Review of the application of metal-air battery principle in water treatment

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Abstract: The reaction principle of metal air battery is expounded, and the application of this method in the field of water treatment as an emerging technology is introduced. The recent research results of domestic and foreign researchers using metal-air battery technology to treat pollutants in water (such as removing arsenic, reducing COD and collecting algae in water) are introduced, and the actual treatment effects of these methods are briefly described. The feasibility of applying metal-air battery technology to water treatment is analyzed. At the same time, the problems of water treatment using metal-air battery are pointed out. The broad application prospects of this technology are analyzed and prospected.

1 Introduction

With the development of society, demand for energy is increasing. Traditional energy sources such as coal and oil have serious pollution problems. Therefore, electric energy and wind energy are receiving more and more attention as clean energy. Fuel cells are devices that generate electricity [1], whose principle is to convert chemical energy into electricity [2]. Metal-air battery is a new type of fuel cell. Compared with other fuel cells, it has high theoretical energy density and power density. It is very convenient to operate, the product is non-toxic or less toxic, and it is environmentally friendly. At present, there are practical applications in the fields of new energy vehicles [3], military [4] and navigation [5].

However, there are few studies on removal of pollutants in water by using metal-air batteries devices in the field of water treatment. At present, most of the water treatment methods are still by adding chemical agents [6-8] (such as adding flocculants, coagulants and oxidants, etc.) additional energy [9-10] or biodegradation of microorganisms to achieve the purpose of removing pollutants [11-12]. Metal-air batteries can achieve the purpose of removing certain pollutants in water while producing electricity [13], it is an emerging technology with great development potential. This article describes the reaction principle of metal-air batteries and introduces the application of metal-air batteries in water treatment.

2 The reaction principle of Metal-air battery

The metal-air battery is a special kind of electricity generating device between the primary battery and the fuel cell [14]. This type of battery is similar to the primary battery, but has some characteristics of the fuel cell [15]. It is a kind of electricity generating device between the primary battery and the fuel cell, which is called a "semi-fuel" battery [16]. Metal-air battery not only has some advantages of the fuel cell, but also overcomes some shortcomings of the fuel cell. Metal-air batteries can be classified into non-aqueous metal air batteries and water-based metal air batteries according to the materials between the electrodes [17]. Non-aqueous metal air batteries mainly use non-aqueous gel electrolyte as a substance between two electrodes [18]. It was less studied and applied; water-based metal-air batteries are more common, and the electrolyte between the electrodes is aqueous solution. At present, there are many researches on water-based aluminum-air batteries [19], lithium-air batteries [20] and magnesium-air batteries [21], and some achievements have been made.

The metal-air battery structure is shown in Fig 1. The anode is made of a metal material (such as iron, aluminum, zinc, magnesium, etc.), the cathode is an air electrode. The electrolyte is between two electrodes. The metal anode and the air cathode are connected by a wire to generate electric energy. The metal electrode of the metal-air battery acts as an anode, it loses electrons and undergoes oxidation reaction [22]. The oxidized metal becomes an

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ion electrode form and enters the electrolyte solution. The air electrode acts as a cathode. It is also a place for oxygen reduction [23]. The air cathode usually has a waterproof gas permeable layer, a gas diffusion layer, a conductive layer and a catalytic layer from the outside to the inside. The waterproof and breathable layer is a waterproof porous material, which has good gas permeability and avoids water oozing. The material is mostly Poly Tetra Fluoro Ethylene (PTFE); the gas diffusion layer is a place where the air is in full contact, and the layer has good gas permeability, so that the air can easily reach the catalytic layer for oxygen reduction reaction. The gas permeability makes it easier for the air to reach the catalytic layer for oxygen reduction reaction; the conductive layer is composed of a metal mesh, generally composed of a metal having good conductivity such as nickel, platinum and copper mesh, and the conductive layer plays a conductive role at the same time. The electrode can be supported to maintain a relatively fixed shape by conductive layer; the catalytic layer is important in the overall structure, the main component is a catalyst for catalyzing the oxygen reduction reaction, and the composition and types of the catalytic layer are various and non-fixed forms. Many people still work to study more effective catalysts.

![Fig.1. Diagram of Metal-air battery structure](image)

It is precisely because of the continuous oxidation of the metal anode that the oxygen reduction reaction of oxygen at the air cathode produces a continuous transfer of electrons, so that the device has a continuous current generation. In the half-reaction of the two electrodes, the metal anode is oxidized to a metal ion, which is similar in form to an ordinary battery, and the reaction occurring at the air cathode is an oxygen reduction reaction, and the electrode itself is the reaction and the ions in the electrolyte solution are not participating in the reaction. The element that reacts is oxygen in the air, which is similar to the one-half reaction in the hydrogen-oxygen fuel cell. It is because the positive and negative reaction forms are half similar to the original battery and the other half is similar to the fuel cell, so it is called a "semi-fuel" battery. The specific reaction principle is slightly different for different metals and different electrolyte systems, but overall it is still the oxidation of the metal electrode and the oxygen reduction reaction on the air electrode. The principle of some metal-air batteries is shown in Table 1.

![Table 1 The principle of some metal-air batteries.](image)

According to the reaction principle, the fundamental reason for the continuous reaction is the different potential between the anode and cathode. The different potential promotes the continuous flow of electrons [24]. Some of the metal standard potentials of the metal-air battery anode are shown in Table 2. The oxygen reduction standard potentials in different conditions of the air cathode are shown in Table 3.

![Table 2 Standard potential of some metal anodes](image)

![Table 3 Oxygen reduction standard electrode potential](image)

3 Application of metal-air battery in the field of water treatment.

For metal-air batteries, current research focuses on optimizing their electrical performance, including the use of alloyed metal anodes to slow the metal's self-corrosion; treating the electrolyte to slow the formation of flocs to maintain the discharge performance of the battery; Develop more efficient air cathode oxygen reduction catalysts. However, a small number of people have used metal-air batteries to conduct research and exploration in the field of water treatment, which proves that metal-air batteries also have the function of water treatment while treating the electrolyte to slow the formation of flocs to maintain the discharge performance of the battery. Currently, metal-air batteries are used in the field of water treatment. The research is relatively rare and has not yet formed systematic analysis and research methods. According to previous research contents, the research content of metal-air battery in water treatment is divided into the removal of heavy metals in water, the collection of substances in water and the treatment of wastewater and domestic sewage.
3.1 Treatment of heavy metals in water by Metal-air battery

Arsenic is a heavy metal with strong toxicity. It is usually in the form of trivalent and pentavalent in nature. If it contains excessive arsenic in drinking water (>0.01mg/L), it will cause serious damage to human body. Therefore, it is very important to effectively remove excess arsenic from water. At present, the methods for removing arsenic include adsorption method [25-26], membrane separation method [27-28], biological treatment method [29-30], and coagulation and sedimentation method [31-32], etc. All of the above methods have a certain removal effect, but some of them are costly, some require related processing facilities, and some require longer processing time. The use of a metal-air battery device to treat arsenic in water is a novel method. The specific operation method is to pass arsenic-containing wastewater as a metal-air battery electrolyte into a metal-air battery, and use the battery reaction to generate substances and wastewater. The reaction can not only effectively remove arsenic from the wastewater, but also produce electricity simultaneously. The use of a metal-air battery device for processing not only requires simple equipment, but also achieves good processing results.

The factors affecting the power generation performance of the battery involve many aspects, and the type of the electrolyte solution has a significant influence on the power generation performance. Joo Yang Park et al. [33] respectively configured the same concentration of NaCl, NaHCO\textsubscript{3} and Na\textsubscript{2}SO\textsubscript{4} as electrolytes that contain a certain concentration of arsenic for iron-air batteries, and explored the effects of different electrolytes on electricity production and arsenic removal. It was found that when using NaCl as the electrolyte, it has the best arsenic removal effect. Although Na\textsubscript{2}SO\textsubscript{4} has better electricity production performance, the effect of arsenic removal is not the best. It is speculated that SO\textsubscript{4}\textsuperscript{2-} will compete with AsO\textsubscript{4}\textsuperscript{3-} for iron and thus affect the arsenic removal effect. The use of NaHCO\textsubscript{3} will cause the iron electrode to quickly produce a passivation layer, which results in poor electrical performance and arsenic removal. Subsequently, Jung Hwan Kim et al. [34] also constructed an iron-air battery, analyzed the reaction products, and explored the effect of arsenic removal under different pH conditions. It is judged by Raman spectroscopy that the reaction products of the iron electrode are mainly iron hydroxides such as fibrite and maghemite. In the arsenic removal effect, the arsenic removal effect is better between pH 4 and 6 with acidic pH, and the arsenic concentration in water can be reduced to less than 0.01 mg/L within 6 hours. According to previous studies, arsenic can form a bifurcated surface complex with fibrite through surface complexation, which explains the reason why iron-air battery can remove arsenic. In addition to the effect of the electrolyte on the removal of arsenic, the air electrode composition of the metal-air battery also has a certain influence on the electricity generation and arsenic removal. Yanxiao Si et al. [35] also constructed an iron-air battery to investigate the effects of changes in air electrode composition on electricity production and arsenic (trivalent and pentavalent) removal effects. It was found that when activated carbon, carbon black mixed activated carbon and carbon black were used as air electrode materials, the best electrical performance was obtained when activated carbon was used as air electrode material alone. Carbon black mixed activated carbon was the second. The battery has the worst discharge performance when carbon black was used as air electrode material. In the removal of arsenic, the removal of pentavalent arsenic was best when activated carbon was used, and the removal of trivalent arsenic was the best when carbon black was used. When activated carbon mixed carbon black is used as the air electrode material, the removal effect of arsenic is between the former two. At the same time, it is found that when using activated carbon as the electrode material, the four-electron reduction of oxygen mainly produces OH\textsuperscript{-}, so it has better electrical properties. When carbon black is used as the electrode material, the main reaction is the two-electron reduction of oxygen. The electricity generation performance is poor. However, H\textsubscript{2}O\textsubscript{2} and hydroxyl radicals are generated near the air cathode, which oxidizes trivalent arsenic to pentavalent arsenic, which makes the pentad arsenic and iron oxidation products combine to be more effectively removed. Hubdar Ali Maitlo et al. [36] constructed a dual-chamber iron-air battery. The basic structure of the dual-chamber battery is basically the same as that of the single chamber. The difference is that an ion exchange membrane is added in the middle to divide the device into two chambers. In this study, the arsenic-containing wastewater was added to the anode chamber, and NaCl and HCl were used as the cathode chamber electrolyte. The effects of different catholytes on the electricity production performance and arsenic removal efficiency were investigated. The study found that the performance of electricity production and arsenic removal are better when HCl was used as the catholyte. When using a dual-chamber battery, the reaction between the two electrodes can be better controlled, and the discharge performance of the battery is more stable, but the structure is slightly more complicated than that of the single-chamber battery. Currently, there are no related studies comparing the arsenic removal and electricity generation performance of single-chamber and dual-chamber batteries. The impacts of battery structure on arsenic removal and electricity production performance are not clear, and further research is needed. Byung Min An et al. [37] constructed an aluminum-air battery to treat arsenic in water and explored the effects of different electrolytes on the electrical performance and arsenic removal of aluminum-air batteries. It is found that when three different electrolytes of NaCl, NaHCO\textsubscript{3} and Na\textsubscript{2}SO\textsubscript{4} are used, the electricity production performance and arsenic removal efficiency are optimal when using NaCl electrolyte. This is because the presence of Cl\textsuperscript{-} can destroy the passivation layer of the aluminum anode. The aluminum anode can be continuously reacted, and the effect of removing arsenic is directly related to the amount of Al(OH)\textsubscript{3} formed after the reaction of the aluminum anode. The HAsO\textsubscript{4}\textsuperscript{2-} and HASO\textsubscript{4}\textsuperscript{2-} in the water are co-precipitated and adsorbed by Al(OH)\textsubscript{3}. According to previous studies, the effect of arsenic removal is basically related to the discharge performance of the battery, and the discharge performance of the battery
is related to many factors. The type of electrolyte solution affects the performance of the battery. When the electrolyte contained Cl\textsuperscript{-}, the electrical performance and arsenic removal effect of the metal air battery are better, because Cl\textsuperscript{-} can destroy the passivation of most metals and weaken the polarization of the metal anode, so that the potential of the metal anode does not rise rapidly. Other ions such as HCO\textsubscript{3}\textsuperscript{-} and SO\textsubscript{4}\textsuperscript{2-} not only do not destroy the passivation layer of the metal, but also adversely affect the combination of HASO\textsubscript{4}\textsuperscript{2-} and HASO\textsubscript{4}\textsuperscript{2-} with the metal hydroxide, which not only causes the battery discharge performance to deteriorate, but also reduce the effect of arsenic removal. The pH value of the electrolyte also affects the electricity production performance and the arsenic removal effect. The different pH value will cause different oxygen reduction reactions in the air cathode. The oxygen reduction reaction potential under acidic conditions is higher, so the battery discharge performance is better. At the same time, different pH values will also result in different floc morphology, which will result in different adsorption properties; the material of the air cathode also affects the power generation and arsenic removal effect of the battery. When the oxygen reduction reaction of the air cathode is more inclined to four-electron reduction, the battery has better electricity generation performance. When the oxygen reduction reaction of the air cathode is more prone to two-electron reduction, hydroxyl radical and hydrogen peroxide are generated, which is beneficial to the oxidation of arsenic in water, making it easier to precipitate and adsorb. This is a factor that needs to be weighed, and it is necessary to produce electricity more efficiently while effectively removing arsenic; The structure of the battery may also affect the treatment effect, but the effects of different battery structure on arsenic removal and electricity generation have not yet been studied, and subsequent experiments are needed to prove.

In addition to arsenic, heavy metal pollution in water also contains elements such as lead, nickel, chromium, cadmium and mercury. These heavy metals also have a certain degree of harm to human health and are also needed to be removed in time. The study on the removal of heavy metals from water by using metal-air battery devices has focused only on the removal of arsenic, and a wide range of studies have not yet been carried out. According to the metal-battery reaction mechanism, the metal hydroxide or metal oxidation product formed in the device should also have a certain degree of adsorption, coagulation and oxidation to other heavy metals. For example, the use of aluminum salts can remove chromium from water; the use of iron salts can remove mercury and nickel from water. Subsequent research can be carried out using aluminum-air batteries to treat chromium-containing wastewater and iron-air batteries for treating wastewater containing heavy metals such as nickel and mercury. These methods are theoretically feasible, and the actual processing effects and design of the device require further research and exploration.

### 3.2 Collection of substances in water by Metal-air battery

Some substances in the water can be collected by adding coagulants or electrolysis to form precipitation, and separating them from water. These methods can not only improve water quality, but also achieve the purpose of reusing certain substances, this method meets the requirements of sustainable development of economic and environmental protection. A small number of researchers have tried to use metal-air battery to collect substances in water.

Phosphorus is a common element in water. If the content exceeds the standard, it will cause a series of problems such as eutrophication of water. Excessive phosphorus can be combined with magnesium salt in water to form struvite (MgNH\textsubscript{4}PO\textsubscript{4}), which will be collected. Dae Hwan Lim et al. [38] constructed a magnesium-air battery with magnesium anode and air cathode, and used phosphorus-containing wastewater as a battery electrolyte to collect phosphorus in water. The study found that when the phosphorus concentration in water is low, the addition of NaCl in the electrolyte can also have a higher collection efficiency, but when the phosphorus concentration is gradually increased, a passivation layer composed of struvite will be formed on the surface of the magnesium anode, resulting in a decrease in the dissolution rate of the magnesium anode. The rate of phosphorus collection is reduced because of low dissolution rate of magnesium anode. After the addition of NaCl to the electrolyte, the power generation performance of the battery and the phosphorus collection efficiency are all improved with the increase of the NaCl dosage, which is mainly because Cl\textsuperscript{-} destroyed the surface passivation layer of the magnesium anode. However, when the phosphorus concentration in the water is too high (more than 0.05 mol/L, in terms of HPO\textsubscript{4}\textsuperscript{2-}), a passivation layer is generated on the surface of the magnesium anode, so that the collection efficiency is greatly reduced. It will no longer improve the surface condition of magnesium anode by adding Cl\textsuperscript{-}. Therefore, it is able to collect struvite using a magnesium-air battery at an appropriate phosphorus concentration.

There are also various algae in the water. Some algae are difficult to collect, need to add chemicals and consume a lot of time. Liu Qing et al. [39] collected the Dunaliella salina in the water by constructing an aluminum-air battery. The water that contain Dunaliella salina acts as an aluminum-air battery electrolyte, and the flocs produced by the metal anode combine with the algae to form a precipitate in order to achieve the purpose of collecting Dunaliella salina. The study found that after the water containing algae enters the battery as an electrolyte, the zeta potential changes from a negative value to a positive value when the algae is combined with the anode product. When the discharge current of the battery is increased, the collection rate is also increased correspondingly. When the algae concentration is below 0.8g/L, the collection efficiency is as high as 97%, which proves that this is a very effective method.

According to previous studies, it is known that there are few studies on the use of metal-air battery devices to
collect substances in water. And most studies are limited to one substance. Different types of metal-air batteries can be used for collecting substances in water in the future.

3.3 Treatment of production and domestic wastewater by metal air battery

Aquaculture wastewater has the characteristics of high COD, high chroma and high ammonia nitrogen. If the wastewater is directly discharged into the water, it will cause serious pollution problems. The aquaculture industry is widely distributed in various rural areas in China. The treatment facilities in most villages are not perfect. Many of the aquaculture wastewaters are discharged into the water without treatment, causing serious pollution of local water. At present, some people have used metal-air batteries to study the treatment effect of aquaculture wastewater, and some results have been achieved. Zhao et al. [40] treated the swine wastewater by constructing an aluminum-air battery and an iron-air battery. They studied the treatment effect and electricity production performance of swine wastewater by setting different conductivity (adjust by adding different concentrations of NaCl), different pH values and different metal anode. The study found that the higher the conductivity, the better the battery's electrical performance, and the different pH values have an effect on the device discharge, but the impact is small. While exploring the conventional influencing factors, it also innovatively built an air battery with a bimetal electrode. The battery with double metal anode configuration has the best performance. The metal-air battery of bimetallic anode was studied in the experiment. It was found that the battery with aluminum-iron double anode has the best performance. Subsequently, the swine wastewater was treated by the experimental device. Two methods were used, one was to add wastewater as an electrolyte to the experimental device for treatment, and the other was adding solution in the device to the wastewater as a coagulant for treatment. The study found that when the swine wastewater is added to the battery as an electrolyte, the treatment effect can be better than the other method. However, the removal mechanism of COD, ammonia nitrogen and other pollutants in aquaculture wastewater is basically not involved. Based on previous studies, it is speculated that the floc of aluminum and iron produced by the battery should have an adsorption effect, so it has a certain treatment effect on the aquaculture wastewater, the specific reasons still need further research.

The use of metal-air batteries for the treatment of aquaculture wastewater has certain treatment effects, as well as the advantages of simple installation and short start-up time, which is suitable for remote villages and places where water treatment facilities are imperfect. With the deepening of research, the power generation performance of metal-air battery will also be improved, and the power can be supplied while treating the wastewater. As a power supply in remote areas, it is a promising method for treating aquaculture wastewater.

According to the reaction mechanism, the metal-air battery can be used in the treatment of other sewage and wastewater, in addition to the production wastewater of aquaculture wastewater. For example, due to various factors, remote water treatment facilities are not perfect. Many domestic sewages are discharged into water without treatment, causing serious pollution problems to natural water. The discharge of domestic sewage in remote areas is relatively scattered, and it is difficult to collect and treat. The metal-air battery is easy to start and operate as a small-scale device. It is highly feasible to use it as a pretreatment device for domestic sewage. The treatment of air battery devices can reduce ammonia nitrogen, COD and total phosphorus in domestic sewage. Most industrial production wastewaters are less biodegradable, some industrial wastewaters have high salinity, high heavy metal content and heavy pollution. High salinity water as an electrolyte is beneficial to the discharge of metal-air batteries, enabling the device to produce more metal anode products, so the metal-air battery as a pretreatment unit is more feasible and can be used as a preliminary treatment unit for reducing pollutant concentration. At present, the treatment of production and domestic wastewater by metal-air battery devices has only been studied on aquaculture wastewater, and research on wastewater treatment of other properties has not been carried out. For different water quality, how to choose the appropriate metal electrode for different pollutants, suitable battery structure, etc. need further exploration and research.

4 Conclusions and prospects

Metal-air battery is a new technology in the field of water treatment. At present, preliminary research has been carried out in the fields of removing arsenic from groundwater, collecting phosphorus in water, collecting algae in water and treating aquaculture wastewater, and have also achieved certain Effect. However, the research of the predecessors found that there are still some problems in the metal-air battery, such as the overall low power generation performance during the processing and the harsh requirements for the solution to be treated. Moreover, the research scope of the predecessors is relatively limited. Most people still pay attention to the treatment effect and power generation. The analysis of the material form of the electrode is less, and the research on the air cathode product is less. Only one person has proved it. The possibility of producing oxidizing substances (hydrogen peroxide and hydroxyl radicals) on the air cathode of different materials. As a battery, it can generate energy, but it has not been able to utilize the energy generated yet.

Subsequent in-depth research can be conducted in the following directions: Try to use metal-air batteries to test the removal of more pollutants; explore the possibility of using metal-air batteries in more water treatment processes; The generated electric energy could be designed a metal-air battery-electrolysis coupling process, multi-step processing to form a better process of processing effect; the catalyst in the air cathode can be optimized to make the device produce more oxidizing substances, resulting in better removal of pollutants.

In summary, metal-air battery as a new technology in
the field of water treatment has the advantages of simple structure, fast starting speed, etc. It does not require input of energy, and can generate electricity. It is a green technology and worthy of further study.

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