the scalable manufacture and recycling of HEMs, as well as offer incentives for the development of sustainable batteries.

1.10 Summary and Outlook

A novel approach of developing next-generation energy storage systems has been brought about by the development of HEMs. These materials are intriguing options for solid electrolytes, battery electrodes, and interface engineering because they use combinations of multiple primary elements to increase ionic transport, phase stability, and electrochemical characteristics. The basic ideas, synthesis methods, compositional tweaking, electrochemical performance, and characterization methodologies of HEMs in energy storage applications have all been thoroughly covered in this book chapter. An introduction to HEMs and the function of configurational entropy in stabilizing single-phase solid solutions kicked off the conversation. In order to prevent phase segregation and guarantee uniform ionic and electronic conduction, the idea of entropy-driven phase stabilization was examined. The chapter went on to discuss compositional engineering techniques, emphasizing the ways in which multi-element interactions affect the redox activity, defect chemistry, and electronic structure of electrodes and electrolytes. High-entropy electrodes, including HEOs, HESs, HEPs, and HEAs, accounted for a large amount of the chapter. These materials are appealing for LIBs, SIBs, Li-S, and ASSBs due to their better cycling stability, multi-electron redox capabilities, and programmable electrochemical potentials. The function of HEEs was also explored, including how compositional disorder and interface engineering can improve electrochemical stability, inhibit dendritic development, and increase ionic conductivity. The chapter additionally covers advanced characterization methods that are necessary for studying HEMs, such as in situ/operando analysis, transmission electron microscopy (TEM), XRD, neutron scattering, XPS, and EIS. These methods allow for the logical design of high-entropy battery components by offering crucial insights into phase evolution, defect chemistry, interfacial phenomena, and long-term cycling performance. HEMs have several advantages, but a number of issues prevent them from being commercialized. These issues include cost considerations, interfacial compatibility, electrochemical stability, and synthesis scalability. To overcome these obstacles and hasten the industrial deployment of HEM-based battery materials, techniques such as sustainable synthesis methods, ML-guided material discovery, and enhanced interface engineering have been put forth.

1.11 Outlook

In energy storage research, HEMs mark a paradigm shift by providing new avenues for creating battery materials that are efficient, long-lasting, and reasonably priced.

To fully realize the promise of HEMs and make their commercialization easier, several important research avenues need to be explored in the future.

1.11.1 Accelerating High-Throughput Computational Discovery

Advancements in ML and AI-assisted materials design will play a significant role in adjusting the compositions of HEMs. DFT simulations, MD simulations, and computational high-throughput screening can be employed to accelerate the discovery of novel HEMs with predesigned electronic, ionic, and mechanical characteristics. Phase stability, defect energy of formation, and electrochemical performance can be estimated through AI-assisted models, and hence trial-and-error experimentations can be reduced significantly.

1.11.2 Interface Engineering for Next-Generation Batteries

For HEMs to be successfully integrated into commercial batteries, interface compatibility with electrolytes and current collectors must be optimized. Strategies such as thin-film coatings, artificial SEI layers, and grain boundary engineering will be crucial in reducing interfacial resistance and enhancing charge transfer kinetics. In SSBs, improving electrolyte–electrode contact through engineered interfaces will be key to achieving high-energy density and prolonged cycle life.

1.11.3 Sustainable and Cost-Effective Material Development

The economic viability of HEMs depends on their scalability and cost-effectiveness. Future research should focus on replacing expensive transition metals with earth-abundant elements while maintaining high electrochemical performance. Additionally, low-energy synthesis techniques, such as aqueous processing, solvent-free synthesis, and roll-to-roll manufacturing, should be explored to minimize production costs. Developing recycling and upcycling strategies for HEM-based electrodes and electrolytes will further contribute to sustainability and circular economy initiatives.

1.11.4 Integration with Emerging Energy Storage Technologies

Beyond LIBs and SIBs, HEMs hold promise for next-generation energy storage systems, including multivalent batteries (Mg, Zn, Al-based systems), supercapacitors, and hybrid energy storage devices. Their intrinsic phase stability, multi-electron charge storage capabilities, and defect-tolerant structures make them attractive candidates for high-energy density and high-power applications. Further research should explore the potential of HEMs in advanced battery chemistries, enabling breakthroughs in energy storage beyond current lithium-ion technologies.

1.11.5 Industry–Academia Collaborations and Commercialization Pathways

To accelerate the transition of HEMs from research laboratories to commercial-scale production, strong collaborations between academic institutions, industry leaders, and government agencies will be essential. Establishing standardized testing protocols and performance benchmarks will facilitate the comparison of HEMs with conventional battery components. Additionally, government policies, funding initiatives, and industry investments will play a crucial role in driving the commercialization of HEM-based energy storage solutions.

HEMs have shown great potential as electrolytes for next-generation batteries because they are more stable, have customizable electrochemical characteristics, and perform better than traditional materials. Energy storage issues including capacity fading, interfacial degradation, and safety can be resolved by these materials by utilizing entropy-driven stabilization and compositional flexibility. However, before HEMs can be widely adopted in the commercial sector, much research is still needed to overcome scalability, interfacial, and cost obstacles. Optimizing high-entropy compositions for useful energy storage applications will need the combination of computational modeling, sophisticated characterization methods, and sustainable material design. HEMs are expected to transform energy storage as developments proceed, opening the door to the creation of long-lasting, high-performing, and ecologically friendly battery technologies. Their successful commercialization could pave the way for the next generation of high-efficiency energy storage devices by bringing about innovations in portable electronics, grid-scale storage, and electric cars. HEMs could influence battery technology in the future and help create a more sustainable, energy-efficient, and electrified society by resolving outstanding issues and utilizing new scientific discoveries.

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