Pilot-scale combustion studies with kraft lignin in a powder burner and a CFB boiler

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ABSTRACT: Processes have been developed to produce a solid biofuel with high energy density and low ash content from kraft lignin precipitated from black liquor. Pilot-scale tests of the lignin biofuel were carried out with a 150 kW powder burner and a 12 MW circulating fluidized bed (CFB) boiler. Lignin powder could be fired in a powder burner with good combustion performance after some trimming of the air flows to reduce swirl. Lignin dried to 10% moisture content was easy to feed smoothly and had less bridging tendencies in the feeding system than did wood/bark powder. In the CFB boiler, lignin was easily handled and cofired together with bark. Although the filter cake was broken into smaller pieces and fines, the combustion was not disturbed. When cofiring lignin with bark, the sulfur emission increased compared with bark firing only, but most of the sulfur was captured by calcium in the bark ash. Conventional sulfur capture also occurred with addition of limestone to the bed. The sulfur content in the lignin had a significantly positive effect on reducing the alkali chloride content in the deposits, thus reducing the high temperature corrosion risk.

Application: This research is of interest for pulp and paper and energy and utilities companies that want to understand how kraft lignin can be used to replace fuel oil or coal in many combustion applications.

One way of exploiting the energy surplus of a modern kraft market pulp mill is to extract lignin from black liquor. Lignin extraction has the additional advantage of providing incremental capacity in the chemical recovery area, and can thus be used to off-load the recovery boiler or to avoid an expansion when pulp production is increased. For such a concept to be feasible, the extracted lignin must be handled and fired in combustion equipment other than conventional recovery boilers, such as lime kilns and other applications where powder burners are common, or in fluidized bed boilers frequently used for biomass combustion.

Methods to process kraft lignin precipitated from black liquor and to produce a solid biofuel with high energy density and low ash content have been developed in research programs. The process concept is now being verified on a scale of about 4,000 metric tons/year at a demonstration plant in Bäckhammar, Sweden (**Fig. 1**), operated by LignoBoost AB. As part of the ongoing FRAM2 (future resource-adapted pulp mill) Swedish national research program, several full-scale combustion trials will be carried out to evaluate the effects of firing the lignin biofuel in various types of boilers and in lime kilns.

In preparation for the full-scale trials, tests were carried out at pilot scale in a 150 kW powder burner at Energy Technology Center (ETC) in Piteå and in a 12 MW fluidized bed boiler at Chalmers University of Technology in Göteborg. Approximately 300 kg of lignin powder was fired in the tests with the powder burner. The lignin was first dried and milled to give a particle size below 1 mm. A mixture of bark and wood powder was used as the reference fuel. About 3 metric tons of lignin was fired during a 3-day trial in the circulating fluidized bed (CFB) research boiler at Chalmers. The lignin was cofired with bark pellets, which was also used as the reference fuel. The objectives of the tests were to study fuel feeding properties, effects on emissions (carbon monoxide (CO), sulfur dioxide (SO_2), and nitrogen oxides (NO_x)), fireside deposits on heat transfer surfaces, in-bed sulfur capture with limestone addition, and effects on the sintering properties of the bed material.

1. Interior view of the demonstration plant, showing the chamber filter press used for dewatering and washing of the lignin.

METHODS AND MATERIALS

Lignin prepared at the demonstration plant in Bäckhammar was used for combustion tests in the powder burner, together with a reference powder (**Table I**). Filter cake with an original moisture content of about 30%–40% was air dried to a moisture content of 18%. It was then milled in a RAN 70N grater/shredder (Alexanderwerk) using a working cylinder with 1.5 mm holes. Some of the lignin was further air dried to a moisture content of 10% at ETC. The lignin powder was finer than the reference fuel, with no particles larger than 1 mm and 85% of the particles smaller than 250 µm. In the reference fuel, 4.2% of the particles were larger than 1 mm and 57% were smaller than 250 µm. About 300 kg of lignin powder was fired in the tests.

Fuel		Lignin powder	Bark/ wood powder
C	% of dry solids	65.3	49.3
H		5.8	6.1
N		0.1	0.3
S		1.8	0.06
O		25.6	42.6
Ash		1.3	1.6
Lower heating value	MJ/kg fuel (wet basis)	22.9	18.8
Moisture content	% of fuel	10	5

I. Fuels used in the powder burner trial.

Fuel		Lignin filter cake*	Bark pellets	15% lignin + 85% bark
Moisture content	%	29.3	10.3	12
Higher heating value	MJ/kg dry solids	26.3	21.0	21.7
Lower heating value	MJ/kg fuel	17.0	17.7	17.7
Ash	% of dry solids	1.4	3.6	3.3
C		62.7	51.9	53.3
H		5.7	6.1	6.1
O		27.6	37.9	36.5
S		2.5	0.04	0.4
N		0.15	0.4	0.4
Cl		<0.01	0.02	0.02
Ca	mg/kg dry solids	1085	9324	8253
K		543	2138	1931
Na		1666	310	486
Na+K	mmol/kg dry solids	86	68	71
Na/(K+Na)	mole ratio	0.8	0.2	0.3
Ca/S		0.03	18.7	1.7
S/Cl		>278	2	22
Theoretical SO_2	ppm at 6% O_2	2100	41	332

* Note that the lignin data is based on one sample only and is higher with respect to ash content than the average of all the large bags used in the trial.

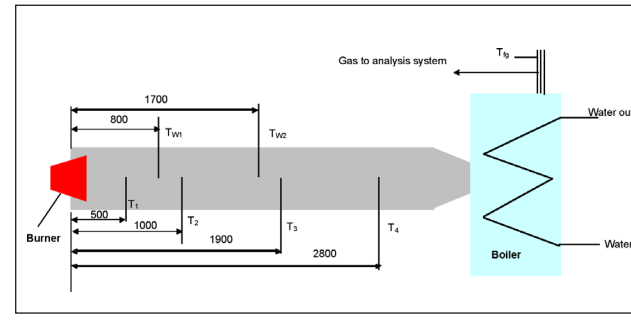
II. Fuels used in the circulating fluidized bed boiler trial.

We used bark pellets as the base fuel in the CFB boiler trial. The lignin fuel was delivered as broken filter cake in large bags. The lignin had been produced in a pilot plant trial in an earlier project leading up to the construction of the demonstration plant, and moisture content (9%-34%) and ash content (0.5%–1.4%) varied. To even out the differences, the lignin with high moisture content was fed together with lignin with low moisture content. **Table II** shows the fuel characteristics for the bark pellets and the lignin fuel.

The free-flame burner (VärmeTeknisk Service AB (VTS), Nyköping, Sweden) has a refractory lined cone at the front of the burner. The combined oil-powder burner is dimensioned for a maximum power of 150 kW. The powder nozzle is in the center and a conical mass in front of the powder nozzle is used as a distributor. This mass was removed during the last two combustion tests with lignin. The burner is equipped with three air inlets: primary, secondary, and tertiary. All the inlets are formed to give a rotational movement to combustion air and form a swirl to establish good mixing between the fuel and air.

The burner fires into a 3.3 m long combustion channel with a rectangular cross-section of 550×550 mm². The channel is made of 3 mm iron plates and has four layers of insulation materials. We measured four gas temperatures (T_1–T_4), the flue gas temperature (T_{fg}), and two wall temperatures (T_{w1},T_{w2}) in the combustion channel. The inner wall temperature was measured by thermocouples placed inside the refractory and about 20 mm from the inner wall of the combustion chamber. **Figure 2** shows the positions of the thermocouples. The fuel nozzle of the powder burner is about 300 mm inside the channel.

A screw feeder was used to deliver the powder. The flow

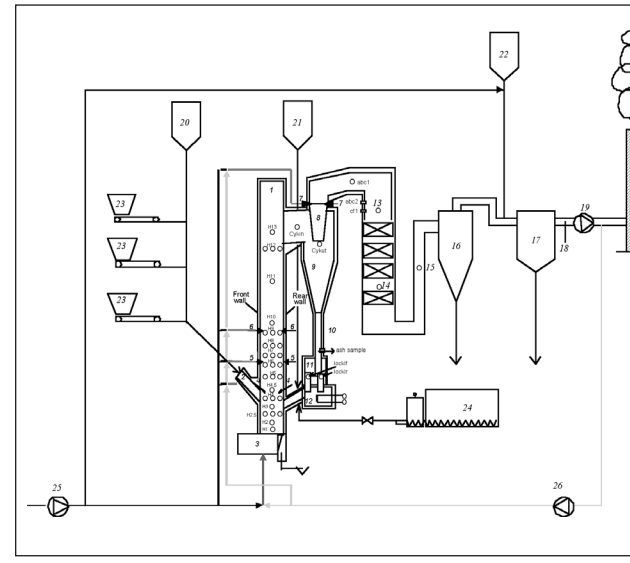


2. Schematic view of the combustion furnace with a 150 kW powder burner. The figures (500,800) are referred to the distance between the position of the thermocouples relative to the outlet of the fuel nozzle.

rate of the powder was measured by calibration of the screw feeder and by continuous weighing of the feeding bin. The air flow was measured by a mass flow meter. The composition of the flue gases was measured with respect to oxygen (O_2), CO, and carbon dioxide (CO_2). NO_x was recorded by an emission analyzer (Testo, Flanders, NJ, USA; 350 XL). The powder-particle mass size distributions were determined by collecting size-classified particle samples with a 13-stage low-pressure cascade impactor (Dekati Ltd., Tampere, Finland).

The powder combustion was started when the wall temperatures reached about 900ºC, after preheating with the oil burner. The temperature and the composition of flue gases were measured continuously during the tests. Total dust in the flue gases and the size distribution of the dust particles were measured when stable temperatures of the gas and wall were obtained.

The CFB boiler at Chalmers was built for research, but has all the features of a commercial unit (**Fig. 3**). The combustion chamber (1, in Fig. 3) has a cross-section of 2.25 m and a height of 13.6 m. Fuel is fed to the bottom of the furnace by a fuel chute (2). The circulating bed material is separated in the primary cyclone (9) and returned back to the combustion chamber. Primary air is supplied through the air plenum located below the bottom of the combustion chamber where the fluidizing nozzles also are found. Secondary air was supplied during the present tests through an air register (4) 2.1 m above the bottom. After the combustion chamber, the flue gases enter the convection path, where the gases are cooled down to 150°C. Fly ash is separated from the flue gas in two stages, first by a secondary cyclone (16) and finally by passing a bag house filter (17).



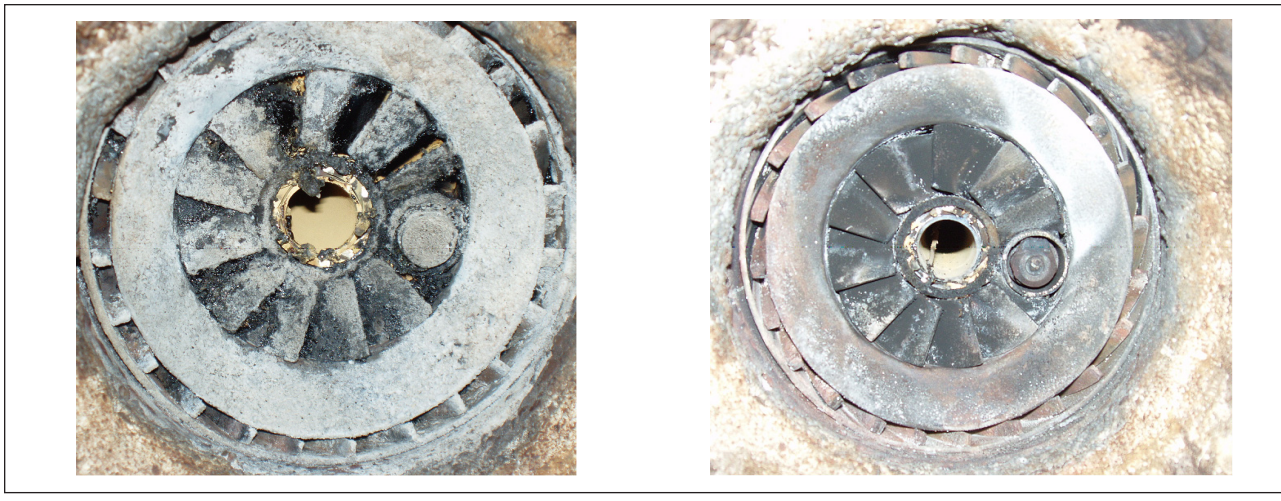
3. Flow diagram of the 12 MW circulating fluidized bed boiler.

NO_x, SO_2, and CO emissions were measured at a position after the bag house filter (18). Ash samples were taken from the bottom bed in a position named "H2" (Fig. 3) and from the particle return leg (10). Fly ashes were taken from the secondary cyclone (16) and from the bag filter (17). To measure gaseous alkali chlorides, an in-situ alkali chloride monitor [1,2] was situated before the convection section. Deposits also were collected with a cooled probe (500ºC) inserted after the cyclone (flue gas temperature = approximately 800ºC)

The laboratory unit used to study bed agglomeration at SP Swedish National Testing and Research Institute consists of a steel reactor (inner diameter = 0.07 m) surrounded by an electrically heated oven. Air is used as fluidization gas during the combustion step. When an appropriate quantity of fuel has

Fuel	Bark/wood powder		Lignin powder		
Test number	**R1**	**R2**	**L1**	**L2**	**L3**
Fuel flow, kg/h	20.8	20.6	20.8	19.8	20.0
Fuel moisture content, %	5	5	18	18	10
Burner thermal power					
(lower heating value), kW	109	108	124	118	119
Air flow, L/min	1280	1055	1530	1550-1950	2100
Air temperature, °C	Room temp.	360 (tertiary air)	360 (tertiary air)	Room temp.	Room temp.
O_2, %	3.5	5.4	N.A.	3.8	2.8
NO_x, ppm	111	67	N.A.	139	110
CO, ppm	34	433	N.A.	298	61
Dust, mg/ Nm^3	237	312	N.A.	679	396
Average gas temp, °C	1,175	1,121	N.A.	1,228	1,214
Test duration, h	7.0	5.1	0.6	4.8	5.0

III. Powder burner tests conditions and results.



4. The figure at left shows the burner after test L2, where there was recirculation of lignin powder toward the hot burner surfaces. At right, some soot can be seen on the vanes of the secondary zone in test L3, but no clumps of char on either the primary or the secondary zone. The refractory lined cone is not shown in this figure.

been combusted, the fuel feeding is stopped and the temperature of the reactor is reduced to 750ºC. During the agglomeration stage, the fluidization gas is changed to a mixture of 85% nitrogen (N_2) and 15% CO_2. At the same time, the temperature is increased by 3.5ºC/min. The pressure in the fluidized bed and the bed temperature is continuously measured. When the bed agglomerates, partly or totally, the pressure decreases; the temperature at which this happens is defined as the agglomeration temperature. The accuracy of the agglomeration temperature is estimated to ±10ºC.

RESULTS

Two tests (R1 and R2) were carried out with the wood/bark powder reference fuel. Three tests were carried out to burn kraft lignin powder (L1, L2, and L3). **Table III** shows the main results from the tests.

Tertiary air was preheated to about 360ºC in test R2, with the objective of simulating the temperature of secondary air in a lime kiln. Stable wall and gas temperatures were achieved after about 2 h. The temperature of the gas reached 1,100ºC–1,200ºC (maximum 1,223°C). Wall temperatures were lower (average 1,071ºC). NO_x emissions were about 111 ppm during R1 and about 70 ppm during R2. NO_x emissions were almost stable in both tests.

Carbon monoxide emissions were very low during R1, but less stable and much higher in R2 (average of 433 ppm). During R2, the air flow was lower, to reduce the O_2 content in the flue gases to 2%–3%, again to simulate the situation in the lime kiln. The result of this reduction was that there were more fluctuations in O_2 and CO concentration during R2 than R1, resulting in higher average O_2 and CO concentrations. This variation is related mostly to differences in the feeding rate of the fuel. In earlier tests on wood powder in the same equipment, O_2 content less than 3% in the flue gases gave high CO content, possibly because lower air flow causes less swirl and leads to poorer mixing in the burner.

Test L1 was carried out with a lignin powder with a moisture content of 18% and under the same conditions as in test R2. The test was stopped after only 35 min because of irregular fuel feeding. The fuel built clumps between the powder outlet and the conical distributor in front of the outlet. The clumps fell down when they became big enough, which resulted in unstable combustion.

The conical distributor was removed before starting L2. The heating of tertiary air was stopped also. L2 went better than L1, and a continuous combustion of lignin powder could be achieved. Char tended to build on the vanes that guide the combustion air, especially the vane of the primary and secondary air. This was a result of burner design. The burner is built to generate a strong swirl, which forms a recirculation zone. This resulted in lignin powder being recirculated, heated, and sticking to the guide vanes, forming char. The char closed partially (or in some cases totally) the gaps between the vanes, severely disturbing the air distribution (**Fig. 4**).

We measured the temperature at the outlet of the fuel nozzle during L2 to examine the effect of preheating of the combustion chamber on lignin feeding. This was important to address concerns before the test that lignin could start to melt in the pipe leading up to the burner. The combustion channel was heated with oil before powder feeding. Air was supplied at 200 L/min at room temperature during the heating stage. The temperature at the outlet of the fuel nozzle was about 200ºC before lignin feeding. The temperature decreased to about 40ºC when the lignin feeding was started. The lower temperature can be a result of a cooling effect from the lignin powder and a reduction of the radiation effect on the thermocouples.

Extensive melting of the lignin thus could be ruled out as a cause of the problems experienced during L1 and L2. It was instead decided to focus on making the lignin flow more easily into the burner and to avoid recirculation of the lignin once it had entered the furnace. Ways of reducing the swirl were

discussed with the burner supplier, leading to changes described below. In separate cold feeding tests it was noted that there were less fluctuations in the lignin flow rate when the moisture content was decreased. Another test to determine the bridging tendency of lignin and wood powders in a silo pointed in the same direction, e.g., lignin powder at 10% moisture content flowed more easily than wood powder at 5% moisture content.

In the third test (L3), the moisture content of the lignin powder was reduced to 10%, and some modifications of the air supply were made to reduce the risk of lignin powder recirculating. The feeding air was increased to amplify the powder velocity by 50% compared with tests L1 and L2, and the primary air was reduced. The aim was to reduce the swirl generated by the vanes of the primary air supply. More air was supplied through the secondary zone. The feeding system also was modified to give a more even feeding rate. All these changes contributed to give a stable combustion with low emission levels.

Combustion test R1 was more stable than R2, and L3 was the most stable lignin combustion test. The O_2 concentration during R1 was 25% higher than during L3, while the gas temperature was 3% higher during L3. The peak gas temperature during L3 was 1,371°C and 1,223°C during R1.

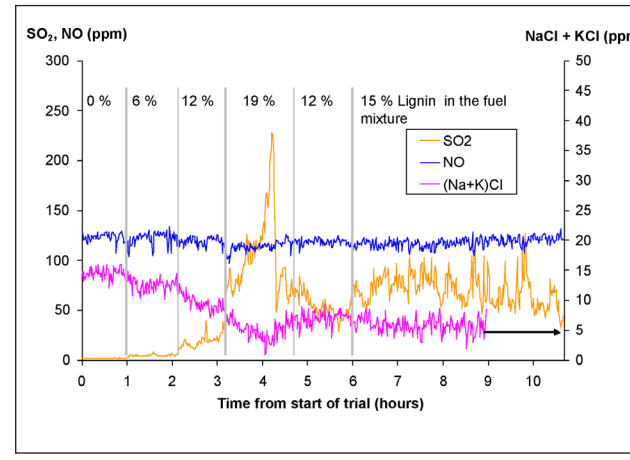
The nitrogen content was 0.3% in the reference fuel and 0.1% in the lignin. We could thus expect that NO_x emissions would be higher during R1, but the average NO_x emissions were the same during R1 and L3 (Table III). Higher peak gas temperature during lignin combustion might explain the difference in this trial. The carbon monoxide concentration was 34 ppm during R2 and 61 ppm during L3. The higher concentration of carbon monoxide during L3 can be explained with some occasions where the carbon monoxide concentration was very high, due to uneven feeding of the lignin powder.

The dust content in the flue gases during L3 was about 50% higher than that in R1. The higher dust content can be explained by the finer size distribution of the lignin compared with wood/bark powder. However, fine particulate matter (PM 2.5) was 99.7%–99.9% of the dust in L3, but 67% during wood/bark powder combustion.

Sulfur dioxide emissions could not be determined accurately. However, most of the sulfur in the lignin will form SO_2 when there is no excess of sodium (Na), potassium (K), or calcium (Ca) (see the following discussion of the CFB boiler trial).

For cofiring lignin filter cake with bark in the CFB boiler, the fuel was 100% wood pellets before the 3-day test period started. We changed the fuel to 100% bark pellets the morning of the first day. Lignin was then introduced and the lignin fuel feed was increased in three steps up to 19% on mass basis. The major part of the test was then carried out with 15% lignin and 85% bark.

Firing bark pellets led to a concentration of alkali chlorides of 17 ppm (at 6% O_2, dry gases). This is at least 3 times higher than during wood pellets or wood chips combustion where the concentration is typically below 5 ppm [3]. In both wood and bark, there are excess amounts of calcium present to capture the sulfur. Adding excess amounts of sulfur will completely prevent any formation of alkali chlorides. The chlorine (Cl) in the fuel is instead converted to hydrogen chloride (HCl) according to the following reaction:



5. Cofiring of lignin and bark.

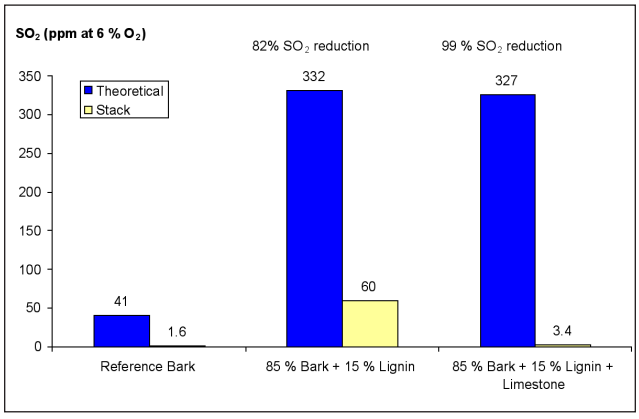
$$4\ KCl(s) + 2\ SO_2(g) + 2\ H_2O(g) + O_2(g) \leftrightarrow 2\ K_2SO_4(s) + 4\ HCl(g)$$

The introduction of lignin increased the SO_2 emissions from a very low level to about 75 ppm, at a lignin proportion of 15% (**Fig. 5**). As a result, the level of sodium and potassium chloride decreased by about 50%. It is well known that sulfur in the fuel or addition of ammonium sulfate in the flue gas has this effect. Instead of forming sodium/potassium chloride the corresponding sulfates are formed, thus reducing the risk for chlorine induced corrosion in the boiler. Although the SO_2 concentration in the flue gas increased, it is not expected to lead to increased corrosion at these levels. No measurable effect on the NO_x level could be seen when introducing the lignin.

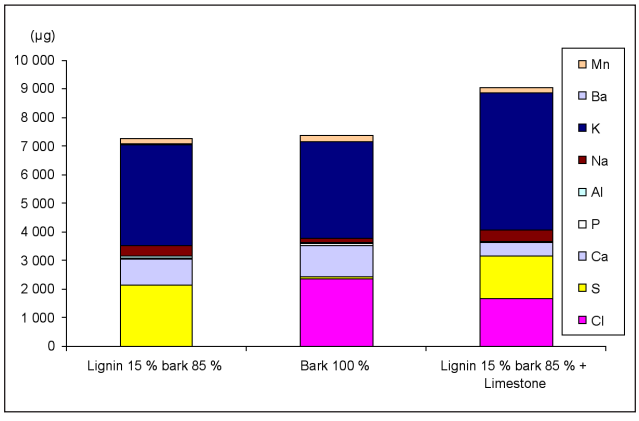
We conducted a reference test with 100% bark during the second day of the test period. The feeding of lignin was stopped before the reference test and after some hours the NO_x level increased markedly. One explanation is that the bed was saturated with free calcium oxide (CaO) because of the lime content in the bark ash. It is well known that calcium oxide can catalyze the NO_x formation in a system with low sulfur dioxide levels [4]. It could also be noticed that the sodium/potassium chloride level increased again when the sulfur emissions disappeared.

The limestone addition started 1 h after the start of the lignin feeding at a rate corresponding to a molar ratio of Ca:S = 2. When accounting for the calcium content in the bark ash, the total Ca:S ratio was 3.6:1. The sulfur capture was efficient with very low sulfur emissions (**Fig. 6**). However, when limestone was added the alkaline chloride content in the flue gas increased again. In an actual case, it would be better not to reduce the SO_2 emission in the bed as much. The limestone addition also increased the NO_x level somewhat.

The deposits from the probe rings were dissolved and the chemical composition of the total deposits was analyzed.



6. SO_2 capture by bark ash and limestone addition.



7. Composition of deposits.

Figure 7 shows that, in agreement with the flue gas measurements, the deposits from bark firing contained chloride; in the case when bark was cofired with lignin, the chloride content was very low, while the sulfur content increased. In the case when limestone was added, Cl was again present in the deposit. These results show that cofiring with lignin will reduce the Cl content in deposits. However, addition of limestone reduces this effect. It is also important to keep in mind that if more chlorine is introduced into the fuel mixture, the alkali chloride in gas form would increase [5].

Sintering tests

One of the main concerns when firing fuels containing alkali metals is the increased risk for bed sintering. The lignin produced within the FRAM2 project has a low ash and alkali content, and because of that, the risk for sintering problems should be low. However, the lignin ash contains about 15% alkali, mainly sodium. If enough ash is accumulated (e.g., in the bed) without proper regeneration of the bed, sintering problems might still be a risk. To study this, samples were taken from the bottom bed and cyclone particle seal and the sintering behavior was tested in a laboratory unit. The results showed no clear pattern. There is a declining trend in sintering temperatures (**Table IV**), but it is difficult to conclude that this effect depends on the lignin fuel. For the bottom bed material, the sintering temperature was lower the second day compared to the first day when bark was the only fuel. The cyclone ash had the same sintering temperature both the first and second day.

Sample		Estimated sintering temperature (°C)
Bottom bed	Bark + lignin	984
Bottom bed	Reference bark	924
Bottom bed	Bark + lignin + limestone	930
Cyclone	Bark + lignin	958
Cyclone	Reference bark	960
Cyclone	Bark + lignin + limestone	831*
*two measurements		

IV. Sintering temperatures measured on samples from the circulating fluidized bed boiler during the lignin trial.

When limestone was added for sulfur reduction the last day, the sintering temperature for the bottom bed material was the same as when firing bark only. For the cyclone bed ash however, there was a sharp decrease in sintering temperature. This clear drop in sintering temperature could depend on the limestone addition, but more studies have to be carried out to verify this explanation.

CONCLUSIONS

The pilot-scale combustion trials showed that lignin can be fired as moist filter cake and as a dry powder. Based on the results, we can proceed to full-scale trials. Some important information was obtained during the pilot trials that will be necessary to consider in the next step. With regard to powder firing, the following can be noted:

- No clogging or melting was observed during lignin feeding when the moisture content was reduced to about 10%. This suggests that no cooling other than transportation air is required for the feeding system, but that a moisture content of less than 10% is needed for smooth feeding of lignin powder.
- Dry lignin powder shows less bridging tendency than wood/bark powder, so it should flow more easily out of a storage silo.
- A stable and continuous combustion of kraft lignin is possible.
- Lignin powder is finer than wood powder produced in the same grinding equipment.
- Recirculated lignin powder will stick to the burner, causing a disturbance in the air supply and combustion process.
- Fine powders need less residence time for combustion. A burner with strong swirl produces a strong recirculation zone. The swirl should therefore be reduced when firing lignin powder.
- Dust emission during lignin combustion is at the same level as for wood powder combustion, but the dust particles are finer.
- NO_x emissions were slightly higher during lignin combustion than during combustion of the reference bark/wood

powder, despite the lower nitrogen content in lignin compared with bark/wood. Higher peak gas temperature during lignin combustion can explain the difference in this trial.

With regard to the cofiring of lignin and bark in a fluidized bed boiler, we determined the following:

• Lignin was easily handled and cofired together with bark. Although the filter cake was broken into smaller pieces and fines, the combustion was not disturbed.

• The combustion performance was not influenced by the lignin.

• The sulfur content in the lignin had a significantly positive effect on reducing the alkali chloride content in the deposits, thus reducing the risk for sticky deposits and high temperature corrosion.

• When cofiring lignin with bark, the sulfur emission increased compared with bark firing only, but most of the sulfur was captured by calcium in the bark ash. Conventional sulfur capture with addition of limestone to the bed also occurred.

• The lignin addition had no measurable effect on the sintering properties of the bed material. **TJ**

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ABOUT THE AUTHORS

Lignin isolated from black liquor can become an important biofuel inside and outside pulp mills, with a potential to replace oil and coal in many applications. Lignin has interesting properties that distinguish it from other solid biofuels, such as a high heating value, hydrophobicity, and a homogeneous composition. Our aim with this study was to evaluate the combustion of lignin under well-controlled conditions on a small scale, before moving on to large-scale trials.

In biofuels combustion, feeding is often a challenge. This was an area that we addressed carefully in the trials (e.g., by doing cold feeding tests before the actual combustion experiments).

Before the trials, we expected that a cooled feeding system might be needed for the lignin powder, but temperature measurements proved that this was not the case; a simple but important finding that will facilitate the work on larger scale.

We think that mills that are planning for lignin extraction in the future will find the information from our study very useful when it comes to using the lignin as a fuel in lime kilns or power boilers.

The next step is to evaluate lignin combustion in full-scale trials in a lime kiln and a boiler.

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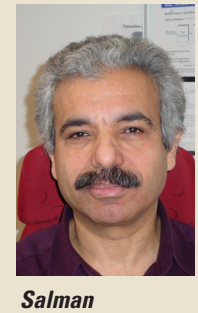
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