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Characterization and density separation of coal gasification residues generated from the Zecomix research infrastructure

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ABSTRACT

This paper presents the results of characterization investigations carried out on the solid residues produced during coal gasification tests performed in the Zecomix (Zero Emission of CarbOn with MIXed technology) research infrastructure. In this pilot-scale plant, coal is gasified in a steam/oxygen-blown bubbling fluidized bed containing olivine. The solid residues, collected both directly from the solid bed (bed ash) and downstream from it (mixed ash), were characterized in terms of their main physical, chemical and mineralogical properties with the aim of identifying suitable management strategies for each of them within the Zecomix process. Thus, an experimental protocol was also developed to separate the organic and inorganic fractions of both ash types. The main constituents of the bed ash were Mg, Si and Fe, which represent the elemental components of olivine. The total organic carbon content of the bulk bed ash was of 5%, while that of the bulk mixed ash proved to be significantly higher (24–27%). Finally, the particle size and density separation procedure developed in this work showed to be effective for separating the organic and inorganic fractions of the bulk samples of both types of residues, allowing to reach separation efficiencies higher than 90%.

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1. Introduction

Security of energy supply and CO_2 emissions are two growing concerns for an increasing number of countries. Coal, an abundant worldwide resource, will still play an important role in the domestic energy portfolio mix of the near future. Because of the high emissions associated to its utilization, however, countries are under strong pressure to exploit clean technologies for coal use and limit CO_2 emissions. Carbon capture and storage (CCS) appears as a promising technology option in the framework of decarbonized energy production. The research of new technologies based on more efficient materials and more efficient design for integration of CO_2 capture technologies in power cycles is a promising way to ensure, in the medium term, costs and energy performances comparable to those of current power plants without CCS. Zecomix (Zero Emission of CarbOn with MIXed technology) represents an Italian initiative led by ENEA, the National Italian Agency for New Technologies, Energy and Sustainable Economic Development, aimed at combining clean coal technologies and CCS in order to substantially reduce the environmental impacts related to coal use [1]. Zecomix is an integrated pilot-scale platform where coal is gasified and the yielded synthetic gas is decarbonized in order to be used as a H_2 rich fuel in a gas turbine. The experimental activities of the Zecomix platform, with regard to coal gasification, started within the framework of a program funded by the Italian Ministry of Economic Development [2]. The main aim of the project is to demonstrate, through a series of modeling and experimental activities, the feasibility of an innovative process for producing electricity and hydrogen. In the Zecomix project, a mix of different processes, such as coal gasification, syngas treatment, capture of CO_2 and combustion of hydrogen in a gas turbine was integrated in a single research infrastructure. The key factor for the high energetic and environmental performance of the overall process relies in the optimization of the integration of the different technologies described above. The Zecomix experimental pilot plant was designed and constructed to test each of these technologies independently. The plant was presented in international research frameworks (e.g. CSLF, ZEP and EERA) as one of the advanced technological initiatives of the European and worldwide scientific community. Moreover, the Zecomix plant was evaluated and inserted both in the first Italian Roadmap towards large research facilities conceived by the Ministry of Research and in the Meril database for the mapping of the European research infrastructure landscape.

The main units of the platform, as shown in Fig. 1a, are: (i) the coal bubbling fluidized bed (BFB) gasifier, (ii) the calcium looping (CaL) decarbonizing reactor and (iii) the 100 kWe micro-turbine modified to accommodate a mixture of H_2 and air.

The coal gasifier is a 500 kW_{th} steam/oxygen blown bubbling fluidized bed reactor. The coal feed system was designed for a nominal load of 1.4 t/d of coal. The system includes a 2 m^3 unit for coal storage, which allows the stationary operation of the gasifier for up to 36 h, and two screws driven by an engine connected to an inverter. The

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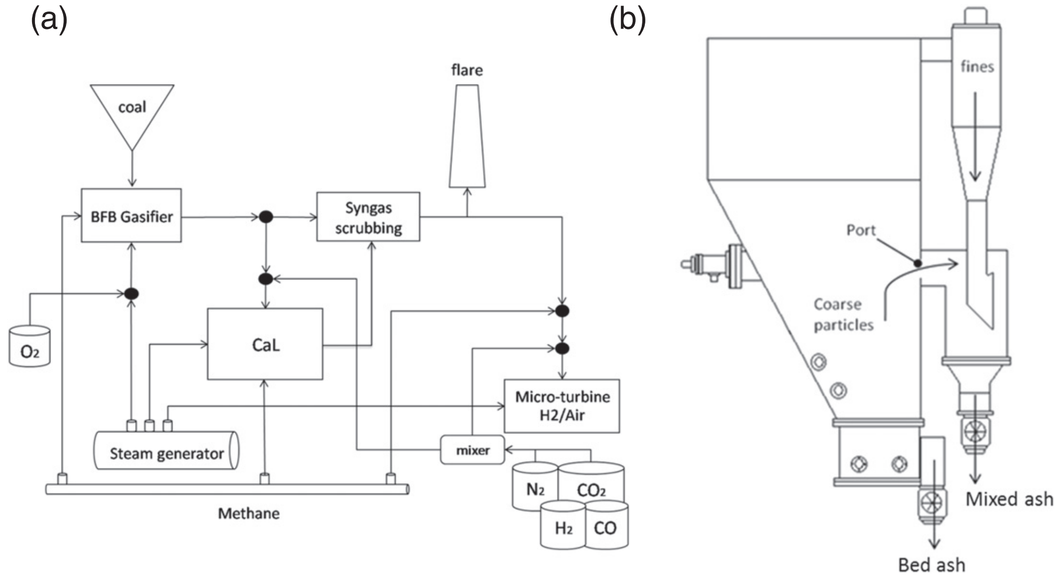

Fig. 1. (a) Schematic layout of the Zecomix platform; (b) section of the BFB gasifier unit showing the locations from which the two types of ash analyzed in this work were sampled.

particulate solid bed of the coal gasifier is made up of olivine ($[Fe,Mg]_2SiO_4$) which acts as a catalyst agent for tar cracking [3,4]. The coal is gasified by means of a steam and oxygen flux, which is fed at different points of the reactor, in order to control the hydrodynamics of the solids and the reaction rate in the entire reactor. The fuel obtained as a product of the gasification process, defined as synthetic gas or syngas, is composed of H_2, CO, CO_2 and steam at temperature of 820 °C, and is sent to a regenerative heat exchanger which reduces its temperature to around 600 °C. After this device, the syngas should be sent to the decarbonizing reactor and then treated, or directly cleaned by scrubbing, before being sent to the micro-turbine for electricity production.

In this paragraph, a short description of the decarbonizing unit installed downstream the gasifier, as well as of the other main units making up the Zecomix platform, is provided. Among the various technologies for separating (or capturing) CO_2 from gas streams, the calcium looping (CaL) option with CaO-based solid sorbents was implemented in the Zecomix platform. One of the key advantages of this option, which makes it a valuable route for gas decarbonization, is the low cost and wide availability of the feed material (a naturally occurring sorbent such as limestone or dolomite) and its high reactivity with CO_2. Moreover, CaO-based solid sorbents are more environmentally benign compared to other state-of-the-art reagents (e.g. amine-based liquid solvents). In the CaL process, CaO is converted into $CaCO_3$ by reacting with gaseous CO_2, then the spent solid sorbent is regenerated in a calcination step at 800–850 °C leading to the release of the captured CO_2 in a pure stream that can be sent to final storage. In addition, in the decarbonizing unit, which is a fluidized bed reactor, CO-shift and steam reforming are also carried out in order that the syngas is fully decarbonized and is mainly made up at the outlet of this unit by a mixture of steam and hydrogen. Syngas cleaning consists in two-stage water scrubbing; then the H_2-rich gas is dried and compressed to make it suitable as a fuel for a 100 kWe micro-turbine. Moreover, a water steam generator is envisaged for feeding the gasifier, the carbonator and the micro-turbine (see Fig. 1a). Finally, there is an oxygen pre-heater for preventing condensation of water in the oxygen/steam mixture and a flare for the combustion of the syngas generated by the coal gasifier when the micro-turbine is not employed.

Besides syngas, also solid residues are generated during coal gasification, namely bottom or bed ash or slag, the material left in the reactor after the thermal process, and fly ash, obtained from the cyclone placed downstream the gasifier for particulate removal from the produced syngas. Gasification bed ash is a typically amorphous and inert solid, mainly made up of mineral matter of the feed coal, unburnt carbon due to incomplete coal gasification and other solid materials employed in the process. Hence, the characteristics and composition of these by-products are strongly affected by the properties of the feed material and the operating conditions of the specific gasification technology adopted, such as feedstock dimensions and the operating temperature and pressure of the gasification chamber [5,6]. Gasification bed ash is generally characterized by a high content of glassy compounds (SiO_2 and Al_2O_3) related to the mineral constituents of the coal and, from an environmental point of view, by an overall limited release of potentially toxic metals compared to other types of thermal treatment residues, due to its vitreous character [7,8]. However, depending on the coal conversion grade of the gasification technology employed, the slag sample may present various percentages of a carbon-rich fraction, called char. There are three main types of gasifier configurations depending on syngas and coal flow characteristics: entrained flow, fixed (or moving bed) and fluidized bed [6]. The slag removed from an entrained bed gasifier is usually made up by a coarse, vitreous and inert fraction characterized by a low carbon content (around 30%), and a fine more porous ash presenting a higher unburnt carbon content (around 60%) compared to the coarse slag [9,10]. Heterogeneous and irregularly shaped clinker samples, with a diameter greater than 1 cm, consisting of grey stone in which a black glassy matrix is included, as well as coarse particles of unburnt carbon-rich particles, are the main output stream of the Sasol Lurgi fixed bed gasifier [11–13]. The residues resulting from a fluidized bed gasifier, such as the one installed in the Zecomix plant, are usually made up by both the mineral material that makes up the fluidized bed and acts as a catalyst (e.g. silica sand or olivine) [3], and residual unconverted coal presenting a significant carbon content (more than 75% and depending on the coal used as feedstock) [14]. The incomplete carbon conversion of the fluidized bed gasification technology is the consequence of the lower operating temperatures employed (typically in the range 800–1050 °C) that are kept well below ash fusion temperatures of the fuels in order to avoid ash melting and hence clinker formation [6]. As for coal gasification fly ash, these residues are characterized by a particle size varying from 4 to 153 μm [14] and present a predominant Al–Si glass matrix and an amorphous character, along with different crystalline reduced phases, such as sulfides [7].

It is estimated that a gasifier operating in a 425 net MW Integrated Gasification Combined-Cycle (IGCC) plant burning a coal characterized by a 10% ash content generates about 450 to 500 t/d of solid residues [15]. So, considering that the coal gasification technology should be

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