

Reaction of SO₂ with sodium chlorate powder triggering oxidation of NO and Hg⁰Youngchul Byun^a, Jeonghyun Kim^b, Dongnam Shin^{c,*}^a School of Chemical Engineering and Analytical Science, University of Manchester, Manchester, M60 1QD, UK^b Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, PR China^c Environmental Research Department, Research Institute of Industrial Science and Technology, Hyoja Dong, Pohang, Kyungbuk, 790-600, Republic of Korea

ARTICLE INFO

Article history:

Received 30 August 2011

Received in revised form 2 February 2012

Accepted 3 February 2012

Keywords:

Sodium chlorate
Sulfur dioxide
Nitrogen monoxide
Elemental mercury
Removal
Oxidation

ABSTRACT

This study investigated the reaction of powdered sodium chlorate (NaClO₃(s)) with air pollutants. 200 ppm SO₂, 200 ppm NO, and 260 μg/N m³ Hg⁰ in 2.6 L/min of N₂ balance were introduced into a reactor containing 2.4 g of NaClO₃(s). Results showed that SO₂ has reacted exclusively with NaClO₃(s) to produce Na₂SO₄(s) above 220 °C, whereas NO and Hg⁰ themselves have no reactivity toward NaClO₃(s). However, oxidation of NO and Hg⁰ occurred when they were introduced with SO₂ into the reactor containing NaClO₃(s). NO₂ as a radical scavenger was introduced at the forward and backward positions of reactor to determine which species are responsible for the oxidation of NO and Hg⁰ during the reaction of SO₂ with NaClO₃(s), where ClONO₂, ClNO₂, and OClO species were observed. Kinetic simulations were also conducted to elucidate reactions between chlorine species, where final products were nearly Cl₂ on both OClO + Cl and OClO + ClO reactions when reaction time was 2.4 s. These suggest that OClO, ClO, and Cl generated during the reaction of SO₂ with NaClO₃(s) can oxidize NO to NO₂ and Hg⁰ to Hg²⁺.

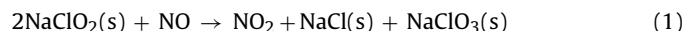
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1. Introduction

Sodium oxychlorides (NaClO_x (x = 1, 2 and 3) are used widely in many industrials due to their high oxidizing power. Among them, sodium hypochlorite (NaClO) and sodium chlorite (NaClO₂) has been studied extensively to control air pollutants, bleach textiles, fiber, pulp, and paper, purify water, whereas NaClO₃ is not used due to its toxicity to humans; chlorate ion (ClO₃⁻(aq)) is listed by the World Health Organization as a chemical of health significance in drinking water [1–6]. However, NaClO₃ is also a powerful oxidant having toxic effects on a wide range of organisms. Therefore, NaClO₃ has been intermittently applied to agriculture, e.g. as a weed controller, defoliant and soil sterilant as well as for producing chlorine dioxide (ClO₂), which has been used for a bleaching and water purification [2,7].

NaClO and NaClO₂ have been used to control air pollutants (NO_x, SO₂, and Hg⁰) in forms of liquid and solid. Aqueous NaClO and NaClO₂ solutions have been used as chemical additives to enhance absorption of SO₂ and oxidation of NO in wet scrubbers. We also investigated applications of NaClO₂(s) to oxidation of NO and Hg⁰. Results showed that NaClO₂(s) oxidizes NO to NO₂ with conversion to NaClO₃(s) [8,9]. Regardless the number of studies on the

reactions of NaClO and NaClO₂ with air pollutants, there are no reports of the application of NaClO₃ to control of gaseous air pollutants. There are two methods for applying NaClO₃ to the control of gaseous air pollutants. One is to use an aqueous NaClO₃ solution for scrubbing air pollutants. However, this method is unsuitable because, as mentioned above, the ClO₃⁻ ion dissolved in water is toxic to humans. The other is to directly inject NaClO₃(s) into a duct containing gaseous air pollutants. This method has following three benefits. The first is that if NaClO₃(s) can control air pollutants, it generated by the reaction of NaClO₂(s) and NO, can be reused [8,9] because NaClO₂(s) also have an applicability for the control of NO as following reaction,



The second is that the injection of NaClO₃(s) can avoid the generation of ClO₃⁻ ions. The third is that the injection of NaClO₃(s) can be applied simply in existing duct of plants.

To the best of our knowledge, there has, prior to this work, been no experimental study on the reaction of non-dissolved NaClO₃(s) with air pollutants. Representatives of air pollutants were NO, SO₂, and Hg⁰ emitted during combustion processes; we conducted the application of NaClO₃(s) for the control of gaseous NO, SO₂, and Hg⁰ with kinetic simulations to elucidate which species are responsible for the oxidation of NO and Hg⁰. We demonstrated that NaClO₃(s) can remove SO₂ at temperatures above 220 °C by formation of solid Na₂SO₄ (Na₂SO₄(s)) with production of chlorine-containing species

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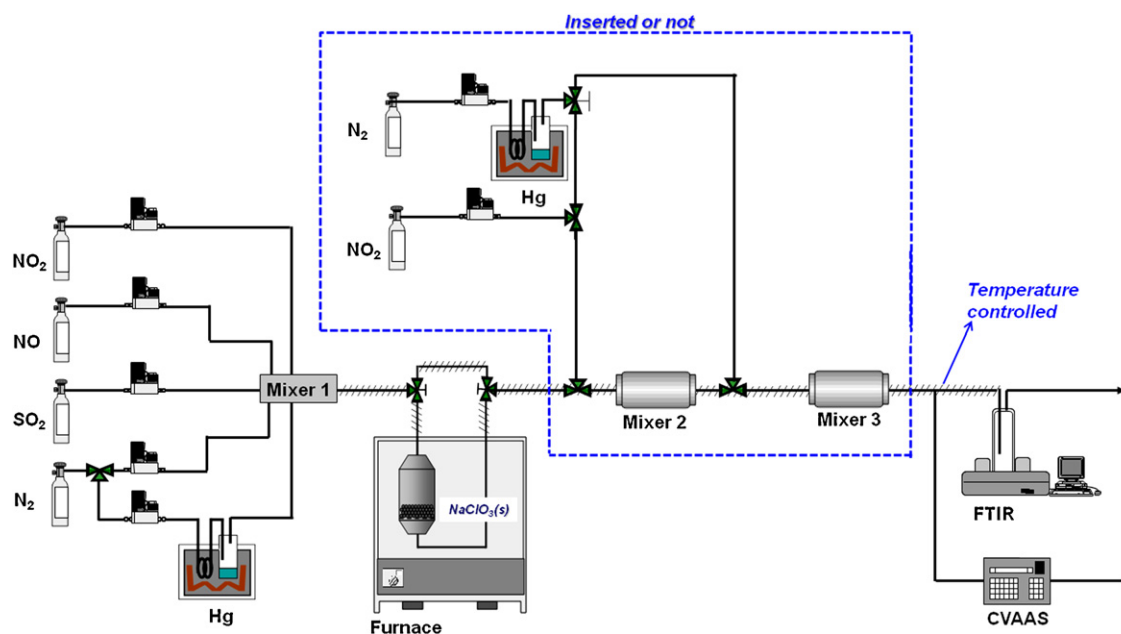


Fig. 1. Schematic diagram of experimental apparatus.

(gaseous OClO, ClO, and Cl). These can then oxidize NO to NO₂ and Hg⁰ to Hg²⁺.

2. Experiment

2.1. Experimental apparatus

In an experimental system (Fig. 1), gas flow rates were controlled using a set of mass flow controllers (MFC, Brooks 5850E) connected to an MFC readout box (Brooks Instrument Model 0154E). Test gases were prepared by mixing N₂ with NO (2000 ppm; N₂ balance, BOC gases), NO₂ (2000 ppm; N₂ balance, BOC gases), and SO₂ (2000 ppm; N₂ balance, BOC gases) at mixer 1. Apparatus for Hg vapor preparation involved saturating, metering and mixing Hg with the simulation gases. Portions of N₂ gas passed through two stages of Hg saturators made by ours and were then mixed with buffer air to prepare simulation gas stream comprising the desired Hg concentration. Hg saturators were immersed in a temperature-controlled digital oil bath (vWR Scientific Model 1156). All experiments were carried out at atmospheric pressure. NaClO₃(s) was packed in a quartz reactor (inner diameter: 15 mm and length: 160 mm) that was kept in a furnace to maintain the desired temperature. All lines consisted of Teflon and stainless steel tubes wrapped with heating tape. All gases except Hg⁰ were analyzed using a Fourier transform infrared (FTIR, BIO RAD) spectrometer, in which temperature of gas cell was set to 80 °C. Hg⁰ concentration was monitored by a lab-made Hg⁰ analyzer which adapts a principle of cold vapor atomic absorption spectrometry (CVAAS) [10]. SO₂ entering into the CVAAS interferes in analysis of Hg⁰. In this study, the concentration of SO₂ injected with Hg⁰ into CVAAS was maximum 40 ppm which responds to Hg⁰ signal about 2–3 μg/N m³. Therefore, we ignored the interference of SO₂ in CVAAS because 260 μg/N m³ Hg⁰ was used in this study; the reason why relatively higher Hg⁰ concentrations in this study were used is to reduce the possible experimental errors mainly attributed to SO₂. After the reaction of NaClO₃(s) with SO₂, used NaClO₃(s) was analyzed by X-ray diffraction (XRD). The XRD patterns were obtained using a D/max-2500 XRD (Rigaku) instrument, equipped with a graphite monochromator and Cu K as the X-ray source. The tube voltage and current was set to 50 kV and 180 mA, respectively. Scan ranged from 5° to 90° 2θ.

2.2. Experimental conditions

Five sets of experiments were conducted (Fig. 2 and Table 1). In experiment sets 1, 2, and 3, SO₂, NO or Hg⁰ in N₂ balance was injected directly to the reactor containing NaClO₃(s). In experiment sets 4 and 5, SO₂ in a N₂ balance was injected directly into the reactor, and the gases produced by the reaction with NaClO₃(s) were fed into mixers 2 and 3, and mixed with the injected NO₂ or Hg⁰ at 1 L/min in a N₂ balance. According to the injection position of NO₂ and Hg⁰, we inserted mixers 2 and 3 or not to fix the residence time before entering the FTIR and CVAAS. This was performed to

Table 1
Experimental conditions of each experimental set.

Experimental sets		
Set 1	Total flow rate	2.6 L/min
	Temperature	25–240 °C at 4 °C/min rate
	Packed NaClO ₃ (s)	2.4 g
	Hg ⁰ concentration	260 μg/N m ³
	NO concentration	200 ppm
Set 2	SO ₂ concentration	200 ppm
	Total flow rate	2.6 L/min
	Temperature	25–225 °C at 3 °C/min rate
	Packed NaClO ₃ (s)	2.4 g
Set 3	Hg ⁰ concentration	260 μg/N m ³
	SO ₂ concentration	5–40 ppm
	Total flow rate	2.6 L/min
Set 4	Temperature	25–265 °C at 3 °C/min rate
	Packed NaClO ₃ (s)	2.4 g
	NO concentration	200 ppm
	SO ₂ concentration	40 ppm
Set 5	Total flow rate	2.6 L/min
	Temperature	225 °C
	Packed NaClO ₃ (s)	2.4 g
	SO ₂ concentration	500 ppm
Set 5	NO ₂ concentration	15–700 ppm
	Total flow rate	2.6 L/min
	Temperature	25–220 °C at 3 °C/min rate
	Packed NaClO ₃ (s)	2.4 g
Set 5	SO ₂ concentration	40 ppm
	Hg ⁰ concentration	260 μg/N m ³

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