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Editorial

Brief Introduction on Manufacturing and Characterization of Metallic Electrode and Corresponding Modified Materials

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Abstract: As an important part in new energy storage devices, electrodes containing metals or their corresponding derivatives are widely used due to the diversity of material types, existing forms and assembly methods. In order to obtain novel energy storage components with superior performance, new technologies and studies on the improvement of electrode materials are emerging in recent years. This editorial paper aims to summarize the classical and latest research highlights on manufacturing, characterization and modification of metallic electrodes, especially new materials.

Keywords: energy storage device; metal batteries; metallic electrode materials; experimental synthesis and manufacturing; advanced characterization methods

Citation: Huang, Q.; Wang, S.; Chen, Y.; Liu, C.; Li, Q. Brief Introduction on Manufacturing and Characterization of Metallic Electrode and Corresponding Modified Materials. *Metals* **2023**, *13*, 703. https://doi.org/10.3390/met13040703

Received: 12 February 2023
Revised: 28 February 2023
Accepted: 15 March 2023
Published: 3 April 2023

1. Introduction

The energy storage devices with metal electrode materials and relating techniques have been greatly developed in recent years [1], especially based on the metal batteries including lithium (Li) [2] /sodium (Na) [3] /potassium (K) [4] /zinc (Zn) [5] /aluminum (Al) [6] /magnesium (Mg) [7] /calcium (Ca) [8] cells, metal/air batteries, secondary batteries, solar energy storage and catalytic hydrogen production, which have played significant roles in a wide range of application scenarios. However, with the increasingly prominent global energy problems, the demand for the development of new energy storage devices and technologies might continue to increase.

At present, there are still some defects in metallic electrode materials, especially in large-scale manufacturing. In order to meet the requirements of energy storage, many scholars have explored the new high-performance metal electrode material system, and some improved preparation ways have been put forward, which provided a basis for developing the large-scale production process for the next generation of chemical energy storage devices. In addition, advanced characterization methods were constantly emerging, which offered basic support for regulating the structure and properties of metallic electrode materials.

In this paper, pure/abnormal metal and metallic compound electrodes would be emphasized, respectively.

2. Metal Electrode

In recent decades, lithium-based batteries have dominated the development of high-performance batteries because of the low density, high voltage and excellent electrochemical equivalent to 3860 mA·h/g for lithium metal among various negative electrode materials [9]. Lithium metal cells are difficult to commercialize even nowadays, which is mainly due to the problems of poor cycle stability and low coulombic efficiency (CE) of lithium anode during charging and discharging processes. On the one hand, the root cause of the above defects is that there is a serious side reaction between lithium anode and electrolyte, and another reason is the obvious volume change during the cycling of lithium anode [10]. In unit time, the flux of metal ion diffusion to different positions of the electrode would change while the current density and deposition speed at various positions on the surface of the metal electrode also differ. When the current density is high, the deposition speed of metal electrode is fast, and the tip growth occurs.

The unwanted deposition of lithium dendrites (as shown in Figure 1) and Li metal on foil electrode is the main cause of damage to the performance of lithium metal cells. The replacement and alloying reactions between special ionic liquid and lithium foil were applied to construct Li alloy anode to solve the use problems caused by lithium dendrite growth inside lithium metal batteries [11]. Coralloid carbon fiber (CFs) was prepared by electroplating silver (Ag) particles on the surface of CFs in a published work [12]; then, the composite lithium metal anode CF/Ag-Li could be obtained by siphoning molten lithium, and the synthesized electrode could adjust the nucleation and growth of lithium during lithium plating as well as reduce the deposition degree of lithium during the reactions.

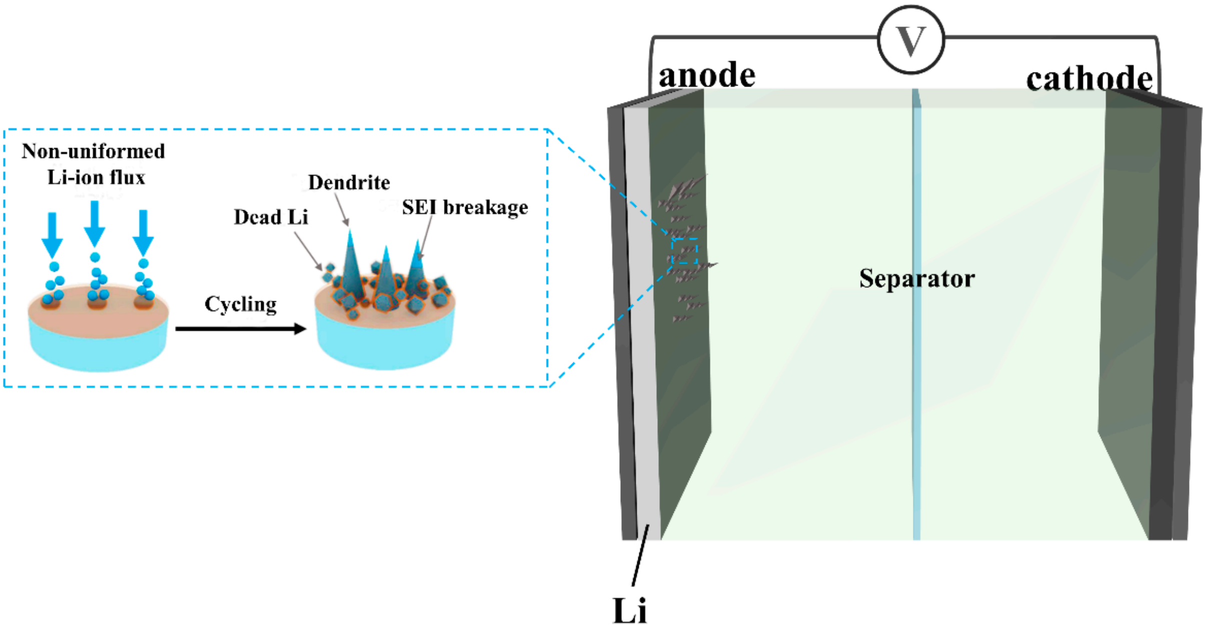

Figure 1. Formation processing of lithium dendrites in lithium metal batteries.

The working principle of Na metal batteries is similar to that of Li ones. With high specific capacity of 1166 mA·h/g and low redox potential of −2.71 V, normal hydrogen electrode (NHE), as well as abundant sodium element and much lower price than that of lithium, sodium-ion battery (SIB) is expected to replace lithium-ion battery (LIB) as the most promising next-generation energy storage equipment. On the contrary, the growth of sodium dendrites in Na metal cells is always out of control, and the unstable solid electrolyte interface (SEI) layer would also limit the development of Na-relating cells [13].

Compared with lithium dendrites, there are more serious disadvantages to those of sodium and SEI layer inside Na metal cells. Currently, the new combination of metallic salt and solvent to obtain advanced electrolyte has become the research focus to realize the stable cycle of sodium metal battery [14]. By studying new electrolytes [15] or changing the concentration of solute [16], the cyclic performance of sodium metal battery could be enhanced and stabilized.

The working principle of potassium battery is similar to that of lithium and sodium cells. K metallic electrode has high theoretical specific capacity and extremely low electrode potential, but it still faces many problems when modified and used in potassium-ion batteries (PIBs). Potassium has high reactivity, which requests super-clean assembly environment for batteries containing it [17]. At the same time, during the cycling, the side reactions on the interface between negative electrode and electrolyte are usually violent, which leads to the loss of active substances. In addition, the formation of potassium dendrites causes the premature failure of the battery [18]. The interfacial side reactions caused by solvent and active oxygen can be alleviated by in situ synthesis of film on the surface of metal potassium [19]. It is also possible to synthesize an alloy SEI layer such as K-Hg to adjust the interface reactions [20].

Although zinc and aluminum are bivalent and trivalent, respectively, their metal cells are facing problems and challenges similar to those of lithium, sodium and potassium batteries, and the solutions are basically referable [21,22].

Magnesium–sulfur (Mg-S) battery is a strong competitor of lithium–sulfur (Li-S) one, and many scholars have studied their performance and stability [23]. The interfacial reactions between electrode and electrolyte will cause serious capacity decline, and adding additives can improve the performance and compatibility of cathode materials [24]. Besides electrolyte, designing suitable sulfur carrier to avoid the electrical insulation performance of sulfur is another obstacle for Mg-S battery [25].

Perovskite solar cell is a new generation in photovoltaic cells, and its performance is mainly related to the distribution of $CaTiO_3$ compounds existing in perovskite films [26]. Controlling the distribution of perovskite thin films by depositing perovskite via various physical and chemical methods is the focus of research in recent years [27,28].

To sum up, some physical and chemical parameters of several pure metal electrodes are listed in Table 1.

Table 1. Some physical and chemical parameters of several pure metal electrodes [13–24].

Metal	Redox Potential (V vs. NHE)	Mass Specific Capacity (mA·h/g)	Capacity Density (mA·h/cm^3)	Melting Point (°C)	Crustal Abundance (ppm)
Li	−3.045	3860	2061	108	18
Na	−2.714	1166	1128	97.72	22,700
K	−2.928	685	591	63.65	21,000
Mg	−2.372	2204	3835	648	27,640
Ca	−2.868	1337	2072	842	46,660
Zn	−0.762	819	5851	419	70
Al	−1.662	2979	8043	660	83,000

3. Abnormal Metal Electrode

3.1. Foamed Metal Battery

Metal foam material has high porosity and large specific surface area, so it can be filled with more active substances, which increases the capacity of the battery and greatly reduces the real current density of the electrode. Therefore, it is an ideal material for manufacturing electrodes of various batteries such as storage batteries/accumulators, air batteries, fuel cells and solar cells, and it provides the possibility for the development of lightweight batteries with low internal friction, long service life and high specific energy [29]. Nowadays, the commonly used metal foam electrodes mainly include foamed nickel (Ni) [30], foamed lead (Pb) [31], and foamed zinc (Zn) [32].

The performance of metal foam electrode mainly depends on the amount of active substances it carries [33], which depends on the porosity of the foamed metal electrode [34]. In the recent years, a lot of methods have been proposed to produce metal foam with high porosity which could reach 70~90% after adding porosity agent by using loose powder sintering method [35]. The porosity of foamed metal electrode material produced by electrodeposition could increase to be 80~99%, the pore structure was evenly distributed, and the opening rate was high [36].

Metal foam electrode is of great significance for the development of batteries. Nevertheless, a large amount of research on metal foam electrodes is in the experimental stage, which is still far from large-scale application.

3.2. Liquid Metal Battery

Typical liquid metal battery includes two liquid metal electrodes and molten salt electrolyte, while the three types of liquid substances are naturally divided into three layers because of the density difference and incompatibility from each other [37], as illustrated in Figure 2.

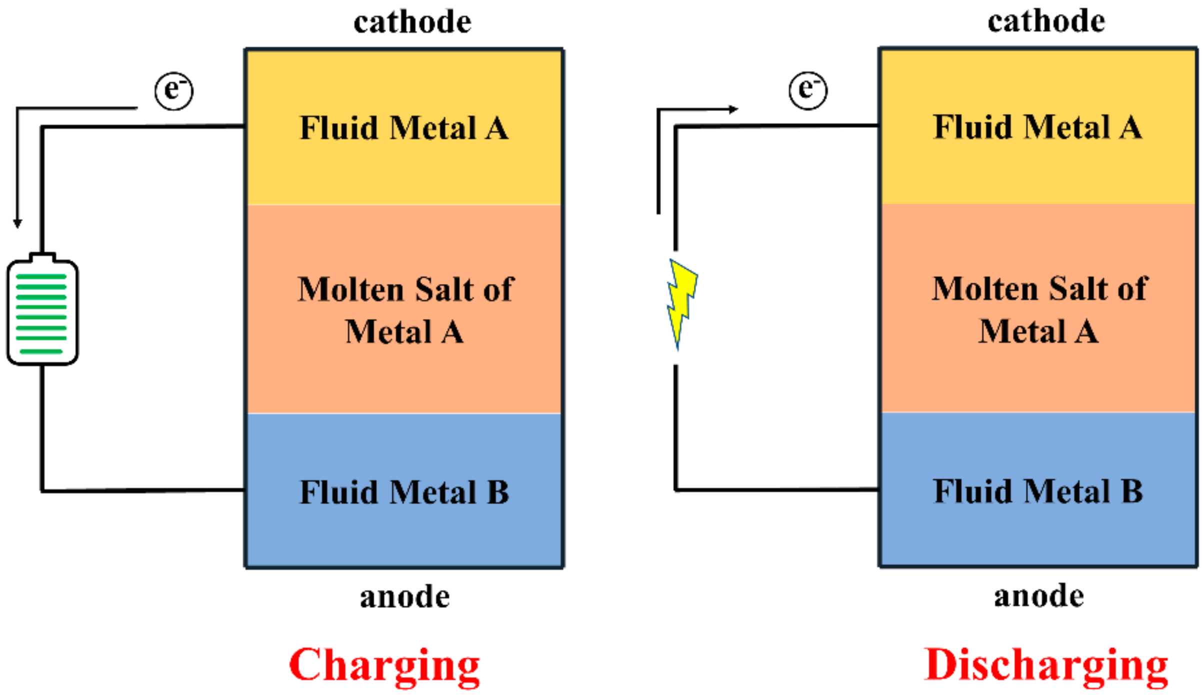

Figure 2. Charge and discharge processing of liquid metal battery.

Because of its unique structure and electrical conductivity, liquid metal electrode has the following advantages in the application of energy storage battery:

(1) During the charging and discharging process, liquid metal electrode can effectively avoid dendrite growth, phase transition and grain size change, thus the battery has a long energy storage life [37].

(2) Liquid metal electrode has good conductivity and rapid electrode interface dynamic response, which can meet the needs for high-power application fields [38].

(3) The structure of liquid electrode is simple, and the electrode interface is easy to construct; therefore, the period of time for battery assembly process is short, so the manufacturing cost is also relatively low [39].

Liquid metal batteries mainly include lithium-based battery [40], sodium-based battery [41], zinc-based battery [42] and aluminum-based batteries [43], magnesium-based battery [44], and calcium-based battery [45], the equilibrium voltage of which mainly depends on the activity difference of active components in metal electrodes A and B [46], as presented in Figure 2. The negative electrode is generally the liquid metal mentioned above such as Li, Na, Zn, Al, Mg, and Ca. Therefore, selecting appropriate metal as the positive electrode is a main mean to guarantee the performance of liquid metal batteries. Besides considering the balance voltage between the two electrodes, the cost should also be taken into account. Generally, Sb, Bi, Sn or Pb could be chosen as the positive electrode, and their applicability is very strong [47]. In addition, the electrodes used in liquid metal batteries are not strictly limited to pure metals; sometimes, alloy electrodes are applied, and the goal of electrode alloying is to reduce the melting point of the electrodes without obviously decreasing the voltage or increasing the material cost [48]. The elements added during alloying should not have negative effects on electrodes, electrolyte, insulators, current collectors and other assembly materials, and the alloyed electrodes should still meet the requirements of maintaining the density of three liquid layers [49].

4. Metallic Compound Electrode Materials

Layered metallic oxides, metallic nitrides and metal sulfides could be applied as metal-derived electrode materials whose energy storage principle is mainly Faradaic capacitance generated by highly reversible redox reactions or chemical adsorption/desorption, which have hence gained significant concerns among various new energy storage materials.

4.1. Metallic Oxide Electrode Materials

Metallic oxides have attracted much attention because of their high energy density, and their theoretical specific capacitance can reach 10–100 times that of carbon materials, and their stability is great [50]. The commonly used metallic oxide electrode materials include ruthenium oxide (RuO_2) [51], manganese oxide (MnO_2) [52], cobalt oxide (Co_3O_4) [53] and nickel oxide (NiO) [54]. Furthermore, the conductivity of bimetallic oxide electrode materials is several orders of magnitude higher than that of monometallic oxides, which can promote the transfer of electrons and ions [55] to achieve higher output power density. Moreover, multi-metallic oxides are more diversified than bimetallic oxides, which can effectively reduce the band gap width of electrode materials and improve the electric conductivity [56], and the diversity of components can regulate the electrochemical activity and stability of cells [57]. To realize the synergistic effect in components and obtain excellent comprehensive energy storage performance, the development of multi-metallic and bimetallic oxides is the current research hotspot [58]. Taking $ZnMn_2O_4$-Li battery as an example, the charging and discharging processes of batteries with typical metallic oxide electrodes is indicated in Figure 3.

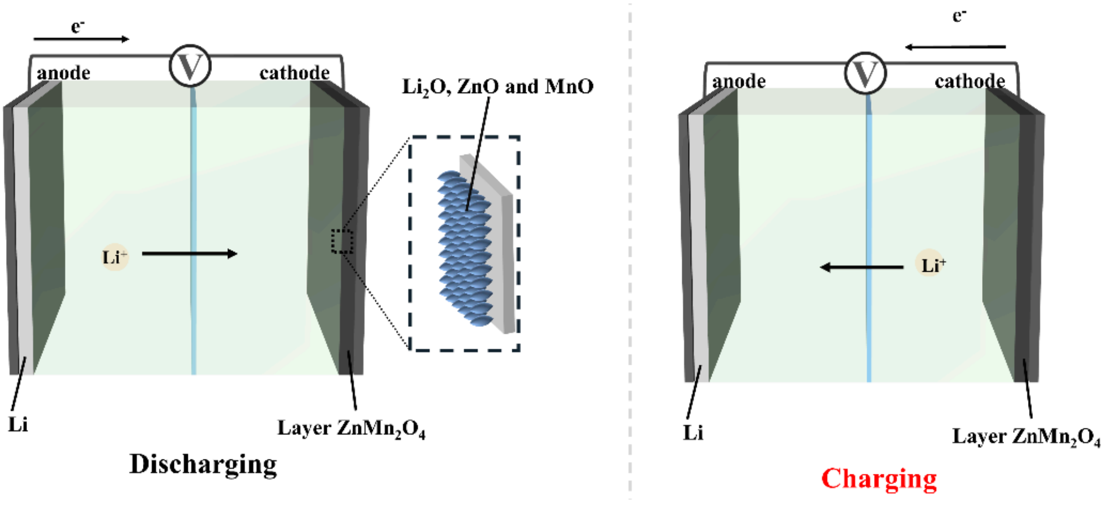

Figure 3. Charge and discharge processes of $ZnMn_2O_4$-Li battery with metallic oxide electrode.

4.2. Metallic Nitride Electrode Materials

The metallic nitrides produced from urea or ammonia gas have high state density because of the metal lattice deformation, which contributes to the excellent electrical conductivity. The energy storage performance can be effectively improved by transforming metallic oxides into highly conductive metallic nitrides via nitriding treatment under high temperature. At present, the commonly used metallic nitride electrode materials are titanium nitride (TiN) [59] and multilayered porous aluminium nitride (AlN) [60].

4.3. Metallic Sulfide Electrode Materials

Compared to oxygen atoms in metallic oxides, sulfur atoms in transition metallic sulfides have stronger electronegativity, and there are more abundant anions when the reversible redox reactions are active, so metallic sulfides could be ideal electrode materials due to their large theoretical capacity and long cycle life. Metallic sulfides generally exist in the form of nano-arrays, such as porous cobalt sulfide nano-sheet arrays [61]. Similar to metallic oxides, bimetallic sulfide electrode materials have superior properties than monometallic ones [62]. Regardless of whether the bimetallic sulfides were obtained from the combination with carbon nanomaterials or simple and easy-to-operate means such as hydrothermal method, they have been proved to be superior electrode materials after experiments [63–68].

5. Conclusions

Electrode materials containing metals and metallic derivatives with huge reserves showing redox activity, which is precisely the reason why metallic electrode materials have been the research focus in the field of energy storage devices since the last century. The constantly emerging innovative and efficient preparation and modification methods can greatly broaden the application scenarios and development prospects of new energy storage devices in the future. This paper mainly introduces various elemental metal electrode materials, those existing in abnormal conditions, metallic oxides, nitrides as well as sulfides, which provides some basis for valuable aspects to be investigated.

Manufacturing as well as characterization of metallic electrode materials are worthy and important themes in various fields in the past, present, and future. This Special Issue aims to promote further research activities in power supply/energy storage relevant fields, and accelerate the industrialization of high-performance metallic electrode materials. We sincerely invite high-quality contributions including but not limited to original research articles/reviews/case reports/communications/perspectives/viewpoints that present innovative and significant findings and experiences on this topic.

Author Contributions: Investigation, Q.H.; Writing—original draft preparation, Q.H.; Methodology, S.W.; Conceptualization, Y.C.; Writing—review and editing, C.L.; Project administration, Q.L. All authors have read and agreed to the published version of the manuscript.

Funding: This study has been sponsored by National Natural Science Foundation of China (No. 12202410 and No. 51906238), Research Project Supported by Shanxi Scholarship Council of China (No. 2022-139), Natural Science Foundation of Shanxi Province (No. 20210302123017) and Fund Program for the Scientific Activities of Selected Returned Overseas Professionals in Shanxi Province (No. 20220012). Also, this work was supported by Changsha Municipal Natural Science Foundation (No. kq2208277). The authors gratefully acknowledge this support.

Conflicts of Interest: The authors declare no conflict of interest.

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