

## Electrochemical Oxidation of Graphite Electrodes Contaminated with Common Coal Minerals in a Molten Carbonate Electrolyte

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The electrochemical oxidation of carbon under molten carbonate conditions has had renewed interest over the past two decades as the increasing energy demands, depletion of fossil fuel resources and trends toward greener technologies have stimulated research surrounding devices that satisfy these requirements. The conditions and reactions under which carbon can be electrochemically oxidized in molten carbonates have been investigated so as to implement easily within direct carbon fuel cell (DCFC) technology, providing rapid growth in the technology, and the inventory of lower CO<sub>2</sub> producing, greater coal efficiency, “greener” energy technologies.

Many studies have focused on the electrochemical effects of the contaminant phases by the addition of oxides; i.e., Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, MgO, Fe<sub>2</sub>O<sub>3</sub>, and CaO, which make up the ash component of coals after the proximate analysis technique has been used [1,2]. However, when coals are introduced into a DCFC there is no clear indication that the mineral phases will be pure oxides, but could remain as the original mineral; i.e., quartz, clays, feldspars, carbonates and sulfur containing compounds.

The more commonly found mineral components are silicates such as quartz and clay minerals, sulfides such as pyrite and galena, carbonates and sulfates such as calcite and gypsum (both usually found in the associated water within coal rather than in the actual coals themselves) and some oxides and hydroxides, such as anatase and boehmite [2]. On a by-mass basis, mineral contaminants such as clays, silicates and sulphides (kaolin, quartz and pyrite respectively) make up the major contaminant phases, with some oxides and waterborne minerals making up the minor phases. Priority during selection was given to the major contaminant phases as it was thought they would have the most significant effects on the electrochemical reactions within a DCFC running on coal as a fuel source, as they would accumulate within the cell at a greater rate than the minor phases. Ultimately the selected contaminant phases were quartz, kaolin, montmorillonite,  $\alpha$ -alumina and pyrite.

By taking advantage of low-temperature ashing and XRD techniques, the common components of several coal samples were identified, and chemical or mineralogical equivalents of the phases were introduced into a specifically designed graphite working electrode, and ultimately into an electrochemical cell which could be maintained at DCFC like conditions.

From the results and findings in this study, the addition of common coal mineral contaminants had a discernible effect on the electrochemical performance of carbon

oxidation within the electrochemical test cell. With the exception of quartz, all the contaminants increased the current output of the test cell when introduced into the carbon phase of the working electrode, as shown in Figure 1.

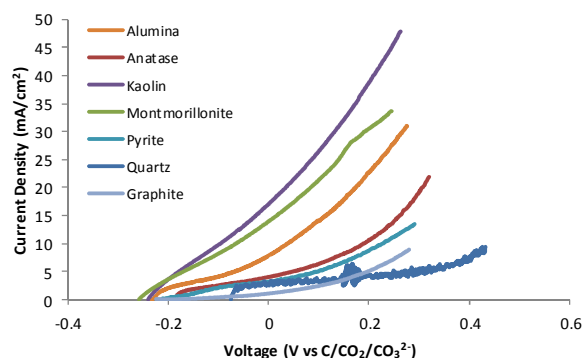


Figure 1. Overlaid view of the contaminated working graphite electrodes with 50% contamination from the selected mineral components from coal.

The increase in performance from the clay, alumina and anatase contaminated electrodes is attributed to the contaminant phase affecting the carbon oxidation mechanism by facilitating the adsorption of the O<sup>2-</sup> species at the reactive site on the carbon that is suspected to be the slowest step in the oxidation mechanism according to literature.

The comparison of the electrochemical results from quartz, alumina and kaolin contaminants indicates that the properties and structure of the contaminant phase plays a significant role in the facilitation of the oxidation reaction. Furthermore, the comparison of these contaminants has highlighted that the supplementation of actual mineral phases found in coals, with their similar oxides i.e., replacing clay with SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and other alkali/alkali earth oxides, it not truly representative of the original contaminant behaviour during the carbon oxidation.

[1] G. A. Hackett, J. W. Zondlo, R. Svensson, Evaluation of carbon materials for use in a direct carbon fuel cell, *Journal of Power Sources*, Volume 168, Issue 1, 25 May 2007, Pages 111-118

[2] Australian Standard AS 1038.3 - 1989, Coal and coke - Analysis and testing Part 3: Proximate analysis of higher rank coal, Originated as part of AS CK2.3 - 1949, North Sydney: Standards Association of Australia; 2000.

[3] Colin R Ward, Analysis and significance of mineral matter in coal seams, *International Journal of Coal Geology*, Volume 50, Issues 1-4, May 2002, Pages 135-168