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Lignite as a fuel for direct carbon fuel cell system

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ABSTRACT

Lignite, also known as brown coal, and char derived from lignite by pyrolysis were investigated as fuels for direct carbon solid oxide fuel cells (DC-SOFC). Experiments were carried out with 16 cm^2 active area, electrolyte supported solid oxide fuel cell (SOFC), using pulverized solid fuel directly fed to DC-SOFC anode compartment in a batch mode, fixed bed configuration. The maximum power density of 143 mW/cm^2 was observed with a char derived from lignite, much higher than 93 mW/cm^2 when operating on a lignite fuel. The cell was operating under electric load until fuel supply was almost completely exhausted. Reloading fixed lignite bed during a thermal cycle resulted in a similar initial cell performance, pointing to feasibility of fuel cell operation in a continuous fuel supply mode. The additional series of experiments were carried out in SOFC cell, in the absence of solid fuels, with (a) simulated CO/CO_2 gas mixtures in a wide range of compositions and (b) humidified hydrogen as a reference fuel composition for all cases considered. The solid oxide fuel cell, operated with 92%CO + 8%CO_2 gas mixture, generated the maximum power density of 342 mW/cm^2. The fuel cell performance has increased in the following order: lignite (DC-SOFC) < char derived from lignite (DC-SOFC) < CO + CO_2 gas mixture (SOFC) < humidified hydrogen (SOFC).

Introduction

The direct, electrochemical conversion of coal to electric energy has been discovered in the middle of 19th century. Only in the last decade renewed interest in the direct carbon fuel cells (DCFCs), supported by the unmatched global electricity generation from coal, has led to a significant progress in the field. In a DCFC based on solid oxide fuel cell technology (DC-SOFC), electrochemical oxidation of carbon is combined with electrochemical cathodic reduction of oxygen. The overall electrochemical reaction yields CO_2 as a product, similar to the conventional combustion process. The fuel and oxidant streams are separated in the process, providing additional system design benefits. When carbon monoxide unutilized in the fuel cell is combusted with oxygen, the exhaust gas stream consists of only pure CO_2. The expensive process of CO_2 separation from flue gases can be avoided this way.

In the DCFC cell, the reverse Boudouard reaction is proceeding in the bulk of the coal bed:

$$\text{coal bed:} \quad C + CO_2 \rightarrow 2CO \tag{1}$$

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The electrochemical oxidation of carbon with oxygen ions takes place at the SOFC anode in parallel 2-electron or 4-electron reactions:

$$\text{anode:} \quad C + 2O^{2-} \rightarrow CO_2 + 4e^- \tag{2}$$

$$\text{anode:} \quad C + O^{2-} \rightarrow CO + 2e^- \tag{3}$$

$$\text{anode:} \quad CO + O^{2-} \rightarrow CO_2 + 2e^- \tag{4}$$

When combined with cathodic reaction of oxygen reduction:

$$\text{cathode:} \quad O_2 + 4e^- \rightarrow 2O^{2-} \tag{5}$$

the overall electrochemical reaction yields CO_2:

$$\text{overall electrochemical:} \quad C + O_2 \rightarrow CO_2 \tag{6}$$

From the thermodynamic efficiency viewpoint, reaction (2), exhibiting transference number equal to 4, is the most desirable. The theoretical thermodynamic efficiency of 100.22% is calculated for the reaction (2) as the ratio between the Gibbs free energy change of the reaction and the reaction enthalpy change. It is nearly independent of temperature.

The energy conversion efficiency, defined as the ratio between the useful electric power output and the total LHV energy input of the fuel, is targeted in the 50%–80% range for the DCFC system. In DC-SOFC reactor, electric power generation is possible using coal, biomass, carbon black, graphite, activated carbon, char, and other solid fuels containing significant amount of elemental carbon. One should note that term "direct carbon fuel cell" is not consistent with traditional fuel cell classification convention, based on the electrolyte type rather than the fuel supplied to fuel cell.

Lignite, also known as brown coal, is a low rank coal, formed naturally from peat. It is commonly used for conventional, stationary electric power generation with energy conversion efficiencies approaching mid-40s in the modern power plants. It accounts for a significant portion of generated electric power, reaching 50% in some countries. However, conventional combustion of coals raises challenges over GHG emissions, prices of the emission rights in some regions of the world and decreasing social acceptance.

The majority of the DCFC installations are implemented in a laboratory scale. Most of them are based on the molten carbonate fuel cell technology (MCFC) and solid oxide fuel cell technology (SOFC) [1,2]. Other configurations utilize molten hydroxides of alkali metals [3]. There are hybrid approaches also investigated [1,4]. In DCFC approach based on SOFC technology (DC-SOFC), pulverized solid fuels can be supplied to the anode compartment utilizing a fluidized bed [5], fixed bed, moving bed and other means of fuel supply.

A wide range of solid fuels has been tested in the DC-SOFC, including coal, lignite and biomass [5,6]. The elemental carbon content of dry solid fuels, their heating value and sulfur concentration in the fuel decrease, in general, in the following order: coal > lignite > biomass. In the absence or diminished presence of hydrogen fuel, exhibiting the highest exchange current density, carbon and CO electrochemical oxidation with O^{2-} ions are the primary paths of electric current generation in the DC-SOFC cell. The importance of each path in a specific DC-SOFC configuration is a complex function of the operating temperature, fuel gas atmosphere, catalytic properties and structure of anode material [7]. It is also influenced by the pre-processing methods of the carbonaceous fuel [8], method of solids supply to anode compartment, absence or presence of the anode inlet gas supply stream (CO_2), geometry of the anode compartment and other factors.

Several approaches to implement electric current generation based on SOFC technology and lignite as a fuel have been investigated (Fig. 1):

(a) fuel mixture of CO and CO_2, derived from lignite or lignite char in a dry gasification process can be used, as a gaseous fuel, in an SOFC cell (Configuration I),
(b) lignite received from a coal mine, dried and milled, can be directly fed to anode compartment of the DC-SOFC (Configuration II),

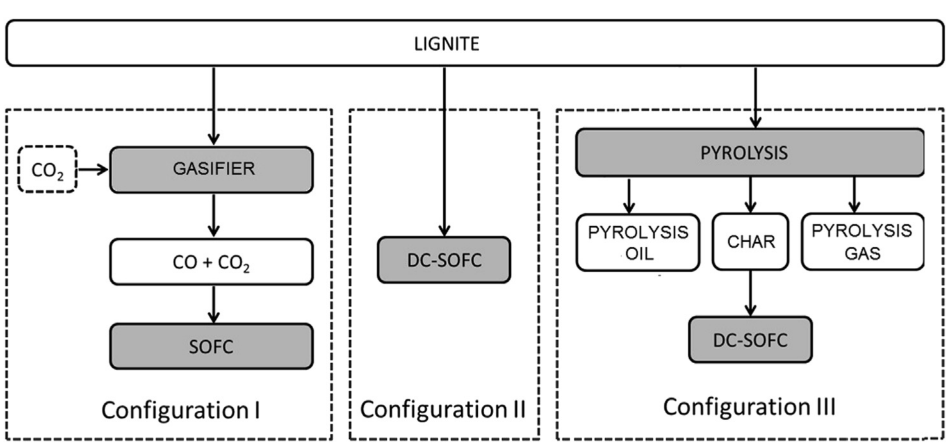

Fig. 1 – Schematic diagram of the configurations considered for the lignite fuelled SOFC.

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