Applied Thermal Engineering 130 (2018) 822-829

Contents lists available at ScienceDirect

Applied Thermal Engineering

journal homepage: www.elsevier.com/locate/apthermeng

Research Paper

A system including enriching coal bed methane by solar energy and selective catalytic reduction



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HIGHLIGHTS

• A system that combines enriching CBM with solar energy and SCR of NO_x is proposed.

- The enriched CH₄ is utilized as the reductant in SCR with catalyst La_{0.8}Sr_{0.2}MnO₃.
- The concentration of CH4 can be enriched to 80% through 200 enriching pipe units.
- In the CH₄-SCR system, the maximum conversion ratio of NO can reach to 80%.

ARTICLE INFO

Article history: Received 3 October 2016 Revised 10 November 2017 Accepted 12 November 2017 Available online 13 November 2017

Keywords: CBM Enrichment Solar energy SCR Perovskite structure catalyst

ABSTRACT

Enriching CBM (coal bed methane) and then making good use of the enriched CH₄ have great significance in protecting the environment. This paper proposes a new system that uses solar energy to enrich CBM and then utilizes the enriched CH₄ as the reductant in SCR (selective catalytic reduction) of NO_x with a new catalyst (La_{0.8}Sr_{0.2}MnO₃). Theory research, numerical simulation, experiments were done to investigate the key factors of the enrichment such as the initial CH₄ concentration, the temperature field. Results indicate that when the temperature difference $\Delta T = 450 - 300 = 150K$, 200 enriching pipe units as in this experiment model can make the final mole fraction of CH₄ reach to 80%.

Besides, a new SCR of NO_x by CH_4 with catalyst $La_{0.8}Sr_{0.2}MnO_3$ is presented in this paper and the enriched CH_4 is the reducing agent. Experiments were done to investigate the efficiency of the new catalyst. In the new CH_4 -SCR system, the maximum conversion ratio of NO can reach to 80% and the mass concentration of NO at the outlet can be lower than 20 mg/m³ in a proper condition. This new system can make good use of the solar energy to enrich CBM and present an efficient way to use the enriched CBM, which is friendly to the environment.

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1. Introduction

As we know, there are abundant methane resources on the coal bed (CBM) and the solar resources is convenient in China. Enriched CBM is a potential green energy supply for solving the worldwide energy crisis and it has been widely used in the energy and chemical industry. New technology and research are urgently needed to make good use of CBM. All those advantages can help us to enrich CBM with the temperature gradient caused by solar energy. Meanwhile, the typical reducing agent of SCR (selective catalytic reduction) of NO_x in power plant is NH₃, which has many shortcomings.

* Corresponding author. E-mail address: qlzhou@mail.xjtu.edu.cn (Q. Zhou). Therefore, it is urgent to promote a new reducing agent with proper catalyst to improve the efficiency of SCR.

Many researches on the physical process that temperature gradient causes mass transfer were done. In 1918, Chapman and Dootson proved that the mass diffusion of thermal effect also works in gas mixture [1]. In 1928, Eastman proposed the theory of Soret effect which describes the mass diffusion caused by thermal effect [2]. In 1980, Rosner investigated the influence of Soret effect on mass transfer rate on the two-phase interface [3]. Then Mortimer studied elementary transition state theory of the Soret and Dufour effects [4]. And Dulal Pal researched the influence of chemical reaction and thermal radiation on mixed convection heat and mass transfer with Soret and Dufour effects [5].

The present adopted NH₃-SCR has its weaknesses such as its expensive catalyst (V_2O_5/TiO_2) and releasing unreacted ammonia







which may bring second pollution, flog and haze [6]. In recent years, the experimental studies and previous theoretical efforts have paid much attention to new reducing agents of SCR. Tao Qiu proposed a method to estimate the temperature field in SCR catalyst [7]. SCR using hydrocarbons as reducing agents (SCR-HCs) is a technology that is still found in its development stage. Byoung adopted a numerical simulation which is accurate and efficient for the prediction of NO_x conversion in diesel exhaust situation [8]. An interest is kept in the utilization of CH₄ as reducing agent in order to replace NH₃.

Many researchers investigated simple metal oxide mixture (supported on zeolitic materials) as catalyst of SCR. However, that kind of catalyst has its problems when applied in engineering because of its sensitivity to SO₂ and water. ABO₃ (metal oxide with perovskite structure) has good oxidation activity and thermos ability and it can be stable in high temperature over 1273 K [9]. The metal ions are fixed in the structure while oxygen ions can move in lattice [10]. ABO₃ can catalyze the combustion of CH₄, in which lattice oxygen reacts with CH₄ and CO absorbed on the catalyst while O₂ in air replenishes the vacant lattice. ABO₃ can also catalyze the decomposition of NO, in which lattice oxygen plays a role in oxygen desorption and N₂ production after the combination of NO molecules [11]. Therefore, it may become the new catalyst for CH₄-SCR. There are many researches about catalyst with perovskite structure. Teraoka investigated the process that La_{0.8}Sr_{0.2}-CoO₃ and La_{0.6}Sr_{0.4}Mn_{0.8}Ni_{0.2}O₃ catalyze NO decomposition [12]. Tofan studied the way $La_{1-x}Sr_xM_{1-y}O_{3-\delta}$ (M = Co, Ni, Cu) catalyzes NO decomposition [13]. Giannakas studied NO-CO reactions with catalyst LaFeO3 series where La0.8Ce0.2FeO3 has the best work in catalyzing NO-CO reactions [14,15]. La_{0.8}Sr_{0.2}MnO₃ which may bring zero greenhouse emissions has outstanding performance in the SCR of NO_x by CH_4 [16].

In this study, we propose a system that enriches CBM during its transit using solar energy and then utilizes the CH₄ in the selective catalytic reduction of NO_x with a new catalyst ($La_{0.8}Sr_{0.2}MnO_3$). Theory research, numerical simulation and experiments have been done to investigate the key factor of enrichment such as the initial CH₄ concentration, the temperature field. Experiments have been

done to investigate the efficiency of CH_4 -SCR with $La_{0.8}Sr_{0.2}MnO_3$ in this work.

2. System description

This new system includes two parts: the enriching part and the SCR part. The enriching part is driven by solar energy and the SCR part with new catalyst uses the enriched CBM as the reducing agent. Fig. 1 illustrates the schematic of the entire system including CBM line (dark blue line), flue gas line (black line) and control line with feedback and commands (dash line).

In the enriching part, a pipe unit consists of a gas pump (CBM Jumbo), a mass flow meter (MFM), snake-spread pipes covered by solar absorbing coating and a CH₄ holder. The air-cooling facilities such as air pumps, air flowmeters and explosion-proof equipment are left out in the drawing. Three enriching pipe units are showed here to represent the multilevel enriching pipe units. Multilevel enriching pipe units can help to enrich CH₄ to a certain concentration. According to the feedback from the CH₄ detectors before the CH₄ entrance of SCR, control center can make commands to MFMs to obtain CH₄ with appropriate and steady concentration at the exit of the enriching part. Before the desulfurizer, flue gas flows through two-stage catalyst layers, which are the main parts of the SCR. Enriched CH₄ is injected into the catalyst layers to reduce NO_x. The second catalyst layer can help to burn the uncompleted burned carbon (UBC) and reduce residual NO_x by regulating the flow rates of CH₄ and air.

3. Research on enriching CBM

3.1. Theories analysis

Soret effect describes a physical process that the temperature gradient can cause mass transfer. Steady temperature field can make the gas molecules move to the higher or lower temperature zone according to their different molecule weight. The key factors of Soret effect are the temperature gradient field and physical



Fig. 1. Schematic diagram of the new system.

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