

THE LIGNOBOOST PROCESS

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A process for lignin removal from alkaline pulping liquors (black liquors) has been developed within the FRAM2 R&D Program (Future Resource Adapted Pulp Mill, part 2). The lignin product (Fig. 1) from a demonstration plant, owned and operated by Innventia (formerly STFI-Packforsk), was characterized and used in different combustion trials with good results – co-firing of lignin and bark in a fluidized bed boiler, co-firing with coal in a PFBC (Pressurized Fluidized Bed Combustion) boiler and firing of lignin in a full-scale lime kiln. The process development and operation of the demonstration plant has displayed good results. Runnability in the demonstration plant and the lignin quality have been both very good. The work done on the investment and operational costs showed great potential for improving the concept of economy, which is very promising.

Keywords: kraft pulp, lignin, biofuel, combustion

INTRODUCTION

One way of exploiting the energy surplus of a modern kraft pulp mill is to extract lignin from the black liquor. This gives the pulp mill a great opportunity to develop new economical revenues when the new by-product, lignin, is commercialised or used within the pulp mill to reduce production costs. Lignin extraction has the additional advantage of providing an incremental capacity in the chemical recovery area, so that it can be used to off-load the recovery boiler or to avoid expansion, when pulp production is increased. One way to handle the bulk flow of produced lignin could be to use the dry lignin powder as a biofuel in the lime kiln and swap fossil fuel. Other ways could be to use lignin in other burners/boilers, where fossil fuel is normally used, boilers in which corrosion from chlorides is a problem or, generally, 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, is an idea originating from the R&D Program entitled KAM (the Ecocyclic Pulp Mill), developed within the R&D Program

entitled FRAM (Future Resource-Adapted pulp Mill). The process concept, called LignoBoost, has been verified on a scale of about 4,000 tonnes per year, in a demonstration plant located in Bäckhammar, Sweden, owned and operated by Innventia.¹ The LignoBoost process, including patents and patent applications, is today owned by Metso.

Within the FRAM2 R&D Program (Future Resource-Adapted pulp Mill, part 2), the development of the LignoBoost process was carried out to reduce costs. Furthermore, full-scale combustion trials were carried out to evaluate the effects of firing the lignin biofuel product in two different types of boilers^{2,3} and in a lime kiln. The FRAM2 Program, with management and project management teams from Innventia, included work done by many partners, *e.g.* universities and companies. The main contributors of the R&D work were Innventia, Chalmers University of Technology and ÅF Process. FRAM2, initiated in October 2005 and completed in December 2008, brought about new knowledge and know-how, some of which is presented in this paper.



Figure 1: Lignin produced in the demonstration plant of Bäckhammar – filter cake and lignin pellets

RESULTS AND DISCUSSION

The LignoBoost concept, according to the development within FRAM2

The traditional process of lignin precipitation and separation from kraft black liquors causes severe problems, related to complete or partial plugging of the filter cake and/or of the filter medium. Plugging of these types of filter cakes results in an extremely low level of the washing liquor flow through the cake and, consequently, to the need for extremely large filter areas. Partial plugging of the filter cake also leads to high levels of impurities in the lignin.⁴ It has been shown that these negative effects on the filtration process are caused by changes in lignin solubility, due to an excessive pH level and to the ionic strength gradients in the lignin filter cake during the washing process. Such changes result in restructuring the lignin particles, e.g. returning to a colloidal state, or in dissolution and re-precipitation of lignin in the filter medium (Patent applications 1794363, 1797236 and PCT/SE 2008/000142).

In the LignoBoost Process, a stream of black liquor is taken from the black liquor evaporation plant (Fig. 2), then lignin is precipitated by acidification (the preferred acid is CO₂) and filtered (“chamber press filter 1”, Fig. 2). Instead of washing lignin immediately after filtration, as in traditional processes, the filter cake is re-dispersed and acidified (“cake re-slurry”, Fig. 2). The resulting slurry is then filtered and washed by means of displacement washing (“chamber press filter 2”, Fig. 2).

When the filter cake is re-dispersed in a liquid, at pH level and temperature values approximately equal to those of the final washing liquor, the concentration gradients during the washing stage will be low. The change in the pH level, most of the change in ionic strength and any change in lignin solubility will then take place in the slurry, and not in the filter cake or in the filter medium during washing.

It should be noted that the filtrate from chamber press filter 1 (filtration and dewatering stage, Fig. 2) will be recycled to the black liquor evaporation plant, after the point at which the feed stream to LignoBoost is taken. This should avoid any decrease in lignin concentration in the stream fed to the LignoBoost operation. The filtrate from chamber press filter 2 (filtration, washing and dewatering stages) should be recycled to the weak black liquor. In some cases, this filtrate can be also used for washing the unbleached or oxygen delignified pulp.

The LignoBoost process therefore makes it possible to extract lignin efficiently from the black liquor in kraft mills. The major advantages, compared to the previous technology, are the following:

- the filter area and the volume of acidic washing water can be kept at lower values, resulting in lower investment costs,
- the addition of sulfuric acid can be also kept at a lower level, resulting in lower operational costs,
- the yield of lignin is higher,
- the lignin has a lower ash and carbohydrate content,
- the lignin has a higher content of dry solids.

LignoBoost process investment costs (drying, pulverizing, pelletizing and storage are excluded)

Within the FRAM2 Program, the investment costs were estimated according to an assumed process configuration (Fig. 2) and to the budget prices (from 2008) for the equipment in Scandinavia. The total investment cost was calculated, excluding the cost for a general contractor to coordinate the project, the costs for evaluation and taking risks, and the performance guarantees. The investment costs are related to what is believed to be the cost after the first installations had been established.

The size of the filters is based on the experience gained in the demonstration plant of Bäckhammar. Compared to the studies made on lignin precipitated from black liquor in many other pulp mills, the Bäckhammar lignin has a somewhat higher filtration resistance. It has been shown that the lignin

precipitated from liquors from other mills has a lower filtration resistance, while requiring only one third of the filter surface. This will naturally have a corresponding effect on the cost of the filters in the installation.

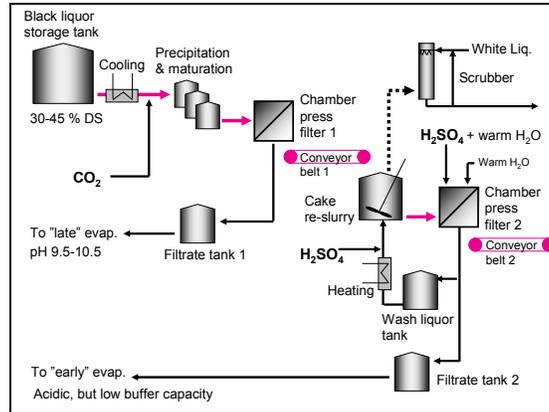


Figure 2: General layout of the LignoBoost lignin removal process (post-treatment, drying and pulverizing are excluded)

In a commercial LignoBoost plant, the costs may be different, due to the layout of the mill and to other local factors. The layout of a site, in which the different pieces of equipment are positioned, has not been optimized in our example.

The investment cost estimate below assumes that:

- the lignin plant can be placed close to the evaporation plant and to the recovery boiler,
- there is a weak gas system that can take care of the foul smelling gases from the lignin,
- there is sufficient capacity for the electrical installation, so that only low-voltage equipment is required,
- no further processing of the lignin cake, *e.g.* drying or pelletizing, is included.

The investment costs, related to the second half of 2008, refer to a lignin plant capacity of 50,000 tonnes/year (7 tonnes/h), as follows:

- direct costs, including civil equipment (spare parts included), process controls, electrical (low voltage), piping and valves and construction were estimated at SEK 88 million,
- indirect costs, including freight, temporary facilities, engineering, commissioning, the start-up and unspecified costs have been estimated at SEK18 million.

The accuracy of the total investment costs for a LignoBoost plant (SEK 106 million), with the particular current design, should lie within +/- 20 % of the estimated cost. It has been assumed that the lignin plant will operate 350 days/year, at an utilization rate of 85%, producing 50,000 tonnes of lignin per year (calculated as a dry product).

LignoBoost operating costs (drying, pulverizing, pelletizing and storage excluded)

The operating cost is mainly related to chemicals (CO_2 , H_2SO_4 and NaOH), although minor increases in the black liquor evaporation (2.2-2.5 m^3 /tonne of lignin) and in the power consumption are needed.

A major operating cost item is CO_2 . A large part of the CO_2 cost is for its transportation. In our calculations, it has been assumed that the CO_2 will be purchased at a rate of SEK 800/tonne, an estimate based on discussions carried out with a gas supplier in Sweden and related to a consumer of large quantities of CO_2 . A preliminary study of the costs for producing in-house CO_2 from lime kiln flue gases indicates that the estimated costs for CO_2 might be possibly reduced by 40 to 50%.

Other major operating costs should be for H_2SO_4 and NaOH . Acid is used in the washing operations to minimize the sodium content in the final lignin product. It is an

advantage to use internal sources, such as chlorine dioxide spent acid in the “re-slurry of cake” dilution stage. However, in the displacement washing on chamber press filter 2, we suggest that fresh acid should be used, since the acid consumption there is very low. The higher operating cost given below can be regarded as very conservative, since calculation is based on using 100% fresh H₂SO₄ (no spent acid). This intake of sulfur would consequently interfere with the Na/S balance, causing a need for the discharge of the recovery boiler dust, resulting in a need to add NaOH for maintaining the Na/S balance. The lower operating cost, given below, shows the potential utilization of the spent acid in the “re-slurry of cake” dilution stage and whether CO₂ is produced from the lime kiln flue gas.

In addition, ongoing studies have resulted in a promising method that will substantially reduce the demand for acid in the “re-slurry of cake” operation. A new patent application has been performed for this method.

The expected operational costs for the LignoBoost process range from SEK 230 to SEK 580 per tonne of lignin (SEK 30-80/MWh). The lower value presents a reasonable potential, when:

- the spent acid from the pulp mill is sufficient to meet almost the complete acid demand and spent acid can be used, essentially without any additional costs for NaOH,
- CO₂ is collected from the flue gases and concentrated before being used in a LignoBoost process.

The costs for H₂SO₄ (SEK 400/tonne) and NaOH (SEK 2,700/tonne) have increased, then dropped slightly during the last few years, which is in line with rising energy costs and developments in the world economy. We have assumed that, in future, these prices will drop down to the levels we have used in our calculations (2007 levels).

Drying and pulverizing of LignoBoost lignin

Drying of LignoBoost lignin is a way of reducing transportation costs and of meeting the specifications for the demands of different products. It is particularly important to dry and pulverize the lignin for its use in lime kiln burners, for example. It is also beneficial to dry lignin before pelletizing or

co-pelletizing it with sawdust or with other raw materials, for making biofuel pellets.

The theoretical demand on energy to obtain completely dry lignin is between 0.3 and 0.4 kWh/kg of DS lignin, depending on whether the drying operation starts at 70 DS lignin or 65% DS lignin.

The aim was to find a solution to the process in the drying operation, where an existing heating source could be used (Fig. 3). A general approach was identified for selecting the type of dryer to be used for drying moist lignin cakes from the second press filter in the LignoBoost process. One alternative was to create small particles to improve the heat and moisture transportation to and from the moist lignin. Smaller particles, however, result in an increased risk of dust explosions. The way to handle this is to lower the oxygen content in the drying medium to a level at which the risk of an explosion is avoided. This is possible, for instance, by using flue gases from the lime kiln operation as a heating medium. The other alternative is to avoid creating small particles and, therefore, minimize the risk of an explosion. In this case, indirect drying could be a safer and more reliable way of drying the lignin. We explored several technologies. Considering the drying result and the expected investment costs, the most promising ones are:

- a belt drying technology, where dust formation is minimized and the drying cost is low. This technology is traditionally used for the drying of sludge, timber and lumber;⁵
- a “Swirl Fluidizer” technology, which gives a very fine lignin powder. This technology is traditionally used for drying of potato starch and similar products;⁶
- a rotary drum dryer, which is a robust technology that worked well for lignin. It is, however, potentially more expensive.⁷

In early studies, the temperature during lignin drying was limited to an interval between 70 and 80 °C. Efficient drying, however, is the result of a combination of temperature, retention time and of how effectively the moisture is transported from the lignin. Our studies showed that it is possible to use higher drying temperatures if the retention time is short and if moisture is effectively removed before the lignin starts to soften (melt).

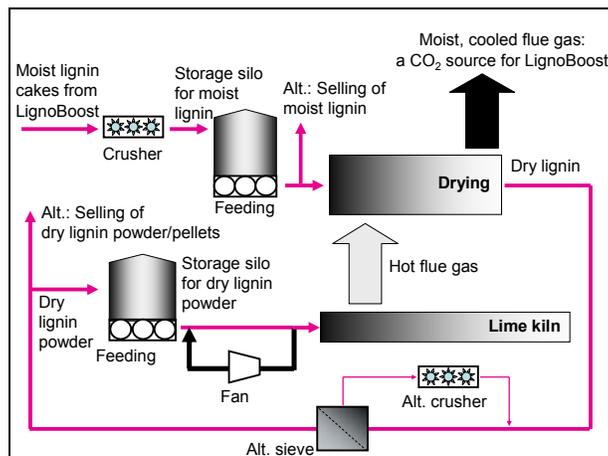


Figure 3: General principles for lignin post-treatment and use in a lime kiln (or for selling)

Characteristics of lignin from the LignoBoost Demo plant

Lignin from the kraft process contains more carbon, compared to other biofuels. Lignin also has a higher heating value. The heating value of the lignin fuel samples was determined as being higher (HHV) in a bomb calorimeter. The lower heating value (LHV) was calculated, based on HHV and basic analysis. On the average, the HHV is 26.7 MJ/kg of dry lignin. This can be compared with 18 to 22 MJ/kg of DS for wood or bark. The LHV of the dried lignin powder, used in the combustion trials described below, in which a lime kiln was operated with lignin powder as a fuel (at approx. 4% moisture content), was of 24.4 MJ/kg fuel.

The average dry solid content of lignin coming directly from the LignoBoost demonstration plant (*i.e.* filter cake) was of approx. 63%, varying between 60 and 70% DS during the first year of operation. The bulk density of the various forms of lignin (filter cake, dried powder, milled powder,

etc.) was determined during various trials. The most representative bulk density data for the lignin powder was of approx. 650-750 kg/m³.

The mean ash content of the lignin produced for biofuel purposes in the LignoBoost demonstration plant was of approx. 0.8% during the first year of operation. The ash content determined from 90 lignin samples, collected in the first year, varied from 0.3 to 1.2%. As expected, the basic composition of the ash varied considerably among the different lignin powder samples. The major ash components were sodium, potassium, calcium and aluminum (Table 1).

The relatively high amount of sulfur bonded to the lignin from the kraft process makes this biofuel attractive as an additive to fuels where, due to a large amount of alkali chlorides, corrosion may raise problems. The sulfur present in lignin reduces the chloride content in ash by releasing HCl.

Table 1
Analysis of kraft lignin from the LignoBoost demonstration plant in Bäckhammar (some examples)

Parameter	Median values	Variation in samples		No samples
		min.	max.	
<i>Content in fuel (mass-%)</i>				
Moisture ^{a)}	32.3	29.3	40.0	3
Ash (dry)	1	0.2	1.4	5
<i>Heat value (MJ/kg)</i>				
HHV (dry ash free)	27.1	26.6	27.3	5

HHV (moist) ^{a)}	18.2	15.9	18.6	3
LHV (dry ash free)	25.9	25.3	26.0	5
LHV (moist) ^{a)}	16.6	14.2	16.9	3
<i>Elemental analysis (% dry ash free)</i>				
C	65.1	63.6	66.2	5
H	5.8	5.7	6.2	5
O	26.1	25.9	27.5	5
S	2.5	1.8	3.2	5
N	0.1	0.1	0.2	5
Cl	0.01	0.01	0.01	2
<i>Ash analysis</i>				
Element	mg/kg ds	mg/kg ash, average value	mg/kg ash, min. max.	No samples
Na	1 666	151 500	119 000 227 152	5
K	543	38 800	24 000 76 154	5

^{a)} Data from filtered lignin. Other data are based on both filtered and dried lignin

The sulfur content in the LignoBoost kraft lignin is normally between 1 and 3%, with 2.5% as a typical value in kraft lignin from the demonstration plant.

Compared to traditional biofuels, lignin possesses some unique properties that have to be taken into consideration when being handled, *i.e.* transported and stored:

- the moist lignin powder (>10% moisture) has a lower softening temperature, when compared to dry lignin powder. Moist lignin starts to soften at a temperature as low as 70-80 °C;
- the dry lignin powder is very reactive, due to the small particles it contains. The equipment involved must be ATEX classified, to minimize the risk of dust explosions.

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