

A kinetic study of gaseous alkali capture by kaolin in the fixed bed reactor equipped with an alkali detector

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Abstract

In this study, the interaction between gas phase potassium species and kaolin was investigated in a fixed bed reactor equipped with a surface ionization detector, which is capable of detecting alkali metals in gas phase at ppb level. The effects of mass transport, space time, sorbent temperature and concentration of KCl on the rate of potassium adsorption on kaolin were studied in air. Kaolin, mainly composed of kaolinite— $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, was found to be very efficient in removing gaseous alkali species from hot flue gases at fluidized bed combustion temperatures. The removal efficiency increased as temperature was decreased or KCl concentration was increased. The capture of potassium by kaolin was irreversible with formation of both water-soluble and water-insoluble products. Kaolin captured KOH almost as effectively as KCl, but K_2SO_4 was captured much less effectively than KCl.

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Keywords: Alkali capture; Biomass combustion; Kaolin; Solid sorbents; Kinetics; Surface ionization detector

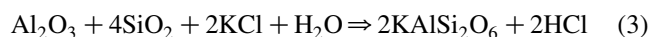
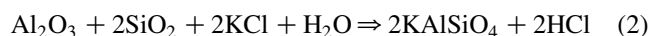
1. Introduction

Kaolin has been identified as one of the most promising sorbents for gas phase alkali metal capture at high temperatures [1–3]. During biomass combustion, the addition of kaolin has been found to increase the first melting temperature of ash [4–7]. Kaolin refers to a group of clay minerals, whose most common mineral is kaolinite— $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$. When kaolin is heated, water is released at temperatures of 450–600 °C, and an amorphous mixture of alumina and silica called meta-kaolinite is formed



The reaction between kaolin and gas phase sodium at high temperatures has been extensively studied [1–4,8–11]. Kaolin has been found to capture gaseous NaCl irreversibly and very effectively. The weight gain for NaCl capture was consistent with the formation of nephelite and carnegite, sodium aluminosilicate polymorphs represented by

the chemical formula $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ (=NaAlSiO₄) [1–3]. The capture of potassium compounds, which are of greater concern in combustion of biomass, has also been studied but to a lesser extent than that of sodium [1,5,12]. Kalsilite (KAlSiO_4) and leucite (KAlSi_2O_6) have been identified among products from a reaction between solid KCl and kaolin at high temperatures [5]. The overall reactions from meta-kaolinite to produce KAlSiO_4 and KAlSi_2O_6 can be presented as



Recently, a fixed bed reactor equipped with a surface ionization detector for on-line measurement of alkali metals in gas phase has been developed for studying the capture of gaseous alkali by solid sorbents at conditions relevant to biomass combustion [13]. The detector is capable of measuring alkali metal concentrations of the order of 1 ppb. This present work utilizes the technique for a more detailed study into the interaction between kaolin and gas phase KCl, KOH and K_2SO_4 . The thermodynamics

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and kinetics as well as the mechanism of potassium capture are discussed.

2. Experimental

Fig. 1 presents schematically the experimental set-up employed in this study. The reactor assembly consists of three major components: an alkali metal source, a fixed bed for holding the sorbent and an alkali metal detector. The fixed bed has an inner diameter of 12 mm and is situated in the centre of the reactor. The alkali metal cup is fixed on top of a movable thermocouple, which measures the temperature of the alkali metal source. By moving this thermocouple up and down, the alkali source temperature can be varied. The alkali metal concentration in the off-gases from the reactor is monitored directly by an alkali detector. The detector is based on surface ionization of alkali metals on the hot surface of the Pt filament. The filament potential was kept at +400 V and the temperature of the filament at 1500 K. More details on the equipment are described in [13], including test and calibration results.

The kaolin used in this study was French kaolin (MERCK, CAS-No: 1332-58-7), supplied by Merck Euro-lab. A TriStar 3000 Gas Adsorption Analyzer (Micro-metrics) was used for the determination of the surface areas of the kaolin particles. Some physical parameters for the kaolin are given in Table 1.

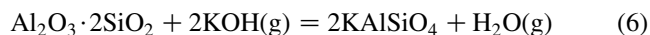
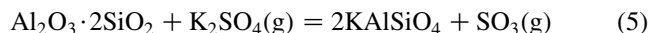
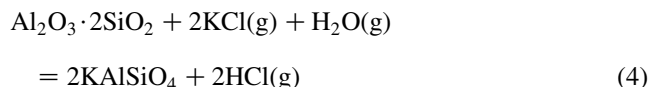
The original kaolin powder particles were too fine for this study due to a high pressure drop over the sorbent bed and difficulties related to the formation of a defined backed bed of the particles in the reactor. Therefore, the powder was made into a slurry with de-ionized water, allowed to dry naturally at room temperature, then crushed and sieved to retain the particles with desired sizes. Four different sizes,

0.5–0.71, 0.71–1.0, 1.0–1.4 and 1.4–2.0 mm in dynamic diameter (d_p), were selected for experiments.

The experiments were carried in air with a water content of no more than 20 ppm (AGA Gas AB, gas 0811). Conversion data reported here were collected when a steady state was established in the system, 24 h after the start [13].

3. Thermodynamic considerations

The thermodynamic software Outokumpu HSC Chemistry® for Windows, version 5.1 was used for equilibrium calculations. Results from chemical reaction calculations for a temperature range from 600 to 1000 °C indicate that reactions (4)–(6) are thermodynamically favoured. In addition, while KCl needs water vapour to react with meta-kaolinite (reaction (4)), K_2SO_4 and KOH do not (reactions (5) and (6)).



The equilibrium constants of the K capture reactions decrease with increasing temperature. Thus, the higher the temperature is, the lower the conversion of potassium and kaolin at equilibrium. Calculations were also made with different potassium species concentrations. Increases in gas phase potassium concentration will increase the amount of KCl converted at equilibrium, but the equilibrium conversion of the K gases to sorbed products decreases.

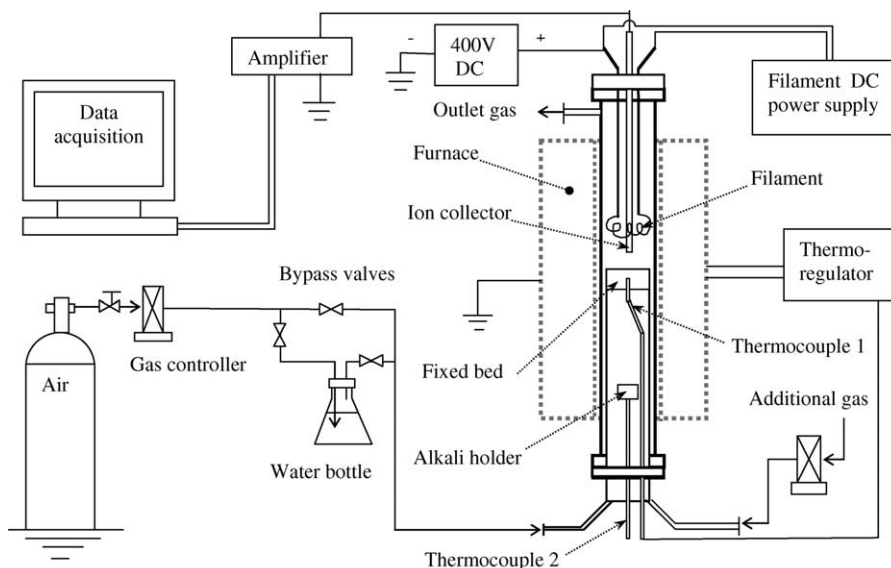


Fig. 1. Fixed bed reactor and schematic of the experimental setup.

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