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Investigation of hydrogen and power co-generation based on direct coal chemical looping systems

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ABSTRACT

This paper evaluates hydrogen and power co-generation based on direct coal chemical looping systems with total decarbonization of the fossil fuel. As an illustrative example, an iron-based chemical looping system was assessed in various plant configurations. The designs generate 300–450 MW net electricity with flexible hydrogen output in the range of 0–200 MW_{th} (LHV). The capacity of evaluated plant concepts to have a flexible hydrogen output is an important aspect for integration in modern energy conversion systems. The carbon capture rate of evaluated concepts is almost total (>99%). The paper presents in details evaluated plant configurations, operational aspects as well as mass and energy integration issues. For comparison reason, a syngas-based chemical looping concept and Selexol®-based pre-combustion capture configuration were also presented. Direct coal chemical looping configuration has significant advantages compared with syngas-based looping systems as well as solvent-based carbon capture configurations, the more important being higher energy efficiency, lower (or even zero) oxygen consumption and lower plant complexity. The results showed a clear increase of overall energy efficiency in comparison to the benchmark cases.

1. Introduction

The world is facing the challenge of reducing its anthropogenic greenhouse gas emissions and securing its primary energy supply to satisfy the growing energy needs, while improving the economic competitiveness. In particular, the European Union (EU) is committed to reduce its greenhouse gas emissions by at least 20% compared to 1990 levels by 2020 and by 85–90% by 2050 [1]. Also, other relevant EU targets by 2020 are aiming to increase the renewable energy sources share in the energy mix at 20% and reducing by 20% the energy consumption by increasing the energy efficiency. The transition to a low-carbon economy can only be realized through the acceleration of development of a diverse portfolio of low-carbon energy technologies, which, in turn, will enable the timely commercialization and large-scale deployment of these technologies in the energy sector [2–5].

Currently, fossil fuels (coal, lignite, natural gas, oil) are the backbone of the European energy system, supplying more than 50% of power generation capacity. Coal provides for about 60% of fossil fuel electricity, while natural gas provides for most of the balance [6]. Furthermore, all projections show that fossil fuels will remain the main source for electricity generation in Europe at least in short to medium term [7], despite the significant ongoing efforts to promote renewable

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energy technologies and energy efficiency. In this respect, carbon capture and storage (CCS) technologies applied in high-efficiency power plants represent a critical component of the low-carbon technology portfolio for the transition to a low carbon economy since the utilization of fossil fuels in power generation is the main source for greenhouse gas emissions [8–10].

Many overviews of various carbon capture technologies can be found in the literature [4,5,11–16]. Today, the most technologically mature carbon capture methods are based on post-combustion gas–liquid absorption using alkanolamines. Pre-combustion capture is representing also, a promising alternative for energy conversion technologies based on partial oxidation processes (reforming, gasification). One of the major drawback of carbon capture technologies based on gas–liquid absorption is the significant energy penalty imposed by heat duty to regenerated the solvent. The energy penalty of post-combustion capture options is of the order of magnitude about 10 net electricity percentage points [13,17–19]. Pre-combustion capture options show slightly lower energy penalties in the range of 8–9 net electricity percentage points [12,20,21]. In this respect, proposing other innovative carbon capture methods for reducing significantly the energy penalty is of paramount importance.

One of these innovative carbon capture technologies which are very promising in reducing the energy penalty is chemical looping [22]. In a chemical looping system, an oxygen carrier (usually metallic oxides of iron, copper, nickel, mangan etc.) is used to provide the oxygen needed for fuel total or partial oxidation. The reactor is which the fuel is oxidized by the oxygen carrier is called fuel reactor. The gas stream leaving the fuel reactor contains mainly CO_2 and H_2O. After water condensation and removal, a captured CO_2 stream results ready to be sent to geological storage sites. The reduced form of the oxygen carrier is then oxidized back to its initial oxidation state. As oxidant agents air, steam or a combination of both can be used. When only air is used as oxidation agent for oxygen carrier reoxidation, the whole process is called chemical looping combustion (CLC). Most of previously evaluations of chemical looping systems were geared along this line in direct comparison to combustion processes. In case of CLC, heat and power are the only energy vectors to be generated.

In contrast, when steam is used for partial reoxidation of the oxygen carrier (the rest of the reoxidation process being done, as in case of CLC, with air), hydrogen is produced. The main advantages of chemical looping for hydrogen (CLH) production are: supperior energy efficiencies for instance in case of iron and nickel-based oxygen carriers [23], very high hydrogen purity at any given production scale [24] and the capability of generating various energy vectors (hydrogen, power, heat, synthethic fuels etc.) [25].

There are significant characteristics of the chemical looping systems which are promising in respect of reducing energy penalty of the carbon capture process [25,26]. Firstly, as mentioned above, chemical looping systems provide an inherent carbon capture process. The gas leaving the fuel reactor is cooled down and, after water condensation and condense removal, it contains the captured CO_2. No additional solvents are needed. Secondly, chemical looping systems are promising a very high carbon capture rate (close to total decarbonization of the used fuel). Thirdly, since the fuel, steam and air reactors are operated at high temperature (600–1000 °C) the potential of heat recovery at high temperature is significantly enhanced. This aspect will be reflected in superior energy efficiencies compared with conventional energy conversion technologies. Various chemical looping configurations are proposed and evaluated in the literature [22,27–31]. Up to now, most of the evaluations were concentrated mainly for converting gaseous fuels (natural gas or syngas resulted from reforming and gasification processes) in a chemical looping combustion arrangements. These systems are much simpler than the corresponding ones which are using solid fuels in the sense that the solid phase (oxygen carrier in various oxidation states) is not contaminated with ashes. Significant challenges are laying ahead for development of solid fuel chemical looping systems but considering the superior efficiencies and wider reserves of these fuels more efforts have to be done to promote the research in this field [32,33]. In addition, the ability to produce other energy vectors than heat and power (as for CLC) is also of great importance.

This paper evaluates in details direct coal chemical systems looping for hydrogen and power co-generation. As evaluated oxygen carrier, an iron-based system was investigated. The choice of iron oxide as oxygen carrier is based on good fuel conversion yields as well as the possibility to use the spent solid carrier in metallurgical sector [33]. Various plant configurations were proposed for generation of 300–450 MW net electricity with flexible hydrogen output in the range of 0–200 MW_{th} (based on hydrogen LHV). A flexible power plant architecture offers significant techno-economic and environmental advantages considering the emerging hydrogen market and developing low-carbon applications (e.g. PEM fuel cells for mobile applications, other hydrogen-based energy technologies).

2. Plant configurations for direct coal chemical looping systems

The first hydrogen and power co-generation concept (noted Case 1) is based on direct coal chemical looping system in which the fuel (Romanian high grade coal) was oxidized in the fuel reactor (FR) with iron oxide (hematite) according to the reaction:

$$Fe_2O_3 + Coal(C_xH_yO_zN_mS_n) \rightarrow Fe/FeO + CO_2 + H_2O + N_2 + SO_2 \quad (1)$$

This case do not use any other oxidant except the oxygen carrier to convert the coal. The reduced form of the oxygen carrier (Fe/FeO) is oxidized back in a separate reactor using steam (named subsequently steam reactor – SR) to partially regenerate the iron oxide (in form of magnetite – Fe_3O_4) and to produce hydrogen according to the reaction:

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2 \quad (2)$$

To provide the needed heating balance for Reaction (1) which is endothermic, the magnetite is further oxidized