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Research Progress of Electrocatalysts for Carbon Dioxide Reduction

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Abstract. The problem of global warming has become increasingly serious due to increasing carbon dioxide emissions caused by the burning of fossil fuels. Hence, achieving carbon reduction and carbon neutrality by means of the resource utilization of CO_2 has become a priority for more in-depth research. The electrocatalytic reduction of carbon dioxide (CO_2RR) is a promising way and a key technology to convert CO_2 into valuable chemicals and fuels using renewable energy. This paper briefly summarizes the CO_2 sources, the fundamentals of CO_2RR, and summarizes the recent advancements in the metal-based, carbon-based, and composite catalysts, discussing merits, limitations and application prospects. From the above review, it is seen that metal-based catalysts are very efficient in terms of their catalytic activity; carbon-based catalysts are cost-effective and offer structural flexibility and good conductivity; whereas composite catalysts combine the advantages of multiple materials, combining high catalytic efficiency, product selectivity, and stability, thus showing promising prospects for further development. Future work should focus on advancing electrocatalytic mechanisms, the development of new catalysts, enhancing active site regulation for long-term reaction stability, and enabling the scale-up of industrial applications. The present work offers valuable knowledge for designing and optimizing highly active electrocatalysts, as well as for their practical application in CO_2RR technologies.

Keywords: Carbon dioxide, Electrochemical reduction, Electrocatalyst, Carbon neutrality

1. Introduction

The two industrial revolutions brought completely new types of energy for mankind and quickly improved the productivity, accelerated industrialization, and caused substantial increases in world populations. It also increased human dependence on fossil fuel energy sources like coal, oil, and natural gas. As the major greenhouse gas, CO_2 plays a key role in global warming, while increased atmospheric CO_2 also results in ocean acidification, a greater frequency of extreme weather events, and disruption to the balance of the natural carbon cycle [1]. This is compounded by the heavy dependence of mankind upon fossil fuels for its needs. In the opening years of the new millennium, with increasing international awareness of environmental concerns, nations have increasingly pursued goals including carbon emission peaking and carbon neutral status, to mitigate global warming by decreasing greenhouse gas emissions and increasing carbon use-efficiency.

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In fact, carbon dioxide in the atmosphere is not merely a liability for Earth and humanity. Rather, it can be utilized as a carbon resource. Simply reducing carbon emissions is insufficient to fully address the environmental challenges posed by greenhouse gases [2]. Alternatively, renewable and non-polluting energy sources ought to be used to transform CO_2 into valuable chemicals or fuels, including CO, methane, methanol, formic acid, and various other hydrocarbons. In contrast with conventional carbon capture and storage (CCS) methods, carbon capture, utilization, and storage (CCUS) not only stores carbon but also enables the conversion of CO_2 into feedstocks needed throughout upstream and downstream industrial sectors, which can in turn lower manufacturing expenses.

The CO_2RR technology is essential to the use of CO_2 resources, and the C=O covalent bond in the CO_2 molecule is very strong, so the catalyst is very important. Currently, there are four catalytic processes: thermal catalysis, biocatalysis, photocatalysis and electrocatalysis, with the latter being very promising. The CO_2RR process can operate under relatively mild conditions such as ambient temperature and pressure, which can minimize the operating cost, and can also utilize renewable electricity generated using solar and wind energy, which can further improve environmental benefits [3].

Although many of these catalytic avenues have been explored, CO_2RR remains with many challenges. The first problem is that the CO_2 molecule is thermodynamically stable and is rather inert kinetically, due to its strong covalent bonds. CO_2RR typically requires a high overpotential and complex reaction pathways, multi end products and complex selectivity control.

Apart from the intrinsic limitations of carbon dioxide molecules, CO_2RR has several other technical challenges. The major issues are competition with hydrogen evolution reaction (HER), number of intermediate species, poor selectivity for the desired product, and low long-term stability [4]. In particular, HER consumes electrons and active sites, which reduces the effectiveness of desired product formation, and CO_2RR is a multi-step reaction with successive intermediates. Furthermore, coupled reactions with multiproton and multielectron participation produce a large number of products that compete, complicating the control of the product and hindering the commercialization. The use of CO_2RR to the fullest extent is hindered by these problems and great efforts are being made to create more efficient electrocatalysts.

This research review begins with an introduction to the background of CO_2 emissions, based on the above-mentioned research background. It then presents the fundamentals of CO_2RR. The review subsequently summarises recent advances in various types of electrocatalysts and addresses the catalyst design for various desired products. Lastly, the current problems and future industrial application of CO_2RR is discussed on the basis of which conditions high-performance electrocatalysts can be developed.

2. Sources of CO_2 emissions

2.1. Natural sources of CO_2

The carbon dioxide has played an important role in chemistry reactions, physical processes, and biological evolution since the dawn of life on earth until the Industrial Revolution. For thousands of years, the natural carbon cycle was dynamic with continual emissions of CO_2 from natural sources to the atmosphere. There are several natural sources of CO_2 emissions, most notably respiration, decomposition, volcanic activity, and ocean-atmosphere exchange [5]. However, these natural cycles are not the main driving force behind the present-day increase in CO_2 concentration. Human-made emissions have upset that balance.

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2.2. Anthropogenic sources of CO_2

In general, the CO_2 emissions released to the atmosphere from humans can be split into domestic and industrial emissions. Residential emissions are relatively small environmental problems, but are closely associated with daily life. Transportation, residential energy use, and residential heating depend on fossil fuels every day and are significant sources of anthropogenic CO_2 emissions. However, with the development of renewable energy technology, clean energy like solar, wind, hydrogen and nuclear energy are more and more applied in daily life and thus partly alleviate this issue. However, industrial activities remain the primary source of CO_2 emissions.

The emission of carbon dioxide is almost inevitable during industrialization. The significant part of CO_2 emissions from industry is due to the reliance on fossil fuel energy sources. Industry has grown fast and has been a higher consumer of energy, particularly in the thermal power industry, the cement industry, steel industry, petrochemicals and non-ferrous metals, which are significant consumers of fossil fuels [6]. The use of coal, oil and natural gas has continued and remains the dominant source of energy, despite the increasing contribution from renewable clean energy sources like wind, solar, hydrogen and biomass. Many fossil fuels are also used as feedstocks or process agents in industry, in addition to their use for power generation. Burning of fossil fuel is thus a major contributor of CO_2 in the atmosphere. Rising CO_2 levels in the atmosphere add to global climate change, ocean acidification and disruption of the global carbon cycle. Too much carbon dioxide in ocean water can change the chemistry of the carbonates that can reduce the ability of marine creatures to grow shells. In the near term, industrial emissions cannot be avoided, so CO_2 should also be utilized. Industrial production still relies on energy and carbon-based feedstocks; therefore, merely reducing carbon emissions is insufficient—CO_2 must also be converted into valuable resources.

2.3. Significance of CO_2 utilization

Human approaches to utilizing CO_2 resources are varied and long-standing. Advances in agricultural techniques could be seen as CO_2 recycling since very early on in agriculture, when agriculture was first developed, it was possible to improve the technique applied. In industrialization the only way to create an economic benefit was to cut carbon, and sometimes governments were unable to design carbon reduction policies without subsidies [7]. However, high concentrations of CO_2 in the atmosphere cannot be solved by simply reducing emissions, but can be addressed by renewable or circular energy, which transform CO_2 into valuable chemicals and fuels thereby improving industrial supply chains and possibly reducing production costs. The interest in catalytic CO_2 reduction methods, particularly CO_2RR, has grown significantly, alongside technological development, due to their high potential.

3. Electrocatalysts for CO_2RR

The idea behind CO_2RR is that the activation energy for the reduction will be lowered by the presence of catalysts, thus making the reduction more efficient. On a molecular level, the covalent bonds in CO_2 are highly thermodynamic stable and are highly kinetic inactive, which makes direct reduction hard. Electrocatalysts are used to provide pathways to allow electron and proton transfer to reduce the activation energy of the reaction. Therefore, it is one of the key research directions for CO_2RR to develop better electrocatalysts, and metal catalysts are the most intensively studied type.

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3.1. Metal-based catalysts

Metal catalysts have unique surface-active sites and they are electrically conductive which lower the activation energy for reduction reactions. A number of elements can be used as catalysts, such as copper, silver, gold, tin, bismuth, indium, zinc and palladium. In order to make the use of low cost metal catalysts for industrial application, these costs have to be reduced. These metals prefer different products due to differences in their electronic structure and surface adsorption. Copper, for instance, is more efficient for hydrocarbon and C_2+ product formation, while silver, gold, and zinc are more efficient in reducing CO_2 to carbon monoxide and zinc and post-transition metals are more efficient in reducing CO_2 to formate and formic acid [8].

The electrocatalysts of copper are some of the most extensively explored metal electrocatalysts, as they can be used to make a variety of substrates, are highly efficient for multicarbon compounds, and have a relatively low cost. Cu is one of a few metals that can reduce CO_2 to various carbon-based products including C1 compounds such as CO, formic acid, methanol and methane. These products have various applications. CO is a key industrial feedstock, reaction intermediate, reducing agent and a major component of syngas. Liquid chemicals use formic acid and formates as feedstocks and preservatives, and methanol and methane are primarily used as fuel. Copper can also produce C_2/C_2+ species such as ethylene, ethanol, acetate and propanol. However, some metals such as Ag, Au and Zn can easily form CO, but are not very efficient at forming multicarbon products. The specialness of Cu is that it can adsorb and convert CO intermediates, and is one of the few metals that can generate products of more than one carbon. C_2+ products are of higher energy density and are of higher industrial value [9]. Comparing copper with gold or silver, it is also cheap and has a low manufacturing cost, so that it can be used in the commercial sector more widely. The benefits have led to the extensive research of Cu-based catalysts for CO_2RR.

Copper catalysis, however, does have a few drawbacks: complex reaction intermediates, high overpotential, low product selectivity and lack of long-term stability. There are a number of reaction intermediates that can be formed during the reaction, which gives rise to complex reaction mechanisms with Cu catalysts. Some of the products Cu can produce are CO, formic acid, methanol, methane, ethylene, ethanol, acetate and propanol, some of which are formed in several intermediate steps, as mentioned before. This leads to the formation of many intermediates (build-up and competition) at the later stages of the reaction, yielding a wide product range, complex product mixture, high product cost and low selectivity of the target products. Furthermore, copper reduction reactions have a large over potential which results in a decrease of energy efficiency. Cu catalysts can also undergo the processes of sintering, agglomeration or restructuring during extended electrolysis, compromising their stability.

Silver, gold, tin and bismuth are other metal catalysts with different characteristics than copper. Ag and Au demonstrate high selectivity towards CO production at low overpotential, and high Faradaic efficiency, while Sn and Bi primarily formic acid or formate. Another advantage of Bismuth-based catalysts is that they are also strong inhibitors of HER (1) and can be produced at a low cost, with low toxicity and good stability [10].

3.2. Carbon-based catalysts

Because carbon-based catalysts are cost effective and may be less polluting than metal-based electrocatalysts. There are several types of major classes such as graphene, carbon nanotubes, porous carbon, activated carbon, carbon aerogels, heteroatom-doped carbon, and defect-engineered carbon [11]. The site of their structural tunability is a range of pore architecture, heteroatom identity

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and defect density that can be intentionally engineered to influence CO_2 adsorption and electron distribution and intermediate binding. As a result, this flexibility makes it possible to use carbon materials as active catalytic platforms, with selectivity which can be engineered, as well as the use as supports.

Some of the unique properties of carbon catalysts are that they have relatively low production costs, a large amount of carbon resources available, high electrical conductivity, high specific surface area, adjustable pore structure, and can be heteroatom doped, highly flexible structure and excellent stability. Carbon-based catalysts are much cheaper than precious metals like gold, silver and platinum. The high electrical conductivity facilitates electron transfer and the large surface area provides a larger number of active sites, thus providing greater contact with CO_2. Moderable changes in the porestructures may increase the CO_2 adsorption and activation properties and the control of the key intermediates. The doping of heteroatoms such as nitrogen, sulfur, or phosphorus re-distributes charge, generates extra active sites, and can enhance the catalytic activity or product selectivity. The long-term stability can also help to lower the frequency of catalyst replacement and maintenance on production lines.

However, carbon-based catalysts have many drawbacks. The catalytic activity of their intrinsic materials is comparatively low than metal catalysts are, due to the relatively uniform electron distribution in undoped carbon, that is, there are not enough active sites. Doping of the heteroatom and defect engineering can increase the activity but could be quite complicated and expensive. There are also drawbacks: They are not very selective in their products; some catalytic mechanisms are not fully understood, and there are ongoing scale up challenges in industry. These restrictions have stimulated the interest in composite catalysts, such as those containing metals and carbon materials.

3.3. Composite catalysts

A composite catalyst is a catalyst that is composed of two or more materials that together exhibit properties that are not shown by the individual materials. A representative example is the so-called Metal-N-C catalysts, in which transition metal atoms are dispersed on nitrogen doped carbon, changing the electron density at the local level and enhancing the catalytic activity. If these metals are atomically dispersed, then these are known as single-atom catalysts (SACs).

There are a number of intrinsic advantages of composite catalysts. Metal-N-C SACs have high atom use efficiency, high catalytic activity, high selectivity for products, and the synergy between the metal and carbon components. Metal-N-C catalysts can reach virtually 100% utilization of the metal atoms in comparison to the conventional metal nanoparticle catalysts [12]. The stable M-Nx coordination complexes alter the electronic structure of the metal center, boost the adsorption and activation of CO_2, facilitate electron transport, and boost catalytic performances. Thus, these catalysts present a high activity of metals and the conductivity and stability of carbon material.

Technically, Metal-N-C catalysts have several advantages over single-component catalysts, but some significant industrial issues must be addressed. The major issues are regulation of the active site, scaling up, structural/activity relationships, and lack of long-term stability. Precise control of the metal oxidation state and the neighboring carbon structure is especially important in the case of active-site control.

4. Prospects for CO_2RR catalysts

Catalyst design should shift from a performance screening of empirical recipes to mechanism-driven control of electronic structure, geometry of active sites and intermediates binding. Current systems

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still suffer from poor energy efficiency, selectivity, activity and long-term stability. The most helpful improvements will link theory with operando characterization and repeatable synthesis, rather than record the best single performance in an ideal lab condition.

In addition, the most promising future direction is not one catalyst family, but a co-design of the catalyst, reactor and pathway to the product. Research should focus on products with an apparent market value and the ease of separation rather than those that simply transfer carbon or energy costs. Solid carbon may be appealing if it offers long-term storage and useful material, but the benefits in terms of life cycles will be determined by the established benefits, the ability to control the morphology, and the realistic demand for energy. Industrial progress should be judged by a sustained carbon efficiency, the value of the product, and the system level emissions, not just Faradaic efficiency.

5. Conclusion

This paper outlines the current status in development and future perspectives of CO_2RR electrocatalysts, global sources of CO_2 emissions, the significance of CO_2 utilization and the conversion of CO_2 into carbon-based products. Metal based catalysts have high catalytic activity, and the presence of copper is advantageous for producing valuable multicarbon products. Carbon-based catalysts are considerably less expensive and more structurally tunable but need to be improved in terms of intrinsic activity. Composite catalysts are comprised of complementary components to achieve greater activity, selectivity and stability. In general, next-generation catalysts are characterized by the need to focus on controllable electronic structure, clearly defined active sites and high durability in industrially relevant conditions.

The use of renewable electricity to convert captured CO_2 to valuable carbon products through CO_2RR is a good path. But true advances in carbon neutrality go beyond the lab—catalyst performance needs to be coupled with mechanistic studies, scalable synthesis, efficient reactors, product separation, and life cycle assessment. Repeatable selectivity, long-term operation, energy efficiency at system level and economically viable products should therefore be the focus of future research. These priorities can guide CO_2RR from laboratory demonstrations to real world and environmentally sound deployment.

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