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Performance assessment of natural gas and biogas fueled molten carbonate fuel cells in carbon capture configuration

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HIGHLIGHTS

- MCFC is evaluated for carbon capture purpose downstream a thermoelectric plant.
- Impact of NG and biogas fuels on electric and CO_2 capture performance is evaluated.
- MCFC reached high CO_2 capture levels with both fuels.
- NG showed slightly better performance both in CO_2 capture and power production.
- At parity of MCFC installed power NG feeding resulted in a lower capital cost.

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ABSTRACT

The ability of MCFCs as carbon dioxide concentrator is an alternative solution among the carbon capture and storage (*CCS*) technologies to reduce the CO_2 emission of an existing plant, providing energy instead of implying penalties. Moreover, the fuel flexibility exhibited by MCFCs increases the interest on such a solution.

This paper provides the performance characterization of MCFCs operated in CCS configuration and fed with either natural gas or biogas. Experimental results are referred to a base CCS unit constituted by a MCFC stack fed from a reformer and integrated with an oxycombustor. A comparative analysis is carried out to evaluate the effect of fuel composition on energy efficiency and CO_2 capture performance.

A higher CO_2 removal ability is revealed for the natural feeding case, bringing to a significant reduction in MCFC total area (−11.5%) and to an increase in produced net power (+13%). Moreover, the separated CO_2 results in 89% (natural gas) and 86.5% (biogas) of the CO_2 globally delivered by the CCS base unit. Further investigation will be carried out to provide a comprehensive assessment of the different solutions eco-efficiency considering also the biogas source and availability.

1. Introduction

During the last decades, the growing concern about global warming and the simultaneous awareness of fossil fuels scarcity have pushed scientific and industrial research towards innovative solutions in the field of carbon capture and sequestration (CCS) and renewable energy. So far, strong efforts have been done in multidisciplinary contests (e.g. experimental and numerical studies focused on power systems design and optimization) to find out the best technical alternatives for facing the increasing energy demand and the urgent topics related to environmental sustainability and energy efficiency. Several studies have shown how these critical topics could find a promising solution in Molten Carbonate Fuel Cell (MCFC) technology, thanks to its excellent efficiency and intrinsic operation.

Fuel cells represent one of the most efficient devices for direct energy conversion. Amongst them, MCFCs can be operated flexibly with different fuel compositions and can also be exploited as carbon dioxide concentrators. Indeed, the electrochemical process that takes place within MCFCs (Eqs. (1) and (2)) involves the migration of CO_2 molecules from the cathode to the anode of the fuel cell (FC). This process plays a key role in the FC power generation and can be useful for separating a pure CO_2 stream from some kind of exhausts

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gases fed to the cathode side, following a concept already investigated in several simulation [1–5] and experimental [6–9] works, for large and small scale applications. Moreover, on the anode side, the combination of high operating temperature (~650 °C) and catalytic metals within FC layers (Ni-based) fosters the internal fuel reforming, enabling the fuel cell operation with natural gas, biogas or other gaseous fuels directly.

Fuel flexibility is widely acknowledged as one of the most attractive features of these devices and gives way to the utilization of renewable and other unconventional fuels. This solution has been widely discussed in literature where information about some demonstration pilot and industrial FC modules are available, for instance the thermal integration of MCFCs and anaerobic digesters to produce heat and power in waste water treatment stations facilities [10] wherein a moderate voltage loss with respect to pure natural gas operation must be acknowledged owing to the mixture of biogas and natural gas that is supplied to the anode [11].

Despite this efficiency reduction, which is naturally associated to the different fuel composition, several publications recognize the enormous synergy potential between fuel cells and biogas systems, considering the wide deployment of biogas production facilities and the possibility of providing a de-centralized power production by means of a sustainable and clean primary energy supply [12]. One of the critical point of this joint development is the design of a proper and cost-effective fuel processing system. Biogas clean-up processes involve the installation of ultra-efficient purification technologies, for reasons associated to the severe poisoning effect of H_2S, mercaptane, siloxane and other minor trace gases on MCFC catalytic materials [12–15]. In particular, sulfur traces are a well recognized issue for high temperature fuel cells, whose performances and durability are strongly affected by the poisoning mechanisms that arise on the anode and cathode surfaces (nickel sulphide formation), and also within the electrolyte (sulphate formation) [16]. For this reason the tolerance on H_2S (on the anode side) and SO_2 (at the cathode) is commonly assumed to be very close to 1 *ppm* [17–19].

Small size fuel cell power plant fed by natural gas/biogas have already been considered a promising solution for CO_2 separation when integrated in a wastewater treatment facility [1]. In this framework, the unusual working conditions that arise from both CCS assessment (high CO_2 utilization factors) and utilization of alternative fuels (different H/C ratios with respect to NG), require a deep investigation on MCFC behavior inasmuch as these could meet operational limits on waste heat management or critical overpotential losses (due to high U_{CO_2}). In this work, the particular operative conditions of this kind of power plants is considered which could influence the system design and plant performances in terms of electric and CO_2 capture efficiencies; these aspects are herein evaluated through comparative analysis.

Natural gas and biogas feeding options are analyzed and compared in Ref. [20] but only for MCFC installations in wastewater treatment facilities with trigeneration scopes. Nevertheless, the study does not consider the CCS application and is based only on modeling results. Other authors approach the integration of wastewater treatment facilities with MCFC units [21]; not including CCS application and considering biogas utilization only; unfortunately, this work does not rely on experimental data. MCFCs powered by biogas are further investigated in Ref. [22] through model simulations considering both CCS and trigeneration scopes. Comparative assessments based on experimental data and relative to the exploitation of biogas and city gas are finally provided in Refs. [23], but they deal with a different application based on a MCFC/micro gas turbine hybrid plant which does not include CCS.

This literature review therefore evidences the lack of studies on biogas-fed MCFC based on a specific experimental investigation. The main contribution of the present study is thus to approach a comparative analysis based on experimental tests performed at lab-scale on MCFCs operated in CCS configuration and fed with either natural gas or biogas. Moreover, these tests were carried out with cathodic streams reproducing real exhaust gases from conventional internal combustion engines.

The specific target of the present work is hence to characterize and compare the utilization of MCFCs in CCS application with natural gas or bio gas fuels. In order to obtain a generalized result, this analysis is performed on a reference base CCS unit installed downstream of a reciprocating internal combustion engine (ICE). The fuel cell CCS subsystem is in practice constituted by a MCFC stack directly fed from a reformer and integrated with an oxy-combustor and ASU assembly (*Air Separator Unit*). A comparative analysis is carried out to evaluate the effect of fuel composition on energy efficiency and CO_2 capture performance of the considered base CCS unit, as well as on its sizing. Furthermore, improvements that could be attained in terms of CO_2 performance under particular operating conditions and potential reductions of investments costs are investigated for the operation on natural gas, biogas and specific fuel blends reported in Ref. [1] as result of system optimization.

From a practical standpoint, all the evaluations are based on results from preliminary experimental activities aimed at investigating (through the determination of polarization curves) the fuel cell behavior in CCS applications. Overall, three different reformed fuels (obtained by a proper combination of natural gas and biogas) and three different cathode gases, all derived from assessments relative to plant layouts discussed in Refs. [1], are considered for the experimental activity whose test campaign is then scheduled to fully characterize the MCFC behavior in terms of H_2, O_2 and CO_2 utilization factors, resulting in the functional relations between electric efficiency, CO_2 capture rate and current density. These laboratory simulations are then exploited for a first theoretical design of the base CCS unit to be ideally integrated as a carbon concentrator in a sewage treatment facility.

Finally, this work has been further extended with the techno-economic simulation of the most interesting case studies among those initially investigated. The techno-economic evaluations aim to identify the most profitable MCFC feeding conditions and, therefore, to optimize plants layouts previously considered for the retrofitting of a wastewater treatment plant equipped with reciprocating engines. Specifically, techno-economic evaluations are approached by assuming two alternative design criteria (constant installed MCFC electric power or constant exhausts flow rate to be processed), both assessed on the basis of the most promising configurations emerged from the experimental activity.

The main findings are the ability of the CCS unit (MCFC + oxycombustor) to store 89% and 86% of the treated carbon dioxide for the natural gas and biogas fueled systems respectively. The former solution produces 0.485 $kWh \cdot kg_{CO_2}^{-1}$ (*kWh* for kilogram of stored CO_2) whilst the latter reaches 0.41 $kWh \cdot kg_{CO_2}^{-1}$.

Moreover, the study confirms the extreme importance of MCFC capital cost on the competitiveness of this type of installations, which enables the efficient capture of CO_2 even if only after a wide deployment of the technology that ensures mass production and hence lower costs; otherwise the solution is technically feasible but not economically appealing.

2. Carbon capture from an existing plant

2.1. MCFC: operating principle

MCFCs are DC power units that exploit the following electrochemical reactions to generate current (even if other secondary reactions may contribute to generate electricity):