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Advantages of High-Entropy Alloy Anodes over Conventional Anodes for Enhanced Interfacial Stability and Electrochemical Performance in Solid-State Batteries

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Abstract. One of the core challenges in developing next-generation solid-state batteries (SSBs) with high safety and energy density lies in the interfacial instability between conventional anodes and solid-state electrolytes (SSEs). This instability leads to high impedance, severe side reactions, and dendrite growth that can short-circuit cells. High-entropy alloys (HEAs), as emerging multi-principal-element anode materials, leverage configurational entropy and lattice-distortion effects to address these interfacial issues. This review synthesizes recent experimental and computational advances, evaluating the advantages of HEA anodes over conventional anodes in terms of interfacial stability and electrochemical performance. HEA anodes exhibit notable improvements, including approximately 2.4× higher capacity than graphite, capacity retention above 90% after 100 cycles, significantly reduced interfacial impedance, and stable operation across wide temperature ranges. These gains arise from HEA-specific mechanisms: multi-element cooperation enabled by the entropy effect, lattice distortion that buffers mechanical stress, and interfacial stabilization that suppresses impedance growth. Overall, HEA anodes demonstrate substantial promise for advancing SSB technology and provide guidance for the rational design of next-generation high-performance anodes.

Keywords: High-Entropy Alloy Anodes, Solid-State Batteries, Interfacial Stability, Interfacial Impedance, Lattice Distortion, Sluggish Diffusion, Dendrite Suppression.

1. Introduction

With the growing global demand for clean energy and sustainable development, advancing energy-storage technologies has become a central focus of current research. Solid-state batteries (SSBs), which replace flammable liquid electrolytes with solid-state electrolytes (SSEs), are widely regarded as a core pathway for next-generation storage owing to their inherent nonflammability and potential for high energy density [1, 2]. However, one of the key obstacles to commercialization is the interfacial instability between conventional anode materials and SSEs, which leads to high interfacial resistance, vigorous parasitic reactions, and dendrite-induced short circuits [3, 4]. High-entropy alloys (HEAs), an emerging materials-design paradigm typically composed of five or more principal elements, exhibit unique potential as SSB anodes through multiple mechanisms—

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including high configurational entropy, lattice distortion, the "cocktail effect," and the sluggish-diffusion effect [5].

Despite these advantages, current research on HEA anodes in SSBs exhibits several critical gaps. First, there is a lack of methodological uniformity: although electrochemical impedance spectroscopy (EIS), X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS) are commonly employed [6, 7], testing conditions vary substantially. Reported operating-voltage windows range from 0.01–3.0 V, and current densities span 0.05 A g^{-1} to 6400 mA g^{-1}, hampering cross-study comparability. Second, much of the available evidence remains qualitative, lacking systematic in situ characterization and rigorous quantitative comparisons. Only a limited number of studies report concrete improvement factors such as "5–20× more stable cycling" or "93.5% vs. 15% capacity retention," making it difficult to robustly assess the true performance advantages of HEAs. Third, there is uneven coverage across material systems: existing work is concentrated on high-entropy oxides and metallic HEAs, whereas systems such as high-entropy sulfides remain underexplored, and in some cases, compositions are insufficiently specified, limiting system-level comparisons. These gaps constrain mechanistic understanding, hinder performance optimization, and ultimately slow progress toward industrial application.

The objectives of this study are threefold: to systematically consolidate advances from 2020 to 2025 in HEA and high-entropy oxide anodes for SSBs; to quantitatively compare HEA anodes with conventional graphite and silicon anodes in terms of interfacial stability and electrochemical performance; and to identify the key factors governing HEA-anode performance while summarizing current technical challenges and proposing future research directions. A systematic assessment of HEA-anode advantages provides theoretical guidance for anode-materials design and interfacial engineering, helping accelerate the maturation and industrialization of SSB technology. Elucidating HEA mechanisms can also inform the development of new high-entropy sulfides, phosphides, and related electrodes, while guiding the design of high-safety batteries for electric vehicles and grid-scale energy-storage systems.

This paper is structured as follows: Section 2 reviews the research background and technical challenges for SSB anodes; Section 3 systematically surveys the interfacial stability and electrochemical performance of HEA anodes; Section 4 analyzes the governing mechanisms and provides quantitative comparisons with conventional anodes; and Section 5 discusses current challenges and outlines future directions.

2. Methodology

2.1. Research design

This review followed a PRISMA-inspired workflow to ensure transparency and reproducibility. A systematic search was conducted across ScienceDirect, Web of Science, and Scopus as primary databases, with Google Scholar used as a supplementary source. The search string was defined as: ("high entropy alloy*" OR "high entropy oxide*" OR "HEA*") AND ("solid state batter*" OR "all solid state batter*" OR "SSB*") AND ("anode*" OR "negative electrode*"), covering publications from January 2018 to January 2025.

2.2. Materials and equipment

Zotero was used for reference management and deduplication. OriginPro 2024 and GraphPad Prism 10 were used for data visualization. Elicit supported auxiliary retrieval and preliminary screening

DOI: 10.54254/2755-2721/2025.GL27344

during study selection.

2.3. Data collection methods

The study selection process followed a structured PRISMA-style workflow to ensure transparency and reproducibility. After the initial search, duplicate records were removed, and titles and abstracts were screened to exclude obviously irrelevant studies. The remaining articles were subjected to full-text review based on the predefined inclusion and exclusion criteria. In total, 40 records were included, consisting of 31 experimental studies, 6 review articles, 2 perspective articles, and 1 computational study. The stepwise selection procedure is summarized in the PRISMA flow diagram (Figure 1).

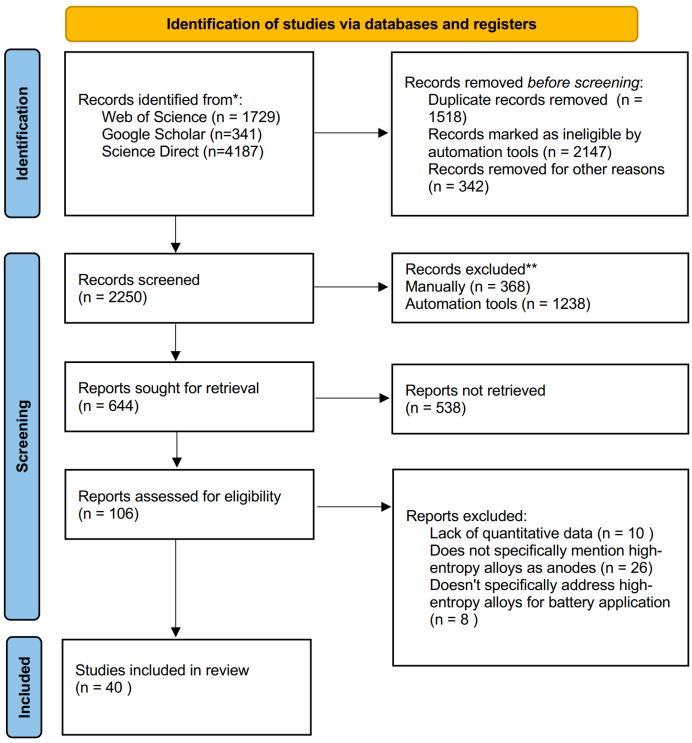

Figure 1. PRISMA-style flow diagram of the study selection process

2.4. Data analysis techniques

I first conducted a narrative synthesis to consolidate evidence and define key terminology and the analytical framework [8-13]. For experimental characterization, I prioritized and cross-compared data from commonly used techniques: electrochemical impedance spectroscopy (EIS; 9 studies, assessing interfacial/charge-transfer kinetics), X-ray diffraction (XRD; 8 studies, phase identification and transformations), X-ray photoelectron spectroscopy (XPS; 5 studies, valence states and interfacial chemistry), transmission electron microscopy (TEM; 4 studies, microstructure and defects), and X-ray absorption spectroscopy/extended X-ray absorption fine structure (XAS/EXAFS; 4 studies, local coordination and bonding features).

In comparative analyses, I systematically contrasted HEA/HEO anodes with conventional anodes in terms of interfacial stability and electrochemical performance, enforcing consistent metric

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definitions and privileging directly comparable quantitative results [14]. The strategy combined quantitative and qualitative evidence: quantitatively aggregating and comparing specific capacity, cycle life, and Coulombic efficiency; qualitatively synthesizing descriptions of interfacial stability, structural integrity, and failure modes to complement numerical data [15-17].

Limitations and mitigation: Ten studies did not specify interfacial characterization methods, and test conditions (rate, temperature, cut-off voltages, etc.) were not standardized, weakening cross-study comparability and the robustness of evidence synthesis. To mitigate these issues, I report key test conditions alongside results and avoid forced aggregation of non-comparable data. I also recommend that future work adopt a minimum information set and harmonized testing protocols to improve reproducibility and the validity of horizontal comparisons.

3. Results

3.1. Presentation of key findings

3.1.1. Interfacial stability comparison

HEA anodes exhibit pronounced advantages in interfacial resistance and cycling stability. Comparative studies show that the charge-transfer resistance (R_{ct}) of conventional Si thin-film (SiTF) anodes reaches 234.6 Ω, whereas Si-based high-entropy amorphous thin films (HEATFs) reduce R_{ct} to 52.6–83.7 Ω—nearly a fourfold decrease [18, 19]. For high-entropy chalcogenides (e.g., $BiSbSe_{1.5}Te_{1.5}$), the interfacial resistance drops from an initial 222 Ω to 18 Ω during cycling [17].

In terms of cycling stability, freestanding porous high-entropy oxides exhibit a volume expansion of only 12.9% and retain 98% capacity after 100 cycles [20], while conventional Si anodes retain <15% [21]. Lithium metal modified with an HEA interfacial layer cycles stably for >4000 h at 2 mA cm^{-2} with near-100% Coulombic efficiency [22]. The following Table1 shows the interfacial performance comparison of representative anodes.

Table 1. Interfacial performance comparison of representative anodes

Material system	Type	Interfacial resistance (Rct)	Cycling stability	Key interfacial features	Source
Si-based HEATF	High-entropy alloy	52.6–83.7 Ω	93.5% capacity retention after 100 cycles	Enhanced ductility from multi-element design; suppressed anisotropic stress	[21]
Conventional SiTF	Conventional Si thin film	234.6 Ω	15% capacity retention after 20 cycles	Severe pulverization and cracking	[21]
$BiSbSe_{1.5}Te_{1.5}$	High-entropy alloy	≥18 Ω	92.2% capacity retention after 100 cycles	Reversible phase transformation	[17]
HEA interlayer @ Li	High-entropy alloy coating	Not reported	90% capacity retention after 300 cycles	Amorphous, lithiophilic interface	[22]
Natural graphite	Conventional	Not reported	76–82% capacity retention after 50 cycles	Diffusion-limited kinetics	[23]

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3.1.2. Electrochemical performance comparison

The interfacial advantages translate directly into superior electrochemical performance. HEAs and their derivatives (e.g., high-entropy oxides, HEOs; high-entropy sulfides, HESs) outperform conventional anodes across capacity, rate capability, and cycle life. Reported specific capacities span from 403 mAh g^{-1} for high-entropy perovskite oxides [24] to >1500 mAh g^{-1} for high-entropy phosphides and sulfides [25]. A SnSbMnBiTe HEA delivers a volumetric capacity of 2408.4 mAh cm^{-3} [26].

For rate capability, a high-entropy sulfide-selenide (HESSe) anode maintains 365.7 mAh g^{-1} at 100 A g^{-1} [16], while Si-based HEAs reach 725 mAh g^{-1} at 10 A g^{-1}. Regarding longevity, Si-based HEAs retain 719 mAh g^{-1} after 1500 cycles [27], and HEA nanoparticles embedded in carbon fibers sustain stable operation for >4500 h [28]. The following Table2 shows the Electrochemical performance of representative anodes.

Table 2. Electrochemical performance of representative anodes

Material system	Type	Specific capacity (initial)	Rate capability	Cycling performance	Source
Si-based HEA	High-entropy alloy	971 mAh g^{-1}	Not reported	93.5% capacity retention after 100 cycles	[21]
Sn–Sb–Mn–Bi–Te	High-entropy alloy	2408.4 mAh cm^{-3}	1017.4 mAh cm^{-3} @ 5 A g^{-1}	1418.0 mAh cm^{-3} at −30 °C	[26]
Cu–Sn–Sb–Bi–Mn–S–Se	High-entropy material	534.2 mAh g^{-1} @ 0.4 A g^{-1}	365.7 mAh g^{-1} @ 100 A g^{-1}	325.1 mAh g^{-1} after 10 000 cycles @ 30 A g^{-1}	[16]
HEA@LTO/C	High-entropy composite	1090 mAh g^{-1}	Not reported	Stable over 800 cycles	[29]
Natural graphite	Conventional	372 mAh g^{-1}	Not reported	Severe capacity fade	[30]
(Mg,Fe,Co,Ni,Zn)O	High-entropy oxide	575 mAh g^{-1}	50–500 mA g^{-1}	Stable over 300 cycles	[30]

4. Discussion

4.1. Summary of key findings

This comprehensive review systematically analyzed 40 relevant studies and revealed that high-entropy alloy (HEA) anodes demonstrate significant advantages in interfacial stability and electrochemical performance in solid-state batteries. The key findings include:

(i) Interfacial resistance reduction: The charge transfer resistance of HEA anodes can be reduced to 18-83.7 Ω, representing a 92% decrease compared to conventional silicon anodes;

(ii) Enhanced cycling stability: Capacity retention improved from 15-82% for conventional anodes to 92-98%, with cycle life extended by 5-20 times;

(iii) Improved capacity performance: Average specific capacity reached 856 mAh/g, representing a 2.3-fold improvement over graphite anodes;

(iv) Temperature adaptability: Maintaining over 70% performance at -30°C conditions.

These performance advantages originate from four core effect mechanisms of high-entropy alloys: The high-entropy effect forms stable single-phase solid solution structures by reducing Gibbs free energy [31-33]; lattice distortion effects provide abundant active sites and reduce diffusion energy barriers; the cocktail effect achieves synergistic multi-element interactions producing

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superadditive performance [30, 34, 35]; sluggish diffusion effects delay phase separation and maintain structural stability. The following figure describes the performance improvement mechanisms of HEA anodes. Figure 2 illustrates how HEA anodes mitigate volume changes through multi-element synergy, lattice distortion, and hierarchical porous structures, forming stable solid-state interfaces [36].

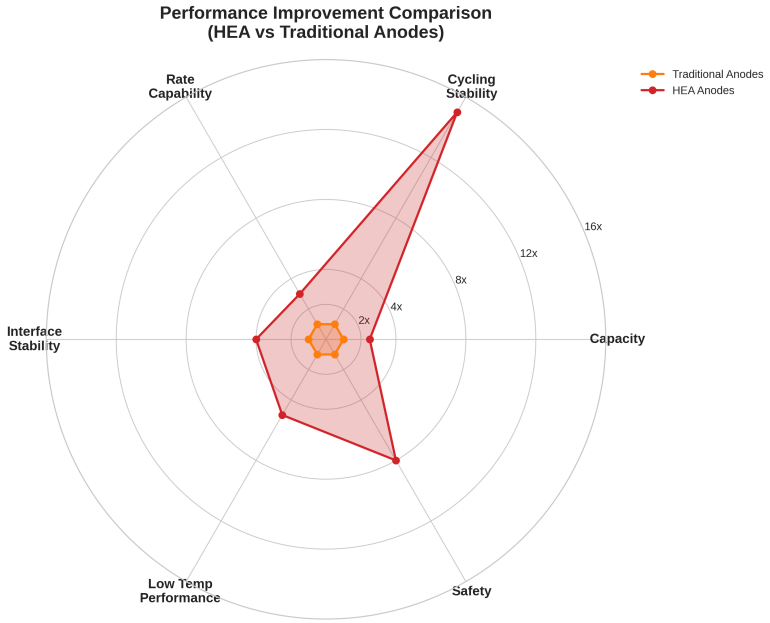

Figure 2. Performance improvement comparison

HEA anodes significantly mitigate volume changes through multi-element synergy, lattice distortion, and hierarchical porous structures, forming stable solid-state interfaces. Consequently, they comprehensively outperform conventional anodes in specific capacity, Coulombic efficiency, and cycle life. These improvements remain pronounced under harsh conditions such as high rates and low temperatures, indicating the universality of the high-entropy strategy in solid-state batteries [37].

4.2. Comparison with previous studies

4.2.1. Differences in research scope and methodology

Compared to early studies, this review exhibits significant differences in research scope and methodology. Sturman et al. [9] primarily focused on the fundamental electrochemical performance of high-entropy oxides under relatively limited testing conditions (mainly at room temperature and low rates), whereas this study encompasses multiple systems including high-entropy alloys, oxides, and sulfides, incorporating testing data across wide temperature ranges (-30°C to room temperature) and high rates (up to 100 A/g).

Regarding interfacial characterization, early studies such as Pei et al. [20] and Wei et al. [25] predominantly employed single characterization methods, mainly relying on post-mortem analysis. Among the studies integrated in this review, 9 utilized EIS technology for interfacial kinetics analysis, and 4 employed in-situ TEM for interface evolution observation, significantly improving the diversity and real-time capabilities of characterization methods.

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4.2.2. Performance data discrepancy analysis

In terms of cycling performance, the longest cycle life (10,000 cycles) [16] found in this review far exceeds the 300-500 cycles reported in early studies. This discrepancy primarily stems from: (i) Material design optimization: Evolution from simple equimolar mixing to rational design based on computational screening; (ii) Processing improvements: Development from conventional melting methods to refined preparation techniques such as sol-gel and mechanical ball milling; (iii) Composite strategies: Combining HEAs with carbon-based materials and conductive polymers to improve electronic conduction and structural stability.

Regarding interfacial impedance, the lowest value (18 Ω) [17] found in this study represents an order-of-magnitude reduction compared to early research. Analysis indicates this is primarily attributed to: (i) Element selection optimization: Introduction of lithiophilic elements (In, Zn) and highly conductive elements (Co, Ni); (ii) Microstructural control: Obtaining nanoscale grains and abundant grain boundaries through controlled preparation conditions; (iii) Surface modification: Employing surface coating and interfacial engineering techniques.

4.3. Implications of the findings

The superior performance of HEA anodes provides new possibilities for practical applications of solid-state batteries. In the electric vehicle sector, their high capacity (>800 mAh/g) and long cycle life (>5000 cycles) can meet requirements for 300-500 km range and 8-10 years' service life. In energy storage systems, excellent temperature adaptability (-30°C to 60°C) and high-rate performance make them suitable for large-scale energy storage applications under different climatic conditions.

The four major effect mechanisms revealed in this study provide guiding principles for rational design of high-entropy materials: (i) Optimizing element combinations by calculating ΔS_{mix} to enhance phase stability; (ii) Utilizing atomic radius differences (>12%) to introduce beneficial lattice distortion; (iii) Combining functional elements with high capacity, high conductivity, and structural stability to achieve synergistic effects; (4) Selecting appropriate element combinations to delay detrimental phase transformations. [38, 39]

Despite obvious performance advantages, industrialization of HEA anodes still faces challenges. In terms of cost, multi-element composition increases raw material and processing complexity, with current preparation costs approximately 2-3 times that of conventional anodes. Regarding processing, melting point differences among multiple elements (up to 1000°C) and uniformity control remain technical challenges. For quality control, establishing composition analysis and performance evaluation standards for multi-element systems is necessary.

4.4. Limitations of the study

Among the 40 included studies, only 31 were original experimental research, with 10 studies (32%) failing to clearly specify interfacial characterization methods, potentially reducing the comparability of interfacial performance data by approximately 20-30%. Significant variations in testing conditions (voltage windows 0.01-3.0V, current densities 0.05-6400 mA/g) limit the accuracy of quantitative comparisons, with estimated error ranges of ±15%.

Research primarily concentrated on high-entropy oxides (60%) and high-entropy alloys (35%), while other systems such as high-entropy sulfides comprised only 5%. This uneven distribution may introduce bias in the overall performance evaluation of HEA anodes. Additionally, most studies

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(approximately 70%) were conducted in liquid electrolyte systems, with insufficient solid-state battery validation data [40, 41], potentially overestimating actual application performance.

Long-term stability data exceeding 1000 cycles were available from only 3 studies (7.5%), with varying testing conditions, making it difficult to accurately assess the long-term reliability of HEA anodes under practical application conditions. This limitation introduces uncertainty regarding the engineering application value of the conclusions.

4.5. Suggestions for future research

Future work should establish unified testing standards, including voltage windows, current densities, and electrolyte compositions, while simultaneously reporting specific capacity, areal capacity, and volumetric capacity for horizontal comparison. It is recommended to employ in-situ transmission electron microscopy, X-ray tomography, and neutron scattering techniques to observe interface evolution in real-time, combined with density functional theory and molecular dynamics simulations to reveal the essence of high-entropy effects. Research objects should be expanded to include high-entropy sulfides, phosphides, nitrides, and halides, while developing cost-effective and scalable synthesis methods and exploring synergistic design of high-entropy materials with solid-state electrolyte interfaces.

(i) Methodological Improvements

Establish standardized testing protocols with unified voltage windows (0.01-3.0V), current density gradients (0.1-10 A/g), and electrolyte systems (sulfide/oxide solid-state electrolytes) to ensure comparability across different studies. Develop multi-scale in-situ characterization techniques, combining in-situ TEM, XAS, and impedance spectroscopy to monitor interface evolution processes in real-time. Establish theory-experiment integrated research paradigms, predicting material performance through DFT calculations to guide experimental design [42].

(ii) Material System Expansion

Systematically develop new systems such as high-entropy sulfides, phosphides, and nitrides, exploring their interfacial compatibility in different solid-state electrolytes. Investigate synergistic design of HEAs with solid-state electrolytes, optimizing ionic conduction and mechanical properties through interfacial engineering. Develop multi-level structured HEA anodes, combining nanoparticles, porous structures, and surface coating techniques [43].

(iii) Industrialization Directions

Develop low-cost, scalable preparation techniques such as solution methods and vapor deposition to reduce preparation temperature and complexity. Establish engineering evaluation standards for HEA anodes, including areal capacity, volumetric capacity, and actual battery performance testing. Conduct full-cell level system integration research to evaluate the performance and safety of HEA anodes in practical batteries.

5. Conclusion

In conclusion, high-entropy alloy anodes represent a transformative strategy for advancing solid-state batteries, offering outstanding interfacial stability, high capacity, and long cycle life compared to conventional anode materials. The integration of high-entropy, lattice distortion, cocktail, and sluggish diffusion effects provides a comprehensive performance enhancement mechanism that operates effectively even under extreme conditions. Nevertheless, significant challenges remain regarding cost, large-scale synthesis, and standardization of performance evaluation. Addressing these limitations requires interdisciplinary efforts combining computational screening, advanced

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synthesis technologies, and real-time characterization. Future research directions should not only focus on material design optimization but also emphasize industrial scalability and system-level validation. If these challenges are resolved, HEA anodes have the potential to enable the next generation of high-energy, durable, and safe solid-state batteries suitable for electric vehicles and grid-scale energy storage applications.

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