



The electrocatalytic properties of lithium copper composite in the oxygen reduction reaction



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ABSTRACT

A new carbon supported lithium-copper cathode catalyst (gel@LiCu) is composed of activated carbon (@) supported gel, LiNO₃ and CuCl₂ mixture is investigated in the oxygen reduction reaction. The gel@LiCu cathode, which is selected from different configurations of electrodes, is found to have the best material for the cathode. The molar ratio of lithium-copper (1.0:1.5) and the amount of glycine given in terms of grams are determined in order to find the most active material. The catalytic activity of the gel@LiCu electrode is evaluated by cyclic voltammetry, potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The morphology and the phase structure of the gel@LiCu electrode are characterized with scanning electron microscopy (SEM), transmission electron microscopy (TEM) and X-ray diffraction (XRD) spectrometer. The catalyst shows good catalytic activity for oxygen reduction reaction (ORR), as well as oxygen evolution reaction (OER).

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1. Introduction

In recent years, batteries have been keenly investigated because of the development of transportable electric vehicles. New technology items run with high-capacity batteries. Lithium air batteries, which use oxygen molecules of the atmosphere, have higher energy than other battery types and they have been studied as promising candidates for high-energy density secondary batteries [1–5]. The rechargeable lithium-ion (Li-ion) batteries have been used for portable electronic appliances such as mobile phones, music players, etc. As the electrical devices have been developed, the energy density of Li-ion batteries has become insufficient [6]. Li-ion batteries have other disadvantages as well. For instance the specific capacity of lithium ion batteries are limited by the weight of the anode and cathode materials such as typically graphite (170 mAh g⁻¹) for the anode and metal oxide such as LiCoO₂ (130 mAh g⁻¹) for the cathode [7,8]. The cathode materials used for Li-ion batteries are restricted in nature. Therefore, most of the cathode materials are prepared by high-temperature solid-state reactions, which results in consumption of energy and CO₂ emission [9]. What is more, cathodes of conventional lithium-ion batteries are not renewable (or reusable)

[10]. It seems that the solution for these challenges requires developing a new concept of the lithium battery. In 1996, Abraham and Jiang first reported the Li-air battery using an organic electrolyte [3]. Lithium air batteries are attracting more and more attention because of their energy and power density.

There are four types of Li-air batteries; aprotic, aqueous, solid state and mixed aqueous/aprotic. We used the aprotic type for our battery tests. The aprotic type consists of four parts; a lithium metal anode, a solid electrolyte interface, an aprotic electrolyte and an air cathode [11]. The battery functions as follows:



when Li₂O₂ is formed at the cathode during discharge, and



when Li₂O is formed at the cathode, which has also been reported under certain conditions [12–15].

During the charge process both the above reactions turn vice versa, which plays an important role in the lithium oxide decomposition (mainly lithium peroxide) and the oxidation of peroxide anions (oxygen evolution reaction, OER). These batteries have several restrictions of use in electrical devices, such as slow

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kinetics of the ORR and OER during discharge and charge, respectively [16,17]. Recent studies show that the battery performance can be affected by catalysts [18,19].

Porous structures are preferred in the cathode section of lithium air batteries. Carbon materials have been widely studied as cathode materials for Li-air batteries due to their large pore volume, high surface area and electrical conductivity [20–22]. The cathode catalysts are usually carbon black or carbon loaded with a noble metal, which is used to catalyze the air reactions [23]. Pores of carbon materials can act as a path for oxygen access and an electrolyte reservoir, and they can also be holders of the discharge product (Li_2O_2) on space for non-aqueous Li-air batteries [24].

Many studies have been performed to improve OER kinetics and reduce charge overpotential [25–28]. Different types of materials such as carbon-based materials [26,28], noble metals [18,29], metal oxides [30,31] and nitrides [32,33], perovskites [34,35], and pyrochlores [36,37] have been used as catalysts in nonaqueous LiO_2 batteries. Shao-Horn et al. [38] have studied the effects of Pt–Au nanoparticles on the voltage gap between the ORR and OER as a bifunctional catalyst. They have found that Au enhances the ORR and Pt decreases the OER voltages.

Transition metals tender cheaper cost alternative catalysts. Copper is used as the renewable cathode material, because it reduces the oxygen of air and it shows a high capacity in lithium air batteries. The copper nanoparticles play two different roles in the catalyst. First, copper nanoparticles improve the conductivity of the catalysts. Second, the metallic copper can accelerate the ORR at its surface, starting from the copper corrosion in aqueous solution [39,40]. The concept of Li–Cu cathode using in Li–Air battery may provide a new direction for the study of future lithium batteries [9]. CuSO_4 , CuV_2O_6 , $\text{CuO-V}_2\text{O}_5$, Cu_6Sn_5 , perovskite loaded with copper nanoparticles, Co_3O_4 microspheres loaded with copper and Li–Cu have been used as cathode materials in different studies [9,23,39,41–44]. When lithium ions move into the spinal matrix, the electrocatalytic activity enhances toward the OER [45–47].

The aim of this study is to prepare a lithium-copper cathode catalyst, including activated carbon, which has a high catalytic efficiency when used in lithium air battery cells. Therefore, gel, LiNO_3 and CuCl_2 were used to build a matrix for bifunctional metals and to improve the ORR and OER performance, respectively.

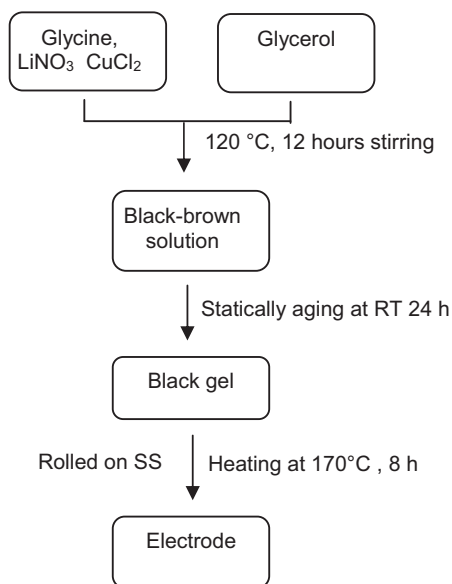


Fig. 1. Scheme for preparation of cathode catalyst.

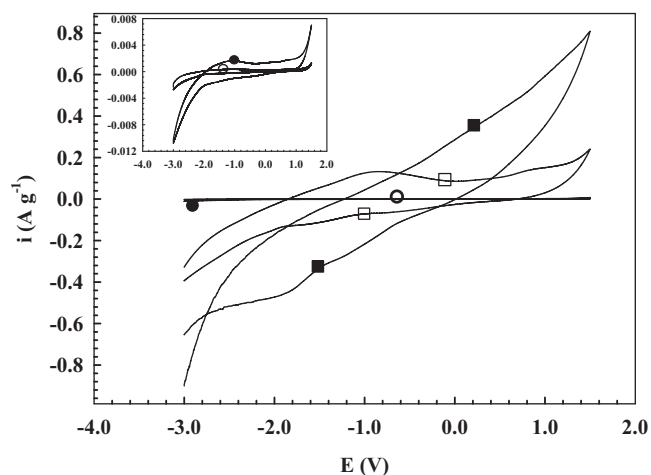


Fig. 2. Cyclic voltammograms of gel (○), gel/Li (●), gel/LiCu (□) and gel@LiCu (■) electrodes at 100 mV s^{-1} scan rate in LED solution.

2. Experimental

All used chemicals, which were purchased from Merck Millipore Co., Ltd. Germany were of analytical or chemical grade purity. The catalysts were prepared by the sol-gel method using glycine, glycerol, LiNO_3 and CuCl_2 . The flow chart of catalyst preparation is shown in Fig. 1. Glycine, glycerol, ammonia, lithium and copper salts were mixed in a pressure resistant Schlenk tube. Then different temperatures and times were applied to obtain a viscous mixture. 12 hours and 120°C were selected as the most suitable temperature and time. After that the viscous mixture was statically aging for 24 hours at room temperature. Following this process, one part of the gel was rolled and left to paste on a stainless steel plate (SS) at 170°C for 8 hours in order to make an electrode. Gel was prepared from glycine and glycerol, which were used as the chelating agent and the polymerization precursor, respectively.

The catalysts were characterized by using cyclic voltammetry (CV), potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) techniques. Gamry (interface 1000) model electrochemical analyzer (serial number: 2,009) was used for electrochemical measurements under computer control. A double-wall one-compartment cell with three electrode configuration was

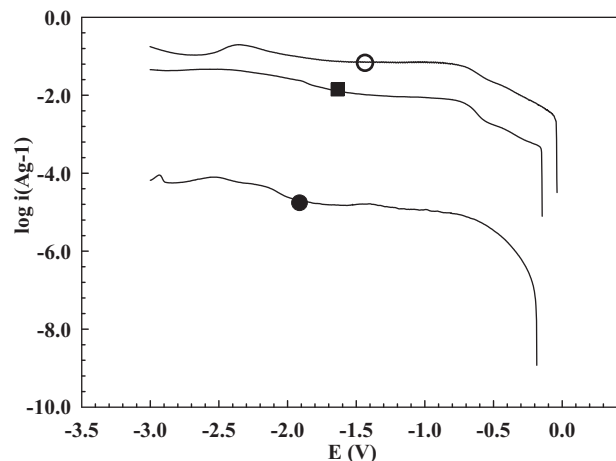


Fig. 3. Current-potential curves of SS (●), gel@Li (□) and gel@LiCu (○) electrodes under oxygen atmosphere at 1 mV s^{-1} scan rate in LED solution.

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