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Investigation of Electrode kinetics for the Electro-oxidation of Coal Slurries

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Introduction

Coal is an important traditional and cheapest source of energy due to its diversity, availability, chemical stability and competitive price. It is reported that approximately 38% of the worldwide electricity is produced from coal. In the conventional method of gasification of coal reported elsewhere [1], the gasification furnace is kept at high temperature (800°C) by means of combustion heat generated from the oxidation reaction of carbonaceous materials with oxygen. Since this method requires high temperature, which means high temperature sustaining materials of construction and at the same time the process releases SO_x and NO_x. This method also requires the purification of the gases.

In 1979 [2] a new method called electrochemical gasification of coal water slurry using platinum electrodes was carried out, which generated pure streams of CO_2 and hydrogen at anode and cathode respectively free of tar and sulfur compounds. The authors [2] have also reported that the reversible thermodynamic potential of the oxidation of coal is only -0.21 when compared to conventional water electrolysis which is -1.23 V, thus making coal slurry electrolysis more competitive.

The authors [2] have suggested the following reaction scheme taking place during the electro-oxidation of coal-water slurry. The anodic reaction is mainly CO_2 liberation:

$$2H_2O(l) + C(s) \longrightarrow CO_2(g) + 4H^+ + 4e^- \quad (1)$$

The cathodic reaction is hydrogen liberation:

$$4H^+ + 4e^- \longrightarrow 2H_2(g) \quad (2)$$

The overall reaction is:

$$2H_2O(l) + C(s) \longrightarrow CO_2(g) + 2H_2(g) \quad (3)$$

The above steps show the reactions without much in-depth explanation about the reaction mechanism. Since the reactions involve four electrons transfer, which means the intermediate elementary steps have to be determined. Hence the actual mechanism is still unclear and is a matter yet to be explored.

Several authors [3,4] have proposed that the iron content in the coal is mainly catalyzing the reaction. Dhooge et al. [3] tried to elucidate the mechanisms associated with the electrolysis of coal that seems to be in agreement with the observations reported by Coughlin and Farooque [2]. But the authors did not explain that the coal particles have to be in contact with the anode for the electro-oxidation to take place. We are trying to propose a better explanation as to how the reaction of iron takes place at the electrode surface as well as on the coal particle surface.

Within this context, the objective of this work is to determine the reaction mechanism and give an explanation to the above uncertainty. Classical polarization on rotating disk electrode (RDE) will be performed to achieve the objective. The polarization curves will be modeled, and parameter estimation combined with modeling discrimination techniques will be used to elucidate the mechanism.

Experimental

The electro-oxidation of coal water slurries was studied using a Pt RDE. The electrochemical measurements were conducted at 40 °C in a glass cell containing aqueous solution of coal (size 74-105 μm) suspended in 1.0 M H_2SO_4 as electrolyte using Pt rotating disk electrode (0.5 mm diameter). The counter electrode was made of platinum or Pt-Ru, while SCE was used as the reference electrode. The measurements were done using a potentiostatic monitoring system from ARBIN Instruments. To study the effect of mass transfer, experiments were also carried out at different rpm of the electrode. The effect of the catalytic oxidation of iron has been tested by adding known quantities of iron sulfates in the solution. Pittsburgh seam No. 8 was used for the analysis. The coal was characterized before and after any measurement was performed to determine any morphological changes.

Results and Discussion

Figure 1 shows the polarization performance of a coal water slurry solution (0.12 g/ml) containing 100 mM Fe^{3+} swept at 50 mV/s in acidic medium (1.0 M H_2SO_4) using Pt RDE at 40°C for different rpm. The curves indicate increase in limiting currents with increasing rpm and this happened even in baseline (1.0 M H_2SO_4) solution except that the limiting currents are negligible indicating almost no reaction. The maximum limiting current was close to 7.3 mA/cm^2 observed at a potential of approximately 0.6 V and 2100 rpm. It is possible that the limiting currents observed could be mainly due to the oxidation of the iron present in the coal slurry. We will use these results to diagnose the mechanism and to quantify kinetic parameters. Basically we will propose a hypothesis for the mechanism, predicting experimental behavior on the basis of the hypothesis, and comparing the predictions against the results. Further analysis and results will be presented.

References

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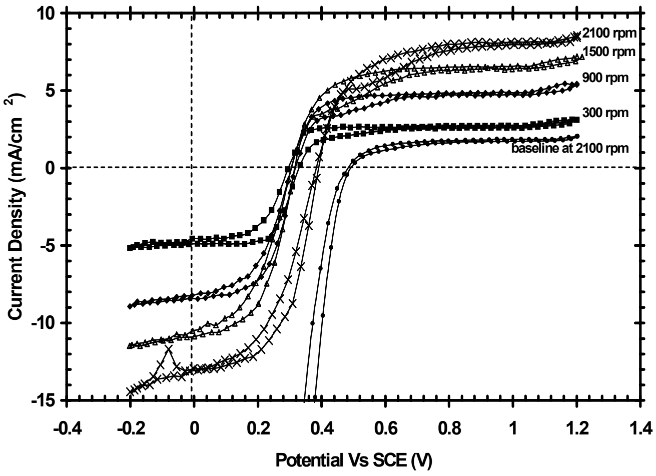

Figure 1. Polarization curves for Pt RDE at scan rate of 50mV/s, coal concentration 0.12g/ml, 1 M H_2SO_4 and 100mM Fe (III) operating at 40°C.