Environmental Science & Technology

Article

Cite This: Environ. Sci. Technol. 2019, 53, 12113−12120

pubs.acs.org/est

In-Furnace Control of Arsenic Vapor Emissions Using Kaolinite during Low-Rank Coal Combustion: Influence of Gaseous Sodium Compounds

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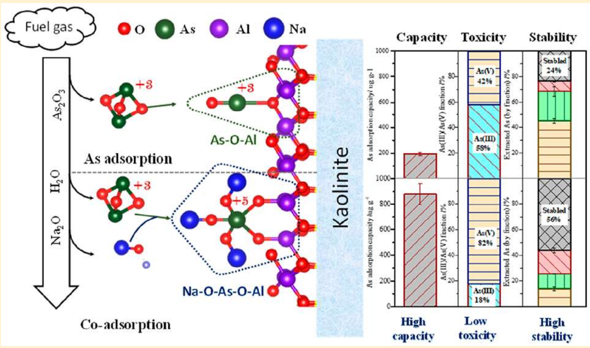
Supporting Information

ABSTRACT: Using additives in the in-furnace control of arsenic emissions is promising for reducing the impact on the downstream selective catalytic reduction system and blocking the spread of arsenic pollutants into the environment. The study quantifies the arsenic adsorption capacity of kaolinite at high temperature and clarifies its fixation pathway with and without the existence of sodium vapor, which is easily adsorbed by kaolinite. Experiments about Al-coordination and acid sites of products, as well as calculations of thermodynamic equilibrium and the adsorption energy based on density functional theory were performed. During separated arsenic adsorption, nearly 40% of trivalent arsenic [As(III)] is oxidized to pentavalent arsenic [As(V)] and bonded to kaolinite, forming an As−O−Al structure. In this respect, the arsenic adsorption capacity of kaolinite is 200 $\mu g\ g^{-1}$, with 24% of arsenic shown to be well-crystallized Al-bound. During the co-adsorption process, 82% of As(III) is oxidized to As(V) and connected to the Al surface of kaolinite, and the O−Na groups bond to As around the As−O−Al structure, thereby forming Na−O−As−O−Al. The arsenic adsorption capacity increased to 878 $\mu g\ g^{-1}$ with well-crystallized Al-bound arsenic accounting for 56%. This study demonstrates the potential for the application of kaolinite as an arsenic adsorbent in the actual furnace.



1. INTRODUCTION

Arsenic is a semi-volatile trace element that has received an increase in public attention due to its teratogenicity, bioaccumulative tendency, and potential carcinogenicity.[1−3] Coal-fired power plants in China emit approximately 550 tons of arsenic per year and are thus one of its main anthropogenic sources.[4,5] Statistics have shown that the average concentration of arsenic in Chinese coal is nearly 3.18 mg kg^{-1}, and a higher arsenic level (mostly ranging from 4 to 25 mg kg^{-1}) has been reported in low-rank coal mined from western regions.[6] When burning this coal, most of the arsenic is released into the flue gas as As_2O_3(g),[7] part of which goes through the furnace and enters the selective catalytic reduction (SCR) system, where it is easy for As_2O_3(g) to diffuse and be adsorbed on the catalyst surface, resulting in the poisoning of denitrification catalysts.[8] However, As_2O_3(g) that is not captured by the air pollution control devices eventually escapes into the atmosphere.[9] It is thus crucial to control arsenic emissions from furnaces, both to reduce the impact on the downstream SCR system and to block the spread of arsenic pollutants into the environment.

Numerous studies have shown that aluminosilicates, CaO and Fe_2O_3, are effective additives for use in capturing arsenic-containing gas under heating conditions.[10−13] Among them, aluminosilicates (such as artificial zeolite and natural kaolinite) are potential commercial adsorbents, owing to their low-cost, abundant porous structure, and nontoxicity. Du et al. investigated the adsorption performance of As_4O_6(g) on zeolites experimentally and theoretically, and the results of density functional theory (DFT) calculations showed that a low Si/Al ratio is favorable for capturing As_4O_6.[11] As the positive effects of Al substitution suggested an intrinsic connection between arsenic adsorption and Al atoms in zeolites, our group recently tested the capacity of kaolinite and meta-kaolinite in the capture of arsenic vapors at 573 and 873 K in As_2O_3/air atmosphere, and was the first to report the potential arsenic adsorption capacity of kaolinite and its derived meta-kaolinite.[10] However, the temperature of the coal-fired furnace is usually much higher than 873 K, and it has not yet been clarified whether kaolinite or its derivatives are

Received: May 29, 2019
Revised: August 7, 2019
Accepted: September 5, 2019
Published: September 5, 2019

DOI: 10.1021/acs.est.9b03195
Environ. Sci. Technol. 2019, 53, 12113−12120