



# Investigation of additives for preventing ash fouling and sintering during barley straw combustion



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## HIGHLIGHTS

- Kaolin and zeolite 24 have high capacities to capture KCl.
- KCl capturing efficiencies of kaolin and zeolite 24 decreased at a high temperature and with long reaction time.
- Barley straw ash has a high sintering and melting tendency.
- Addition of kaolin and zeolite 24A increased melting temperature of the barley straw ash.
- High melting points K–Al-silicates formed in the barley straw ash upon additive addition.

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## ABSTRACT

Formation of potassium chloride reduces ash sintering temperature and causes fouling deposits in biomass combustion applications. In the present work, the capacity of two mineral additives zeolite 24A and kaolin to capture KCl were investigated. A series of thermogravimetric experiments were carried out to measure fractions of KCl retained in the two additives as function of reaction temperature and heating time. The residues from additive-KCl mixtures after heating treatment were analyzed by X-ray diffractometry (XRD). When heated at 900 °C for 1 h, the overall KCl capturing efficiencies of the two additives were 60% and 45% for zeolite 24A and kaolin respectively, which slightly decreased to 50% and 43% as the heating time increased to 12 h. At 1000 °C, the fractions of KCl captured by zeolite 24A and kaolin significantly decreased from 50% and 40% to 26% and 17%, as the KCl-additive mixtures were heated for 1 and 12 h, respectively. The decrease in of the overall KCl capturing efficiencies is mainly attributed to reduction of surface areas and chemically active compounds of the two additives with increasing temperature and heating time. The XRD analysis results showed that both zeolite 24A and kaolin can react with KCl to form different potassium aluminium silicates. It indicates that chemical reactions play an important role in the overall capturing process. The effects of zeolite 24A and kaolin on sintering behaviors of the barley straw ash were also investigated. The residues from sintering tests were analyzed by a combination of X-Ray diffractometry (XRD) and scanning electron microscopy equipped with energy dispersive X-Ray analysis (SEM-EDX). The barley straw ash melted intensively at elevated temperatures. Together with XRD analysis, the SEM-EDX analysis results revealed that severe melting of the barley straw ash was due to formation and fusion of low temperature melting potassium silicates. Addition of kaolin and zeolite 24A significantly reduced the sintering tendency of the barley straw ash. Upon additive addition, high temperature melting potassium aluminium silicates formed in the barley straw ash as revealed by XRD and SEM-EDX analyses. Formation and presence of the refractory potassium aluminium silicates partly explain the improved sintering behaviors of the ash-additive mixtures.

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## 1. Introduction

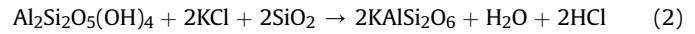
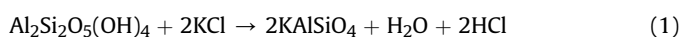
Due to the rapidly increasing demand of bioenergy, biomass materials such as residues from the agricultural sector are entering the market for heat and power production. Currently, combustion is still the most widely used technology for converting agricultural

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residues for energy production purpose [1]. However, combustion of agricultural residues is often challenging due to different ash related operational problems, including ash sintering, fouling and high temperature corrosion on heat transfer tubes [2]. Agricultural residues normally have high contents of ash forming matters, which can be 5–20 times higher than those of conventional woody biomasses [3]. Such high ash content often implies a high tendency of ash related problems and needs for ash removal. Moreover, concentrations of certain ash forming elements such as K, Cl, P and S in agricultural residues are generally high. The major fraction of ash forming elements in biomass fuels are nutrients needed for plants growth, which are often enhanced as a result of utilization of fertilizers [4]. Contamination of inorganic impurities during the transporting and storage processes may also result in enrichment of ash forming matters such as silica in the agricultural residues. In addition, a major portion of the ash forming elements in agricultural residues is organically associated with the fuel matrix or present in water soluble form [5]. Ash forming elements with such association readily release and react with other ash elements to form low temperature melting species, causing ash sintering, fouling and high temperature corrosion [6]. Among the major ash forming elements in agricultural residues, potassium is the most important one in initiating and boosting ash related operational problems in combustion applications [7]. During combustion, part of the potassium in agricultural residues readily releases and ends up as gaseous potassium containing compounds (i.e., KCl, K<sub>2</sub>SO<sub>4</sub> and K<sub>2</sub>CO<sub>3</sub>). Presence and condensation of the potassium containing gases cause formation of sticky fly ash particles that deposit on heat transfer surfaces and reduce heat transfer efficiency consequently [8]. On the other hand, a significant fraction of the potassium in the fuel may be retained in the solid combustion residues and reacts with silicon/silica to form low temperature melting potassium silicates, initiating and promoting ash sintering and slagging [9]. In addition, significant amounts of potassium phosphates form during combustion of phosphorus rich agricultural residues. Some of the potassium phosphates will also melt and lead to ash sintering at normal biomass combustion temperatures [10]. Ash sintering and fouling are main barriers limiting further utilization of agricultural residues for combustion applications [11]. It is necessary to obtain more detailed knowledge about ash transformation behaviors during the combustion of agricultural residues and to find ways to abate ash related operational problems.

There are several approaches to abate ash related operational problems during agricultural residues combustion, including (1) fuel leaching, (2) fuel mixing and (3) using additives. Utilization of additives has been proved promising and efficient, and applicable in biomass combustion systems [12]. One main objective of using additives is to bind and/or convert the low temperature melting potassium containing compounds (mainly potassium chloride) into species with high temperature melting points. Moreover, addition of additive can alter ash chemistry and promote formation of less troublesome species in combustion residues left in the bottom section of boilers [13]. According to major elements contained in additives, they can be approximately grouped into aluminum silicates based, sulfur based, calcium based, and phosphorus based additives [12]. Different aluminum silicates based additives have been investigated in past decades. Kaolin has been proven the most effective one to capture potassium chloride during combustion of biomass fuels [14]. In comparison with other additives, kaolin contains a more chemically reactive compound kaolinite (Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>). Kaolinite can bind and thereby convert the potassium chloride into less troublesome species through the following irreversible chemical reactions [15]:



The main products from the above reactions are KAlSiO<sub>4</sub> (kaolinite) and KAlSi<sub>2</sub>O<sub>6</sub> (leucite), which have high melting temperatures of about 1600 and 1500 °C, respectively [16]. In addition, the kaolin is normally presented in powder form and has porous structures and a large surface area. Therefore kaolin particles also can physically adsorb gaseous potassium chloride, which can be carried out from the furnace by the flue gas [8]. In one previous study, different additives were tested to capture potassium chloride formed during wheat straw combustion, which caused severe fouling, corrosion and agglomeration in a fluidized bed boiler. Kaolin showed superior ability compared to bauxite and emaltheite for capturing potassium chloride by a combination of physical adsorption and irreversible chemical reaction [17]. Zeolite is a promising additive candidate to capture alkali species during biomass combustion. Zeolite refers to a group of aluminum silicates with different Si/Al molar ratios that depend on its nature and synthetic origins [18]. The zeolite can be described with an overall formula M<sub>x/m</sub> [(AlO<sub>2</sub>)<sub>x</sub>·(SiO<sub>2</sub>)<sub>y</sub>]·zH<sub>2</sub>O, where M is various cations contained in the zeolite. One kind of zeolite, Doucil 24A, has been tested as an alkali getter during combustion of a mixture of 80 wt% wood and 20 wt% straw as pellets in a 12 MW<sub>th</sub> circulating fluidized bed boiler [18]. Upon addition of the zeolite Doucil 24A, about 80% of the potassium released from the fuel gas was reduced. At the same time, the amount of fouling deposits formed on the convection section of the boiler was significantly decreased [18]. In the same boiler, another zeolite ((Na<sub>2</sub>CaO)·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>·4H<sub>2</sub>O) was tested and showed ability to reduce concentrations of potassium chloride in the flue gas and deposits forming on a sampling probe as well [19]. Further analyses on fly ash collected by a cyclone indicated that the added zeolite captured the potassium containing species (mainly potassium chloride) in the fuel and bind them into insoluble alkali aluminum silicates [19]. In addition, zeolite is the most important constituent of washing powder, and becomes largely contained in the municipal waste water. During processing of the municipal waste water, the zeolite mostly ends up in the sewage sludge that is a main solid by-product from waste water processing plants [20]. With zeolite as a main compound, sewage sludge has showed promising effects regarding reducing fouling deposits during combustion of high potassium content biomass fuels [18]. However, for both kaolin and zeolite, previous studies mainly focused on adding them to reduce fouling deposits and ash slagging during combustion of biomass fuels. Only a few studies have been carried out to quantitatively study capturing of gaseous potassium chloride by kaolin at a high temperature. Interactions of gaseous potassium chloride with kaolin were studied in a fixed bed reactor equipped with an alkali detector [16]. It was found that more than 90% of vaporized potassium chloride was captured by the studied kaolin pellets. The capture efficiency of the potassium chloride by kaolin decreased with increasing heating temperature. However, the experiments were only carried out at the temperature range 750–950 °C and with a short contact time (seconds) between kaolin and potassium chloride vapors. More detailed understanding about reaction between kaolin and potassium chloride at both high temperature and longer reaction time is required. Moreover, studies about reactions between zeolite and potassium chloride are rarely reported in available literature. Therefore, one objective of this work was to characterize the capacity of kaolin and zeolite to capture KCl by applying a thermogravimetric method. The possible mechanisms of zeolite and kaolin to capture of KCl were investigated.

In addition to reducing fouling deposits, additives have also been studied for their ability to abate sintering of biomass ashes. In a comparative study, seven additives including clay minerals, lime-

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