

PILOT-SCALE COMBUSTION STUDIES WITH KRAFT LIGNIN AS A SOLID BIOFUEL

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ABSTRACT

Processing of kraft lignin precipitated from black liquor to produce a solid biofuel with high energy density and low ash content has been developed in research programs by STFI-Packforsk and partners. In preparation for full-scale combustion trials, tests were carried out on pilot scale in a 150 kW powder burner and in a 12 MW 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 wood/bark powder. In the CFB boiler lignin was easily handled and co-fired together with bark. Although the filter cake was broken into smaller pieces and fines the combustion was not disturbed. When co-firing lignin with bark, the sulfur emission increased compared to 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 was also demonstrated. 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.

INTRODUCTION

One way of exploiting the energy surplus of a modern kraft market pulp mill is to extract lignin from the 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 it is required that the extracted lignin can be handled and fired in combustion equipment other than conventional recovery boilers, e.g. in lime kilns and other applications where powder burners are common, or in fluidized bed boilers frequently used for biomass combustion.

Processing of kraft lignin precipitated from black liquor to produce a solid biofuel with high energy density and low ash content has been developed in research programs by STFI-Packforsk and partners. The process concept is now being verified on a scale of about 4000 tonnes per year at a demonstration plant in Bäckhammar, Sweden (Figure 1), operated by LignoBoost AB. As part of the ongoing FRAM2 (Future Resource-Adapted pulp Mill) 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 on 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 were 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 three tonnes of lignin were fired in a three-day trial in the circulating fluidized bed (CFB) research boiler at Chalmers. The lignin was co-fired 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 (CO, NO_x, SO₂), fireside deposits on heat transfer surfaces, in-bed sulfur capture with limestone addition, and effects on the sintering properties of the bed material.



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

METHODS AND MATERIALS

Lignin used in the powder burner. Lignin prepared at the demonstration plant in Bäckhammar was used for combustion tests together with a reference powder (Table 1). 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 from Alexanderwerk, using a working cylinder with 1.5 mm holes. Some of the lignin was finally air dried to a moisture content of 10 % at ETC. The lignin powder was finer compared to 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 . Some 300 kg of lignin powder were fired in the tests.

Table 1. Fuels used in the powder burner trial.

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	22.9	18.8
Moisture content	% of fuel	10	5

Lignin filter cake and bark pellets used in the CFB boiler. Bark pellets were used as base fuel in the trial. The lignin fuel used as additional fuel was delivered as broken filter cake in big 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 was therefore of varying moisture content (9-34 %) and ash content (0.5-1.4 %). To even out the differences the big bags were fed in a specified order. In Table 2 the fuel characteristics are given for both the bark pellets and the lignin fuel.

Table 2. Fuels used in the CFB boiler trial.

Fuel		Lignin filter cake*	Bark pellets	15 % lignin + 85 % bark (wet fuel)
Moisture content	%	29.3	10.3	12
Higher Heating Value	MJ/kgDS	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/kgDS	1085	9324
K	543		2138	1931
Na	1666		310	486
Na+K	mmol/kgDS	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 @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 big bags used in the trial.

Powder burner. The burner supplied by VTS is of a free flame burning type, and a refractory lined cone is placed at the front of the burner. It is a combined oil/powder burner and is dimensioned for maximum power of 150 kW. The powder nozzle is in the centre. A conical mass is situated in front of the powder nozzle and 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.

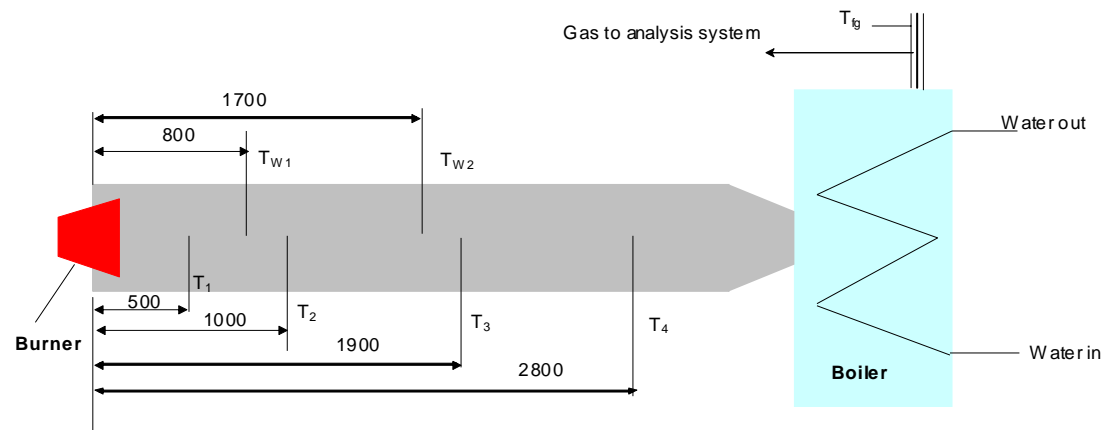


Figure 2. Schematic view of the combustion furnace with a 150 kW powder burner at ETC.

The burner fires into a combustion channel with a rectangular cross-section of $550 \times 550 \text{ mm}^2$. The length of the channel is 3.3 m. It is made of 3-mm iron plates and insulated by four layers of insulation materials. Four gas temperatures were measured in the combustion channel (T_1 - T_4), as well as the flue gas temperature (T_{fg}), and two wall temperatures (T_{w1} - T_{w2}). The inner wall-temperature was measured by thermocouples placed inside the refractory and about 20 mm from the wall. The positions of the thermocouples are shown in Figure 2. The fuel-nozzle of the powder-burner is placed about 300 mm inside the channel.

A screw feeder was used to feed the powders. The flow rate of the powder was measured by calibration of the screw feeder and also 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 O_2 , CO , and CO_2 . NO_x was recorded by a chemical cell instrument (Testo, model 350 XL). The particle mass size distributions were determined by collecting size-classified particle samples with a 13-stage low-pressure cascade impactor (Dekati).

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

Circulating fluidized bed boiler. The boiler at Chalmers was built for research but has all the features of a commercial unit. The combustion chamber (1) has a cross section of 2.25 m^2 and a height of 13.6 meter. 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 also the fluidizing nozzles are found. Secondary air was supplied during the present tests through an air register located 2.1 meter (4) above the bottom. After the combustion chamber, the flue gases enter the convection path where the gases are cooled down to 150°C . The 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).

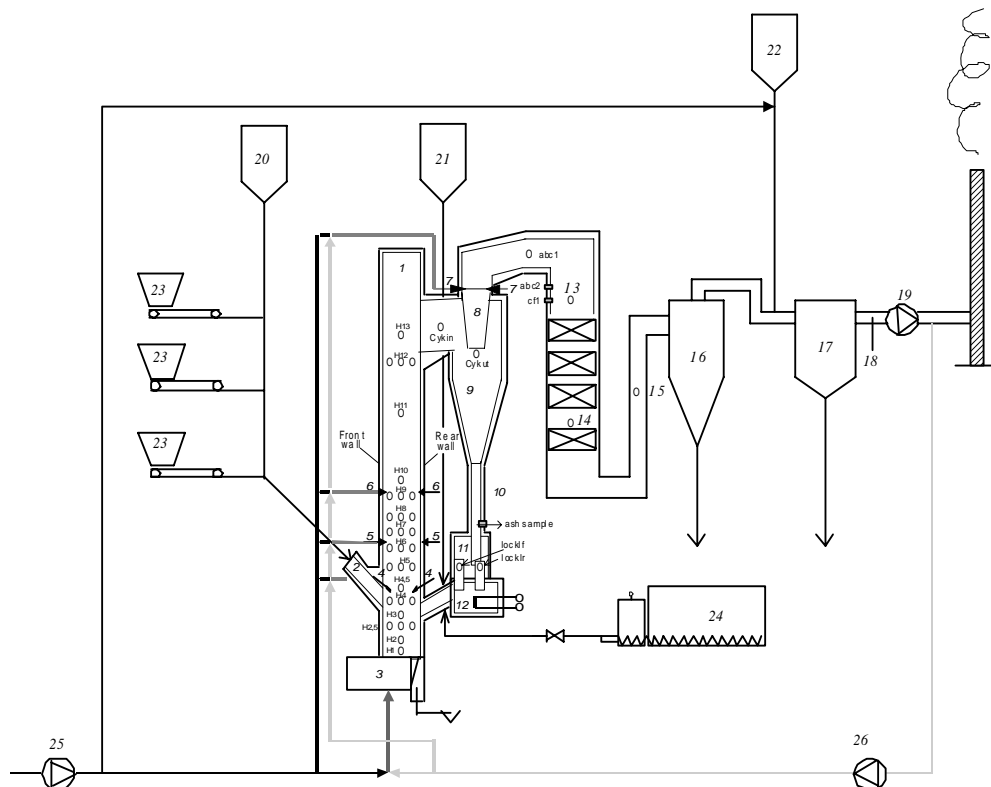


Figure 3. Flow diagram of the 12 MW CFB boiler at Chalmers.

The emissions of NO_x, SO₂ and CO were measured at a position after the bag house filter (18). Ash samples were taken from the bottom bed in a position named "H2" in Figure 3 and from the cyclone leg indicated below (10). Fly ashes were taken from the secondary cyclone (16) and from the bag filter (17). In order to measure gaseous alkali chlorides an In-situ Alkali Chloride Monitor (IACM) [1,2] was situated before the convection section.

RESULTS

Firing of Lignin Powder

Two tests (designated 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 3 shows the main results from the tests.

Bark/wood powder reference tests. The tertiary air was preheated to about 360°C 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 hours. The temperature of the gas reached a level of 1100-1200°C. Wall temperatures were lower (1000-1100°C). NO_x emissions were about 120 ppm during R1 and about 70 ppm during R2. NO_x emissions were almost stable in both tests.

CO-emissions were very low during R1, but less stable and much higher in R2 (average of 450 ppm). The difference between R1 and R2 is the air flow which was reduced in the second test. The aim of this measure was to reduce the O₂-content in the flow gases to 2-3% to simulate the situation in the lime kiln. It can be seen that there are larger variations in O₂ and CO contents in the flue gases during R2 than R1. This variation is related mostly to the variation in the feeding rate of the fuel. It has been observed in earlier tests on wood powder in the same equipment that O₂ content less than 3% in the flue gases gives high CO content. An explanation to such phenomena is that lower air flow causes less swirl and leads to poorer mixing in the burner.

Combustion with lignin powder. Test L1 was carried out on a lignin powder with a moisture content of 18 % and with the same conditions as in test R2. The test was stopped after only 35 minutes 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 too. L2 went better than L1 and a continuous combustion of lignin powder could be achieved. There were tendencies to char building on the vanes that guide the combustion air, especially the vane of the primary and secondary air. This can be explained by the burner design. The burner is built to generate a strong swirl, which will form 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 (Figure 4).

The temperature at the outlet of the fuel nozzle was measured during L2. The combustion channel was heated with oil before powder feeding. Feeding air of 200 L/min was supplied at room-temperature during the heating stage. The temperature at the outlet of the fuel nozzle was about 200°C before starting lignin feeding. When the lignin feeding was started the temperature decreased to about 40°C. 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. This information is important, since there was concern before the test that lignin could start to melt in the pipe leading up to the burner.

A third test (L3) was performed with a lignin powder with a moisture content of 10 %. Some modifications of the air supply were made during this test. The feeding air was increased to amplify the powder velocity by 50 % compared with tests L1 and L2. At the same time 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 was also modified to give a more even feeding rate. All these changes contributed to give a stable combustion with low emission levels.

The length of test L3 can be compared with the tests that were carried out on the reference fuel. Combustion test R1 was more stable than R2, and L3 was the most stable combustion test done on lignin. It is therefore the R1 and L3 tests that will be commented and compared.

Table 3. Conditions and results, powder burner tests.

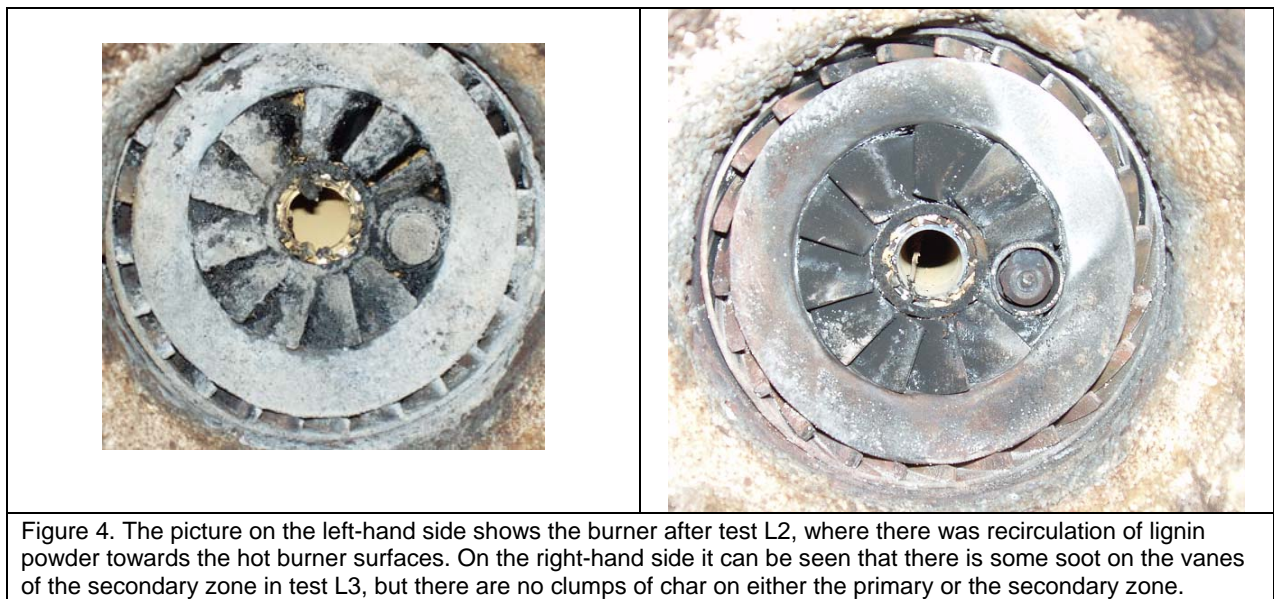
Fuel	Test number	Bark/wood powder		Lignin powder		
		R1	R2	L1	L2	L3
Fuel flow	kg/h	20.8	20.6	20.8	19.8	20.0
Fuel moisture content	%	5	5	18	18	10
Burner thermal power (LHV)	kW	109	108	124	118	119
Air flow	L/min	1280	1055	1530	1550-1950	2100
Air temperature	°C	Room temp.	360 (tert.air)	360 (tert.air)	Room 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	1175	1121	n.a.	1228	1214
Test duration	h	7.0	5.1	0.6	4.8	5.0

The O₂-concentration during R1 was 25 % higher than during L3, while the gas temperature was 3 % higher during L3. The nitrogen content in the reference fuel is 0.3 % while in lignin it is 0.1 %. It could thus be expected that NO_x emissions would be higher during R1, but as shown in Table 3, the average NO_x emissions are the same during R1 and L3. Higher gas temperature during lignin combustion can explain the difference in this trial.

CO concentration was 34 ppm during R2 and 67 ppm during L3. The higher concentration of CO during L3 can be explained with some occasions where the CO 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, PM_{2.5} is 99.7-99.9% of the dust in L3 while it is 67% during wood/bark powder combustion.

SO₂ emissions could not be accurately determined. It can, however, be expected that most of the sulfur in the lignin will form SO₂ when there is no excess of Na, K or Ca (cf. CFB boiler trial below).



Co-firing Lignin Filter Cake with Bark in the CFB Boiler

The fuel was 100 % wood pellets before the three-day test period started. In the morning of the first day the fuel was changed to 100 % bark pellets. Lignin was then introduced and the lignin fuel feed was increased in three steps up to 19 % on mass basis (approximately equivalent on energy basis, since the lignin filter cake had higher moisture content than the bark pellets). The major part of the test was then carried out with 15 % lignin and 85 % bark.

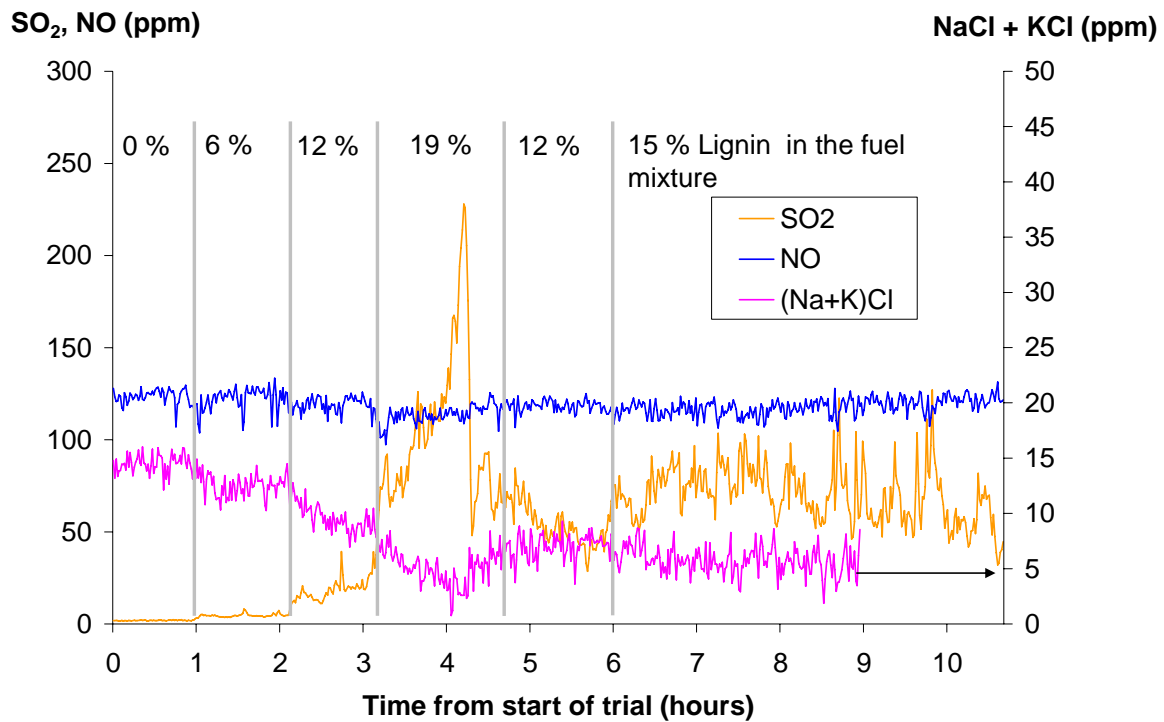
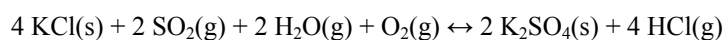


Figure 5. Co-firing of lignin and bark.

Effects on combustion and emissions. Firing bark pellets only led to a concentration of alkali chlorides of 17 ppm (@6%O₂, 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 [4]. 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 Cl in the fuel is instead converted to HCl according to the following reaction:



As can be seen in Figure 5, the introduction of lignin increased the SO₂ emissions from a very low level to about 75 ppm at a lignin proportion of 15 %. 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. No measurable effect on the NO_x level could be seen when introducing the lignin.

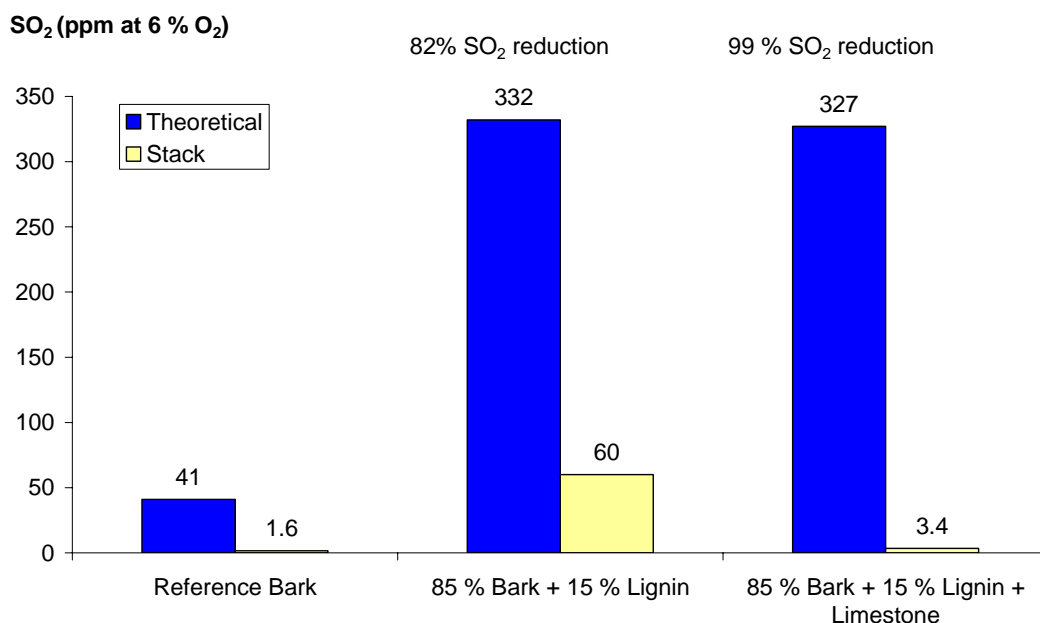


Figure 6. SO₂ capture by bark ash and limestone addition

During the second day of the test period, a reference test with 100 % bark was carried out. The feeding of lignin was stopped in the evening prior to the reference test and after some hours the NO_x level increased markedly. One explanation is that the bed was saturated with free CaO due to 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 [3]. It could also be noticed that the sodium/potassium chloride level increased again when the sulfur emissions disappeared.

Limestone addition. The limestone addition started one hour 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. The sulfur capture was efficient with very low sulfur emissions, see Figure 6. However, when limestone was added the alkaline chloride content in the flue gas increased again. In a real case it would be better not to reduce the SO₂ emission in the bed as far. The limestone addition also increased the NO_x level somewhat.

Deposit composition. The deposits from the probe rings were dissolved and the chemical composition of the total deposits was analyzed. It can be seen from Figure 7 that, in agreement with the flue gas measurements, the deposits from bark firing contain sodium and potassium chloride, but in the case when bark is co-fired with lignin the chloride is replaced by sulfur (in the form of sulfate). In the case when limestone is added, Cl is again present in the deposit. These results show that co-firing with lignin will reduce the Cl content in deposits. However, addition of limestone reduces this effect. It is also important to have in mind that if more chlorine would be 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 for example in the bed without proper regeneration of the bed there could still be risk for sintering problems. Based on the results from a laboratory study no increased sintering tendency was expected from the lignin addition. To confirm 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 4) 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.

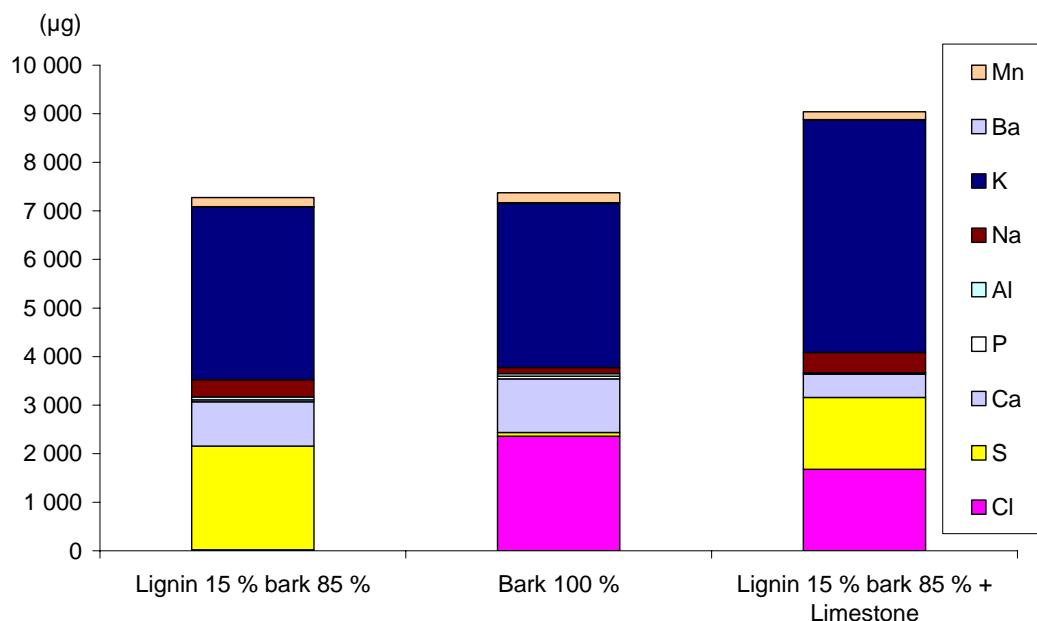


Figure 7. Composition of deposits.

The last day when limestone was added for sulfur reduction, 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.

Table 4. Sintering temperatures measured on samples from the CFB boiler during the lignin trial.

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

CONCLUSIONS

As a general conclusion, the pilot scale combustion trials have shown that lignin can be fired both as moist filter cake and as a dry powder. Based on the results it will be possible to 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:

- A moisture content of less than 10 % is needed for a smooth feeding of lignin powder.
- Dry lignin powder shows less bridging tendency than wood/bark powder, i.e. flows more easily.
- No clogging or melting was observed during lignin feeding. It can be concluded that no cooling other than the transportation air is required for the feeding system.
- A stable and continuous combustion of kraft lignin is fully possible.
- Lignin powder is finer than wood powder produced in the same grinding equipment.

- Recirculated lignin powder will stick to the burner and cause 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.
- The 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 gas temperature during lignin combustion can explain the difference in this trial.

With regard to the co-firing of lignin and bark in a fluidized bed boiler

- Lignin was easily handled and co-fired 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 co-firing lignin with bark, the sulfur emission increased compared to 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 was also demonstrated.
- The lignin addition had no measurable effect on the sintering properties of the bed material.

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