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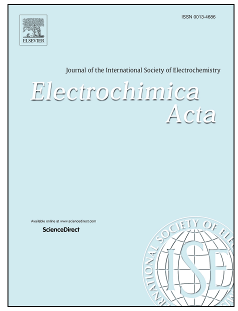

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Confined and Covalent Sulfur for Stable Room Temperature Potassium-Sulfur Battery

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Abatract: Potassium-sulfur (K-S) battery represents one of the most promising electrochemical energy storage devices because of the low cost and abundance of potassium and sulfur resources, but the inferior cycle stability and high operating temperature seriously impede its applications. Herein, a high energy and stable room temperature K-S battery was developed with a confined ad covalent sulfur cathode, which could deliver an energy density as high as ~445 Wh Kg^{-1}, a Coulombic efficiency close to 100%, and a superior cycle stability with a capacity retention of 86.3% over 300 cycles at a voltage cut-off of 0.8-3.0 V. This work displayed a basic study on the rechargeable potassium-sulfur batteries and may pave the way for designing sulfur based electrode materials for potassium based ion batteries.

1. Introduction

Lithium ion batteries have been commercialized for decades since their first launch by Sony in the early 1990s and are becoming more and more popular in our daily life. However, the limited lithium resources and high cost of lithium ion batteries have aroused severe concerns over their long-term development.[1] Therefore, tremendous efforts have been dedicated to exploring alternative rechargeable metal ion batteries, including potassium ion batteries (PIBs) [2-27], sodium ion batteries (SIBs) [28-33], magnesium ion batteries (MIBs) [34], and aluminum ion batteries (AIBs) [35]. Among them, PIBs are considered one of the most promising rechargeable batteries, not only due to the plenty and low cost of potassium resources, but also the benefits coming from the lower standard electrode potential of K (-2.93 V *vs.* E^0) compared with Na (-2.71 V *vs.* E^0), Mg (-2.37 V *vs.* E^0), and Al (-1.66 V *vs.* E^0), implying that the PIBs may possess a higher energy density and a higher discharge voltage in comparison to SIBs, MIBs and AIBs. In addition, sulfur, one of the most efficient cathode materials due to its high capacity and inexpensiveness, has been vigorously investigated in lithium-sulfur (Li-S) batteries and sodium-sulfur (Na-S) batteries[36-49]. Despite of that, the study on potassium-sulfur (K-S) batteries is still in the infancy stage. Therefore, investigation and development of potassium-sulfur (K-S) batteries through incorporating the sulfur electrode into potassium batteries are highly demanded.

K-S batteries are facing the same problem as Li-S and Na-S batteries, the infamous shuttle effect (the transport of soluble polysulfides between cathode and anode), which leads to an inferior Coulombic efficiency and a rapid capacity decay. Besides, the radius of potassium ion (133 pm) is much larger than that of lithium ion (76 pm) and sodium ion (102 pm), causing a huge volume expansion during potassiation/depotassiation processes and possibly destroying the electrode materials, which further deteriorates the electrochemical performance. Even though K-S batteries have been reported by Chen group and Lu group, their poor cycle stability and high operating temperature remain the big challenges[50, 51]. Therefore, it is urgent to develop K-S batteries which have a better cycle stability and are able to work at room temperature. Besides, the sulfur in the previously K-S batteries are main S_8 or K_2S_n, which will result in severious shuttle effect just like Li-S and Na-S batteries, and the performance will be restricted[50, 52]. Drawing on the experience of Li-S and Na-S batteries, the performance of metal-sulfur batteries could be significantly enhanced by confining sulfur in carbon materials or choosing covalent sulfur as the sulfur host materials[40, 53-55]. Therefore, covalent sulfur confined in the host was employed to improve the electrochemical performance of K-S batteries.

Herein, we reported a stable room temperature K-S battery, which was assembled using 1M potassium trifluoromethanesulfonate (KSO_3CF_3) in ethylene carbonate: diether carbonate (EC:DEC, 1:1 *v*/*v*) as the electrolyte, confined and covalent sulfur cathode (CCS, 39.25% sulfur content) as the sulfur host material,