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Research progress and application prospects of iron-based cathode materials for all-solid-state batteries

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Abstract. Fully solid-state batteries, with their inherent high safety and high energy density, are emerging as a core area of development for next-generation electrochemical energy storage technologies. However, cathode materials remain a critical bottleneck in determining their overall performance. Iron-based cathode materials offer advantages such as abundant resources, low cost, environmental friendliness and flexible lithium storage mechanisms, and have seen a series of breakthroughs in the field of all-solid-state batteries in recent years. This review systematically examines the electrochemical reaction mechanisms, performance characteristics and modification strategies of lithium iron phosphate-based insertion materials, halide-based mixed-conducting materials, and sulfide and oxide-based transition materials. It points out that poor solid–solid interface compatibility, slow reaction kinetics and significant volume effects during charging and discharging are common core challenges across these material types, with interface engineering, integrated electrode design and self-healing mechanisms representing key paths to overcoming these bottlenecks. Lastly, looking to the future, the industrialisation of iron-based cathode materials requires a focus on multi-scale co-design, as well as breakthroughs in low-cost, large-scale fabrication processes and full-cell integration technologies, thereby providing the core foundation for the commercial application of high-safety, low-cost all-solid-state batteries.

Keywords: all-solid-state batteries, iron-based cathode materials, interface engineering, conversion reactions, monolithic electrodes

1. Introduction

The global energy mix is undergoing an accelerated transition towards clean, low-carbon sources. Hence, the development of electrochemical energy storage technologies that are highly safe, have high energy density and a long cycle life is a key foundation for supporting the application of new energy vehicles and large-scale energy storage. Conventional liquid lithium-ion batteries pose a safety risk of thermal runaway due to the use of flammable organic electrolytes. Furthermore, their energy density is gradually approaching its theoretical limit, making it difficult to meet future demands for electric vehicles with extended ranges and for large-scale, long-duration energy storage systems [1, 2]. Solid-state batteries replace liquid electrolytes with non-flammable inorganic or polymer solid-state electrolytes. Thus eliminating the risk of fire at source. They are also expected to be compatible with high-capacity lithium metal anodes, enabling a leap in overall battery

energy density. As such, they are widely regarded as one of the key directions for next-generation energy storage technology [3, 4].

Current mainstream cobalt-based and high-nickel trivalent cathode materials face bottlenecks such as resource limitations, high costs and significant environmental toxins, making it difficult for them to meet the dual requirements of low cost and high energy density demanded by all-solid-state batteries [1, 5]. In contrast, iron-based cathode materials have attracted considerable attention due to their unique combination of advantages: iron is abundant in the Earth's crust and the cost of raw materials is significantly lower than that of cobalt-based materials. The multiple valence states of iron ($Fe^{2+}/Fe^{3+}/Fe^{4+}$) support both insertion and conversion lithium storage mechanisms, with a wide theoretical specific capacity range (170–1,007 mAh g^{-1}), enabling a balance between current industrial transition needs and long-term high-energy-density goals [6]. Furthermore, in all-solid-state systems, they effectively circumvent the dissolution issues of transition metals commonly found in liquid batteries, demonstrating good compatibility with solid-state electrolytes.

In recent years, iron-based cathodes have made progress in various aspects of all-solid-state battery technology. However, existing research has largely focused on single-material systems, with a lack of comprehensive comparisons between different technical approaches, as well as a lack of systematic reviews of new strategies such as integrated electrodes and self-healing mechanisms. Accordingly, this review aims to systematically summarise the research progress on iron-based cathode materials for all-solid-state batteries, analyse the common challenges they face and the strategies for addressing them, and outline the future direction of their industrialisation, with a view to providing guidance for research, development and application in this field.

2. Advances in iron-based cathode materials for all-solid-state batteries

According to the lithium storage mechanisms, material properties and industrial maturity, iron-based cathodes for all-solid-state batteries can be divided into four main categories. A full comparison of the core performance characteristics of each material is presented in the Table 1.

2.1. Lithium iron phosphate

Lithium Iron Phosphate ($LiFePO_4$, LFP) has an olivine crystal structure. The theoretical specific capacity is 170 mAh g^{-1} and the average discharge potential is 3.4 V (vs. Li^+/Li). This is the iron-based cathode material with longest research history and highest industrial maturity in all-solid-state batteries [7, 8]. Its main advantage is the structural stability is very good and the volume change is only 6.8% during charging and discharging and it can maintain solid-solid interface contact with the solid-state electrolyte to greatest extent possible [8]. This material can be used as ideal model for testing new electrolytes and interface strategies.

In all-solid-state batteries, ion transport is a difficult problem because no liquid electrolyte exists and the conductivity is low. It is worth noting that 30–40% inactive solid electrolyte and conductive additive are needed to create paths for ions and electrons. However, this can limit energy density of battery.

So most studies focus on interface engineering and electrolyte optimisation. For example, by applying a $LiNbO_3$ coating, the interfacial impedance between LFP and sulfide electrolytes (such as Lithium Phosphorus Sulfur Chloride, LPSC) can be reduced from 380 Ω cm^2 to 110 Ω cm^2 [9]. Also, special electrolyte structures can be designed, such as biomimetic gradient solid-state electrolytes. When used with LFP all-solid-state batteries, these can give a reversible capacity of 136.3 mAh g^{-1} at 1C rate, and capacity retention is 93.8% after 200 cycles [10]. Also, for sustainable development, direct recycling technologies for spent LFP batteries can

work well. In fact, batteries made from recycled materials and sulfide electrolytes can achieve capacity retention of 91.2% after 200 cycles [11].

In short, LFP is key material for commercialisation of all-solid-state batteries. But the theoretical capacity limit makes it hard to get a big increase in energy density. Most studies have focused on laboratory-scale button cells. It is worth noting that there is a severe lack of research on compatibility of high-surface-area electrodes for Ah-class pouch cells and this can be a key area where further breakthroughs are needed.

2.2. Halide-based iron cathodes

LFP and other insertion-type materials are used. Also, the iron-based halide cathodes can provide new options for all-solid-state batteries because they have unique ionic conductivity and higher theoretical capacity in principle, and this characteristic means they can be a good choice for next-generation energy storage systems with better performance than traditional cathodes. In fact, this system can be developed along two tracks: chloride and fluoride. Both approaches have distinct lithium storage mechanisms and core strengths, but at the same time they can face a common challenge that makes it hard to achieve stable compatibility with the solid-state electrolytes when building a full battery system with good performance.

2.2.1. Iron-based chlorides

In iron-based chlorides, $L_{i1.3}Fe_{1.2}Cl_4$ shows some advantages [12]. It has high ionic conductivity (about 2.28×10^{-4} S cm^{-1}) and the electronic conductivity is also high (about 6.98×10^{-5} S cm^{-1}). It is worth noting that it can work as active material and ionic conductor. Also it is electronic conductor at same time. With this property, electrodes can be fabricated without additives and it is integrated. This can help increase active material ratio in the electrode.

It should be noted that this material has self-healing properties. In charged state, the Young's modulus is only about 0.52 GPa and it shows creep behaviour similar to polymer. This helps fill microcracks and interfacial voids generated during cycling. However, in discharged state, the modulus can recover to higher level (about 5.69 GPa) to maintain structural integrity of the electrode [12]. Because of these properties, the batteries based on this material can maintain capacity retention rate of 90% after 3,000 cycles at 5C rate. They can also keep stable performance in wide temperature range from –10°C to 60°C. Also, the electrode cost is 74% lower than conventional electrodes [12]. This design idea also inspired new solid-state electrolytes. For example, in later studies, when reactive oxygen halide electrolytes were paired with LFP, the theoretical energy density of full cell reached 982.1 Wh kg^{-1} [13]. However, such materials can face limitations in practical application. They are sensitive to air and have insufficient oxidative stability at high voltages. This makes it hard to produce them in large scale.

2.2.2. Iron-based fluorides

Different from chlorides, the iron-based fluorides can show a relatively high theoretical specific capacity (712 mAh g^{-1}) because the reaction involves three-electron transfer [14]. But they can have volume changes about 25% during charge and discharge. In fact, the discharge product LiF has very low electronic conductivity and this can cause slow reaction kinetics so capacity can decay fast [14].

These challenges make it hard to get good cycling performance in practical applications. In fact, interface engineering is key. FeF_3 can be coated using fast ionic conductor Lithium Magnesium Zirconium Phosphate (LMZP). This reduces interfacial impedance with sulfide electrolyte from 320 Ω to 145 Ω. Then the battery can keep capacity of 310 mAh g^{-1} after 600 cycles [15]. Also, Another strategy involves interface chemical controls. For example, forming boron-doped electrode-electrolyte interface phase can extend cycle life of FeF_3 electrodes to over 1,000 cycles and the capacity retention rate can reach 70% [16].

2.3. Sulfide and oxide iron-based cathodes

Besides halides, iron-based sulfides and oxides rely on conversion reaction mechanisms. They can attract attention. It is worth noting that the theoretical capacity is high and these materials should be important class for developing all-solid-state batteries with high energy density.

2.3.1. Sulfides

Sulfide cathodes like iron disulfide (FeS_2) can show a high theoretical specific capacity (894 mAh g^{-1}) and pyrite is cheap as raw material [17]. But lithium storage process has multiple phase transitions with volume expansion and particle fragmentation and the reaction kinetics can be slow and the cycling stability is poor [17].

To get better performance, researchers focus on nanoscale modification and composite electrode structures and nanoscale modification of FeS_2 can shorten the ion diffusion path. Then the reversible capacity is increased by about 30% at 0.1C. Also the interfacial impedance with the sulfide solid-state electrolyte is reduced [17]. At the same time, it is crucial to establish a reliable electronic conductive network. In fact, when carbon nanotubes are incorporated to form a 3D conductive structure, the charge transfer impedance of the electrode can be reduced from 210 Ω to 95 Ω [18]. To expand the application range, the combination of FeS_2 with new super-ion conductors (such as LASI-80Si) can work in wide temperature range from –60°C to 60°C. Long cycle life is also obtained at high area capacity (9.05 mAh cm^{-2}) [19].

But the industrialization still faces two challenges about mechanism and process. The migration of insulating Li_2S and polysulfides produced during cycling can be main reason for capacity decay. In fact, the reaction mechanisms and interfacial evolution processes need more study.

2.3.2. Oxides

Iron oxides (like Fe_3O_4) have very high theoretical specific capacity (1,007 mAh g^{-1}), but they can have large volume change during cycling (> 80%) and the capacity drops fast. So they are not used as practical cathodes now [20, 21]. In fact, its main value is as model system for basic research. Because it has good conductivity, it can make ultrathin film electrodes without the conductive additives or binding agents, and then build simplified all-solid-state battery models. Researchers use this method to study basic scientific problems, such as the electrochemical and mechanical behavior at solid-solid interfaces [20, 21].

2.4. Comprehensive comparison of different material systems

In short, iron-based cathodes for all-solid-state battery can store lithium through two mechanisms: intercalation and conversion. These materials have evolved into different systems with distinct roles and complementary advantages. It is worth noting that the Table 1 gives a full comparison across multiple dimensions, including theoretical and practical capacity, core advantages, challenges, and industrialization maturity, and their technical characteristics and current development state can be understood.

The table shows differences in positioning among various material systems. Lithium Iron Phosphate (LFP) has good cycle stability and the highest industrial maturity, so in principle researchers can see it as the most reliable interim choice at this stage. Iron-based chlorides provide new approach to integrated electrode design because they have inherent conductivity and self-healing properties. However, their engineering applications still have problems with stability. Also, sulfide and fluoride systems have advantages in terms of high capacity but they have problems such as volume expansion and interfacial side reactions and they are now at pilot-scale exploration stage and laboratory optimization stage respectively. It is worth noting that iron-based oxides are used as model materials for studying interfacial behavior.

Table 1. Comprehensive comparison of iron-based cathode materials for all-solid-state batteries

Materials System	Representative materials	Theoretical specific capacity ($mAhg^{-1}$)	Actual reversible specific capacity ($mAhg^{-1}$)	Key strengths	Key challenges	Level of industrial maturity
Phosphate-embedded	$LiFePO_4$	170	130-160	Structurally stable, long cycle life, low cost, and mature industrialisation	Low upper limit of energy density, low intrinsic electrical conductivity	Pilot-scale validation completed; highest level of maturity
Chloride-based conductive compound	$Li_{1.3}Fe_{1.2}Cl_4$	145	140-150	Integrated design, self-healing, ultra-long cycle life, extremely low cost	Low capacity and insufficient air stability	Laboratory validation phase, with significant disruptive potential
Fluoride-converting type	FeF_3	712	300-520	Extremely high theoretical capacity, relatively high operating voltage	Significant volume effect and severe kinetic hysteresis	Laboratory optimisation phase
Sulfide conversion type	FeS_2	894	500-890	High capacity, extremely low cost, and excellent adaptability across a wide temperature range	Multi-step phase transitions are irreversible and result in significant volume expansion	In the early stages of pilot testing, there is significant potential in wide-temperature-range scenarios
Oxide-conversion type	Fe_3O_4	1007	300-500	Offering the highest theoretical capacity and an abundance of resources	Extreme volume effect and poor cycling stability	Basic Mechanisms Research Phase

3. Key challenges and regulatory strategies

It is noted that iron-based cathode materials in all-solid-state battery face some key problems, because solid-solid interface compatibility is poor, bulk charge transport is slow, and volume changes during charging and

discharging is significant. These factors interact with each other. It is worth noting that they can limit battery overall performance.

3.1. Solid-Solid interface compatibility challenges and interface engineering

In fact, the solid-solid interface in all-solid-state batteries can affect battery performance. The failure mechanisms include chemical reactions and electrochemical barriers, where at chemical level, mutual diffusion of elements and side reactions can lead to formation of high-resistance interfacial phases, and at the electrochemical level, the space charge layer can restrict ion transport. At the same time, volume changes during cycling can cause interfacial contact failure at mechanical level [6, 22]. For example, sulfide-based LPSCs can undergo oxidative decomposition at voltage around 3.8 V, and the interfacial resistance increases, which shows that interfacial thermodynamic instability is a difficult problem [23].

To control the interface, isolating it and modifying the structure are required so that the interface is expected to become stronger. In fact, forming artificial buffer layers on surface of active particles is a common approach, where $LiNbO_3$ can block side reactions. Then researchers promote formation of in-situ interfacial phases with high ionic conductivity so that the system can achieve self-stabilization, and this method can work together with other strategies to improve the overall performance of the battery system. Also the oxidative stability and mechanical properties of electrolyte itself should be improved [9, 10, 13].

3.2. Charge transfer kinetics hysteresis and optimization of transport networks

Iron-based cathodes have low conductivity. Also, the conversion products (such as LiF and Li_2S) are insulators and can make the charge transport bottleneck worse. Then the rate performance of battery is expected to be poor.

The aim is to build a dual-channel network for ions and electrons. This system should be robust with good efficiency. Common methods include reducing active materials to nanoscale and using three-dimensional conductive frameworks (such as carbon nanotubes [17, 18]) to shorten transport paths, and the conductivity can be improved by element doping. In fact, a key method is to develop inherent mixed conductors, such as iron-based chlorides ($Li_{1.3}Fe_{1.2}Cl_4$), and it can conduct both ions and electrons. Then it can be designed additive-free integrated electrodes [12].

4. Outlook for industrialization

For iron-based cathode all-solid-state batteries to reach practical application, they must overcome multiple barriers spanning from material innovation to engineering integration.

In fact, industrialization now faces several major systemic challenges. In material aspect, it is still hard to find a perfect option that can deliver high energy density and long lifespan at the same time and the cost should also be kept low. At present, each existing system has significant shortcomings. In the battery manufacturing, There remains no well-established solution capable of ensuring the uniform transport of both ions and electrons concurrently and the overall interfacial stability can also be maintained under high areal loading, which is a problem not solved yet. Also, there is urgent need to develop fabrication processes with low cost and good scalability for all-solid-state batteries and it is worth noting that unified standards with good reliability for performance evaluation and safety evaluation should also be established.

Further development should center on several critical fields. At basic research level, in-situ characterization and computational modeling can be employed to reveal the degradation mechanisms of batteries under realistic operating conditions when battery works under high load particularly, because this can help us know

the failure process better. It is worth noting that for technology innovation and studying new stable iron-based compounds. In fact, the design ideas like integration and self-healing should be used in more material systems. At the same time, innovative processes like dry-process electrode technology are being explored and optimized and this can help solve the problems in making thick electrodes. Finally, this technology can be put into market only after passing the cycle life testing for Ah-class pouch batteries and establishing a standardized system, because practical performance must be reliably ensured for real-world applications and this requires collaboration across industry chain which can be supported by the standard system.

5. Conclusion

The research progress of iron-based cathode materials for all-solid-state batteries is systematically reviewed, and according to the lithium storage mechanism, existing systems can be divided into four categories that show different electrochemical behaviors. Lithium Iron Phosphate (LFP) is the most mature intercalation material and because of its stable structure it can be a reliable choice for current industrial transition. Iron-based chlorides have conductive and self-healing properties at the same time and they can pioneer a new paradigm for integrated electrodes and should hold potential for transformative change in battery field. Sulfides and fluorides possess high theoretical capacities. However, their practical application makes it hard to achieve good performance because the conversion reaction causes drastic volume changes and interfacial instability that are difficult to control in solid-state battery system and this leads to fast capacity fading. Also, iron-based oxides are mainly used as model materials for fundamental research. It is worth noting that these materials have different development paths, but they share core scientific challenges including poor solid-solid interface compatibility, slow charge transport kinetics, and there are significant volume effects that limit the cycle life and make the battery fail early. In fact, to improve their performance, material engineering and interface control strategies are expected to become key. For example, artificial buffer layers can be constructed and three-dimensional conductive networks can be designed. Self-healing mechanisms can also be introduced.

Different material systems are compared in this review. The strengths and limitations are examined. The suitable applications of various technologies are also explored to facilitate material selection based on specific objectives. In future, the development of iron-based cathodes should proceed with a coordinated effort in materials innovation and engineering because only through combined approach the existing problems can be solved and it is worth noting that the goal is to complete integration and validation of Ah-level batteries and a standardized evaluation system should be established.. This can help the transition of iron-based cathodes from laboratory to large-scale applications, and it can provide material foundation for all-solid-state batteries with high safety and low cost.

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