Fuel 181 (2016) 1184-1190

Contents lists available at ScienceDirect

Fuel

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

Experiment study on mercury migration across wet flue gas desulfurization slurry under oxy-coal combustion atmosphere

Hui Wu, Bi Zhang, Yong Qiu, Guangqian Luo, Hong Yao*

State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology, Wuhan 430074, China

HIGHLIGHTS

• More mercury enriched in slurry solids of CaSO₃ than CaSO₄.

• CO₂ atmosphere hindered Hg retention in solids and increased gaseous Hg⁰ emission.

• CaSO₃ oxidation by O₂ led to less Hg retained in solid and more Hg⁰ re-emitted.

• More Hg remained in the slurry liquid when Cl⁻ anions presented.

• 6 mM NO_3^- and 5 mM SO_3^{2-} cause Hg transferred from solids and liquid into gas phase.

ARTICLE INFO

Article history: Received 20 October 2015 Received in revised form 4 February 2016 Accepted 31 March 2016 Available online 16 April 2016

Keywords: Mercury Oxy-coal combustion Flue gas desulfurization Gypsum

ABSTRACT

Mercury in oxy-coal combustion flue gas may cause liquid metal embrittlement and material failure to aluminum (Al) heat exchangers, which needs to be removed with high efficiency before CO_2 compression. Oxidized Hg (Hg²⁺) is soluble thus can be co-removed with SO₂ in WFGD (Wet Flue Gas Desulfurization). However, the dissolved Hg²⁺ in slurry may also transform to the insoluble elemental mercury (Hg⁰) form and re-emitted, resulting a reduction of total Hg capture efficiency in WFGD. This paper experimentally examined the Hg migration across WFGD under high CO_2 concentration atmosphere with simulated typical desulfurization slurry. Influences of solids (CaSO₄, CaSO₃) and anions (Cl⁻, NO₃⁻, SO₃²⁻) in slurry are considered. According to the results, solids of CaSO₃ with higher BET surface area are more favored for Hg retention than CaSO₄. Slurry pH under high concentration of CO_2 was a little bit lower than air combustion atmosphere. The decreased pH in turn hindered Hg retention on both solids while the gaseous Hg⁰ emission increased. For anions, 6 mM NO₃ and 5 mM SO₃²⁻ both led to large amount of Hg transformed from CaSO₄ solids and liquid into the gas phase. The existence of 20 mM Cl⁻ can retain more Hg in the liquid, especially when NO₃ and SO₃²⁻ coexisted. However, this effect was quite pH sensitive. As to CaSO₃ slurry, most of the Hg remained in slurry regardless the existence of other anions and atmosphere.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Energy consumption from coal combustion has produced large amount of pollutants, such as NOx, SO₂, as well as trace element mercury (Hg). Hg is a well-known neurotoxin while coal-fired power plants are the main anthropogenic mercury emission sources [1]. Due to the huge volume of combusted flue gas and the extremely low concentration of mercury in flue gas (in the order of parts per billion, ppb), at present there is none costeffective mercury removal technology available for coal fired power plants. Recent years, oxy-coal combustion has showed the

* Corresponding author. E-mail address: hyao@mail.hust.edu.cn (H. Yao). potential of multi-pollutant control technology for CO₂, NOx, SO₂, as well as mercury [2]. However, during flue gas compression for CO₂ liquification, mercury in flue gas may cause liquid metal embrittlement and material failure to aluminum (Al) heat exchangers, NOx and SO₂ can cause equipment corrosion [3]. For the operation safety concern, those impurities must be removed before CO₂ purification and compression. As a reference, there is a Hg concentration limitation of 0.01 μ g/Nm³ for natural gas compression [4]. This implicated that oxy-coal combustion is challenged by very high efficiency Hg removal.

Compared to Hg, the controlling technology for NOx and SO_2 is quite mature and have been commercialized. These controlling technologies also showed co-benefits on Hg removal [5]. With high volatility and very low water solubility, elemental mercury



