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Molten carbonate fuel cell operation under high concentrations of SO₂ on the cathode side

Jarosław Milewski*, Kamil Futyma, Arkadiusz Szczęśniak

Institute of Heat Engineering, Warsaw University of Technology, Poland

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ABSTRACT

Molten Carbonate Fuel Cells (MCFCs) can be used as CO₂ separators in fossil fuel power plants. Fossil fuel power plants emit flue gases with SO₂ contaminants, thus the influence of SO₂ on the operation of MCFC is investigated both theoretically and experimentally. The influence of SO₂ contaminants (up 680 ppm) on MCFC performance was examined. The experimental investigation revealed there is a boundary limit where SO₂ contamination could increase the MCFC voltage.

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Introduction

The European Union has placed limits on CO₂ emissions by Member States as part of its Emission Trading Scheme. The EU's roadmap for long-term low carbon development is to cut carbon emissions by 80% by 2050 compared to the 1990 level in hopes of restricting temperature change to below 2 °C [1]. This impacts fossil fuel power plants to a significant degree, as their emissions are governed by the number of emission allowances they receive from the Member State allocation. Excess CO₂ emissions have to be covered by purchasing extra allowances, which is in effect a penalty. In contrast, under-shooting emission limits enables the emitter to sell CO₂ allowances. The selling price of a traded allowance is estimated at €15/Mg_{CO2}. Presently, fossil fuel based power plants cater to increasing demand for energy associated with improved living standards and industrial expansion [2]. As a result of growing

environmental concerns and the introduction of new technologies (like fuel cells), the power system landscape has changed considerably in recent years [3].

Various types of fuel cells may be distinguished by: different catalysts, different ions as proton carriers, different operating temperatures and different fuels. In general, we may recognize low- (e.g. Polymer Exchange Fuel Cells [4–7]) and high-temperature fuel cells and among the latter Solid Oxide Fuel Cells (SOFC) [8–21] and Molten Carbonate Fuel Cells (MCFC) [22–31]. They both feature high efficiencies and have the highly-desirable feature of methane utilization [32]—already available as a fuel including in bio-fuels. Others need hydrogen feeding [33], which requires a significant amount of energy to produce. Additionally, a high temperature fuel cell can be coupled to a gas turbine for ultra-efficient power generation. Alternatively, small units based on fuel cells can be utilized as power sources in a Distributed Generation system [34].

* Corresponding author.

E-mail address: milewski@itc.pw.edu.pl (J. Milewski).

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Furthermore, MCFCs make it possible to concentrate carbon dioxide, eg. from coal or gas fired power plants [35,36], and might form part of a Carbon Capture and Storage system [37]. To operate, an MCFC requires a flow of CO_3^{2-} as the proton carrier through the electrolyte. This is achieved by feeding CO_2 to the cathode, where it reacts and flows as CO_3^{2-} to the anode. There, after another reaction, it becomes carbon dioxide once again and, after removing water vapor, may be transported as pure gas to the storage point. One might say that MCFCs work as a filter, allowing exclusive flow of CO_2 .

The idea of using a molten carbonate fuel cell to reduce CO_2 emissions was developed by Campanari [38]. In that paper it was shown that an estimated reduction of 77% in CO_2 emissions can be achieved in a steam turbine power plant. A few years later Campanari et al. [39] investigated the possibility of separating CO_2 from combined cycles integrated with Molten Carbonate Fuel Cells. The results obtained show that CO_2 reduction can reach 80% while electrical efficiency remains virtually unchanged, with the power of the cell contributing 17% of the entire system. Amorelli et al. [40] described an experimental investigation into the use of molten carbonate fuel cells to capture CO_2 from gas turbine exhaust gases. They obtained an emission reduction of 50%. Those experiments were performed using a singular cell. Lusardi et al. [41] investigated the application of a fuel cell system for separating CO_2 from thermal plant exhaust. They found that, even without CO_2 separation, the relative emission of carbon dioxide could be reduced to below the Kyoto Protocol limit. If a separator is used, emissions could be reduced by 68%. The use of an MCFC as a carbon dioxide concentrator was investigated by Sugiura et al. [42]. In this work the experimental results of CO_2 sequestration through the use of an MCFC are given. One key conclusion from this work is that the CO_2 removal rate can be obtained by making calculations based on electrochemical theory. In Ref. [43] consideration was given to reducing CO_2 emissions from a coal fired power plant through the use of a molten carbonate fuel cell and in Ref. [44] from a gas turbine power plant and in Refs. [45]—from Fossil Power Plants. Jung-Ho Wee in Ref. [46] analyzed three fields (mobile application, transportation application and stationary application) in terms of CO_2 emission reduction through the use of fuel cells. Only the last of them considered the possibility of using MCFC for the separation of CO_2 . Cryogenic separation of CO_2 from combined cycles integrated with molten carbonate fuel cells was shown by Chiesa et al. [47]. The considered system affords the possibility of separating 80% CO_2 , while the increase in power as a result of the proposed solution is 22% with nearly unchanged efficiency (59% LHV). In Ref. [48] Nansheng Xu et al. presented tests of a new composite—a dual phase mixed carbonation and electron conducting membrane (molten carbonate and silver). The possibility of separation of CO_2 from simulated flue gas was explored, yielding higher CO_2 and O_2 flux densities than molten metal carbonate systems. Based on experimental investigations [49], the achievable CO_2 separation rate from lignite fired boiler flue gas was determined at above 90% by adjusting the cathode inlet flow.

Typical compositions of flue gases from various sources are presented in Table 1. There are several pollutants which can influence MCFC operation as CO_2 separator: primarily SO_x compounds in terms of content. One issue meriting attention

is the effect of sulfur dioxide on the parameters of the MCFC. The sources of sulfur dioxide in flue gas are the sulfur compounds contained in fossil fuels. Although the exhaust gases are desulfurized, they are not deprived of certain quantities of sulfur dioxide. In order to determine the separation capacity with regard to exhaust gas produced by conventional power plants, one must examine the effect of sulfur dioxide on the cathode side.

The MCFC can act as a sulfur scrubber [50], while SO_2 is known to accumulate in the electrolyte as sulfate and is readily transferred to the anode where it can react with hydrogen [51] to form H_2S . This in turn has a strong poisoning effect on nickel based anodes.

The formation of hydrogen sulphide at anode is the most harmful and damaging reaction product in the conversion process [54], and it is advised to remove the sulfur species before the gas enters the fuel cell anode. Hydrogen sulfide poisoning of nickel-based materials can occur through several paths, such as chemical reactions to form bulk nickel sulfides, physical and chemical adsorptions to form a sulfured surface, and electrodic reactions to deposit nickel sulfides through a predominance of electrochemical reactions over chemical reactions [52]. In addition, analysis shows that the poisoning effect is a quadratic function (see Fig. 1a) of hydrogen sulfide which is expressed by the cell voltage, and the drop in voltage asymptotically tends toward a maximum value corresponding to dynamic steady state conditions of poisoning. To investigate these issues, in Ref. [55] an experimental campaign was carried out for the purpose of a preliminary analysis of the effect of H_2S on MCFC performance, highlighting how the main operating parameters affected poisoning phenomena. The results obtained provided new insights for approaching phenomena-reading, confirming the possibility of using MCFCs when a given level of ppm of H_2S is also present in the feeding fuel.

In Refs. [53], the particular effects of the presence of SO_2 on the cathode side of MCFCs were studied. The experimental campaign carried out enabled preliminary analysis of the SO_2 up to 100 ppm—see Fig. 1b—effect on MCFC performance, highlighting the poisoning mechanisms and the influence of the main operating parameters. The poisoning mechanisms proposed in the literature have been confirmed and experimental tests on single-cells showed that SO_2 reacts with the electrolyte on the cathode side forming SO_4^{2-} ions which, after migrating to the anode side, cause the formation of H_2S , whose effects on the anode side are described above. In particular, the effect of SO_2 poisoning was significant even at OCV conditions, suggesting that the chemical reaction for the

Table 1 – Exemplary flue gas composition.

Component	Hard coal	Lignite	Oil	Natural gas
O_2 , vol. %	2.5 ... 10	5	2 ... 6	1 ... 5
CO_2 , vol. %	11 ... 16	10 ... 14	8 ... 10	6 ... 8
N_2 , vol. %	balance	balance	balance	balance
H_2O , vol. %	5 ... 10	16 ... 20	2 ... 6	3 ... 8
NO_x , vol. %	0.1	0.1	0.06	0.05
SO_2 , ppm	<2000	<2600	<1000	0
SO_3 , ppm	11	40	<6	0
dust, mg/m^3	50 ... 150	50	0.05 ... 0.3	0

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