Direct coal fuel cells (DCFC). The ultimate approach for a sustainable coal energy generation.

Pilas de combustible de carbón. Una nueva propuesta para una generación sostenible de energía a partir del carbón mineral.

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Abstract

An alternative for electrical energy generation from coal is presented in this article. The RFCS project *“Efficient conversion of coal to electricity: direct coal fuel cells (DCFC)”* is currently investigating the advantages associated to the technology. To wit: a high energetic efficiency and very low emissions of pollutants. In the midterm report of the project it is shown how the characteristics of the coals used are a key in the behavior and durability of the cell. It seems that coals with high reactivity and low ash content are desirable. Furthermore, the addition of certain catalysts may be important to promote *in situ* gasification of carbon. Currently the work is centered in the upscaling and developments on sealing, interconnections and fuel feeding systems.

Resumen

En este artículo se presenta una alternativa para la generación de energía eléctrica a partir de carbón mineral. El proyecto de la RFCS “*Efficient conversion of coal to electricity: direct coal fuel cells (DCFC)”* está actualmente investigando las ventajas asociadas a la tecnología. A saber: una alta eficiencia energética y muy bajas emisiones de contaminantes. En el último informe del proyecto se muestra cómo las características de los carbones utilizados son una clave en el comportamiento y la durabilidad de la pila de combustible. En principio, son preferibles los carbones con alta reactividad y bajo contenido en cenizas. Por otro lado, la adición de ciertos catalizadores puede ser importante para favorecer la gasificación *in situ* de carbono. Actualmente, el trabajo se centra el escalado y el desarrollo de los sistemas de sellado, interconexiones y alimentación de combustible.

Why combining coal and Direct Carbon Fuel Cells?

Coal is the most abundant, economic and widely distributed fossil resource, presently accounting for more than 30% of the global primary energy consumption [1]. This value is forecasted to remain stable or slightly increased until 2030, implying the primary role of coal in the near future (Figure 1). Nowadays, coal conversion to energy is mainly carried out in conventional power plants, in which, however, the thermodynamically driven low efficiency in conjunction with the increased emissions of environmentally harmful gases per unit of produced energy, render this process unsuitable for a sustainable future. To this end, the development of new technologies for efficient and clean energy production is essential toward a sustainable energy economy [2]. Most likely the solution for assuring future energy supply is to maximise the diversification of energy resources along with energy generation technologies, in the framework of sustainable development.

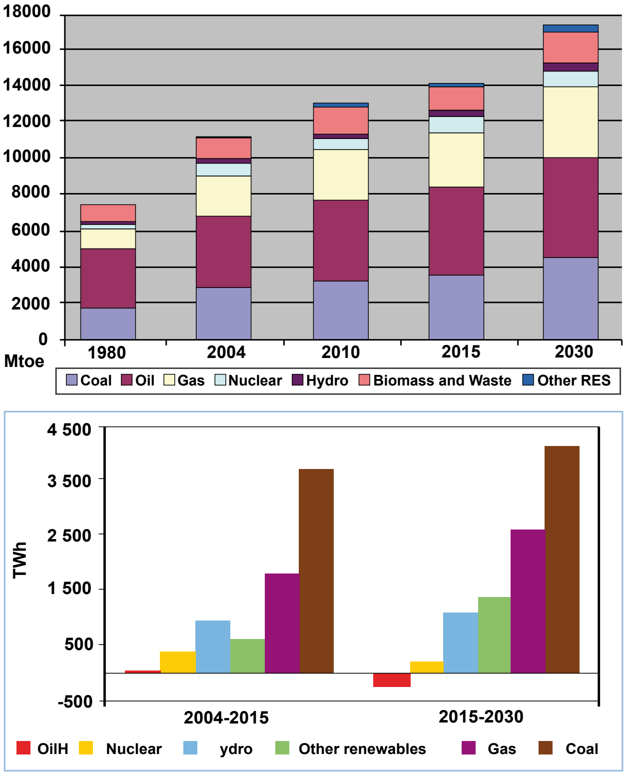

Figure 1. a) Contribution of different energy sources to the CO_2 emissions in the primary world energy consumption according to EU Policy data SEC 2007; b) Forecast coal consumption in electricity generation.

Figura 1. a) Contribución de las diferentes fuentes de energía a las emisiones de CO_2 en el consumo mundial de energía primaria, según EU Policy data SEC 2007; b) Pronóstico del consumo de carbón en la generación de electricidad.

In this context it appears an alternate efficient and environmental friendly approach to produce clean energy from coal, throughout the so called direct coal fuel cells (DCFC), which is an electrochemical device in which the chemical energy of coal is converted to electricity by oxidising carbon into carbon dioxide. This novel technology can effectively exploit the chemical energy of coal in the cleanest and most efficient way among the different energy conversion technologies. Specifically, compared to conventional coal-fired plants, in DCFCs the chemical energy of coal can be directly converted to electricity with a low CO2 footprint per unit of produced energy [3,4]. Moreover, DCFCs have several advantages, compared to conventional power plants and even gas-fed SOFCs, including among others the high efficiency (i.e. theoretical values ca. 100%) [5-7], the high energy density of coal versus other fuels [8] and its abundance and low price, besides ease of handling of the few emissions associated [9].

Background of Direct Carbon Fuel Cells

A DCFC is an electrochemical device in which the chemical energy of carbon is converted to electricity by oxidising solid carbon into carbon dioxide. The first attempt to develop a process for electricity generation from coal has been made by Jacques in 1896 [10,11]. Regarding the electrolytes, various types such as molten hydroxides, molten carbonates and solid oxides have been employed in DCFCs. Among these, the solid oxide DCFCs, such as YSZ-based fuel cells, offer the well established advantages of SOFC technology (chemical and thermal stability, fuel flexibility, etc). However, the weak interaction between solid carbon and solid electrode/electrolyte active interface, is the main barrier toward coal fuel cells development. Therefore, research efforts have mainly been focused on high-temperature liquid electrolyte concepts [7, 12-15], molten hydroxide [8, 9, 24] or molten carbonate electrolytes [12, 25], thus overcoming the restrictions derived from high resistive loss in anode. However, the durability problems related to the corrosive nature of some electrolytes have plagued the development of molten electrolyte fuel cells [12, 25, 26].

For that reason, a method has been developed that combines liquid and solid electrolyte together [16-18], using the solid oxide electrolyte to separate the cathode and anode compartments, while a molten carbonate electrolyte is utilised to extend the anode/electrolyte region and facilitate the diffusion of solid carbon [19-22]. Oxygen is reduced to O^{2-} ions at the cathode and transported across the solid electrolyte membrane to the anode compartment, where carbon is oxidised to CO_2 (see Figure 2).

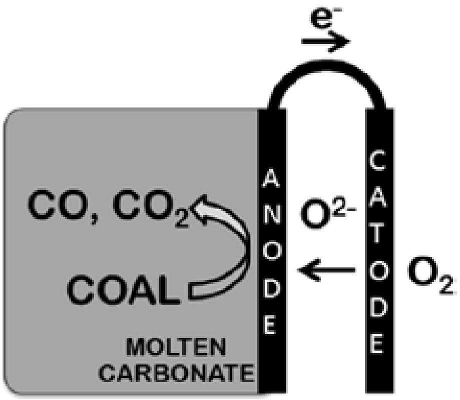

Figure 2. Scheme of a solid electrolyte-based DCFC with coal as a fuel and molten carbonate as a medium to extend the anodic active electrochemical zone.

Figura 2. Esquema de una pila de combustible tipo DCFC en la que se usa carbón mineral como combustible y carbonato fundido, que actúa como medio para ampliar la zona anódica activa.

Various carbon materials have been employed as fuels in DCFCs to reveal their efficacy as energy carriers. It has been found that both the performance and the lifetime of DCFCs are notably affected by the physicochemical properties of carbon fuels [12, 14]. For instance, it was found that the both mineral matter and sulfur may cause a deterioration of DCFC performance [12, 23].

Objectives and project approach

Direct electrochemical conversion of coal to electricity offers very significant increases in efficiency with consequent reductions in CO_2 emissions. Preliminary estimations show that it is possible to double electrical conversion efficiency. The technology also offers easier sequestration of CO_2, than traditional coal combustion, as the exhaust gas is not diluted with nitrogen. Therefore, in the European Project "Efficient conversion of coal to electricity-Direct Coal Fuel Cells (DCFC)" funded by the RFCS, the general objective is to develop DCFC technology. With specific objectives dealing with (i) assessment of different aspects related to the operating conditions, (ii) optimization of stack configurations, (iii) evaluation of different coal sources and processing, (iv) developing new catalysts capable of withstanding the aggressive conditions. Taking into the account all these aspects and the optimization of the durability and efficiency of the DCFC the final objective is to allow a prototype system to be proposed for demonstration studies.

The Project, coordinated by the University of St Andrews, is divided in the following Work Packages (WP):

WP1. DCFC fabrication and optimization

Several aspects regarding cell geometry, sealing, and interconnections are considered in this WP, in order to improve the behavior of the cell and to decrease the ohmic resistance thus allowing the scaling-up of the device.

WP2. DCFC electrocatalysis development

In this WP several catalysts are tested in order to find the most active and stable catalysts under different operating conditions which then will be employed as anodic electrode in DCFC.

WP3. Coal processing and evaluation

This work package aims to exploit different European coals as fuels, to characterize them and eventually to modify them (physically and/or chemically) in order to produce suitable coal fuels for DCFCs. The coal characteristics of the different European sources are correlated with the DCFC behavior assessed in other WPs, in order to make a recommendation of the possible European coal sources for this alternative use.

WP4. Addressing durability

Due to the aggressive nature of DCFC components (e.g. molten electrolyte, impurities, etc) it is necessary to optimize the durability of the devices in the framework of scale-up process. To this end, in this WP several issues related to the corrosion minimization, electrocatalysts stability, etc. are thoroughly considered.

WP5. Dissemination, reporting and management

In this WP several actions are considered to give the more visibility as possible to the project and the results obtained under RFCS funding. Therefore, besides different scientific publications and international meetings, the established reports to the European Commission, etc., a web page has been developed to present the project and also to facilitate the exchange of information between partners (http://dcfc.wp.st-andrews.ac.uk).

Results obtained and next steps

During these 2 years it has been demonstrated the feasibility of using different raw coals (lignites, bituminous and anthracites) as fuel in a hybrid direct coal fuel cell. The results so far obtained clearly revealed that the electrochemical reactivity of coal is highly dependent of its intrinsic properties; thus coal physicochemical characteristics, such as volatile matter, oxygen content and structure disorder greatly enhanced the DCFC performance, probably because they enhance the reactivity of coal. On the other hand, ash and sulphur content inhibit cell performance due to degradation problems. The best DCFC performance is obtained with the bituminous coal, due to its high volatile matter and low sulphur content. Typical results concerning the impact of coal type and its characteristics on the power output are depicted in Figure 3.

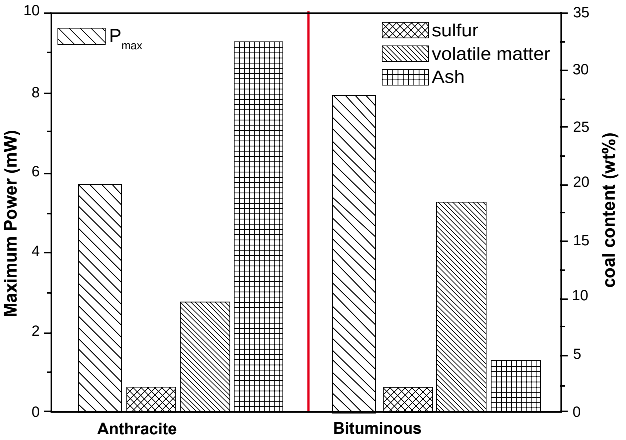

Figure 3. Effect of type of coal, and its characteristics, on DCFC performance at 800oC. Fuel cell type: coal | Cu-CeO$_2$/YSZ/Ag | air; coal loading = 800 mg; CO$_2$ flow = 30 cc/min.

Figura 3. Efecto del tipo de carbón, y sus características en el rendimiento de la celda a 800 º C. Tipo de pila de combustible: carbón | Cu-CeO$_2$/YSZ/Ag | aire; carga de carbón = 800 mg; flujo de CO$_2$ = 30 cc/min.

Low ash and sulphur content is desirable, especially when nickel cermet is used for the electrode in terms of long term stability. Considering the long term operation, demineralisation or another pretreatment of the raw coals should be applied to assure better results.

In this point it is worth underlined the significant process toward the enhancement of DCFC performance though the adoption of a novel, catalyst-aided, internal gasification process. For instance, regarding the commixture of ceria-supported transition metals with coal feedstock results in an increase of cell power of up to 200%, depending on coal type and temperature. The enhanced performance obtained by internally admixing coal fuel with catalyst is ascribed to the pronounced impact of catalyst on the *in situ* coal gasification, and the subsequent electro-oxidation of gaseous products at the anode. Typical results concerning the beneficial effect of catalyst addition to bituminous coal is depicted in Figure 4.

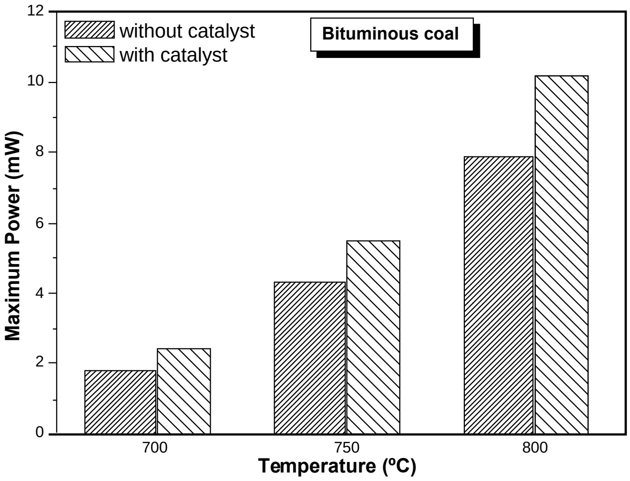

Figure 4. Effect of catalyst addition on the DCFC performance at 700, 750 and 800ºC. Coal loading = 800 mg; catalyst loading = 400 mg; CO$_2$ flow = 30 cc/min.

Figura 4. Efecto del catalizador en el rendimiento de la pila a 700, 750 y 800ºC. Carga de carbón = 800 mg; carga de catalizador = 400 mg; flujo de CO$_2$ = 30 cc / min.

Progress has been made toward scale-up through addressing several aspects related to sealing, interconnections and fuel feeding. New cell architectures have been explored and continuous feed technology has been developed. The particle size of coal is another important issue, since it determines the fuel/electrolyte interaction as well as the rate of carbon transfer in a continuous flow process. In addition, the development of a large fuel entrance is under consideration, in order to ensure the full coverage of the active electrochemical zone by the coal fuel. In Figures 5 and 6 some photographs of different parts of the test-cell are shown.

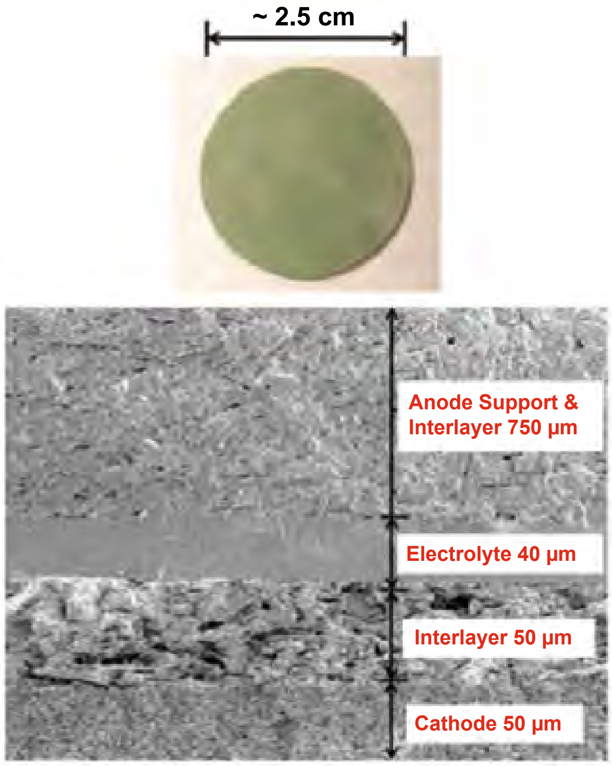

Figure 5. Picture (above) and SEM image (below) of the anode-supported cell.

Figura 5. Fotografía (arriba) e imagen SEM (debajo) del ánodo de una celda.

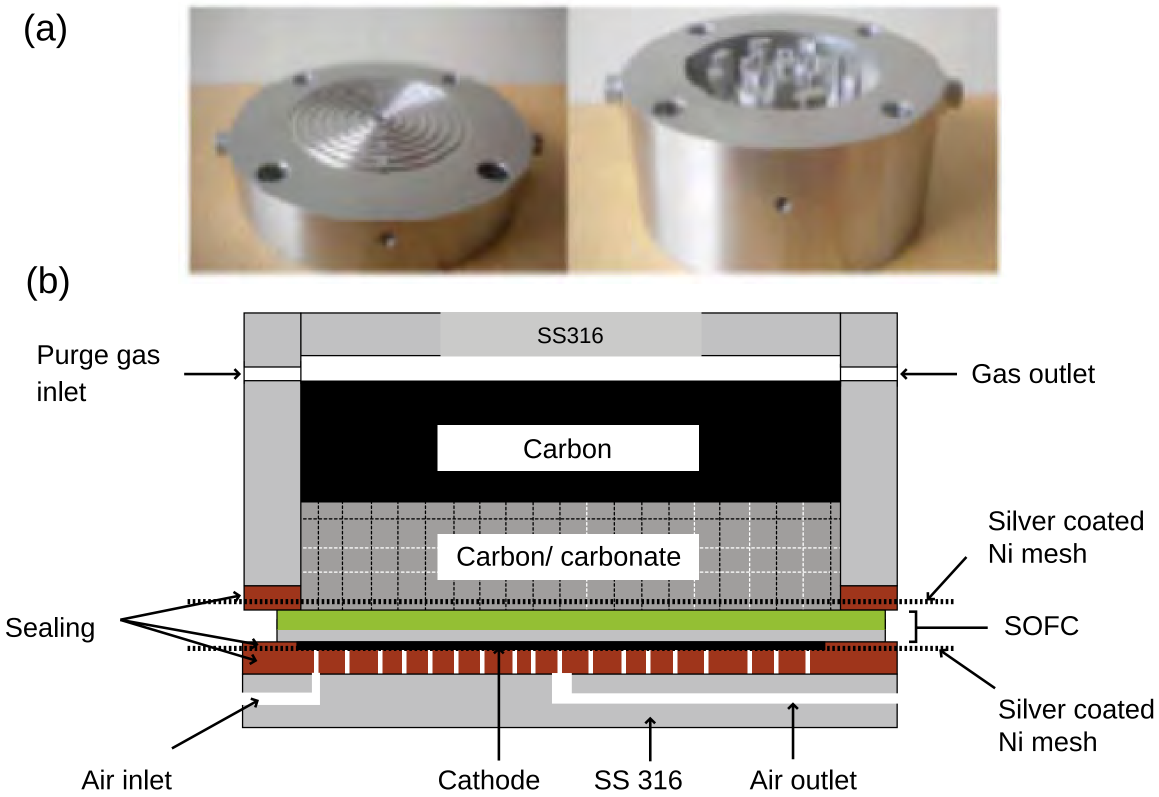

Figure 6. (a) cathode chamber (left) and anode chamber (right), (b) the stack repeat unit for cell testing.

Figura 6. (a) cámara del cátodo (izquierda) y cámara del ánodo (derecha), (b) unidad del empaquetamiento de la celda usada en las pruebas.

At the moment the best initial power production for a given fuel cell reached 1.5 W. Improving current system for continuous power generation and operation of a three-cell stack that produces ~ 4.5 W will be the target in the future.

Acknowledgments

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