INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2014) 1–10

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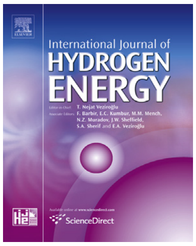

Assessment of CO_2 capture options from various points in steam methane reforming for hydrogen production

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ARTICLE INFO

Article history:
Received 11 July 2014
Received in revised form
24 September 2014
Accepted 29 September 2014
Available online xxx

Keywords:
Steam methane reforming
Hydrogen production
CO_2 emission
CO_2 capture
Oxygen enrichment

ABSTRACT

Steam methane reforming (SMR) is currently the main hydrogen production process in industry, but it has high emissions of CO_2, at almost 7 kg CO_2/kg H_2 on average, and is responsible for about 3% of global industrial sector CO_2 emissions. Here, the results are reported of an investigation of the effect of steam-to-carbon ratio (S/C) on CO_2 capture criteria from various locations in the process, i.e. synthesis gas stream (location 1), pressure swing adsorber (PSA) tail gas (location 2), and furnace flue gases (location 3). The CO_2 capture criteria considered in this study are CO_2 partial pressure, CO_2 concentration, and CO_2 mass ratio compared to the final exhaust stream, which is furnace flue gases. The CO_2 capture number (N_{cc}) is proposed as measure of capture favourability, defined as the product of the three above capture criteria. A weighting of unity is used for each criterion. The best S/C ratio, in terms of providing better capture option, is determined. CO_2 removal from synthesis gas after the shift unit is found to be the best location for CO_2 capture due to its high partial pressure of CO_2. However, furnace flue gases, containing almost 50% of the CO_2 in produced in the process, are of great significance environmentally. Consequently, the effects of oxygen enrichment of the furnace feed are investigated, and it is found that this measure improves the CO_2 capture conditions for lower S/C ratios. Consequently, for an S/C ratio of 2.5, CO_2 capture from a flue gas stream is competitive with two other locations provided higher weighting factors are considered for the full presence of CO_2 in the flue gases stream. Considering carbon removal from flue gases, the ratio of hydrogen production rate and N_{cc} increases with rising reformer temperature.

Introduction

In order to keep global warming to less than 2 °C, some state that the atmospheric CO_2 concentration should not exceed 450 ppmV CO_2-equivalent [1]. Many measures have been proposed to mitigate global warming, including carbon dioxide capture and sequestration (CCS), which some propose as an effective way to stabilise atmospheric carbon dioxide concentrations [2,3]. Fig. 1 presents a breakdown of industrial

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http://dx.doi.org/10.1016/j.ijhydene.2014.09.161

Please cite this article in press as: Soltani R, et al., Assessment of CO_2 capture options from various points in steam methane reforming for hydrogen production, International Journal of Hydrogen Energy (2014), http://dx.doi.org/10.1016/j.ijhydene.2014.09.161

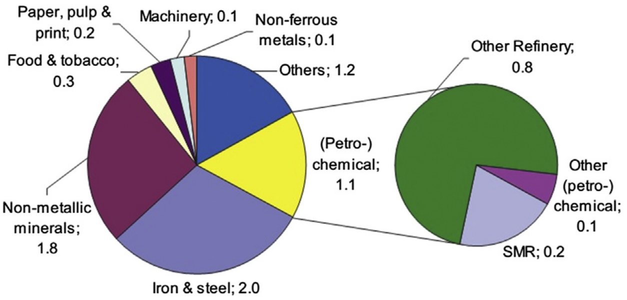

Fig. 1 – Direct global CO_2 emissions of the industrial sector in Gt/yr for 2005 [adapted from 3].

CO_2 emissions based on a 2008 IEA report [3]. There, it can be seen that the (petro) chemical sector in 2005 was responsible for about 16% of industrial CO_2 emissions, and that steam methane reforming (SMR) accounts for a large share. Also, from more recent IEA publications [4,5], the petrochemical sector, after the iron and cement sector, is still the major source of carbon dioxide emissions.

Steam reforming is the commonly used and mature technology for industrial hydrogen production. According to a life cycle assessment of global hydrogen production [6], about 75% of world's total hydrogen is produced by steam methane reforming. Also, a 2008 IEA report estimated the global annual hydrogen production in 2005 at 65 Mt, with 48% from SMR [3]. Other data on the share of hydrogen production from SMR confirm SMR to be the main process for hydrogen production. Although SMR might be replaced in the future by other efficient hydrogen production techniques, e.g. such as thermo-catalytic decomposition [7], it is still expected to be important in the future.

As pointed out earlier, SMR facilities emit on average 7 kg CO_2/kg H_2 [3], which was equivalent to 220 Mt CO_2 globally in 2005 [3]. Compared to total global CO_2 emissions, the share contributed by SMR facilities is small, at around 3% [3]. This share is expected to increase through decarbonising the transportation industry, partly by using fuel cell systems and hydrogen as an energy carrier. The IEA estimates that if fuel cell technology is applied in the transportation sector successfully by 2050, the global hydrogen demand will be as high as 275 Mt per year [8], resulting in emissions of about 2 Gt CO_2 per year if SMR facilities are used. CCS would then be more attractive for decreasing these emissions. One advantage of introducing hydrogen to the transportation sector is that emission control is easier at a central SMR plant than in all cars on the road. When purified, CO_2 has many uses in the chemical industry, in solid form (dry ice), liquid form (e.g. refrigeration equipment) and gaseous form (beverage carbonation). It also is widely used as a reactant in chemical processes and as an inert blanketing gas to prevent oxidation (e.g., for food products) [9]. Recently, it has received attention to help increase oil recovery from depleted or high viscosity oil fields [10].

Numerous studies have been carried out on CO_2 removal from SMR, usually focussing on in-process capture and end-point capture, i.e. capturing furnace flue gases or post combustion capture. The former involves the capture from process streams and is discussed in following sections. Simbeck [11] indicates that carbon capture from the flue gas stream of an SMR furnace is possible using amine-based capture and, in fact, most pilot plants established in the world now use amine-based systems [12,13]. Nonetheless, amine-based technology have been identified is not suitable for low molar fractions of CO_2 (flue gases) [14], and other shortcomings of amine-based technology for flue gases have been described [15], e.g. extensive solvent corrosion issues and energy intensiveness. Removal of CO_2 from flue gases by pressure swing adsorption (PSA) has recently been investigated as an alternative technology for CO_2 removal by Kikkinides et al. [9], Park et al. [16], and Ko et al. [17]. Voss [18] has studied the feasibility of CO_2 removal from different process locations by different technologies and compared them based on various criteria. He concludes that PSA technology is highly competitive to mature amine-based methods due to its comparatively simpler procedure. Also, he states that PSA is favourable for pressurized feed streams, namely synthesis gas and PSA off-gas streams. Economic studies have also been reported, e.g. assessing techno-economically CO_2 capture systems [1,19]. Overall, it is expected that CO_2 avoidance costs at SMR facilities decrease by having high pressure sources of CO_2 with high concentrations. Two main factors dominate capture costs: CO_2 partial pressure and molar concentrations of streams. In this study, mass flows of CO_2 are also considered, as these can motivate investors who seek to avoid potential high CO_2 emission penalties in the future.

Oxygen enrichment can enhance the performance of steam methane reforming and other industrial processes, especially those involving combustion. In case of SMR, oxygen enrichment has been shown to reduce the requirement for natural gas feed [20], mainly due to better heat transfer from the combustion gases to the reformer. This is a consequence of lower nitrogen concentrations in the combustion gases, which decreases the amount of sensible heat lost with flue gases. The other main benefit of oxygen enrichment is, because of the relatively lower nitrogen supply, higher carbon dioxide concentrations are attained in the combustion products. In this study, a fixed hydrogen production rate is considered when analysing the impact of parameter variations and the effect of oxygen enrichment only on the combustion process is examined.

In this research, we analyse an SMR process to determine how the S/C ratio affects the criteria that play major roles in CO_2 capture: partial pressure, concentration, and overall mass flow rate of CO_2. Also, focussing on post combustion capture from furnace flue gases, oxygen enrichment of furnace combustion is investigated to see if it makes CO_2 capture from flue gases reasonable compared to synthesis gases and PSA off gases. Finally, hydrogen production and carbon removal are studied together and compared for varying S/C ratios and reforming temperatures.

Methods considered

General descriptions are provided of the three main processes considered in this study: SMR, CO_2 capture, and oxygen enrichment.

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