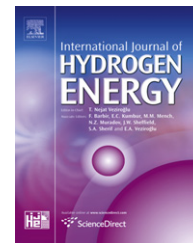


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Electrochemical oxidation of graphite in an intermediate temperature direct carbon fuel cell based on two-phases electrolyte

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ABSTRACT

As a promising intermediate temperature fuel cell, Direct Carbon Fuel Cell (DCFC) with composite electrolyte composed of Samarium-Doped Ceria (SDC) and a binary carbonate phase (67 mol% Li₂CO₃/33 mol% Na₂CO₃) has a much higher efficiency compared with conventional power suppliers. In the present work, SDC powder has been synthesized by an oxalate co-precipitation process and used as solid support matrix for the composite electrolyte. Single cell with composite electrolyte layer is fabricated by a dry-pressing technique using LiNiO₂/Li₂Na₂CO₃/SDC as cathode and 1:9 (weight ratio) graphite mixture with 67 mol% Li₂CO₃/33 mol% Na₂CO₃ molten carbonate as anode. The cell is tested at 600–750 °C using electrolytical graphite mixture as fuel and O₂/CO₂ mixture as oxidant. A relatively good performance with high power density of 58 mW cm⁻² at 700 °C is achieved for a DCFC using 0.8 mm thick composite electrolyte layer. The sensibility of the 1 cm² DCFC single cell performance to the anode gas nature is also investigated. At temperatures higher than 700 °C, both carbon (C) and carbon monoxide (CO) can be considered as reacting fuel for the DCFC system.

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1. Introduction

Demand for electricity production is increasing globally and many R&D groups are working on the development of new power generation technologies. One of these technologies is fuel cell. A fuel cell is an electrochemical device that converts fuel's chemical energy to electricity by electrochemical reactions. As long as fuel is supplied, the cell continues to generate electrical power. Unlike conventional fuel cell that uses hydrogen extracted from natural gas, DCFC consumes solid carbon as fuel.

The overall DCFC reaction is:



The energy conversion efficiency of the DCFC is very high and thus CO₂ emission is correspondingly very low for a given output power. An overall efficiency of about 80% has been reported [1–3].

Ordinary DCFC system operates at very high temperatures, in the range of 800–900 °C in order to generate high power output. The most developed DCFC system is based on molten carbonate electrolyte and solid oxide electrolyte.

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Molten carbonates have been widely used in the molten carbonate fuel cell (MCFC) as electrolytes. They are also attractive for DCFC because of their high conductivity, good stability in the presence of CO_2 produced from the carbon electro-oxidation reaction and suitable melting temperature [1,4]. The molten carbonate salt electrolytes used are K_2CO_3 , Na_2CO_3 and Li_2CO_3 . These carbonates could be used as binary eutectics or ternary eutectics.

The DCFC based on molten carbonate electrolyte operates at a high temperature between 600 and 800 °C to achieve high conductivity of the molten salt. The carbonate ions CO_3^{2-} are considered as charge carrier. Indeed, the ionic transport is governed by carbonate ion transfer. This type of electrolyte is corrosive at high temperatures and it disables the DCFC to operate for an extended period of time without cell degradation [1,2,5].

Weaver et al. [6] presented a molten carbonate electrolyte for direct carbon conversion in 1979. They tested several types of carbon fuels and they reported that devolatilized coal is more reactive than spectroscopic carbon and pyrolytic graphite. They linked the high reactivity to the large surface area and the poor crystallization.

Vutetakis et al. [7] employed a stirred approach to promote various carbons and coal electrochemical oxidation in a molten carbonate electrolyte.

However, Cherepy et al. [1] carried out recently a work using eutectic molten carbonate electrolyte for the direct conversion of solid carbon particulates. They found that the surface area has no strong effects on the carbon discharge rate.

Hemmes et al. [4] tested the electrochemical oxidation of carbon in a 62/38 mol% Li/K carbonate melt using impedance spectroscopy (IS) and cyclic voltammetry (CV). They demonstrated that the electrochemical gasification of coal to CO in a direct carbon fuel cell (DCFC) has thermodynamical advantages through the conversion of heat into power at an efficiency of about 100%.

Li et al. [8–11] confirmed the key results determined by Vutetakis et al. [7] through testing several slurry of carbon samples dispersed in molten carbonate electrolyte. Their result indicates that CO_2 product predominates at 700 °C at 0.1 A cm^{-2} . They found a dependence of the stirring rate on various anode pretreatments that promote wetting. They also investigated raw coal as well as nanofibers produced by catalytic decomposition of methane.

The solid carbon can be wetted by the eutectic molten carbonate, which increases the possibility of the electrochemical oxidation of carbon due to the enhanced active reaction area. The feasibility of the direct conversion of carbon to electricity in the hybrid direct carbon fuel cell (HDCFC), which has an extended anode combining a molten carbonate fuel cell and a solid oxide fuel cell (SOFC) was conceived by Irvine and co-workers [12–14]. They reported good performance by several carbons materials such as graphite, pyrolyzed medium density fiberboard (PMDF) and coal. They recorded a very high OCV at 700 °C after high temperature operation. They demonstrated that low carbonate content in the anode chamber of the DCFC can compromise between the anodic polarization at high carbon loadings and sufficient carbonate for carbon wetting while minimizing the corrosion problems.

Chen et al. [5] reported that anode reaction rate and DCFC output can be improved if carbon particles are pre-dispersed into electrolyte before acting as anode, due to the straightway shift from cathode to anode for CO_3^{2-} provided by the electrolyte soaked in carbon material.

Rady et al. [15] noticed that the HDCFC is a powerful approach that needs further research to overcome the problem of low conductivity at lower temperatures of the used conventional solid electrolyte (YSZ, Yttrium Stabilized Zirconia) in order to improve the output capability of this type of fuel cells.

In addition, reported conductivity of liquid molten carbonate in the literature is higher than that of SDC-carbonate composite electrolyte. However, reports show that the effective conductivity of liquid carbonate electrolyte in a nonconductive porous matrix such as NiO is much lower than the liquid conductivity [16]. So, the carbonate electrolyte effective conductivity seems to be lower than that of the SDC-carbonate composite electrolyte.

For operation at lower temperature, higher ionic conductivities are required [1,2]. Samarium-Doped-Ceria (SDC) has been extensively studied as electrolyte in low-temperature ceramic fuel cells regarding to its high ionic conductivity which can reach $10^{-1} \text{ S cm}^{-1}$ at 800 °C, 200 °C lower than that of the conventional high temperature SOFC electrolyte, (YSZ) having an O^{2-} conductivity of about $10^{-1} \text{ S cm}^{-1}$ at 1000 °C [17–20]. This single-phase electrolyte material is able to ensure super-ionic conduction from low conductive phase to the super-ionic conductive one but, with an accompanying phase structure change [21]. However, doped ceria has some electronic conduction under reducing atmosphere resulting in power loss. At the same time, it also causes material mechanical problems [22].

Recently, ceria–salt composite electrolyte materials have been developed to improve SDC electrolytes. Huang et al. [23] reported a conductivity of 0.13 S cm^{-1} at 500 °C for a composite electrolyte consisted of samarium-doped ceria and Li_2CO_3 – K_2CO_3 carbonates.

The introduction of different inorganic salts could effectively suppress electronic conduction and ameliorate the material ionic conductivity and stability, thus leads to excellent performance at lower temperatures [17]. Compared to the single-phase electrolyte material, the ceria-carbonate two-phase systems ensure the super-ionic conduction through the interfacial regions between the two constituent phases. The super-ionic conduction involves a change in the interfacial properties without an individual phase structural changes. The conductivity of the ceria–salt composite electrolyte materials is then strongly dominated by the coupling and the interactions between the constituent phases. Therefore, the co-existence of the two phases can create the interfacial effects and thus multi-functions known as super-ionic conduction. The ceria–salt composite electrolyte is able to form a dense network and a continuous ion conducting framework and also to maintain good mechanical strength [21].

Various carbon fuels have been tested in different DCFC systems, but the efficiency of carbon fuels remains unclear. The physico-chemical properties of carbon fuels can be seen to highly influence the anodic electrochemical performance in

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