Anode Utilization and Efficiency in Carbon/Air Batteries and Fuel Cells

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We report research concerning the development of carbon/air batteries and fuel cells operating at 750 °C in a molten carbonate electrolyte. Cell voltages and total utilization were measured for paste and porous block anodes in 60-cm^2 laboratory test cells.

Carbon/air batteries and fuel cells are under development for applications requiring high energy density and high efficiency. The net cell reaction is

$$C + O_2 = CO_2 \quad E° = 1.03 \text{ V at } 750 °C \qquad (1)$$

A cell uses a molten carbonate electrolyte and a sintered porous nickel cathode catalyzed with Li-doped nickel oxide. The separator is a porous ceramic matrix. Reaction (1) takes place with a near-zero entropy change, and theoretical efficiency approaches 100%. Our previous research established cell voltages of 0.8 V (theoretical 1.03 V) at current densities of 30-120 mA/cm^2, using reactive carbon materials formed by low-temperature pyrolysis of hydrocarbons. Such "turbostratic" carbons owe enhanced reactivity to random orientation of graphene planes and small crystalline domain sizes. [1]

Total efficiency is the product of theoretical, voltage and utilization efficiencies. Utilization depends on the coulombic efficiency of reaction (1). Losses occur from Boudouard corrosion, $C + CO_2 = 2CO$, which depend on anode configuration and morphology. Hauser measured coulombic efficiencies of 95% (T = 870 °C) and 99.7% (700 °C) for graphite anode rods in carbonate melts at galvanostatic currents between 60- and 100 mA/cm^2 [2]. Weaver *et al.* [3] conducted half-cell polarization studies of calcined compacted coal. They reported coulombic efficiencies above 99% (T = 700 °C) using off gas analysis for anodes at 0.8 V (vs. Au/CO_2, O_2). Current densities for graphite were lower by a factor of 10^3 at the same voltage. Vutetakis [4] measured coulombic efficiencies of slurries of carbon particles in molten salt at an immersed gold anode, by gas analysis, but utilizations were poor (< 20%) because of carbon corrosion in the bulk of the carbon/melt slurry.

This work compares results from a 60-cm^2 rigid anode cell with those from a tilted laboratory cell (anode on top) from which excess electrolyte was drained to leave a dense paste (Fig. 1). At a cell voltage of 0.8 V, acetylene black anodes showed current densities of 76- and 61 mA/cm^2 for 3- and 60 cm^2 area cells, respectively; a petroleum coke sample yielded 0.8 V at 56- and 51 mA/cm^2, respectively [5].

A rigid anode cell makes use of a novel compacted and sintered anode, combining reactive carbons with binders and internal structures imparting strength and conductivity. As with the work by Weaver, the anode achieves high utilization by limiting exposure to the anode surface undergoing polarization.

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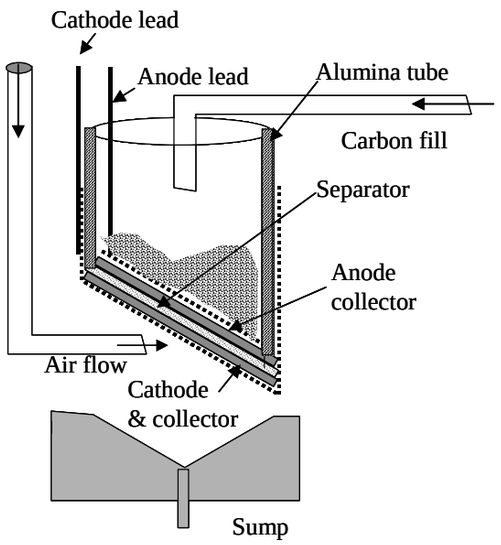

Figure 1. Typical laboratory cells for carbon/air studies consist of a paste of carbon particles and molten salt in contact with a metal current collector. A heavy current collector supports the sintered nickel cathode. The tilted orientation promotes drainage of excess salt from the cell during prolonged discharge, maintaining a dense anode paste. [Patents pending.]

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