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Optimal post-combustion conditions for the purification of CO_2-rich exhaust streams from non-condensable reactive species

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HIGHLIGHTS

- We consider the oxygen reduction in CO2 stream to be sequestrated.
- MILD post-combustion process is feasible to reduce O_2 in CO_2 streams.
- Characteristic Kinetic times of the process are compatible with practical applications.
- We identify the conditions to achieve the target concentration of undesired species.

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ABSTRACT

The presence of a significant amount of oxygen and/or other non-condensable species in flue gases of CO_2-capture-ready combustion systems is a relevant issue to be solved to avoid problems in CO_2 sequestration and storage processes. More specifically, oxygen, as well as other non-condensable species, increases the compression work required for the liquefaction of CO_2. Furthermore, residual oxygen in CO_2 streams used for EOR (Enhanced Oil Recovery) operations reacts with hydrocarbons in oil field both causing an overheating at the injection point and a higher oil viscosity increasing extraction cost.

Post-oxidation process is a feasible and economical possibility to reduce oxygen and non-condensable/oxidizable species (such as H_2 and CH_4) concentration to one digit ppm (or ppb) levels and obtain high purity CO_2 streams that can be used for sequestration or EOR.

This paper presents a numerical study on the post-oxidation processes of a CO_2 rich gas stream, with composition typical of a CO_2-capture-ready system, aimed to outline operative conditions useful to achieve a significant reduction of the gas contaminants below the minimum required level allowing for a useful use of the resulting CO_2 stream for storage or EOR purposes. Attention was focused on output streams of oxy-combustion plants.

High temperatures and elevated levels of dilution of inlet streams make such a post-combustion process work in conditions typical of MILD combustion. Characteristic kinetic times and key species concentrations at steady state were evaluated in order to study the evolution and the completion of the oxidation process.

Such parameters were correlated to main variables that influence the post-oxidation process such as inlet temperature system, composition of feed mixture, fuel and nature of diluent species.

1. Introduction

The presence of oxygen and/or other non-condensable species (i.e. nitrogen, argon, methane, hydrogen) in flue gases of CO_2-capture-ready combustion systems is a relevant issue to be faced with for several reasons. More specifically, such impurities have negative effects on purification, transport, injection and storage of CO_2 stream, because they greatly affect the thermodynamic properties of CO_2 (i.e. vapor–liquid properties, heat capacity, enthalpy and entropy) [1].

Firstly, non-condensable species increase the saturation pressure of liquid CO_2 and decrease the critical temperature. As a result, lower temperature and additional overpressure is required to avoid the risk of two phases behavior during transport and storage of CO_2 [2,3]. It has been highlighted that oxygen as well as other non-condensable species increases the compression work required for CO_2 transport almost linearly with respect to their concentration in the CO_2 stream. The increase in compression work is

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approximately 2.5%, 3.5% and 4.5% for a concentration of 1% by volume of oxygen, nitrogen and hydrogen respectively [4]. Non-condensable species also increase the pressure required for CO_2 injection and reduce the capacity of the storage sites by decreasing the density of CO_2 stream [2].

The residual oxygen in the CO_2 streams used for EOR (Enhanced Oil Recovery) operations may react with hydrocarbons in oil field causing both an overheating at the injection point and an increase of oil viscosity thus implying higher extraction cost and more difficulties in refining operation [4,5]. Another potential effect of oxygen is the increased biological growth even if the relevance of this effect on oil production has not been thoroughly exploited yet. Moreover, oxygen in presence of small amount of water (order of ppm) in the CO_2 stream used for EOR operations, significantly increases corrosion rates in the oil production and downstream processing equipment [4].

Also in amine separation processes for CO_2 capture and sequestration, the presence of oxygen has negative effects on both the efficiency and economics of the process. It degrades most of the amine based solvents not allowing for their regeneration, and also determines the formation of corrosive compounds and surfactants (foaming) poisoning the active adsorbing species [6]. These effects further increase the relatively high cost related to such a separation process.

Even if it has not yet been established unequivocally the maximum acceptable level of oxygen in the currents of CO_2, many studies, carried out as part of European projects focused on CCS technologies, have showed that the maximum volume fraction of all non-condensable species, such as O_2, N_2 and Ar, in CO_2 stream to be stored, in less restrictive conditions, should not exceed 4% in volume. [7]

Companies as Praxair, Kinder Morgan and Statoil involved in the management and development of plants for CO_2 sequestration impose a maximum concentration of oxygen below 10 ppm as pipeline specification [7].

There are generally no significant technical barriers to reduce oxygen and non-condensable species and provide high purity of the captured CO_2 (i.e. physical separation). The most common technique to reduce oxygen down to ppm range is to incorporate a distillation column of the liquid CO_2 in the typical purification process of CO_2 streams for sequestration or EOR [8]. However, such a technique is likely to induce remarkable additional costs and energy requirements resulting in an overall reduction of power plant efficiency. In particular, White et al. [8] showed that separation of residual oxygen increases the difficulties in physical separation of non-condensable species in the stream. Therefore, it would be highly desirable the individuation of alternative, more economical techniques to reduce concentration of oxygen and non-condensable species in the CO_2 captured stream to acceptable levels for transport and storage.

Among the other techniques post-oxidation is worth to be considered as a suitable alternative for reducing oxygen and non-condensable/oxidizable species (such as H_2 and CH_4) concentration to one digit ppm (or to ppb) levels, obtaining high purity CO_2 streams that can be used for sequestration or EOR, without additional physical separation processes for such gaseous species. On the other hand, oxy-fuel combustion based technologies are surely among the most promising capture-ready combustion systems allowing for obtaining higher CO_2 concentration in exhaust streams with respect to other CCS technologies (i.e. pre-combustion and post-combustion) [9]. However relatively high levels of impurities, in terms of residual oxygen and non-condensable species, are expected in the CO_2 stream to be captured [10–12]. For these systems the fraction of residual oxygen in CO_2-exhaust stream to be treated into post-oxidation stage is a function of fuel used in the primary combustion chamber. In particular, the average volume fraction of residual oxygen in the exhaust stream is variable from 2% to 5% by volume depending on whether the plant is fueled with natural gas or pulverized coal [13]. Such an exhaust gas stream at the exit of oxy-fuel combustion system is also characterized by a high temperature and dilution level (due to high concentrations of CO_2 and H_2O). It can constitute the feed of a post-combustion burner where small quantities of fuel can be added in order to convert the oxygen as well as the other non-condensable/oxidable species to CO_2 and/or H_2O. Additionally, the presence of a post burner also increments global plant efficiency by heat recovery, and in peculiar thermodynamic cycles plant electrical efficiency. For example, power efficiency could increase when post-burning is coupled with exhausted heat/mass recirculation and supercritical bottoming strategies [14].

The aim of the present work is to numerically study a post-oxidation process of typical flue gas of CO_2-capture-ready combustion systems to both assess the practical feasibility and identify the optimal working conditions with the target of reducing oxygen and non-condensable/oxidizable species concentrations below their maximum acceptable values. Temperatures and composition of the reacting flows make such a post combustion treatment a MILD combustion process. This denomination refers to a class of novel combustion processes that is identified on the basis of the features of the inlet streams [15]. As better described in the next section, exhaust gases temperature and composition from oxy-fuel combustion systems fall in ranges typical of this new combustion technology.

Characteristic kinetic times (auto-ignition, and oxidation) and key species concentrations at steady state were evaluated to study the evolution and the completion of the oxidation process. These parameters are required for designing and dimensioning post-oxidation stages to be used for oxygen reduction in exhaust streams. Initial conditions considered in the paper are those characteristic of the output streams of oxy-combustion plants. Characteristic times and mole fraction of key species at steady state were correlated to the main parameters that influence MILD combustion processes such as inlet temperature system, fed mixture composition, equivalence ratio (ϕ), fuel and/or diluent nature to provide information about optimal operating conditions of post-oxidation process.

This work has been carried out in the framework of an Italian national project, under a grant provided by Ministry of Economic Development, aimed to improve the performance of pulverized coal combustion systems and CO_2 sequestration [16].

2. Theoretical background

The elimination of O_2 and other non-condensable oxidizable gases present in the flue gases of oxy-combustion systems can be more easily carried out by considering a post combustion section operating in non-standard conditions. More specifically, it is possible to take advantage from the characteristic temperature and composition of such stream to improve the level of conversion of those species to CO_2 and H_2O in a very simple and controlled way.

The reference process to be used to understand actual potentials of this approach is widely explained in the huge literature built up in recent years concerning MILD combustion processes and technologies and well summarized in the review papers Cavaliere and de Joannon [15] and de Joannon et al. [17]. In fact, the high temperature and dilution level of flue gases exiting an oxy-combustion burner correspond to the requirements for fed conditions of MILD combustion systems. In such processes the high initial temperature is required to allow the oxidation evolving despite the high dilution level used. More specifically, the dilution level is so high that the composition of reacting mixtures falls outside the flammability limits so that neither a premixed nor