



Full Length Article

Role of condensed phases in the agglomeration of low rank coal ash in fluidized beds



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ABSTRACT

Fuel diversity is critical in the utilization of fluidized bed combustors. To support the use of diverse fuels including low rank coals and biomass, interactions of chemical components that lead to operational issues such as agglomeration need to be studied. Low rank coals and biomass are generally rich in alkali and alkaline earth metals and their role in the initiation of agglomeration is investigated in this work. A novel modeling methodology to quantify the contribution of these components to the initiation of agglomeration via the condensation of gaseous species was developed. This condensation modeling methodology complements thermodynamic simulations such as those using FactSage and agglomeration models used for high rank coals. The study shows that at temperatures below 800 °C condensed phases play a critical role in the initiation of agglomerate growth. The condensation temperature is computed to be about 30–50 °C lower than the predicted molten slag-liquid formation temperature (740 °C for the lignite and 810 °C for the subbituminous coals studied). The extent of condensation correlates to the amount of alkali metals in the ash. Sodium sulfate forms a major component of condensates for low rank coals with condensation occurring over a larger temperature range for lignite than the sub-bituminous coals studied.

1. Introduction

In the last decade, the search for alternative and economically attractive solutions in the combustion of traditional fossil fuels (anthracites, bituminous and subbituminous coals, natural gas and oil) in thermal or power production energy systems has promoted research interest and development of new technologies that allow the energy valorization of low rank coals, biomass and other waste fuels (culm, plastic residues, municipal solid waste, etc.). Within existing combustion technologies, fluidized beds have aroused high interest due to the numerous advantages that they show.

In spite of the maturity of fluidized bed technologies in coal combustion processes, the heterogeneity and variability in fuel and their availability often leads to changes in the fuel supply including fuel blends. This makes it necessary to continue studying in depth the fundamental aspects of these technologies, especially with respect to understanding the effect of these new fuel types on operating issues.

One of the issues in the operation of fluidized bed is that of ash agglomeration in the reactor. Agglomeration occurs when the particles that form the dense phase of the bed start to stick to each other giving rise to an increase in particle size, and thus modifying the fluid

dynamics characteristics in the bed and consequently affecting the heat transfer and chemical reactions inside the reactor: uneven temperature distribution, lower conversion efficiency and ultimately abrupt defluidization. Agglomeration typically occurs in the dense phase of a fluidized bed where, due to the mixing of solids promoted by the fluidizing agent, solid particles collide against each other. During the collision, the impact force due to the kinetic energy of the particles is dissipated in the form of viscous dissipative forces, cohesive forces, capillarity forces and superficial tension forces. If the sum of the impact forces and the elastic repulsive forces is lower than the sum of dissipative forces, then the particles remain stuck after the collision, initiating the agglomeration process [1]. A detailed discussion of initiation of agglomerate growth in fluidized beds using high rank coals has been made by Khadilkar et al. [2,3]. The model proposed in these earlier studies was also validated with experiments involving agglomerates obtained from fluidized beds [4]. In addition to high rank coals, numerous industrial and laboratory-scale issues have been repeatedly reported in the literature during the utilization of low rank coals as well [5–8]. JEA's circulating fluidized bed boilers clearly demonstrated the industrial relevance of agglomeration forcing frequent shutdowns resulting from alkali-rich fuels [9]. Additionally, an U.S. DOE study

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showed agglomeration in 5 out of 13 industrial fluidized bed boilers operating with low rank coals (North Dakota lignites and Montana subbituminous coals) [10]. Goblirsch et al. [12] presented experimental work conducted in four fluidized bed facilities of different sizes (nominal fuel rate: 15 lb/hr, 110 lb/hr, 500 lb/hr, and 800 lb/hr) with four North Dakota lignites (including Beulah-Zap), six Montana and Wyoming subbituminous coals (including Wyodak), and one Utah bituminous coal. The post-mortem analysis of the agglomerates formed in these reactors showed that the condensation of sodium and sulfur as sodium sulfate caused the increased sodium levels in the bed material and agglomeration [11,12]. The works of Manzoori et al., Vuthaluru et al. and Bhattacharya et al. [13–15] elaborated on similar problems and analyzed the role of inorganic matter in the formation of agglomerates from South Australian brown coals (Lochiel and Bowmans) with high alkali content in fluidized bed combustion and gasification processes. They also referenced that the melt-forming inorganic elements and mineral inclusions in the ash have a pronounced effect on agglomeration and defluidization tendencies and that the release of compounds (mainly sulfates) may cause growth of particles, poor fluidization, and bed agglomeration.

Given the importance of different fuel types discussed above and the agglomeration issues seen with low rank coals, this study attempts to extend the agglomeration model developed by Penn State for high rank coals to the fuel chemistry of low rank coals. Our modeling approach in the current study, is a step towards quantification of the alkali, alkaline-earth metals from fuel and resulting fluidized bed condensates that have been observed to be the culprits causing agglomeration when using low rank coals.

2. Background

The difference in the agglomeration behavior of low-rank coals from bituminous coals and anthracite comes from the composition of the inorganic matter of the fuel. Low rank coals have higher amounts of alkali metals (Na, K), alkaline earth metals (Ca, Mg), as well as Si, Cl, S, P, Fe, that can interact with the bed material (generally silica) forming low melting compounds such as alkali-silicates. Deposition of liquid phase on the surface of the bed particles can occur by melting, condensation or chemical reaction [16,17]. These compounds have adhesive properties thus initiating agglomeration. A discussion of differences in mechanisms due to differences in chemical composition based on a review of the literature has been presented by Khadilkar et al. [18]. In the case of low rank coals these chemical interactions become more critical considering the high content of alkali, alkali-earth metals and sulfur of these fuels and thereby warrant a detailed study, especially under the lower temperatures at which fluidized bed combustors operate.

Agglomeration phenomena may be initiated by two mechanisms. The first mechanism is the melting of fuel mineral matter and this molten phase acts as a glue to adhere to other particles in the bed. This is a non-reactive mechanism, wherein some inorganic compounds melt at the bed operating temperature. During char combustion phase, the surface of the particle exceeds the bulk temperature of the bed by about 100–200 °C due to exothermic reactions depending on the particle size and oxygen concentration at the surface [19]. In the second mechanism, vaporized inorganic species (NaOH, KOH, NaO, KO, Na₂SO₄, K₂SO₄) from the fuel particles condense over cooler bed particles covering their surface and leading to reactions that form low melting point eutectics, which are adhesive. In parallel, these vaporized species may also directly react with the particle surface leading to the same phenomena [20]. Condensation of gaseous species downstream onto heat exchangers and in the loop seal region has led to industrial deposition issues.

The condensation of inorganic species from the gaseous phase onto cooler surfaces, particles, takes place when the vaporized compounds are oversaturated i.e. their partial pressure is greater than the vapor

pressure. For this reason, condensation problems over heat exchange surfaces usually occur when the gases leave the reaction zone (typically in the convective zone of industrial boilers), since the gas temperature decreases, thus decreasing the saturation pressure. Inside the dense phase of fluidized bed reactors, the presence of an enormous effective surface for deposition (all the particles inside the bed and the presence of internals), creates the possibility of the condensation of species inside the bed due to temperature differences between burning char particles and inert bed particles at bed temperatures.

There are two main routes for the condensation of gaseous species: homogeneous nucleation (or aerosols formation) and heterogeneous condensation over other particles or surfaces. Homogeneous nucleation takes place when the gas nucleates to form a new phase, solid or liquid, without the presence of any surface. In this case, the gas molecules converge to form a nucleus, which then forms a particle. Homogeneous nucleation is expected to occur in systems when the saturation ratio, defined as the ratio of the partial pressure of the condensing gas to the vapor pressure of the condensing gas, is much higher than unity. McNallan et al. [21] summarize that the occurrence of homogeneous nucleation depends on the cooling rate of the combustion products, which determines the amount of condensable vapor species in the gas phase, and number and size of existing sites available for condensation, concluding that if surface areas of particles are already present in the system (for example in a fluidized bed), then it is more likely that heterogeneous condensation will occur.

Although thermodynamic equilibrium models are capable of predicting the formation of melt phase by chemical reactions of solid mineral matter in fuel and bed material, their capability in the prediction of condensation is limited to the Scheil-Gulliver cooling model calculations of the condensation of gaseous species. In this work, a condensation model that allows determining the heterogeneous condensation rate of inorganic gaseous species on the bed particles was developed. This enables a study of the role of the condensation mechanism in the initiation and propagation of agglomeration. The implementation of this sub-model in an integrated ash agglomeration model will allow modeling the change in the average diameter of particles with time in the reactor as a result of both agglomeration mechanisms: coating-induced agglomeration and condensation. A model to predict agglomerate growth in high rank coals was presented earlier by Khadilkar et al. [22]. Moreover, the chemistry of low rank coals makes extension of the model critical in the understanding and prediction of fluidized bed agglomeration for these fuel types.

Therefore, specific objectives of this study are:

- 1) To incorporate the effect of the condensation of gaseous species in the prediction of slag-liquid formation in fluidized bed combustors
- 2) To use FactSage thermodynamic simulations with condensation modeling to quantify the occurrence of condensation during low rank coal ash agglomeration
- 3) To identify the components that initiate ash agglomerate growth in low rank coals

3. Materials and methodology

Two low rank coals, Wyodak-Anderson and Beulah-Zap (sub-bituminous and lignite respectively according to ASTM coal classification standards) from the Northern Great Plains province in USA have been selected for the study. The coal samples for this study were obtained from the Penn State Coal Sample Bank (samples APCS-8 – Beulah-Zap and APCS-2 – Wyodak-Anderson). These coals were subjected to chemical fractionation in order to obtain fractions rich in alkali and alkaline earth metals, for this study. The chemical composition of ash (500 °C) was obtained using ICP-AES. FactSage™ version 5.2 was used to determine the slag-liquid formation and the quantity and mass composition of final stable compounds under equilibrium, thereby identifying the components that initiate agglomerate growth in low

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