Ash related effects during co-combusting coal and biofuels in a pilot scale (5kW) fluidized bed boiler

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Askrelaterade effekter vid samförbränning av kol och biobränslen i en fluidbäddspanna i pilotskala (5kW)

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Abstract

The use of fossil fuels for electrical and heat production is a problem due to e.g. CO₂ emissions. One way to reduce the amount of fossil fuels used in heat and power plants is co-combustion of coal with biofuels. This might however lead to an increased risk of operational ash related problems. The two objective with this work were therefore to; i) determine at which fuel mix the bed is self sufficient with bed material (coal ash) and operates satisfyingly and ii) Evaluate the ash related effects in a small pilot scale (5 kW) fluidized bed reactor when co-firing different coals with different solid biofuels. The pilot reactor was used as a research facility to simulate the ash transformation processes of relevance for full scale fluidized bed boilers for heat and power production.

Initially, a short literature review was performed to give an understanding of what has been done in the area of co-combustion of coal and biofuels.

The results from the first “ashing” experiments showed that ash from both coals could presumably be used as bed material. However, only a small fraction of the total ash was suitable for our lab scale combustion experiment. Un-sieved quartz sand was therefore used as bed material in the following pilot scale FBC experiments.

To evaluate the possibility of co-firing two different coals with two biofuels (wood and straw) in a fluidized bed, a number of experiments were carried out with fuel mixtures of 30/70 and 70/30 in weight percent coal/biofuel. During these experiments bed- and ash samples, probe ring deposits, flue gases and particles were sampled for subsequent gravimetrical and chemical analyses. In addition, controlled agglomeration tests were performed.

The results indicated an absence of fine mode particles in all experiments with mixtures of biomass and coals, which is of great relevance from a deposit formation point of view and mechanistically it is connected to condensation of gaseous alkali species. Even at 70 wt-% admixing of straw to both tested coals, no significant fine mode was determined. It must, however, be stated that the interpretation of the deposit rate by the probe measurements in this work are strongly interfered by the deposition of coarse mode bed and ash particles.

The results from the controlled bed agglomeration tests strongly indicate positive effects from mixing of the fuels. For wood, that has an initial agglomeration temperature of around 850 °C, the agglomeration tendencies was in all cases involving mixtures with the coals, reduced considerable, i.e. no agglomeration was observed up to 1050 °C. For straw, that has a very low initial agglomeration temperature around 670 °C, an even more pronounced reduction in agglomeration tendency was observed. For the mixture with 70 wt-% of straw in Coal1 and Coal2 the initial agglomeration temperatures were raised with approximately 300 and 180 °C, respectively.
To obtain enhanced understanding of the ash transformation chemistry, bulk and SEM/XRD analyses on various ash fractions were performed, resulting in the following conclusions:

1) Admixing high levels (up to 70% by weight) of wood to the two studied coals does not impose any major problems from bed agglomeration point of view.

2) Two mechanisms for reduced bed agglomeration problems in co-combustion of straw with the present coals could be suggested: i) reaction of alkali (from the straw) with clay type minerals in the coals under formation of rather refractory alkali alumino-silicates and ii) incorporation of the alkali earth metal oxides (CaO and MgO) into low melting alkali silicate melts, which otherwise is considered to promote bed agglomerations.

3) Assumed high local temperature in the proximity of burning coal fuel particles results in different ash transformation processes e.g. melted Ca-silicates in coarse cyclone ash.
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1 Introduction

The coal consumption in the world has gone up steadily since 1980 and according to projections made by the Energy Information Administration (IAE) [1], it will continue to rise. The non-OECD (Organization for Economic Cooperation and Development) country members are expected to stand for the greatest increase which corresponds to 94 percent of the projected increase in total world coal consumption. Non-OECD Asia stands for the biggest part, caused by rising energy use as a result of strong economic growth. In 2006, coal accounted for 27 percent of world energy consumption. [1]

![Figure 1: World coal consumption by country grouping, 1980-2030. [1]](image)

Except for the fossil CO\textsubscript{2} that are released during coal combustion, also significant air pollutions such as SO\textsubscript{x} and NO\textsubscript{x}, that e.g. leads to acid rain and ozone depletion, are released [2]. In contrast to coal, biofuels are considered CO\textsubscript{2} emission neutral, which means that they release the same amount of CO\textsubscript{2} during combustion as they absorbed during growth. Biomass also normally contains a lot less sulfur that thereby leads to reduced SO\textsubscript{x} emissions compared to coal.

Co-combustion of biofuels and coal in existing fluidized bed boilers for coal is one way to reduce the amount of fossil fuels combusted for electrical- and heat production. When co-combusting with high shares of biofuels, significant changes in the ash transformation chemistry can be expected, which may lead to increased risk of different ash related operational problems. Ash related problems of relevance during co-combustion include bed agglomeration, slagging, fouling and corrosion of surfaces. These problems can lead to reduced efficiency, accessibility of the facility and capacity which results in increased power costs [3, 4]. Emissions of SO\textsubscript{2} and particulate matter (PM) are also affected when mixing fuels and needs concern. How serious these effects are depends on a number of variables where fuel properties and blend choice as well as process conditions during the combustion are of major importance.
1.1 Interest in co-combustion

The interest in co-combustion has rose sufficiently in the last years. In 2003 Leckner [5] reported the number of publications per year dealing with co-combustion or co-firing, independent of fuel kind, extracted from the searchable scientific database ScienceFinder. The number of publications per year has increased from about twenty in 1990 to over a hundred per year in 2003 [5]. These publications deal with every kind of combustion and fuels, and just a few deals with both fluidized bed and co-combustion of coal and biomass.

A literature search in the scientific data bases Web of Science and Science Direct in September 2009 for publications concerning co-combustion of coal and biofuels in fluidized bed boilers gave an overview of the area, see the appendix for a short compilation of the most interesting. The used search terms were co-combustion, co-firing, fluidized bed, fluidised bed, FBC, CFBC, BFB, biomass, biofuel and coal.

Most of the publications that were found in the data-bases dealing with the area of co-combustion of coal and biomass in a fluidized bed seems to be focused on emissions (mainly gaseous) and fuel burnout properties and not so much on ash related issues like bed agglomeration and deposits. However the general opinion seems to be that this is a very important area and more research needs to be done in all the fields.

The publications in figure 2 below illustrate the increased interest in co-combustion of coal and biofuels in fluidized bed boilers. To be noted is that the publications contain both articles, proceeding papers and review papers and may just be mentioning co-combustion of coal with biomass in a fluidized bed and not be focused on it.

![Figure 2: Publications/year dealing with co-combustion of coal and biomass in a fluidized bed boiler, extracted from Web of Science in November 2009.](image-url)
1.2 Objectives

The two objectives with this work were to:

- Determine at which fuel mix the bed was self sufficient with bed material (coal ash) and operating satisfyingly.
- Evaluate the ash related effects in a small pilot scale (5 kW) fluidized bed reactor when co-firing different coals with different solid biofuels. The pilot reactor was used as a research facility to simulate the ash transformation processes of relevance for full scale fluidized bed boilers for heat and power production.
2 Theory

2.1 Fluidized bed combustion

The principle is that a bed of solid particles that initially are stationary in the bottom of the furnace is brought to a fluidized state by a stream of air that flows up through a distribution plate. When the air velocity increases, the pressure difference over the bed increases until a critical value, called “minimum fluidization velocity”, is reached and the pressure difference stops to increase. [6]

![Diagram of fluidized beds classification](image)

**Figure 3:** Classifications of fluidized beds as function of air velocity and bed pressure differential [6].

The bed particles in a bubbling fluidized bed are held in suspension by the air stream [4], this occurs at quite low velocities compared to a turbulent bed, see Figure 3. When the air velocity increases particles starts to leave the bed since the air velocity exceeds the terminal velocity of the particles. The distinct bed surface disappears and the concentration of particles decreases the higher up from the plate you are [4], the bed acts more like a turbulent bed. In a circulating (turbulent) fluidized bed the particles carried out of the bed are separated from the flue gas in an external cyclone and returned to the bottom of the bed [4].
Some positive aspects with fluidized beds are [4]:

- Uniform temperature distribution in the bed because of effective mixing of fuel and bed material.
- Large solid and gas heat exchange area.
- High heat transfer coefficients between heat exchanging surfaces and the bed.
- Possibility to combust various fuels with different sizes and shapes, moisture contents and heat values.
- Stable combustion at low temperature (≈ 850 °C) which inhibits the formation of thermal and prompt NOx.

Negative [4]:

- Requirement of particle separation or gas cleaning due to high dust load in flue gases and carryover of solids by the fluidizing gas.
- Erosion of boiler internals caused by high particle velocities (more severe for circulating fluidized beds than for bubbling).
- Risk for agglomeration of bed material which can lead to defluidization.
2.1.1 Terminal velocity

Generally forces of gravity, buoyancy and drag acts on a particle in a fluid:

![Diagram showing forces acting on a particle moving in a gas.](Image)

**Figure 4:** Forces acting on a particle moving in a gas [7].

Gravitational force – Buoyancy force – Drag force = Acceleration force

A particle falling from rest in a fluid will in the beginning experience a high acceleration. When the acceleration increases the drag force increases causing the acceleration to decrease. An equilibrium state, where the acceleration is zero will eventually be reached. The particle velocity in the equilibrium state is called terminal velocity. Carryover of particles in a fluid bed of uniform size will take place at this velocity.

Gravitational force:

$$F_g = m_p \times g$$  \hspace{1cm} (1)

$m_p$: particle mass
$g$: gravitational acceleration
Buoyancy force:

\[ F_B = m_p \times \frac{\rho_g}{\rho_p} \times g \]  \hspace{1cm} (2)

\( \rho_g \): gas density  
\( \rho_p \): particle density

Drag force:

\[ F_D = \frac{C_D \times A_p \times \rho_g \times U_g^2}{2} \]  \hspace{1cm} (3)

\( C_D \): Drag coefficient  
\( A_p \): Projected area of the particle in the plane perpendicular to the flow direction  
\( U_g \): Gas velocity

Drag coefficient:

\[ C_D = \frac{a_1}{Re^{b_1}} \]  \hspace{1cm} (4)

\( a_1 \): constant  
\( b_1 \): constant  
\( Re \): Reynolds number

<table>
<thead>
<tr>
<th>Range of Re</th>
<th>Region</th>
<th>( a_1 )</th>
<th>( b_1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 &lt; Re &lt; 0.4</td>
<td>Stoke’s law</td>
<td>24</td>
<td>1.0</td>
</tr>
<tr>
<td>0.4 &lt; Re &lt; 500</td>
<td>Intermediate law</td>
<td>10</td>
<td>0.5</td>
</tr>
<tr>
<td>500 &lt; Re</td>
<td>Newton’s law</td>
<td>0.43</td>
<td>0.0</td>
</tr>
</tbody>
</table>

**Figure 5**: Drag coefficient constants for different Reynolds numbers. [12]
Reynolds number:

\[ \text{Re} = \frac{d_p \times U_g \times \rho_g}{\mu_g} \quad (5) \]

\( \mu_g \): gas viscosity

Force balance under steady state:

Gravitational force – Buoyancy force – Drag force = 0

\[ m_p \times g - m_p \times \frac{\rho_g}{\rho_p} \times g - \frac{C_D \times A_p \times \rho_g \times U_t^2}{2} = 0 \quad (6) \]

\( U_t \): terminal velocity

Solving for terminal velocity:

\[ U_t = \sqrt{\frac{2 \times m_p \times g \times (\rho_p - \rho_g)}{C_D \times A_p \times \rho_g \times \rho_p}} \quad (7) \]

For spherical particles:

\[ A_p = \frac{\pi \times d_p^2}{4} \]

\[ m_p = \frac{\pi \times d_p^3 \times \rho_p}{6} \]
That together with equation (7) gives:

\[
U_t = \sqrt{\frac{4 \times g \times (\rho_p - \rho_g) \times d_p}{3 \times \mathcal{C}_D \times \rho_g}}
\]  

(8)

By substituting the different drag coefficient values from Figure 5 in formula 4 together with formula 5 and 8, three cases for calculating terminal velocity, depending on Reynolds number, are obtained:

0 < Re < 0.4, Stokes law:

\[
U_t = \frac{g \times (\rho_p - \rho_g) \times d_p^2}{18 \times \mu_g}
\]  

(9)

0.4 < Re < 500, Intermediate law:

\[
U_t = d_p \times \left(\frac{2 \times g \times (\rho_p - \rho_g)}{15 \times \sqrt{\rho_g \times \mu_g}}\right)^{\frac{2}{3}}
\]  

(10)

500 < Re, Newton’s law:

\[
U_t = \sqrt{\frac{4 \times g \times (\rho_p - \rho_g) \times d_p}{1.29 \times \rho_g}}
\]  

(11)
The terminal velocity for quartz sand and bituminous coal flowing in flue gases depending on particle diameter, calculated with formula 9-11 above, is shown in figure 6.

**Figure 6:** Terminal velocity for quartz sand and coal as a function of particle diameter. The difference in terminal velocity for sand and coal depends on the density difference between the two materials.
2.2 Fuels

When vegetable debris accumulates during a very long time (millions of years) and special environmental conditions, different coals are formed. Such accumulations have been affected by sedimentary influences to produce coals of different ranks. The lowest coal rank is lignitous, which includes coals with a relatively low higher heating value (HHV), low total carbon- and high moisture content. The highest rank is anthracitous which contains coal with high HHV, low moisture content and high amount of total carbon, see table 1 for the general classifications of different coal ranks. [8]

Table 1: Specifications of different coal ranks [9], AF = ash free.

<table>
<thead>
<tr>
<th>Coal rank</th>
<th>HHV [MJ/kg] AF</th>
<th>Volatiles [Wt% AF]</th>
<th>Total carbon [Wt% AF]</th>
<th>Bed moisture [Wt% AF]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lignitous</td>
<td>24-30</td>
<td>45-55</td>
<td>59-75</td>
<td>20-50</td>
</tr>
<tr>
<td>Sub-bituminous</td>
<td>30-32</td>
<td>40-45</td>
<td>75-80</td>
<td>8-10</td>
</tr>
<tr>
<td>Bituminous</td>
<td>33-40</td>
<td>18-40</td>
<td>80-91</td>
<td>0,8-5</td>
</tr>
<tr>
<td>Carbonaceous</td>
<td>38-40</td>
<td>10-18</td>
<td>91-93</td>
<td>1,0-1,5</td>
</tr>
<tr>
<td>Anthracitous</td>
<td>37-39</td>
<td>8-10</td>
<td>93-95</td>
<td>1,5-2,5</td>
</tr>
</tbody>
</table>

Compared to coal, biomass in general has lower density and heating value, contains less carbon and it also contains more volatiles [2]. The moisture content of the fuels may also vary widely. Wood and straw pellets have about twice the volume for the same quantity of energy as coal [5].
Overall, coal and biomass have some typical characteristic differences which are shown in table 2 below.

**Table 2**: Typical differences between coals and biofuels for some important ash forming components according to Leckner, 2007[5].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Coals</th>
<th>Biofuels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulphur</td>
<td>Medium to high</td>
<td>Low</td>
</tr>
<tr>
<td>Chlorine</td>
<td>Medium</td>
<td>Low to high</td>
</tr>
<tr>
<td>Potassium</td>
<td>Bound</td>
<td>Medium to high</td>
</tr>
<tr>
<td>Other alkali</td>
<td>Normal</td>
<td>Low</td>
</tr>
<tr>
<td>Alumina, silica</td>
<td>High</td>
<td>Low to high</td>
</tr>
</tbody>
</table>

The main ash forming elements are Si, P, Al, Ca, Mg, K and Na which at specific combinations and situations, can cause problems with deposits, corrosion and agglomeration [5]. In contrast to biomass where alkali (potassium) is released during devolatilisation and char combustion, alkali species in coal are to a great extent bound in alumino-silicates and therefore not easily available for ash chemical reactions [5,10]. N, S and Cl works as gas emission precursors and can end up as NO\textsubscript{x}, SO\textsubscript{2} and HCl [5], but can also form sulphates and chlorides in deposits and fine particles.
2.2.1 Synergetic effects

Chemical and physical interactions of fuel ashes, both positive and negative, from two or more fuels co-combusted have, in different studies, been shown to occur. Chemical interactions most often are related to the behavior of chlorine and alkalis [11]. In figure 7 below, some previously discussed potential synergy effects are shown.

![Diagram showing synergetic effects between co-firing fuels, according to Leckner, 2007 [5].](image)

As a complement to Figure 7, the effect of Ca on the melting behavior of ashes during combustion are of great relevance for e.g. bed agglomeration as demonstrated by Brus et al, 2005 [12].

Sulfur and silicates or aluminum silicates in coal ash can react with alkali chlorides and bind alkalis and thus forming alkali sulfates and different alkali silicates, leaving the chlorine as HCl [11, 13, 14]. Also alkali hydroxides may react in similar ways as the alkali chlorides [14]. This might occur by the reactions R1, R2 and R3 below.

Sulphation of alkali chlorides, KCl or NaCl [11, 14]:

$$2KCl + SO_2 + \frac{1}{2}O_2 + H_2O \rightarrow K_2SO_4 + 2HCl \quad (R1)$$

Aluminum silicates reacting with alkali chlorides, KCl or NaCl [14,15]:

$$Al_2O_3 * 2SiO_2 + 2KCl + H_2O \rightarrow K_2OAl_2O_3 * 2SiO_2 + 2HCl \quad (R2)$$

HCl is not as harmful for the high temperature parts of a boiler as the alkali chlorides since it doesn’t condense and form fast growing, hard to remove and corrosive deposits.
Aluminum silicates reacting with alkali hydroxides, KOH or NaOH [14]:

$$Al_2O_3 * 2SiO_2 + 2KOH \rightarrow K_2OAl_2O_3 * 2SiO_2 + H_2O$$ \hspace{1cm} (R3)

The presence of calcium in ashes can compete with reaction R1, R2 and R3 by reacting with sulfur oxides and silicates thus forming calcium sulfates and calcium silicates. Calcium might thereby reduce to which extend reaction R1, R2 and R3 occur. [11]

2.3 Agglomeration

Two possible agglomeration pathways are suggested in [16], these are:

- No active layer formation: If the fuel ash contains elements forming low temperature melting compounds, melted ash droplets can begin the agglomeration process without chemical interaction between the fuel ash and the bed material.
- Active layer formation: Due to chemical interaction between the bed particles and the fuel ash a layer is formed:
  - The formed particle layer has a low temperature melting composition, the layer gets sticky and agglomeration starts.
  - The formed particle layer has a high temperature melting composition and agglomeration is postponed but can still occur due to melted ash droplets.
  - Primary, the bed particle layer has a high melting elemental composition, but when the concentration of ash elements increases the melting temperature is lowered. The layer gets sticky with time and the agglomeration starts.

Since the ash consists of a mixture of inorganic compounds it doesn’t have a distinguished melting point, and therefore the melting process is protracted, starting at the initial deformation temperature. Large fluctuations of pressure and the incidence of temperature gradients in the bed are indications of initial agglomeration. Agglomeration leads to defluidization of the bed, resulting in expensive downtime of the boiler. [4]
3 Material and methods

3.1 Fuels

Four fuels have been used in the experiments in this project, two different coals and two biomass fuels. Wood pellets and straw (wheat) pellets were used as biomass fuels.

The coals were one Polish (Coal1) and one Russian (Coal2). Both coals were classified as sub-bituminous/bituminous according to Raask [9]. They were dried in an oven at 120 °C and then sieved to a size distribution between four and fifteen mm to mix well with the biomass pellets in the fuel feeder.

The four fuels and mixtures that were used for the combustion experiments are shown in table 3.

Table 3: Fuel mixtures (weight%) that were used in the combustion experiments.

<table>
<thead>
<tr>
<th>Fuel mixtures:</th>
<th>70%Coal1/30%Wood</th>
<th>70%Coal2/30%Wood</th>
</tr>
</thead>
<tbody>
<tr>
<td>100%Wood</td>
<td></td>
<td></td>
</tr>
<tr>
<td>100%Straw</td>
<td></td>
<td></td>
</tr>
<tr>
<td>100%Coal1</td>
<td>30%Coal1/70%Wood</td>
<td>30%Coal2/70%Wood</td>
</tr>
<tr>
<td>100%Coal2</td>
<td>30%Coal1/70%Straw</td>
<td>30%Coal2/70%Straw</td>
</tr>
</tbody>
</table>
Samples of the fuels were analyzed for characteristics and composition of the main ash forming elements, see figure 8-10.

**Figure 8**: Fuel characteristics for the used fuels, $D_s =$ dry substance and $\% =$ weight percent.

The two coals have higher heating value, lower oxygen level and contain more ash and carbon compared to the biofuels. The big difference in moisture content between the coals and the biofuels depends on the fact that the coals were dried from a moisture level of approximately twelve weight percent for Coal1 and ten weight percent for Coal2.
**Figure 9:** Composition of ash forming elements for the fuels in Wt% of Ds.

**Figure 10:** Fuel composition of ash forming elements for the fuels.
Composition of the ash forming components for the different fuel mixtures, figure 11-14, were calculated from the reference fuel data seen in figure 8-10 above.

**Figure 11:** Composition of ash forming elements for Coal1/Biofuel mixtures in Wt% Ds.

**Figure 12:** Composition of ash forming elements for Coal1/Biofuel mixtures.
Figure 13: Composition of ash forming elements for Coal2/Biofuel mixtures in Wt% Ds.

Figure 14: Composition of ash forming elements for Coal2/Biofuel mixtures.
3.2 Bed material experiments

The initial intension for the work was to use the produced coal ash during combustion as bed material to simulate the real process conditions in full scale fluidized bed boilers, operated by e.g. Vattenfall. To make proper bed material, the coals were therefore incinerated in a suitable furnace to produce ash that could be sieved to the right size distribution to be used as starting bed material. Two different methods were used for evaluating the possibility to produce bed material in a small scale furnace:

3.2.1 Electrical heated furnace

Coal1 and Coal2 were ashed in an electrically heated laboratory-scale furnace, figure 15, under a stoichiometric surplus of air. The temperature in the furnace was set to 700 °C during the experiment.

Figure 15: Electrical heated furnace used during the bed material experiment.

3.2.2 Grate simulation

The fluidized bed, described in chapter 3.3 and viewed in figure 16, was used without a fluidizing medium to simulate grate combustion conditions. Coal1 was ashed during a stoichiometric surplus of air and at 1000 °C.
3.3 Pilot scale FBC experiments

The combustion experiments were carried out in a pilot-scale FBC reactor (5 kW), figure 16. The reactor was made of stainless steel (253 MA). It was 2 m high and had a diameter of 100 and 220 mm in the bed and freeboard sections respectively. A perforated distributor plate with 90 holes and a total open area of 1% was used. [17] For each experiment 542 g un-sieved quartz sand was used as bed material (see results/conclusions from Bed material experiments chapter 4.1 and 5.1).

Figure 16: Pilot-scale Fluidized bed reactor; 1: Propane burner, 2: Distribution plate, 3: Primary air, 4: Secondary air, 5: View window and bed sampling location, 6: Fuel feeder, 7: Electrical wall heaters, 8: Deposition probe sampling location, 9: Cyclone, 10: Lambda probe, 11: In-Situ alkali chloride meter(IACM), 12: Impactor, 13: Fourier-transform infrared spectrometer (FTIR)/Testo.
All fuel mixtures and the reference fuels were combusted at a bed temperature between 850 and 900 °C, except for the 100% straw that had a bed temperature which varied from 670 to 700 °C due to previous knowledge of potential agglomeration problems with straw in general. The furnace walls were equipped with electrical heaters that provided isothermal conditions minimizing the risk of cold wall effects [18]. The electrical wall heaters were set to 850 °C in each experiment except for the straw fuel where 700 °C was used. During all the runs the primary air was set to 80 l/min, corresponding to approximate 10 times the minimum fluidization velocity. The secondary air was set to 10 l/min. The average oxygen level in the flue gas varied in the range of 8-11 %. In the end of each run, a controlled agglomeration test was performed. The fuel feeder was stopped to avoid the uncertainty in the bed temperature that burning particles provides [18]. The bed temperature was increased with 3 °C/min using electrical heaters until bed agglomeration occurred, or until the maximum temperature allowed by the equipment (i.e. 1050 °C) was reached. During the agglomeration test a propane burner was used to simulate the combustion atmosphere in the bed section during the experiment.

3.3.1 Sampling

**Bed material and flue gas cyclone ash (“fly ash”)**

Bed samples were taken with a cyclone sampler in the end of each run, just before the agglomeration test. The whole bed with possible agglomerates as well as the material in the flue gas cyclone (fly ash) was also taken care of after each run.

**Probe deposits**

During combustion, simulation of the deposition on super heater tubes were performed an air cooled probe (Stainless steel), with a length and diameter of 29 mm and 18.7 mm, respectively. The air flow was set to keep the probe temperature at 440 °C during all experiments and sample time was 4.5 hours for each fuel.

**Size fractionated flue gas particulate matter**

To determine the mass size distribution and concentration as well as chemical composition of each particle size, a 13-steps low pressure impactor (Dekati Ltd) was used, which separates the particles from 0.03 μm to 10 μm according to aerodynamic diameter, Aerodynamic diameter is the diameter of a sphere of unit density (1g/cc) that has the same gravitational settling velocity as the particle in question. The inlet temperature of the flue gas was approximately 200 °C and the impactor was heated to 140 °C with external heaters to avoid water condensation in the impactor, see figure 16 for sampling point. Three sets of substrate plates of aluminum were used for each fuel; one greased and two non-greased plates. A mini-cyclone with a cut off diameter of 1.8 μm
was placed before (in series with) the impactor when using non greased plates to collect coarser particles. The greased plates were primarily used to determine the mass size distribution and the non-greased primarily for chemical analysis purposes.

*Flue gases*

CO₂, H₂O, CO, NO, NO₂, SO₂, HCl, and CH₄ was measured with a Fourier-transform infrared spectrometer (FTIR) analysis instrument [17]. Later, another instrument using electrochemical sensors (Testo 350XL) was used instead, due to malfunction of the FTIR. The Testo instrument measures the concentrations of O₂, CO, NO and SO₂, see Testo/FTIR sampling points in figure 16. The concentration of O₂ in the flue gas was also continuously monitored with a lambda probe. To measure the gas phase concentrations of alkali chlorides (KCl and NaCl) as well as SO₂, a special kind of UV-spectrophotometer, i.e. IACM (In-Situ alkali chloride meter) was also used during all experiments, see figure 16 for sampling point. Table 4 shows which of the flue gas analyzing methods that were used in the different experiments.

**Table 4:** Gas measurement methods for each fuel/fuel mixture.

<table>
<thead>
<tr>
<th>Fuel blends</th>
<th>Testo</th>
<th>FTIR</th>
<th>IACM</th>
</tr>
</thead>
<tbody>
<tr>
<td>100%Coal1</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>70%Coal1/30%Wood</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>30%Coal1/70%Wood</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>70%Coal1/30%Straw</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>30%Coal1/70%Straw</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>100%Coal2</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>70%Coal2/30%Wood</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>30%Coal2/70%Wood</td>
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<td>100%Wood</td>
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<tr>
<td>100%Straw</td>
<td>X</td>
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</tbody>
</table>
3.3.2 Chemical analysis of ash samples, deposits and particulate matter

Chemical analysis of ash samples/agglomerates, probe deposits and particulate matter samples were performed with two methods, I) scanning electron microscopy (SEM), combined with energy dispersive X-ray spectroscopy (EDS) in order to determine the elemental composition and morphology (performed on bed ash agglomerates, fine mode (<1 μm) impactor samples and PM1.8 cyclone samples) and II) powder X-ray diffraction (P-XRD) to determine the crystalline phases (performed on bed ash/agglomerates, probe deposits, PM10 cyclone ash, PM1.8 cyclone ash and available fine mode impactor samples).

In addition, special analyses of the bed material were performed with ICP-AES/AAS/MS to determine the amount of e.g. alkali in the bed before the controlled agglomeration tests started. These analyses were performed by ALS Scandinavia in Luleå.
4 Results and Discussion

4.1 Bed material experiments

4.1.1 Electrical heated furnace

The total ash sample, 8 grams, and sieved fractions from combustion of Coal2 at 700 °C in a electrical heated furnace are shown in figure 17 and 18 below.

Figure 17: Total ash sample, some un-burnt, from Coal2 after combustion in an electric heated furnace.

Figure 18: Ash fraction size distribution of Coal2 sample.
Coal2 ash fractions are shown in Figure 19 below.

**Figure 19: Ash fraction size distribution of Coal2.**

Coal2 ash is like a soft powder, at least the size fractions below 200 µm which probably won’t stay in the bed because of too low terminal velocity, and therefore not suitable as bed material. The size fraction of quartz sand bed material normally used in the fluidized bed, used in the Pilot scale FBC experiments, lies between approximately 200-300 µm and taking that to account, less than 10% of the ash from Coal2 can be used.

Coal1 was also combusted in the electrical heated furnace under similar conditions as Coal2 which gave ash that was comparable to the ash from the Grate simulation experiment.
4.1.2 Grate simulation

The total ash sample, 102.1 grams, and sieved fractions from the combustion of Coal1 in the fluidized bed during grate combustions are shown in figure 20, 21 and 22 below.

**Figure 20:** Total ash sample, some unburnt, from Coal1 after combustion in the fluidized bed under grate conditions.

**Figure 21:** Ash fraction size distribution of Coal1 sample.
Figure 22: Ash from Coal1 sieved in different size distributions.

Coal1 ash sample fractions are shown in Figure 23 below.

![Figure 23: Ash fraction size distribution of Coal1 sample.](image)

Coal1 is quite stony and more like the quartz sand normally used in the FBC, and therefore a larger range of size distribution of the ash compared to Coal2 ash will fit as bed material. However, the size fraction between 200 and 300 µm that may be suitable as bed material correspond to approximately 10% for Coal1.

Both coals generated relatively small amounts on ash in size fractions suitable to be used as bed material. Thus, as mentioned earlier in the Method section (3), quartz sand was used as bed material in the following pilot scale experiments.
4.2 Pilot scale FBC experiments

4.2.1 Gaseous emissions

The gas data presented in Figure 24-26 below consists of a mean value of sample points during approximately thirty minutes of steady combustion. It should be noted that, as mentioned in chapter 3.3.1, different gas analysis equipment were used due to malfunction of the original equipment. This may obstruct the comparison of the different combustion experiments. HCl and NOx concentration data are only presented in four of the twelve combustion experiments since only the FTIR measures HCl and NOx.

![Figure 24: Mean (30 min) concentrations of gaseous emission components measured by FTIR (CO, NO\textsubscript{x}, SO\textsubscript{2} and HCl) and Testo 350XL (CO, NO and SO\textsubscript{2}). *=FTIR](image)

When comparing the flue gas analysis with the fuel analysis, see figure 8 in chapter 3.1, one can see that; there is a clear relation between fuel-S and SO\textsubscript{2} emissions, all NO\textsubscript{x} are fuel related and strongly correlated to the fuel content, and HCl is also fuel-Cl related, although influenced also by alkali chloride formation. It is evident that the comparable fuel-S contents in straw and Coal2 doesn’t correspond to the respective concentrations of SO\textsubscript{2} in the flue gases when combusting the pure fuels. The reasons for this may be that the high amount of alkali (K+Na, mainly K) in straw have reacted with the sulfur according to reaction R1 in chapter 2.2.1, and thereby forming K/Na-sulfates, preventing the sulfur to end up as SO\textsubscript{2} in the flue gas emissions.

The relatively low CO in all cases indicates rather complete combustion conditions, which also was confirmed by the fact that not visible “blackening” by soot on particle or deposit samples were seen in any case.
**Figure 25:** Mean (30 min) concentrations of gaseous emission components measured by FTIR (CO, NO$_x$, SO$_2$ and HCl) and Testo 350XL (CO, NO and SO$_2$). *=FTIR, **=Testo

When comparing the emission data in figure 25 and 26 above one can see that the SO$_2$ concentration in the flue gases are higher for Coal1 fuel mixtures compared to corresponding Coal2 mixtures, although a similar trend is seen for the two coals when admixing with the tested biomass fuels.

**Figure 26:** Mean (30 min) concentrations of gaseous emission components measured by FTIR (CO, NO$_x$, SO$_2$ and HCl) and Testo 350XL (CO, NO and SO$_2$). *=FTIR, **=Testo
4.2.2 Size fractionated flue gas particulate matter

The mass size distribution for particles in flue gases captured in the impactor when using greased substrate plates and without a mini cyclone (cut-off at 1.8 µm) is viewed in figure 27.

![Particle mass size distributions and concentrations](image)

**Figure 27:** Particle mass size distributions and concentrations for the different fuel mixtures.

The combustion of 100%Straw resulted in a high concentration of fine particles (<1µm), as expected from previous studies with straw. However, when mixing straw with the two coals the fine particle fraction was almost completely extinguished and instead the concentration of coarse (>1µm) particles dominates. Therefore a mini cyclone with a cut of diameter of 1.8 µm was used in an attempt to collect more fine particles for the chemical analyses also in these experiments.

When looking at the coarse particle concentration one can see that in all cases except the for the fuel mix 30%Coal2/70%Wood, the corresponding Coal2 fuel mixtures to Coal1 mixtures in general have a lower concentration of particles in the flue gases. Higher concentration of coal in the fuel mixes also gives higher concentration of coarse particles in the flue gases.
4.2.3 IACM

The IACM data viewed in figure 28 below consists of non-normalized data unlike the flue gas emission data presented above. The measurement uncertainty was high due to optical problems in the experimental setup.

![SO\textsubscript{2} and KCl+NaCl concentration in flue gas for the different fuel mixtures.](image)

The main conclusions from the application of IACM in the present experiments with co-combustion of coal with biomass can be summarized as:

- The concentrations of KCl and SO\textsubscript{2} for the mixtures were approximately as expected from mixing of the pure fuels.
- The only interactions (even though noisy KCl-signal) were:
  - Coal1 with 30% straw: No significant increase of KCl and SO\textsubscript{2} remained high, indicating KCl sulfation.
- Coal1 with 70% straw: KCl was increased and SO$_2$ was strongly reduced. Available sulfur was probably consumed and KCl+NaCl was thereby present in excess.

- One observation was that SO$_2$ was high and fluctuating which indicates fluctuations in fuel feeding or fuel homogeneity.

- There was some optical problems in the experimental setup that caused high noise level and measurement uncertainties.

4.2.4 Probe deposits

The grow rate of probe deposits for the different fuel blends are shown in figure 29 below.

![Figure 29: Deposit formation on probes for the different fuel mixtures.](image-url)
Fuel mixes containing Coal1 contributed to more probe deposits compared to the corresponding fuel mixes for Coal2. Coal1 blends containing straw gave more deposits than Coal1 blends containing wood at a higher ratio of coal also seems to result in more deposits. For Coal2, a low introduction (30%) of straw resulted in increased deposits, but at higher admixtures the deposits decreased. XRD analyses showed that the 70%Coal2/30%Wood deposits consist of the same material as the 30%Coal2/70%Wood, but was abnormally low. XRD analyses also showed that the high amount of 100%Wood deposits to approximately 40% constitutes of bed material. In addition, a significant amount of fine “dust”, presumably wood dust, was observed on the probe in this experiment. No obvious trend for both Coal1 and Coal2 fuel mixtures was seen.

Pictures of deposits probes are shown in figure 30 and 31 to give an example of how the deposits vary in appearance depending on fuels and mixing ratios.

**Figure 30:** (left); 100%Straw probe deposits, (right); 30%Coal2/70%Straw probe deposits.

**Figure 31:** 30%Coal2/70%Wood
Probe deposits of 100%Straw, shown in figure 30 (left) are, as seen, quite white to the color and has a hard texture which makes them complicated to remove from the probe. In contrast to 100%straw, 100%Coal (both Coal1 and Coal2), are more like a soft powder on the probe and the deposits easily comes off. When mixing the two coals with straw, a mix of hard and soft deposits appears on the probes, see figure 30 (right). Deposits on probes when mixing the coals with wood looks like 100%Coal deposits.

4.2.5 Agglomeration

A controlled agglomeration test, described in chapter 3.3, was performed in the end of each combustion experiment. 8 of the 12 combustion experiments reached the maximum bed temperature of 1050 °C without defluidization. The 4 experiments where defluidization occurred due to agglomeration were; 100%Wood, 100%Straw, 30Coal2/70%Straw and 30%Coal1/70%Straw.

In figure 32-35 below temperature and pressures for the 4 fuels/fuel mixtures that lead to total defluidization of the bed are shown as a function of time.

![Figure 32: Pressure difference and temperature until total defluidization of the bed for 100%Wood.](image-url)
Figure 33: Pressure difference and temperature until total defluidization of the bed for 100% Straw.

Figure 34: Pressure difference and temperature until total defluidization of the bed for 30% Coal2/70% Straw.
Figure 35: Pressure difference and temperature until total defluidization of the bed for 30%Coal1/Straw70%.

When comparing the temperature and pressure behavior for the four combustion experiments in figure 32-35 above, it is clear that during the 100%Wood and 30%Coal1/70%Straw experiments no major complications occurred, and the controlled agglomeration test was successfully performed. In the 100%Straw case, a bed collapse occurred before the controlled agglomeration test but the bed recovered and the test could be performed, this however points at the fact that the temperature during the run was on the verge. When 30%Coal2/70%Straw was combusted, total defluidization occurred right before the controlled agglomeration test was going to be performed, which could depend on to high combustion temperature but also on the increased amount of material in the bed.
Figure 36: Enlarged picture of the pressure difference and temperature during controlled agglomeration test for 30%Coal1/Straw70%.

A classical “text book” appearance of how a controlled agglomeration test should look like, shown in figure 36, to make the calculation of initial defluidization temperature easy and with a small standard deviation interval.
To accurately determine the initial defluidization temperature for each of the four experiments that agglomerated, a multivariate data analysis method using PLS modeling in the software Evince was used (figure 37). The model relates the two temperatures and the two pressures with each other as well as with time separately. Significant deviations of temperatures and/or pressures in the bed are strong indicators for initial agglomerations.

![Figure 37: Initial defluidization bed temperature including standard deviation for the four fuel mixtures](image)

In figure 37 above it can be seen that for these mixtures, the initial defluidization temperature, 860 ± °C for 100%Wood, 674 ± 14°C for 100%Straw and 845 ± 8 for 30%Coal2/70%Straw lies below or slightly above the lower value in the combustion temperature range, 670 °C for straw and 850 °C for the others. This means that the combustion took place at such high temperature that there probably were only a matter of time before total defluidization would occur. The fuel mix 30%Coal1/70%Straw had an initial defluidization temperature of 976 ± 4 °C which means that the combustion temperature interval of 850-900 °C should give no agglomeration problems.
Figure 38-39 shows the bed after total defluidization due to agglomeration for the four fuel mixes.

**Figure 38:** Part of the bed after agglomeration test, (left); 100%Straw, (right); 100%Wood.

**Figure 39:** Part of the bed after agglomeration test, (left); 30%Coal170%Straw, (right); 30%Coal270%Straw.
4.2.6 Bed and cyclone ash

During combustion of the reference (non-mixed) fuels, the bed weight changes at different rates. The difference in bed weight and the amount of collected cyclone ash are shown in figure 40 below.

![Figure 40: Difference in bed weight and weight of particles in the cyclone for used fuels.](image)

Coal1 had the largest bed growth with 56 g/h followed by coal2 with a bed growth at 36 g/h. The references of wood and straw showed small decreases in bed weight, probably because some of the bed particles stick to the inside of the furnace, i.e. a carry-over effect, thus leading to negative bed growth values.
The difference in bed weight and the amount of collected cyclone ash for Coal1 with the different mixtures of straw and wood are shown in figure 41 below.

![Graph](image)

**Figure 41**: Difference in bed weight and weight of particles in the cyclone for Coal1 mixtures.

When comparing the Coal1 reference and the different fuel blends of Coal1 during combustion, the result showed that an increase in Wood- and Straw content in the fuel reduced the bed growth. The reference of Coal1 had a bed growth rate of 56 g/h in comparison with the 70%Coal/30%Straw blend which had a growth rate of 30 g/h. The cyclone ash followed the same trend, an increase of biomass in the fuel blend decreases the amount of cyclone ash in the sampler. During combustion of the Coal1 reference, the increase in bed weight was 20 g/h, in comparison with the 70%Coal/30%Straw blend with an increase of 8 g/h.
The difference in bed weight and the amount of collected cyclone ash for Coal2 with different mixes of Straw and Wood are shown in figure 42 below.

![Graph showing difference in bed weight and weight of particles in the cyclone for Coal2 mixtures.](image)

**Figure 42:** Difference in bed weight and weight of particles in the cyclone for Coal2 mixtures.

The difference in bed weight during combustion of Coal2 showed the same trends as the Coal1 experiments when wood was used as additional fuel, i.e. an increase in wood content reduced the bed growth. During combustion of 30%Coal2/70%Straw fuel blend, the bed weight increase was unrealistically high according to total amount of ash considerations, probably because the combustion temperature was slightly above the initial defluidization temperature during the experiment. Such high temperature makes the bed sticky and might increase the amount of unburned particles in the bed contributing to the increase of the bed weight. The cyclone ash showed the same trends as the combusted fuel mixtures of Coal1, i.e. an increase of biomass in the fuel lowered the amount of cyclone ash.
The weight percent of alkali (K+Na) in bed samples are shown in figure 43 below.

**Figure 43:** Wt % of alkali (K+Na) in bed samples taken prior the agglomeration test.

When comparing the alkali contents in bed samples during combustion of the reference fuels, Straw had the highest alkali content followed by Wood, Coal2 and Coal1 in descending order. During combustion of the Coal/Straw mixtures, the result showed that a higher Straw content gave an increase of alkali in the bed sample.
4.2.7 Chemical analysis of ash fractions and deposits

In the following, a summary of the results from the SEM and XRD analyses are presented.

**SEM-EDS on bed agglomerates**

Analyses of bed material agglomerates were performed on material from the 4 experiments with 100%Wood, 100%Straw, 30%Coal1/70%Straw and 30%Coal2/70%Straw, respectively, i.e. the experiments where agglomeration occurred.

Some of the main SEM-EDS results for the agglomerates in the bed material (after agglomeration tests) are shown in Figure 44-47.

**Figure 44:** SEM images and elemental composition of agglomerated bed material from combustion of 100% Wood.
**Figure 45:** SEM images and elemental composition of agglomerated bed material from combustion of 100% Straw.

**Figure 46:** SEM images and elemental composition of agglomerated bed material from combustion of the mixture with 30% Coal1 and 70% Straw.
Figure 47: SEM images and elemental composition of agglomerated bed material from combustion of the mixture with 30% Coal2 and 70% Straw.

SEM-EDS on PM1.8 cyclone ash

Analyses were performed on samples from all fuels (pure and mixtures), except 100% Wood. In the following figures, some examples are given to illustrate the results.

Figure 48 SEM images and elemental composition of PM1.8 cyclone ash particles from combustion of 100% Coal1 (left) and 100% Coal2 (right).
**Figure 49:** SEM images and elemental composition of PM1.8 cyclone ash particles from combustion of the mixtures 70% Coal1 and 30% Wood.

**SEM-EDS on fine mode (<1 µm) particle samples**

Enough material in the impactor in this size mode was only collected for 100% Straw, and also for 100% Coal1, although in much smaller amounts.

From the analysis of samples from combustion of 100% Straw, it is evident that the fine mode particles consist mainly of K, Cl and S in a 5:3:1 relation (51 At% K, 29 At% Cl and 12 At% S). Since the particles are so small and the samples were treated as a bulk material in the analysis procedure, no images are shown for the sub-micron alkali particles.

The analysis of samples from combustion of Coal1 showed that the fine mode (although small amounts) was dominated by Ca and S.
The above results and observations from the SEM-EDS analyses combined with results from the XRD analyses, i.e. phase composition of the different ash/deposit fractions, gives a rather comprehensive information concerning the ash chemical aspects related to the present co-combustion experiments with coal and biomass. Altogether the information is rather complex, but in the following the main results and observations related to this are summarized:

Coal1 and Coal2:

- "Sand" (quartz and feldspars) is present in all ash fractions (except fine mode particles) of both the Coal1 and Coal2.
- "Clay type minerals" (illite, muscovite) are present in significant levels in cyclone ash (coarse particles) and deposits, especially in Coal1.
- A "surplus" of CaO and MgO exists in cyclone and deposits for both coals, although more pronounced for Coal2 where these phases also were observed in bed ash material.
- Significant amounts of the sulfur is captured as CaSO4 for both coals.
- In all cases involving coals, pure or in mixtures, significant number of “droplet-shaped” coarse particles (up to some 100 um) consisting mainly of Ca and Si, are found in cyclone ash. Normally Ca-silicates melts at rather high temperatures, higher than normally in FB combustion of biomass. Thus, the present findings indicate that the ash forming elements has been exposed to temperatures significant higher than 1100 °C, presumably locally in burning coal fuel particles.

Straw:

- All crystalline material in different ash fractions are dominated or characterized by high amounts of alkali chloride and sulfates.
- From the SEM analysis it is evident that the bed/agglomerate samples contains significant amounts of material that has been melted during combustion and quenched during cooling, thus resulting in glassy amorphous material. The analyses indicate that the bottom ash/agglomerate from straw combustion contains glassy potassium rich silicates that are recognized to cause bed agglomeration/slagging problems. However, it should be noted that in this case a crystalline potassium silicate was found in the cyclone ash (somewhat speculative identification though).

Wood:

- Beside sand, the various ash fractions are characterized as CaO and MgO or minerals derived from those phases, alkali chloride and sulfates and typical bottom ash high temperature silicate minerals as KAlSi2O6 (leucite), Ca3Mg(SiO4)2 (merwinite) and Ca2MgSi2O7 (åkermantite). Based on experiences from many previous investigations, this is a quite expected observation for woody ash features.
Coal1 and Coal2 with Wood:

- Compared to experiments with both pure coals, respectively, the two levels of wood admixtures show large similarities concerning the mineralogy of the various ash fractions. The reason is of course the low ash content of wood compared to the coals. From figure 8 in chapter 3.1 it is evident that admixing wood from an ash point of view mainly can be described as a dilution of the coal ash.
- In the mixtures with Coal1 and wood, the large Ca+Si droplets are present in the PM1.8 cyclone ash. Less of these particles were found in the mixtures with straw.

Coal1 and Straw:

- Straw has much higher levels of ash forming elements compared to wood (about 6 to 10 times higher). Thus some effects on the total ash chemistry are expected from straw on the coal1/straw mixtures.
- Higher levels of alkali sulfates are observed, especially for the 70% straw admixture. Compared with pure straw a striking feature of the mixtures is the absence of KCl in all ash fractions. This may be connected to the appearance of large amounts of leucite (in the 70% admixing case). The reaction between KOH (or KCl) and deteriorated (by high temperature) clay type minerals (see R2 above) is believed to be the reason behind the formation of leucite. This reaction is an analogue to the one between kaolinite (or meta-kaolinite) and KOH(g)/KCl under the formation of leucite or kalsilite, which is recognized as an effective potassium capturing reaction [19]. These phases, leucite and kalsilite, are relatively high melting compounds, that will contribute to enhance the initial bed agglomeration temperature.
- An interesting feature in the high level mixture is the formation of cristobalite (SiO2), which may be interpreted as that the abundant reactive silica in straw is "out-concurred" by the clay type minerals in the competition for alkali (gaseous or melted KOH/KCl). Thus, this silica is left over and will transform to the thermodynamically stable modification, which is cristobalite. Cristobalite is a highly refractory compound that not will contribute to any bed agglomeration tendency.
- The relatively high SO2 emissions in this case may be explained by higher amounts of sulfur and lower amounts of Ca in Coal1 compared to Coal2 as well as higher K-capturing capacity (clay type minerals) in Coal1.

Coal2 and Straw:

- The effect of alkali species reacting with clay type minerals is not as profound for Coal2 as it was for Coal1. One reason may be that the amounts of clay minerals are presumed to be lower in the Coal2. However, some leucite was observed, especially in the high level admixture. Thus, the alkali capturing is believed to be lower and, consequently, rather high level of potassium species (sulfates and chloride) was observed in the fly ash.
fractions of the 70% admixture. Also in these cases, the formation of (minor amounts of) cristobalite was observed.

- The relatively higher levels of CaO and MgO in the Coal2 ash are believed to have positive effects in preventing bed agglomeration tendencies, in that the melting temperatures of the formed potassium rich silicates are raised upon incorporation of these oxides.

- The relatively low SO2 emissions in this case may be explained by lower levels of sulfur and high levels of Ca as well as lesser amounts of clay minerals.
5 Conclusions

5.1 Bed material experiments

Ash from both coals could presumably be used as bed material. However, only a small fraction of the total ash was suitable for our lab scale combustion experiment. To produce the needed amount of bed material ash for the 12 combustion experiments more coal than available in this campaign would have been needed. Due to this fact, unsieved quartz sand was instead used as bed material in the pilot scale FBC experiments.

5.2 Pilot scale FBC experiments

The growth rate of bed material and cyclone material showed different behavior for the admixtures of wood and straw, respectively. When admixing wood to coal, it is clear that the total ash content in the fuel mixture determines, i.e. a question of “dilution” of the coal ash. When admixing straw to coal, however, a more complex and “non-ash content-related” association was seen which can presumably be explained by ash chemical processes that effects the capture/release of different ash forming elements in different ash fractions. This is clearly seen by the high increase in bed material growth for the 30%Coal2/70%Straw experiment.

During combustion, the alkali content of the bed samples increases with increasing admixing of wood and straw to the coals. The alkali concentration in the bed is highest for the fuel mixtures of straw and coal.

As seen in the fuel analysis of the pure coal fuels, Coal1 contains more sulfur than Coal2. Most probably, this contributes to the higher SO$_2$ concentration in the flue gas emissions for Coal1 fuel mixtures compared to Coal2.

The absence of fine mode particles in all experiments with mixtures of biomass and coals are of great relevance from a deposit formation point of view related to condensation of gaseous alkali species. Even at 70 wt-% admixing of straw to both tested coals, no significant fine mode was determined. It must, however, be stated that the interpretation of the deposit rate by the probe measurements in this work are strongly interfered by the deposition of coarse mode bed and ash particles.

Fuel mixtures containing Coal1 contributes to more probe deposits and higher concentration of coarse particles in flue gases, except the mixture of 30%Coal1/70%wood, compared to the corresponding fuel mixtures for Coal2. In general, higher concentration of coal in the fuel mixtures also gives higher concentration of coarse particles in the flue gases.
The results from the controlled bed agglomeration tests are most encouraging. For wood, that has an initial agglomeration temperature of around 850 °C, the agglomeration tendencies were in all cases involving mixtures with the coals, reduced considerable, i.e. no agglomeration was observed up to 1050 °C. For straw, that has a very low initial agglomeration temperature around 670 °C, an even more remarkable reduction in agglomeration tendency was observed. For the mixture with 70 wt-% of straw in Coal1 and Coal2 the initial agglomeration temperatures were raised with approximately 300 and 180 °C, respectively.

Some conclusions that can be drawn from the controlled bed agglomeration tests is that Coal1 compared to Coal2 is more suitable for the combustion conditions in these experiments since only the fuel mixture containing 30%Coal1/70%Straw led to total defluidization and that was with an initial defluidization temperature of 976 ± 4 °C that is well above the operational temperature of 850-900 °C. Coal2 on the other hand had an initial defluidization temperature of 845 ± 8 for the 30%Coal2/70%Straw mix which implicates that all mixes with Coal2 except this one is suitable for the operational temperature of 850-900 °C.

From the SEM/EDS and XRD analysis of the various ash fractions it was concluded that:

- Admixing high levels (up to 70% by weight) of wood to the two studied coals does not impose any major problems from bed agglomeration point of view.
- Two mechanisms for reduced bed agglomeration problems in co-combustion of straw with the present coals could be suggested: 1) reaction of alkali (from the straw) with clay type minerals in the coals under formation of rather refractory alkali alumino-silicates and 2) incorporation of the alkali earth metal oxides (CaO and MgO) into low melting alkali silicate melts, which otherwise is considered to promote bed agglomerations.
- Assumed high local temperature in the proximity of burning coal fuel particles results in different ash transformation processes e.g. melted Ca-silicates in coarse cyclone ash.

Thus, a detailed analysis of the ash mineralogy of a presumptive coal to be used in co-combustion with problematic bio-fuels is recommended prior to utilatization. Based on the mechanisms sketched above and possibly others, suitable matches could be suggested and unsuitable combinations be avoided. However, in order to determine “safe” admixing levels, experiments should be carried out.
In Table 4 below, a summary of the effects on different process aspects when introducing wood and straw to the two tested coal assortments, are given.

**Table 5: Summary of results: Coal1 fuel mixtures compared to Coal1 and Coal2 mixtures compared to Coal2. ↑ means strong increase, ↗ increase, ↓ strong decrease,↘ decrease, → unchanged and Ref =reference.**

<table>
<thead>
<tr>
<th>Fuel blends</th>
<th>Agglomeration temperature</th>
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<th>Cyclone ash growth</th>
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## Appendix

### Literature review

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<td>1</td>
<td>Ash effects during combustion of lignite/biomass blends in fluidized bed</td>
<td>D. Vamvuka et al, 2009</td>
<td>Fluidized bed(lab-scale) combustion and co-combustion. Empirical ash indices for low rank coal.</td>
<td>Bed material: A Na-feldspar NaAlSi3O8, average size 714 µm, was used as inert bed material. Additive: Bauxite.</td>
<td>Lignite, olive kernel and olive tree wood. Also blends of the coal with each biomass, 5, 10, 20%.</td>
<td>Investigate the thermal behavior of the ashes and determine the slagging/fouling and bed agglomeration tendency of these.</td>
<td>Overall the results imply that adding up to 20% wt to the lignite could reduce slagging/fouling and corrosion problems in comparison to firing each fuel alone which could provoke medium/high deposition problems. The same biomass material may be able to play different roles on the burnout yields of the tested lignites. The blends can be co-combusted in the existing systems in the blending ratios. Although blending led to some decreas in the burnout.</td>
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<td>2</td>
<td>Effect of biomass on burnouts of Turkish lignites during co-firing</td>
<td>H. Haykiri-Acma, S Yaman, 2009</td>
<td>Non-isothermal thermogravimetric analysis method using a shimadzu TG 41 thermal analyzer with a cylindrical alumina crucible.</td>
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<td>Turkish lignites: Yozgat-Sorgun, Erzurum-Askale, Tuncbilek, Gediz and Afsin Elbistan. Sunflower seed shell (0, 10, 20% wt).</td>
<td>Addresses the burnout characteristics of low rank Turkish lignites with woody shells of sunflower seed under non-isothermal co-combustion conditions.</td>
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<td>3</td>
<td>Principle, calibration, and application of the in situ alkali chloride monitor</td>
<td>Christer Forsberg, Markus Broström et al, 2009</td>
<td>In situ alkali chloride monitor (IACM).</td>
<td></td>
<td></td>
<td>This paper describes the principle of the IACM as well as its calibration. Measuring gaseous alkali chlorides(KCL and NaCl) and SO2.</td>
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<td>Application of chemical fractionation methods for characterization of biofuels, waste derived fuels and CFB co-combustion fly ashes</td>
<td>Anita Pettersson, Maria Zevenhoven et al, 2008</td>
<td>Chemical fractionation. CFB boiler 12 MWth, Chalmers.</td>
<td>Quartz sand, 99%.</td>
<td>Two digested municipal sewage sludge, Sweden. Straw pellets, RDF pellets and as reference fuels coal and wood pellets. Combustion tests: 79/21 wt% wood/straw, 67/21/12 wood/straw/sewage sludge, 45/14/11/30 RDF. Pure fuels.</td>
<td>In this work, two sewage sludges and a pelletized RDF, as well as coal and biofuel already present in the database, were investigated by the chemical fractionation method. Chemical fractionating is a good predictor of sintering tendencies of coals and most biofuels and mixes of these fuels, estimating the amount of reactive alkali and chlorine. K, Na, Ca and Mg were found in more stable forms in the fly ash, unlike S and to some extent Cl that were more stable prior combustion. When co-firing rapeseed with coal a decrease in CO emissions can be established. The NOx emissions increased with the ratio of rapeseed in the fuel mixture. The fouling rate on the deposits probe increased with the rapeseed mixture in the fuel. About 20% of alkalis were trapped in the bed. No obvious heavy bed sintering or deposits were noticed. These mid-term tests proved successful co-firing of up to 25% rapeseed cake with coal with and without limestone. Longer term effects on boilers need study.</td>
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<td>5</td>
<td>Deposits and emissions during co-combustion of biodiesel residue with coal and biomass in a CFB pilot</td>
<td>Nevalainen H, Leino T et al, 2008</td>
<td>Circulating fluid bed (pilot-reactor 50kW), co-combustion.</td>
<td>Rapeseed, wood chips and bituminous coal. 13/22/33% of rapeseed blended with coal.</td>
<td>Study the emissions, the agglomeration of the bed material and fouling tendencies when co-firing rapeseed and wood with bituminous coal.</td>
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The paper reviews some of the recent progress in our understanding of the fate and behavior of ash forming matter. Ash behavior in boilers using more than one fuel simultaneously may cause additional challenges. Ashes from different fuels may interact with each other. This can lead to surprising non-linear behavior of the ash behavior as function of fuel mix.

Aluminum-silicates found in coal ashes have shown to remove a great deal of the alkali vapors that contribute to deposits on boiler tubes.

Melting characteristics of ash fractions.

Investigates the co-combustion characteristics of peach and apricot stones mixed with lignite coal.

The percentage of apricot stones or peach stones in the fuel mixture should be below 50% in order to obtain the emission limits of EU. High concentrations of Na2O and K2O in the ash, which may cause fouling and corrosion of heat exchange surfaces.
12 Emissions during co-firing of RDF-5 with bituminous coal, paper sludge and waste tires in a commercial circulating fluidized bed co-generation boiler

Hou-Peng Wan, Ying-Hsi Chang et al 2007

Commercial CFBB, 103 MWh and 27 Mwel.

Bed material: silica sand, mean diameter of 0.18 mm. Additives: limestone.

A fuel blend composed of: bituminous coal, paper sludge, waste tires and RDF-5 in different ratios.

Investigate the feasibility of co-firing RDF-5 in a commercial boiler. The properties of ash, bottom ash and pollutants for various fuel ratios are analyzed and discussed.

Results show that Sox and NOx emissions decreased as The ratio of RDF-5 increased, but that CO, LOI, dioxin emissions and Cl content in the fly ash and bottom ash showed a reverse tendency. Co-firing in fluidized beds is to prefer before pulverized fuel boilers. When co-firing a alkali rich fuel with coal. The coal contributes with mitigating the alkali related deposit problems.

13 Co-combustion - a summary of technology

B. Leckner, 2007

Co-combustion in CFB and PF boilers.

Summarizes different technologies, problems and synergy effects when co-firing in CFB and PF boilers.

14 Effect of coal and biofuel with sewage sludge on alkali problems in a circulating fluidized bed boiler


12 MWth circulating fluidized bed, CFB, boiler. Chalmers.

Additives: limestone, chlorine (PVC), hydrated lime. Bed material: quartz sand.


The purpose of the present work is to study alkali-related problems during co-firing of coal and biomass with sewage sludge in a fluidized bed.

Co firing coal and an alkali-rich biofuel decreases the risk for agg and lowers conc. of KCl in the combustion gas compared to firing only bio. Deposits do not contain chlorine. Co firing of coal and/or an alkali-rich bio completely eliminates KCl in the combustion gas and deposits. Co-firing enables biofuels rich in alkali metals and chlorine compounds to be used in coal-fired boilers while avoiding or controlling corrosion and ash deposition problems.

15 Co-firing—a strategy for bioenergy in Poland?

Karin Ericsson, 2006

Pulverized fuel boilers, grate fired and CFB mm

Analysis of the strengths and weaknesses of co-firing biofuels in Poland with respect to technical, environmental, economical and strategic considerations.
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<td>Co-firing coal with rice husk and bamboo and the impact on particulate matters and associated polycyclic aromatic hydrocarbon emissions</td>
<td>Christopher Y.H. Chao et al, 2006</td>
<td>Laboratory-scale pulverized fuel combustion testing facility.</td>
<td>Coal, rice husk and bamboo. 0, 20, 30, 50 and 100% biomass of total mass.</td>
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<td>The potential of co-firing coal with rice husk and bamboo. Focusing on particle and the associated PAH (Polycyclic Aromatic Hydrocarbons) emissions.</td>
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<td>17</td>
<td>Experimental investigation of fluidized bed co-combustion of meat and bone meal with coals and olive bagasse</td>
<td>L. Fryda, K. Panopoulos et al, 2006</td>
<td>Laboratory scale bubbling fluidized bed combustor.</td>
<td>Bed material: Silica sand, 99% SiO2, mean diameter 552 µm. Traces of Al2O3.</td>
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<td>Three primary fuels: high rank Colombian coal, low rank Greek lignite and olive bagasse. MBM (meat and bone meal). 90/10 and 80/10 lignite/MBM, 90/10 and 80/20 Colombian/MBM, 80/20 and 60/40 Olive bagasse/MBM.</td>
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<td>Fuel blends were tested under different combustion conditions to study how primary fuel substitution by MBM affects fuel gas emissions as well as FB agglomeration tendency.</td>
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<td>18</td>
<td>Gas phase alkali chlorides and deposits during co-combustion of coal and biomass</td>
<td>H. Kassman, C. Andersson et al, 2006</td>
<td>Co-combustion in Chalmers 12MW circulating fluidized bed. IACM and SEM-EDX(Analysis of deposits).</td>
<td>Bed material: Polish bituminous coal with high chlorine content and straw. Reference fuel: mixture of wood chips, wood pellets and straw pellets. Three tests: 100% coal with increasing lime supply. Constant mixture of coal/straw with increasing lime supply and coal/straw with constant limestone supply and increasing straw fraction.</td>
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<td>Investigates the possibility of co-firing a fuel with high alkali content such as straw with coal. The Focus was on alkali related problems and the synergy effects of co-firing coal and straw.</td>
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<td>When co-firing coal and straw pellets the concentration of KCl increased. For all tests using coal, high concentrations of Al and Si where found in the deposits. This is a hint of the presence of alumino-silicates that can capture alkali. By adding limestone to the fuel mixture a capture of Cl forming CaCl2 is gained.</td>
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<td>Fouling tendency of ash resulting from burning mixtures of biofuels. Part 1: Deposition rates</td>
<td>Mischa Theis, Bengt-Johan Skrifvars, Mikko Hupa, Honghi Tran, 2005</td>
<td>Lab-scale entrained flow reactor that simulates conditions in the superheater region of a recovery boiler. Finnish peat, oat straw and eucalyptus bark. Pure, peat/straw and peat/bark mixtures. Peat 100-0%. The objective was to establish a lab-scale procedure that can be used to evaluate the deposition behavior of biofuel mixtures with different fouling propensities over the whole mixing range. Focus on equipment setup and recording of deposit rates. The deposition behavior for both mixtures followed a non-linear pattern. The results indicate that it is possible to burn up to 30 wt% bark and up to 70 wt% straw in mixtures with peat without encountering increased deposition rates. Experiments in pilot plants to evaluate would be useful. Gaseous alkali species depends on fuel composition. It is likely that alkalis will condense on flue gas pipe walls, on the bed material and on fly ash particles or/and form aerosol particles in the flue gas. Alkalis that are bound in the minerals in coal are not volatile and will therefore remain in the ash. Fine particles re mostly composed by KCl and K2SO4. When adding SO2 gas or co-firing KCl disappeared from the particles and more K2SO4 is formed. These results agree with an aerosol formation mechanism assumed earlier.</td>
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<td>Alkali metals in circulating fluidized bed combustion of biomass and coal: measurements and chemical equilibrium analysis</td>
<td>Michal P. Glazer et al, 2005</td>
<td>Combustion and co-combustion in a circulating fluidized bed, test rig 25kW. Equilibrium modeling, Factsage. Bed material: standard silica sand, 0.3-0.6 mm. 4 types of straw from Spain: wheat Marius, rape, Brasica Carinata and maize. Columbian hard coal. 100% straw, 100% coal, 20/80 straw/coal and 50/50 straw/coal. The influence of operating conditions and fuel compositions on the release of alkali compounds to the gas phase was researched.</td>
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<td>Effect of co-firing on the properties of submicron aerosols from biomass combustion</td>
<td>Santiago Jiménez, Javier Ballester, 2005</td>
<td>Downfired entrained flow reactor (EFR), with settings so the fuel particles burn in combustion condition similar to pulverized fuel boilers. Addings: SO2. Spanish anthracite coal, coke from the petroleum refining process and Orujillo. Co-combusting 2/3(mass basis) Orujillo with coke and 2/3 Orujillo with coal. Also 100% Orujillo. The effect of combining biomass with sulfur rich fuels, coke and coal, on particulate emissions.</td>
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<td>Fluidized bed combustion of refused-derived fuel in presence of protective coal ash</td>
<td>Eduardo Ferrer et al, 2005</td>
<td>CFB (Small scale, 100kW and 4MW), Co-combustion, XRF (Ash analysis)</td>
<td>Bed material: Natural sand (0.1-0.3mm). Additives: Limestone. Coal Polish and South African. RDF pellets. Pure and co-combustion of RDF (0-68%) with 1 coal at the time. Evaluate different ways to raise the fuel contents of RDF up to 40% without high risk of corrosion and Cl deposition. Aluminosilicates and sulfur in coal ash can act as protective compounds, binding alkalis from alkali chlorides to form alkali aluminum silicates and alkali sulfates. Cl -&gt; HCl which does not concentrate to deposits.</td>
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<td>Biomass-coal co-combustion: opportunity for affordable renewable energy</td>
<td>Larry Baxter, 2004</td>
<td>Coal, subbituminous and bituminous. Biomass, mainly herbaceous. Switchgrass.</td>
<td>This document discusses recent progress in: corrosion, fly ash utilization, impacts on SCR systems and formation of striated flows. There are many issues with co-firing biomass and coal. Results to date indicate that they are all manageable with careful consideration of fuels, boiler operating condition and boiler design. To know more about the behavior when burning fuel mixtures, the analysis methods should be improved. Without good understanding of mixed-firing, the pilot plant result is hard to transfer to full scale.</td>
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<td>Interactions of fuels in co-firing in FBC</td>
<td>Mikko Hupa, 2004</td>
<td>Brown coal, bituminous coal (0-100% mixture with wood) and biomass based fuels</td>
<td>Review of recent research work in the multi-fuel firing in FBC. Investigating the devolutilisation behavior during co-pyrolysis. Conflicting behavior is exhibited depending upon pyrolysis technique.</td>
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26 Evaluation of the influence of biomass co-combustion on boiler furnace slagging by means of fusibility correlations
Marek Pronobis, 2004
Fusibility calculations.
Two Upper Silesian coal bituminous (with lower-LS and higher-HS slagging inclination), straw, wood, dried sewage sludge and bone meal.
The ash properties when co-firing coal and biomass are investigated using fusibility correlations.
In biomass co-firing boilers it is preferable to avoid coals with high slagging inclination. Co-firing of LS coal and wood produces slagging and fouling conditions.

27 Importance of coal ash composition in protecting the boiler against chlorine deposition during combustion of chlorine-rich biomass
Martti Aho, Eduardo Ferrer, 2004
Co-combustion in a circulating fluidized bed (pilot-scale 0.1MW). Ratios.
Bed material: Natural sand, particle size 0.1-0.3 mm, mean diameter 0.20 mm. Additive: Limestone.
German coal with 20/30/40% of MBM, South African coal with 30/40% of MBM and Polish coal with the same fuel mixtures as SAC.
Investigate which of the coals, SAC, PC and GC are the most protective coal so that the portion of MBM is maximized.

28 Demonstration of plasma excited atomic resonance line spectroscopy for on-line measurement of alkali metals in a 20 kW bubbling fluidized bed
Ville Häyrinen, Rolf Hernberg, Martti Aho, 2003
Bubbling fluidized bed (20kW), Co-combustion.
Bed material: Natural sand, average diameter: 0.33 mm.
Pine bark mixed with South African coal and Finnish forest residue mixed with Polish lignite.
To investigate the applicability of the PEARLS method in high ash conditions and to assist in better knowledge of alkalis behavior when co-combusting biomass and coal.
The PEARLS system was successfully demonstrated in high-ash BFB conditions.
Concentrations of alkalis measured by the PEARLS method decreased with increasing share of coal and lignite in the fuel mixture.

29 Assessment of chlorine-alkali-mineral interactions during co-combustion of coal and straw
Xiaolin Wei, Christian Lopez at al, 2001
Equilibrium analysis tool FACT-win. The calculations are based on measurement data from pulverized co-combustion, test rig 0.5 MW.
Bituminous hard coal "Göttelborn", German, fired with regular wheat straw (0-100%), German. Lignite ("brown coal"), Rhenish area German and "DK chlorine-rich" straw (0-100%), Danish.
To investigate the effect of inherent minerals in blended fuels on the release and retention of chlorine and alkali metals.
According to the measurements very low amounts of Cl and S are kept in the bottom, air preheater and cyclone ash. In bag filter the Cl and S contents are relatively high.
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<td>K. Laursen, J. R. Grace, 2001</td>
<td>Fluidized bed co-combustion.</td>
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<td>J. Werther, M. Saenger et al, 2000</td>
<td>Overview paper that discusses various issues related to combustion of agricultural residues.</td>
<td>Mostly biomass, 13 different. Some cofiring of straw with coal, brown and bituminous (black).</td>
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<td>Biomass cofiring: the technology, the experience, the combustion consequences</td>
<td>D.A Tillman, 2000</td>
<td>Overview. Pulverized coal boilers, cyclone boilers, T-fired boiler, FB, spreader-stoker, gasification.</td>
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<td>Investigation of slagging in pulverized fuel co-combustion of biomass and coal at a pilot-scale test facility</td>
<td>T. Heinzel, V. Siegle, H. Spliethoff, K.R.G Hein, 1998</td>
<td>Coal, straw, beech, oats and sewage sludge. Investigates the slagging and fouling tendency when co-combusting biomass and coal. Co-combustion of coal with 25% biomass, did not cause fouling and slagging problems on heat exchange surfaces. The melting temperatures of the mixed fuel are lowered in comparison to coal ash. PF and FBC are well suited for co-combustion of biomass and sewage sludge provided that the fuel feeding system is converted to fit the fuel. By optimizing the sulfur retention when co-combusting high sulfur fuels with biomass fuels, a better than conventional methods was obtained.</td>
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<td>Optimization of sulfur retention in ash when co-combusting high sulfur fuels and biomass fuels in a small pilot scale fluidized bed</td>
<td>Anders Nordin, 1993</td>
<td>Mixture of peat and an average wood residue(Pelletized). Co-combustion of coal and lucerne. Optimize the sulfur retention process in a FBC reactor, to determine the effect of different operating parameters on the retention, and to quantify possible deposit formations.</td>
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