

RECHARGEABLE LITHIUM-SULFUR BATTERY (EXTENDED ABSTRACT)

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The lithium-sulfur system, because of its high volumetric and gravimetric energy densities has attracted interest both for secondary [1 - 3] and primary [5, 6] battery applications. The discharge mechanism of the polysulfides (PS) and the discharge products have been found to be solvent dependent [3, 7]. In THF-T solution at a very low discharge rate (10 - 20 $\mu\text{A cm}^{-2}$), the sulfur utilization is close to 100%, *viz.*, 2 electrons per sulfur atom. However, in dioxolane-based solutions at about 0.1 mA cm^{-2} the discharged product seems to be Li_2S_2 , corresponding to one electron per sulfur atom [7].

The purpose of this work was to assess the feasibility of developing a rechargeable lithium-sulfur system. A spirally wound 2/3A-size cell was used as a test vehicle. It consisted of a 0.3 mm thick lithium anode (2 A h) pressed on a nickel Exmet and a 0.35 mm thick Teflon-bonded porous carbon cathode supported on a nickel Exmet. The electrode area was 50 - 70 cm^2 . The separator was 0.025 mm Celgard microporous poly(propylene). The electrolyte was 0.1 M Li_2S_8 in a supporting electrolyte dissolved in a mixture of ethers, which limits the solubility of PS. The porous carbon cathode was loaded with sulfur. Total sulfur capacity (based on 2e/S) was 1.2 A h. The batteries were cycled using a home-made, computerized battery cycler. They were charged at 50 mA to a predetermined cut-off charge, and discharged across resistive loads at about 0.1 - 1.0 mA cm^{-2} to a predetermined cut-off voltage of 1.5 - 1.75 V.

In most cases, the charging capacity was 20 - 100% higher than the discharge capacity. At room temperature, and at a discharge rate of 6 mA, the initial battery capacity was, typically, 0.45 - 0.55 A h. After 20 - 30 cycles, the capacity (at the same discharge rate) dropped to 300 mA h. The discharge curve of cycle No. 4 is shown in Fig. 1. The charge curve of cycle No. 4 of the same cell is shown in Fig. 2. The maximum number of cycles to failure was 50. At 50 mA discharge rate, the initial capacity was typically 400 mA h (Fig. 3) and it dropped to 280 - 300 mA h after 20 - 30 cycles. Maximum cycle life at this rate was 40. As can be seen in Figs. 1 - 3, the battery exhibits flat discharge and charge curves. Using a PS indicator electrode, it was found that the cell voltage declined at the end of discharge

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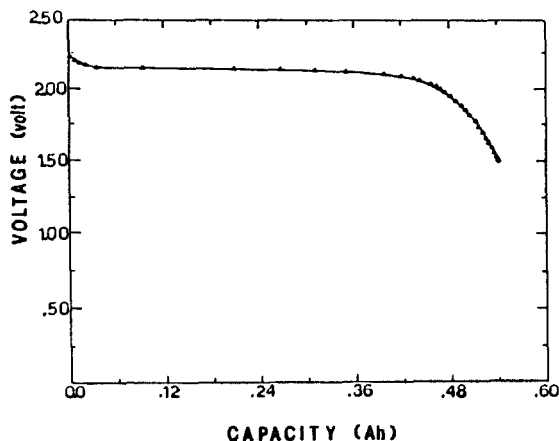


Fig. 1. A discharge curve of cycle number 4 at 0.1 mA cm^{-2} .

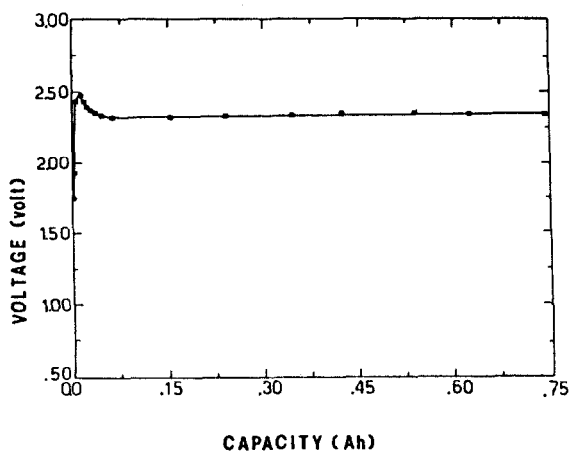


Fig. 2. A charge curve of cycle number 4 at 1 mA cm^{-2} .

due to depletion of PS in the solution. The voltage peak at the beginning of the charge cycle appears to be caused by this PS depletion.

One reason for the decline of capacity with cycling may be an irreversible loss of active cathode material. It is suggested that on cycling some of the PS reduction product is inactive or, at least, difficult to oxidise. We believe this product to be Li_2S . This hypothesis is supported by the following experiment. A cell whose cathode was charged with Li_2S (instead of with sulfur) could not be charged at all and, on discharge, it delivered only the capacity of the $0.1 \text{ M Li}_2\text{S}_8$ dissolved in the solution.

Most cells failed as a result of lithium depletion. The best lithium cycling efficiency was 85%. The present energy density of this cell is 60 and 80 W h kg^{-1} at $C/7$ and $C/70$ discharge rates, respectively. This cell can be overcharged 300% at 50 mA without danger.

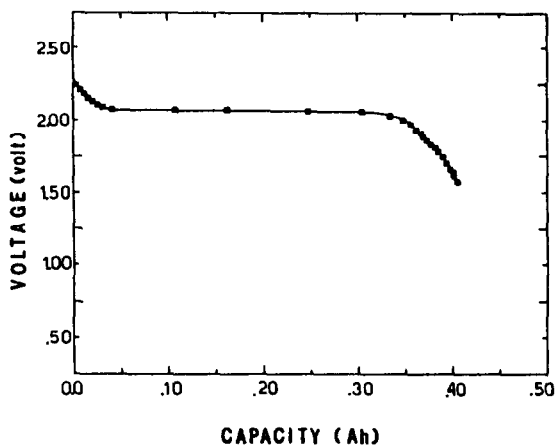


Fig. 3. A discharge curve of cycle number 8 at 1 mA cm^{-2} .

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