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#### Review

### Particle agglomeration during fluidized bed combustion: Mechanisms, early detection and possible countermeasures



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#### ABSTRACT

Particle agglomeration in fluidized bed systems has been observed in several industrial processes. Bed particles tend to agglomerate when the cohesive force between the particles is strong enough to compare with the other acting forces (gravitational, drag). This cohesive force may arise because of different types of interactions: van der Waals, electrostatic, capillary, viscous, sintering, adhesive, chemical reaction, and so on. The most extensively reported case in the literature is concerned with the fluidized bed combustion of biomass, waste or lowrank coals, containing significant amounts of low-melting compounds (typically alkali metals) in the ash. The occurrence of bed agglomeration in such systems implies the unscheduled shut down of the reactor and costly maintenance operations. Therefore, a great deal of research has been devoted to understand and characterize agglomeration during fluidized bed combustion. This short review is focused on this specific system and tries to summarize the present status of understanding of the mechanisms leading to agglomeration, as well as the influence of the different operating variables on this phenomenon. In addition, because of their great practical importance, the possible early detection techniques and operational countermeasures are also briefly described.

#### 1. Introduction

Fluidized bed agglomeration in combustion systems has been reviewed in the literature in the past decade. Bartels et al. [1] first tried to summarize the current understanding on this problem with a specific emphasis on the development of agglomeration detection techniques. Subsequently, Mettanant et al. [2] and Khadilkar et al. [3] reviewed this topic, the latter giving special emphasis on possible agglomeration model development. Niu et al. [4] also devoted a small section of their review on ash-related issues during biomass combustion on agglomeration in fluidized beds. Billen et al. [5] focused their review on equilibrium calculations directed to predict melt formation and, in turn, agglomeration in fluidized bed combustion (FBC). The present review paper, instead, is focused on the detailed analysis of the agglomeration mechanisms during FBC of biomass or of other low-grade fuels, a topic that has been often overlooked in the past. The possible early detection techniques and operational countermeasures to agglomeration are also briefly discussed, because of their practical relevance.

Since the early eighties, operational experience was reported of FBC of low-grade coals, mostly lignite, giving rise to bed agglomeration problems [6-9]. These reports indicated that severe agglomeration in several cases resulted in the defluidization of the bed and consequently in the shutdown of the combustor. It was already recognized at that

time that bed agglomeration was connected to the alkali metals (potassium or sodium) contained in the fuel ash. It was found that alkali tended to build up on the inert bed material (silica sand and/or limestone) at the typical FBC operating temperatures. On the bed particles, as soon as the alkali concentration reached a large enough value, lowmelting-point eutectics with silica and calcium were formed. The formation of a melt at the surface of the bed material enhanced its stickiness, so that an increasing number of particles started attaching together, and large agglomerates were formed. The key parameters of this agglomeration process were found to be the alkali form and content in the fuel ash, the nature of the bed material, the gas superficial velocity, and the temperature and time of combustion [10,11]. The FBC of high-sodium lignite was comprehensively studied by Manzoori and Agarwal [12-14] who first investigated the agglomeration mechanism. These authors found that inorganic elements in the fuel ash formed a molten matrix on the char surface, independently of the fuel particle temperature and size, while sodium vaporization was always limited under FBC conditions. This molten ash was observed to transfer to the surface of the bed material as a result of random particle collisions. It was also reported that no evidence was found that alkali vapor condensation or chemical reaction caused or initiated agglomeration. Also, it was found that the bed temperature did not affect significantly the composition of the ash coating over the bed particles, but it only

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Fig. 1. Photograph of a large sand agglomerate after pilot-scale FBC of biomass [22].

affected its physical properties (*e.g.* viscosity). The rate of ash deposition on the bed particle surface was only dependent on the bed temperature and ash alkali content. Finally, the agglomeration process started after a critical ash coating thickness was reached.

During the last few decades the exploitation of biomass resources as a renewable and  $CO_2$ -neutral energy source has gained increasing interest. FBC has been generally indicated as one of the most suitable technologies for converting this type of fuels, because of its well-known fuel flexibility, the high combustion efficiency and the low environmental impact. However, when burning biomass in fluidized bed systems agglomeration and defluidization problems have been often reported [15–20], even at operating temperatures as low as 650 °C [21] (Fig. 1). This was especially experienced when burning crops cultivated with a high degree of fertilization, as this biomass generally contains higher alkali amounts in the ash.

FBC Operational experience with biomass has shown that defluidization of the bed material is accompanied by a sharp drop of the differential pressure across the bed, because of channeling phenomena through the bed by the fluidizing air [23]. Also, upon agglomeration the in-bed temperatures show a substantial decrease while the lower freeboard section temperature shows a significant increase (since the biomass particles accumulate and burn above the agglomerated bed), making the operation uncontrollable.

## 2. Mechanisms of bed agglomeration and effect of operating variables

As early as in the nineties, Ghaly et al. [24,25] simulated in a muffle furnace the high temperature interaction between straw ash and silica or alumina sand. These authors reported that above 850 °C the straw ash interacted with silica sand forming a melt, because of a low-temperature eutectic in the potassium-silica system, while this did not happen with alumina sand. However, at such temperatures the straw ash fused by its own and covered the alumina particles with a sticky layer. Latva-Somppi et al. [26] and Valmari et al. [27] first investigated the mechanisms of ash-quartz interactions in full-scale FB boilers firing different biomass fuels. The authors report that ash deposition was likely dominated by two parallel mechanisms: 1) the adhesion (promoted by particle collisions) of small micron-sized ash particles on the bed sand surface and their subsequent sintering, leading to the formation of a porous layer; and 2) the diffusion of alkali to the interface with the quartz particle where a compact layer was eventually formed. This second mechanism was imputed both to solid-state diffusion of alkali species from the ash layer to the quartz interface and to the direct attack by volatile alkali. Skrifvars et al. [28,29] highlighted the importance of the bed sintering tendency due to partially molten biomass ash. A method for the prediction of the sintering tendency of biomass ash was proposed, which was based on the combination of compression strength tests and of thermodynamic equilibrium multiphase multicomponent analysis. One limit of this procedure, however, was that it did not take into account the interaction between the ash and the inert bed material. These authors also tested the accuracy of standard ASTM ash fusion tests, which did not succeed in predicting the experimentally measured bed agglomeration temperatures, for the same reason as indicated before (the interaction between ash and sand is not considered in these tests). In conclusion, these results suggested that *ex-situ* methods, which do not recreate the real environment of a fluidized boiler, fail to quantitatively predict the agglomeration characteristics of biomass fuels.

A more realistic controlled FB agglomeration technique for biomass was proposed by Öhman and Nordin [30]. According to this technique a biomass sample was first ashed in the FB, followed by a gradual temperature increase (by external heating) until bed defluidization occurred. This method was extensively applied to many biomass fuels in order to obtain a ranking of their agglomeration tendency [31-33]. While the measured agglomeration temperatures were reasonably accurate and reproducible, it remains unclear how well these results can reproduce bed agglomeration in full-scale FBC units. In fact, since there is no combustion during the temperature increase stage, the local overheating near burning char particles is absent in their tests, and also the progressive accumulation of ash in the bed (which typically occurs during steady combustion) is not accounted for. The analysis of the bed samples collected during their tests clearly showed that a homogeneous layer coated the particles, which was mainly composed of calcium, potassium and silica. In addition, a heterogeneous outermost layer could be detected, suggesting that ash was progressively depositing on the coating layer, and eventually homogenizing with it. The authors also reported that the melting behavior of this coating layer depended on the potassium content and was eventually responsible for particle agglomeration. After extensive SEM-EDX analyses of bed particles collected during lab-scale and full-scale biomass FBC, it was reported that this particle coating could be divided into an inner layer mainly consisting of alkali and silica, and of an outer layer resembling the fuel ash composition [34-37]. The presence of high amounts of phosphorus in selected fuels was also recently shown to enhance agglomeration during FBC, possibly due to the formation of low-melting alkali phosphates or to the influence of phosphates on the availability of calcium in the system [38-43].

An additional significant contribution to shed light on the agglomeration mechanism relies on the work carried out by Lin et al. [44-46] and Lin and Dam-Johansen [47,48] to study the FBC of straw. The effect of the main operating variables was systematically studied in lab-scale FBC tests, showing that bed temperature was the most influencing variable, with defluidization time decreasing exponentially with temperature. Potassium was shown to gradually build-up in the bed, with a rate that was not significantly affected by bed temperature. This result suggested that little potassium compounds were evaporated during combustion, as confirmed by Simultaneous Thermal Analysis on ash [49]. Interestingly, during selected batch experiments the authors noted that few agglomerates already formed few minutes after straw feeding had started. These agglomerates showed a blackish ash core and a shape similar to the straw pellets. The authors suggested that these agglomerates were likely formed because of potassium rich ash transferred by collisions from the burning char particles to the sand. In particular, the higher temperatures close to burning char could have enhanced potassium transfer and subsequent fusion. On the basis of the above results, two different defluidization mechanisms were proposed: 1) at low bed temperatures, as the potassium accumulates in the bed an increasing number of agglomerates is formed during combustion. These agglomerates consist of many bed particles joined together by fused necks, and they progressively build up at the bottom of the bed, eventually leading to the total bed defluidization; 2) at high bed

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