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Research article

Dry syngas purification process for coal gas produced in oxy-fuel type integrated gasification combined cycle power generation with carbon dioxide capturing feature

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A B S T R A C T

Electricity production from coal fuel with minimizing efficiency penalty for the carbon dioxide abatement will bring us sustainable and compatible energy utilization. One of the promising options is oxy-fuel type Integrated Gasification Combined Cycle (oxy-fuel IGCC) power generation that is estimated to achieve thermal efficiency of 44% at lower heating value (LHV) base and provide compressed carbon dioxide (CO_2) with concentration of 93 vol%. The proper operation of the plant is established by introducing dry syngas cleaning processes to control halide and sulfur compounds satisfying tolerate contaminants level of gas turbine. To realize the dry process, the bench scale test facility was planned to demonstrate the first-ever halide and sulfur removal with fixed bed reactor using actual syngas from O_2–CO_2 blown gasifier for the oxy-fuel IGCC power generation. Design parameter for the test facility was required for the candidate sorbents for halide removal and sulfur removal. Breakthrough test was performed on two kinds of halide sorbents at accelerated condition and on honeycomb desulfurization sorbent at varied space velocity condition. The results for the both sorbents for halide and sulfur exhibited sufficient removal within the satisfactory short depth of sorbent bed, as well as superior bed conversion of the impurity removal reaction. These performance evaluation of the candidate sorbents of halide and sulfur removal provided rational and affordable design parameters for the bench scale test facility to demonstrate the dry syngas cleaning process for oxy-fuel IGCC system as the scaled up step of process development.

1. Introduction

1.1. Oxy-fuel IGCC power generation and syngas cleaning processes

Environmentally compatible fossil fuel utilization is inevitable to avoid unrestraint carbon dioxide (CO_2) emission that will incur critical situation concerning to global warming. One of the promising options of the compatible utilization is the high efficiency oxy-fuel IGCC power generation (Romano and Lozza, 2010) that is the affordable technology to reduce the CO_2 emission without large penalty in thermal efficiency (Kobayashi et al., 2014). Oxy-fuel IGCC power generation plant is basically comprised of conventional technologies; oxygen (O_2)-blown coal gasifier, syngas purification process, semi-closed gas turbine, heat recovery steam generator, steam turbine, steam condenser, and compressors (Kobayashi and Akiho, 2016). Key issues to establish the power plant are in the range of feasible development; the main tasks will be the demonstration of the O_2–CO_2 blown gasifier, scaling up of the dry syngas cleaning process, and modification of combustor in the gas turbine for the specific syngas from the gasifier. This work is related to the initial stage of the scaling up of the dry syngas cleaning process. As the oxy-fuel IGCC is basically combined cycle power generation, hydrogen halides and sulfur compounds are major concern for the syngas processing. The pre-combustion syngas cleaning process that comprises dry removal processes for halide and sulfur compounds is exhibited in Fig. 1 which express the whole system configuration of the oxy-fuel IGCC power generation plant. Several issues were taken into consideration to configure the proper syngas cleaning process. The proper order of the syngas processes is determined by their operability in the syngas. Hydrogen chloride is expected to react with zinc element in dry gas desulfurization sorbent. Recent work on simultaneous removal of hydrogen sulfide

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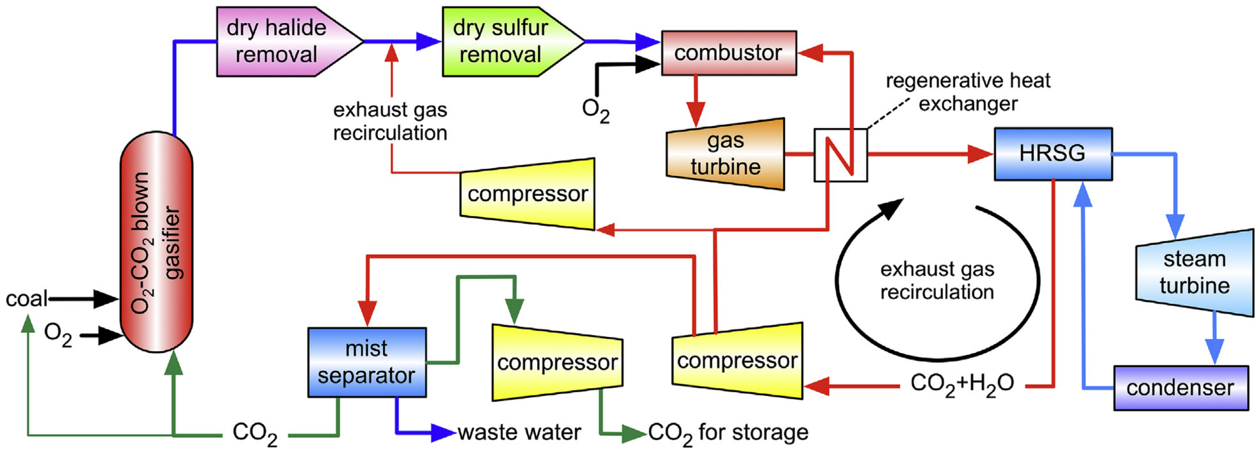

Fig. 1. Conceptual design of the oxy-fuel IGCC power generation plant arranged for exhaust gas circulation to the dry sulfur removal process.

(H_2S) and hydrogen chloride (HCl) by the S Zorb™ SRT sorbent was reported in literature (Schmidt and Sughrue, 2010). The breakthrough of sulfur compounds and HCl occurred at almost same time, which indicates the sulfur and halide removed by the zinc in the sorbent. Because the candidate sorbent for the desulfurization process in Fig. 1 uses zinc ferrite, the sorbent may also react with HCl. Therefore, it is preferred to place halide removal process in upper stream of the desulfurization. Therefore, it is preferred to place halide removal process in upper stream of the desulfurization. Carbon deposition from syngas is fearful and harmful side reaction, which causes fatal destruction of the desulfurization sorbent. The former work of authors (Kobayashi et al., 2014) revealed that the zinc ferrite desulfurization sorbent is applicable to the oxy-fuel IGCC condition when the carbon deposition was retarded by the exhaust circulation to the dry sulfur removal process in the plant. Though carbon deposition slightly occurs during the sorbent reduction step under start-up conditions, it can be released as carbon dioxide during subsequent regeneration steps in the frame of ordinary operation (Kobayashi and Nakao, 2015). It is quite natural to place the exhaust circulation that is essential to inhibit carbon deposition at immediate inlet of the sulfur removal process. The dry halide removal process and sulfur removal process is able to attain the tolerance level of acid gas contaminants of gas turbine as a syngas processing facility. From these multilateral considerations, it is expected that the dry syngas purification process that is comprised of the halide-removal process with sodium aluminate and desulfurization process with zinc ferrite sorbent is applicable to the oxy-fuel IGCC power generation. Then, we can proceed to the process development using the larger scale test facilities that will process actual syngas from O_2—CO_2 blown gasifier to verify process operation of the fixed bed reactor system. When the test facility is established, it is the first-ever demonstration of the dry syngas processing for the oxy-fuel IGCC power generation.

1.2. Scaling up of the dry syngas cleaning process

The reactor type of the halide removal process is expected to be packed bed of pelletized sorbent. Because the spent sorbent cannot be regenerated online, the reactor capacity is important for determining the exchange interval of the sorbent bed. The sulfur removal process comprises of three fixed bed reactors of honeycomb shaped desulfurization sorbent. Those reactors are operated cyclically and continuously according to the desulfurization cycle. Fig. 2 expresses the three reactors system for the process with the desulfurization cycle comprising the steps of, desulfurization, regeneration, and reduction. Each reactor installs honeycomb shaped sorbents in a fixed bed in order to assure the exhaust sulfur concentration as low as the pre-breakthrough concentration that can be achieved with the sorbent. The spent sorbent from the desulfurization procedure is then regenerated by feeding oxygen containing gas to the reactor. Regeneration of the spent sorbent will release sulfur dioxide into the oxygen containing gas. After the regeneration step, the oxidized sorbent bed is preliminary reduced with small amount of coal gas. The dry sulfur-removal process can process syngas continuously by switching the three reactors system according to the operational sequence of the reduction, desulfurization, and regeneration procedures.

Dry syngas cleaning processes are investigated for longer time since the start of development on IGCC power generation plant. The primary advantage of the process for IGCC application is increase in thermal efficiency, which is investigated in various related work. There is a typical thermodynamic assessment of IGCC power generation with cold or hot fuel gas desulfurization derived thermal efficiency of IGCC with hot desulfurization exceeds in 2.3% absolute value (Giuffrida et al., 2010). They also revealed that the efficiency enhancement by the operating temperature increase from 400 to 650 °C is negligible at merely 0.15% absolute value. Higher syngas temperature reduces syngas flow rate, which brings lower coal input and lower power consumption at air separation unit, while gas turbine output remains constant. The steam turbine output however decreases due to lower syngas flow rate, of which compensating effect keeps the thermal efficiency nearly constant. Although the analysis was made for desulfurization of circulating fluidized bed process, the thermal efficiency response does not concern the reactor type. Thus, the result encouraged us to operate our dry syngas cleaning process at 450 °C. As reported in recent review of dry syngas cleaning technologies (Dou et al., 2012), reactor types for dry sulfur removal process are categorized in fluidized bed, moving bed, and fixed bed. Fluidized bed is widely investigated for individual process of dry sulfur removal as well as sorbent injection to gasifier. The sorbent used in the fluidized bed reactor should satisfy the sulfur removal efficiency and the mechanical strength for circulation. The properties of the sorbent tend to fall down in many desulfurization and regeneration cycles (Dou et al., 2012). Moving bed concept is preferably suggested because of the efficient sorbent utilization and sulfur removal performance expected for the counter flow configuration (Yu et al., 2013). Though the moving bed conceptually susceptible of applicable design, it is known that the scaling up is difficult for processing large flow rate of syngas of the power generation plant. Because the