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Inhibition and elimination of carbon deposition in dry gas desulfurization process under oxy-fuel IGCC derived coal gas environment

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HIGHLIGHTS

- Dry gas desulfurization is essential in oxy-fuel IGCC power generation plant.
- The dry gas sulfur removal process is operated by repeating three steps cyclically.
- Carbon deposition is prevented if the exhaust is recirculated to the process.
- Carbon deposition effect is limited while exhaust is not available in the plant.
- Regeneration eliminates the amount of deposited carbon to sufficiently low level.

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ABSTRACT

Oxy-fuel IGCC is a concept of new coal fueled power generation with CO_2 capture capability without large penalty on thermal efficiency. It is estimated that thermal efficiency will attain 44% at lower heating value, LHV along with 99% CO_2 capture capability. An imperative issue to be solved to realize this concept is to reconcile pre-combustion sulfur removal and retaining the inorganic gas composition of the coal derived gas. Although introduction of dry gas sulfur removal is desired to protect gas turbine from corrosion and to preserve the gas composition, the sulfur removal sorbent containing zinc ferrite installed in the process may suffer from the severe side reaction of carbon deposition. Although it was found that mixing the coal gas with the circulating exhaust in the plant would effectively prevent the carbon deposition in the process, exhaust will not be available until gas turbine comes into normal operation after the plant start-up.

The amount of deposited carbon was evaluated and its behavior was examined during the reduction and succeeding regeneration steps performed under expected starting up condition of the plant, which is severe condition for carbon deposition. While carbon deposition occurs during reduction step under start-up conditions, the deposit can be released as carbon dioxide during succeeding regeneration step of scheduled duration. The deposited carbon totalling 0.8 wt% as maximum can be eliminated by regeneration to sufficiently low level below 0.03 wt% for the range of expected operating conditions. The carbon deposition may not affect the desulfurization performance of the zinc ferrite sorbent for the expected condition range at plant start-up. It was confirmed that the carbon deposition and elimination from the sorbent would be adequate measure for operating the dry gas purification process in the oxy-fuel IGCC power generation plant.

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1. Introduction

1.1. Desulfurization sorbent for syngas and carbon deposition issue

Dry gas desulfurization for the syngas treatment is essential technology for various energy conversion processes. Integrated gasification combined cycle (IGCC) power generation is one of the most suitable technology for applying the dry gas desulfurization technology which will bring significant advantage in thermal efficiency of electric power production [1]. The dry desulfurization technology is initially developed for coal-derived syngas cleaning and is expected to utilize in cleaning of biomass-derived syngas. Various sorbents containing zinc, copper, iron, calcium, manganese, and ceria are reviewed for estimating their applicability to the syngas derived from coal or biomass [2]. Besides sulfur,

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other impurities in the syngas should be appropriately removed for downstream fuel application. Dry halide removal, desulfurization, and ammonia decomposition, which are required for IGCC power generation, are summarized from the point of view of development of sorbents and catalyst [3]. Sulfidation reactions of metal oxides are the potential candidates for dry sulfur removal process. According to the specific requirement of operating condition and sulfur tolerance limit, appropriate metal oxide is generally selected by estimating sulfur removal capabilities by chemical equilibrium of sulfidation reaction for those metal oxides [4]. It is also recognized that the online regeneration of desulfurization sorbent is essential [4] to establish the feasible dry sulfur removal process for the application that treat vast amount of syngas, such as IGCC. Desulfurization sorbent containing zinc ferrite is promising candidate for the process because the sorbent has reconciliation of sulfur removal ability and complete regeneration for multiple cycle operation. Zinc ferrite supported on titanium oxide has superior sulfur removal ability in simulated coal gas condition to attain sub ppm concentration for both H_2S and COS at 450 °C for multiple cycle operation [5]. It is well known that the sulfur capacity of regenerable metal oxide sorbent decreased over the multiple cycle regeneration. The sulfur capacity attributed to the zinc portion of the zinc ferrite sorbent was determined by in situ XRD measurements [6]. The stability of the sulfur removal ability of the sorbent was estimated from the sulfur capacity measurements throughout 500 cycles. The result concluded that the sorbent performance is durable to multiple operations over than 500 cycles in a fixed bed reactor when the sorbent maintained its zinc sulfur capacity at around 40% of its initial value [7], which is positively expected from the multiple cycle test result. The weak point of zinc ferrite sorbent is that the iron element in the ferrite may suffer carbon deposition in syngas at the expected operating condition of the dry desulfurization process.

1.2. Carbon deposition in syngas processing

Carbon deposition is a common issue in syngas processing. It is obvious that carbon deposition is investigated for long period, as carbon deposition takes important role in metallurgy of iron making. Then, the carbon deposition is widely investigated concerning to catalytic conversion or utilization of syngas produced from carbonaceous fuel. Catalyst deactivation due to Boudouard reaction is always reported on the methanol synthesis from syngas [8,9]. Iron base catalyst is used for Fischer–Trøpsch synthesis (FT synthesis) from syngas, where the iron carbide phase plays important role on the kinetics of FT reaction and carburization has detrimental effect on the catalyst at the same time [10]. The series of application of syngas from carbonaceous fuel require countermeasure for carbon deposition without exception. Thus, the control of carbon deposition in syngas is an unavoidable issue to explore the conversion of carbonaceous resource at present. Reaction mechanisms and conditions for enhancing or retarding carbon deposition is empirically understood. Carbon deposition over hematite, Fe_2O_3, in pure carbon monoxide occurs through the reaction path that includes reduction of hematite, iron carbide formation, and Boudouard reaction [11]. Carbon deposition on Ni/Al_2O_3 catalyst during CO_2 reforming of methane was studied to find out the presence of hydrogen and carbon monoxide enhance carbon formation from methane [12]. This study revealed that the carbon deposition from CO_2 or $CO_2 + CH_4$ is enhanced by addition of H_2 or $H_2 + CO$, while addition of H_2O can reduce carbon deposition from CO and CH_4. This finding is also explained by the carbon deposition mainly resulting from Boudouard reaction. Thus, it is qualitatively expected that the enrichment of H_2O and CO_2 relative to CO and H_2 would retard carbon deposition in syngas.

1.3. Syngas utilization in power generation and CO_2 abatement

In order to reconcile utilization of coal fuel, and abatement of carbon dioxide emission in the power industrial area, CO_2 capture from fuel gas in the coal gasification power plant, namely pre-combustion capture is intensively researched as alternative to the post-combustion capture in the pulverized coal power plant. The power plant with conventional CO_2 capture, however usually incurs large efficiency loss of 10% points or more in net thermal efficiency [13–15]. The lower plant efficiency consequently causes consumption of increased amount of fuel, which will result in diminishing fuel resources. The efficiency loss is mainly caused by the large heat and power consumption of the CO_2 capture process for fuel gas. There is another concept to lowering the energy consumption for CO_2 separation in IGCC, which is so called "oxy-fuel IGCC" concept [16–18] The key concept of oxy-fuel IGCC power generation is CO_2 separation from CO_2-H_2O mixture exhaust in replacement of CO_2 capture process. The steam condensation procedure from the inert-gas free exhaust will drastically decrease the energy consumption when compared with physical or chemical CO_2 capture process. In order to apply the concept, the power generation system should consists of oxygen-CO_2 blown coal gasification, semi-closed cycle operation of gas turbine with oxy-fuel combustor, and efficient circulation of exhaust. While the exhaust circulation brought advantage in plant efficiency to the oxy-fuel IGCC, the contaminant issue arises from the semi-closed cycle operation. As coal gasification fuel gas contains various contaminants that are basically derived from coal; sulfur compounds, halides, nitrogen compounds, alkaline and alkaline earth metals, and volatile heavy metal are suspected to be contained in coal gas or exhaust. Those contaminants should be properly treated in the plant to protect equipment in the plant and achieve environmental tolerance. Earlier work performed by the authors investigated carbon deposition issue in the dry sulfur removal process that is deployed at pre-combustion stream of the oxy-fuel IGCC power generation [19–21]. The exhaust circulation upstream of the sulfur removal process is effective procedure for inhibiting carbon deposition at all expected conditions in the actual plant operation. Power consumption for the exhaust circulation is small enough to retain those advantages in thermal efficiency and efficient CO_2 separation. The exhaust gas is however not available during the start-up of the plant requiring evaluation of carbon deposition under the start-up conditions to establish countermeasure for abatement of the deposition.

This paper addresses investigation of carbon deposition during the reduction and succeeding regeneration of dry sulfur removal sorbent. The effect of carbon deposition on the performance of sulfur removal process under syngas conditions was also determined.

2. Dry sulfur removal in oxy-fuel IGCC plant

2.1. Oxy-fuel IGCC plant concept

System configuration of the oxy-fuel IGCC plant that includes deployment of dry gas sulfur removal is expressed in Fig. 1. The O_2-CO_2 blown gasifier produces syngas that is processed in the dry sulfur removal process to eliminate sulfur compounds, and then the syngas is provided to gas turbine. Exhaust from gas turbine is mostly circulated to combustor as temperature control media of the combustor, after the heat of exhaust is recovered by regenerative heat exchanger and heat recovery steam generator. The rest of the exhaust is send to the efficient CO_2 separation block where the exhaust is compressed and steam is eliminated by condensation. Obtained CO_2 is then subjected for further compression for storage, besides small amount of CO_2 is circulated to gasifier as gasification media. Net thermal efficiency of the plant on Fig. 1 is evaluated by static system analysis and calculated as efficiency in lower heating

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