ADVANCED BIOFUELS AND CHEMICALS VIA BIOMASS GASIFICATION AND RENEWABLE POWER

Rikard Gebart, Luleå University of Technology

Advanced biofuels and chemicals via biomass gasification and renewable power

Rikard Gebart, Energy Engineering, Luleå University of Technology, Luleå, Sweden
Erik Furusjö, Bioeconomy, RISE, Stockholm, Sweden
Fredrik Granberg, Energy Engineering, Luleå University of Technology, Luleå, Sweden

The political constraints for forest based products are changing as a result of the global ambition to reduce the anthropogenic emissions of greenhouse gases (GHG) to the atmosphere. In particular the transport system will need to undergo a paradigm shift from being completely based on fossil oil to being based on a combination of renewable power and renewable fuels. As an example the Swedish parliament has in 2018 introduced a new obligation for renewable components in gasoline and diesel that has created a significant market pull for renewable fuels with good GHG performance. At the moment the market pull can be met by blending with ethanol and HVO. However, the obligation is designed to grow to 40% in 2030 and 100% in 2045, which requires new production units. Similar initiatives can be expected at the EU level due to the new Renewable Energy Directive (RED II). The pulp and paper industry is in an ideal position to take a leading role in this transition but the industry appears to be hesitating due to the risk of an impact on the existing business. However, this is a questionable policy since other industry segments could easily build new green field plants, which would definitely lead to a competition for sustainably harvested forest biomass. By being pro-active the pulp and paper industry could instead take a leading role in the transition and make good business in a changing industrial landscape.

The present state of the art for black liquor gasification, which is a key technology for fast conversion of a pulp mill into a flexible biorefinery, will be reviewed in the presentation. The advantage with black liquor gasification is that it is a proven process that produces a very clean, low tar syngas that makes it easy to produce a wide range of products, using existing and proven technology from the petrochemical industry. Examples of products with high yield and good market potential are methane, methanol, dimethylether, synthetic gasoline, synthetic diesel and biojet fuels. It is also possible to produce valuable chemicals, e.g. peroxide and ammonia. Many of these products can be produced to final quality at the pulp mill, which simplifies the business case and increases the profitability compared to other concepts that require final upgrading at a petro-refinery.

Black liquor gasification has a huge potential in itself but it is closely coupled to the pulp production, which can sometimes be a drawback. One option to loosen the coupling is co-gasification with other liquid fuels, e.g. fast pyrolysis oil from forest residues. Testing of co-gasification with pyrolysis oil has been successfully done for more than 1 000 hours in the LTU black liquor gasification pilot plant in Piteå (> 25 000 hours with pure black liquor). Another interesting option is to blend the syngas with hydrogen from electrolysis of water. Cost estimates of the different concepts show that the lowest production cost is obtained with pure black liquor gasification, typically 70-90 € per MWh produced fuel. However, both co-gasification with pyrolysis oil and combination with electrolysis only increases the production cost marginally while the total yield increases 2-5 times.

SUOMEN SOODAKATTILAYHDISTYS
FINNISH RECOVERY BOILER COMMITTEE

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Rikard Gebart[1], Erik Furusjö[2], Fredrik Granberg[1]

1: Energy Engineering, Luleå University of Technology, Luleå, Sweden
2: Bioeconomy, RISE, Stockholm, Sweden

June 6, 2019 55th Anniversary International Recovery Boiler Conference

Outline

- Introduction
- Black liquor gasification
- Experience from the Chemrec gasifier
- Boosting with pyrolysis oil
- Combination with electrolysis
- Demo-project with bio-DME and bio-methanol
- Conclusions

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The Renewable Energy Directive (RED II)

- 32% of the energy supply* should come from Renewable Energy Sources (RES) by 2030
- Sub target for transports: 14% of road and rail transport energy should come from RES by 2030
- Advanced biofuels will have a higher value due to double counting
- This creates a powerful driving force for use of biomass in the energy system
- Forest product industry has a competitive advantage compared to green field biorefineries

* app. 1 500 million tonnes oil equivalents in EU 28

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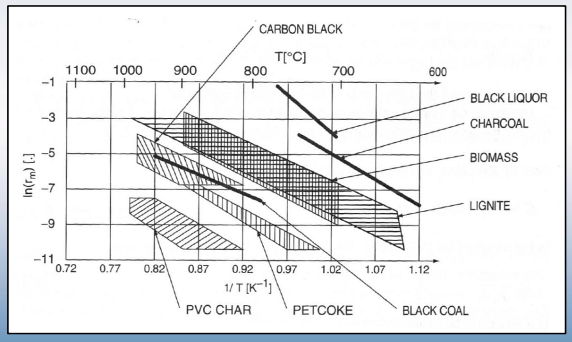
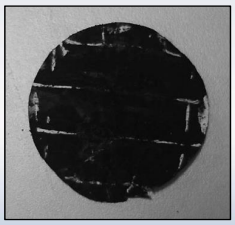
Black liquor is an excellent gasification fuel

- Very high alkali content, 20-30% water
- Very reactive, catalyzed by Na
  - Gasification possible at 1000 C
  - Very little soot and tar
- Very important – it is a liquid that can be pumped

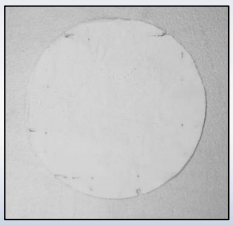
Gasification reactivity



Lab scale experiments @ 1000 °C



Saw dust



Black liquor

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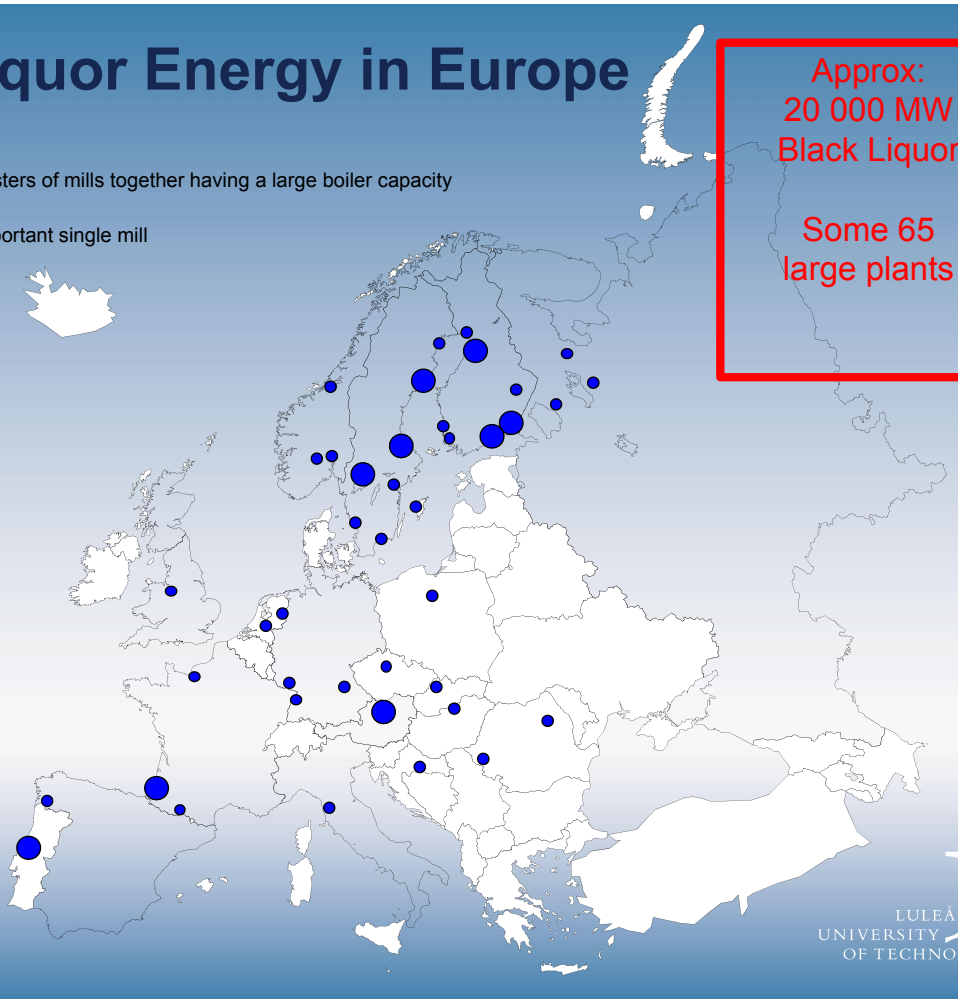
Black Liquor Energy in Europe

= clusters of mills together having a large boiler capacity

= important single mill



Approx:
20 000 MW
Black Liquor

Some 65
large plants

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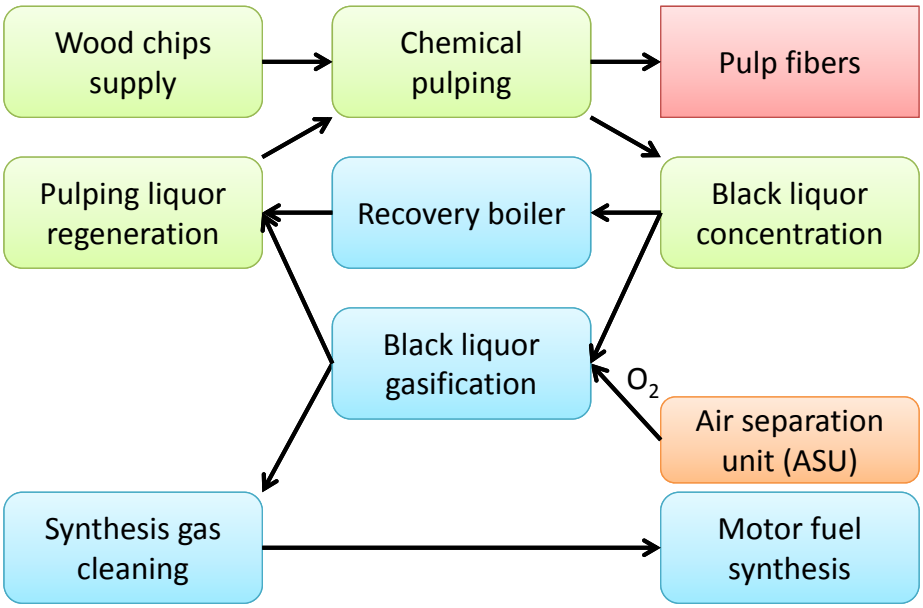
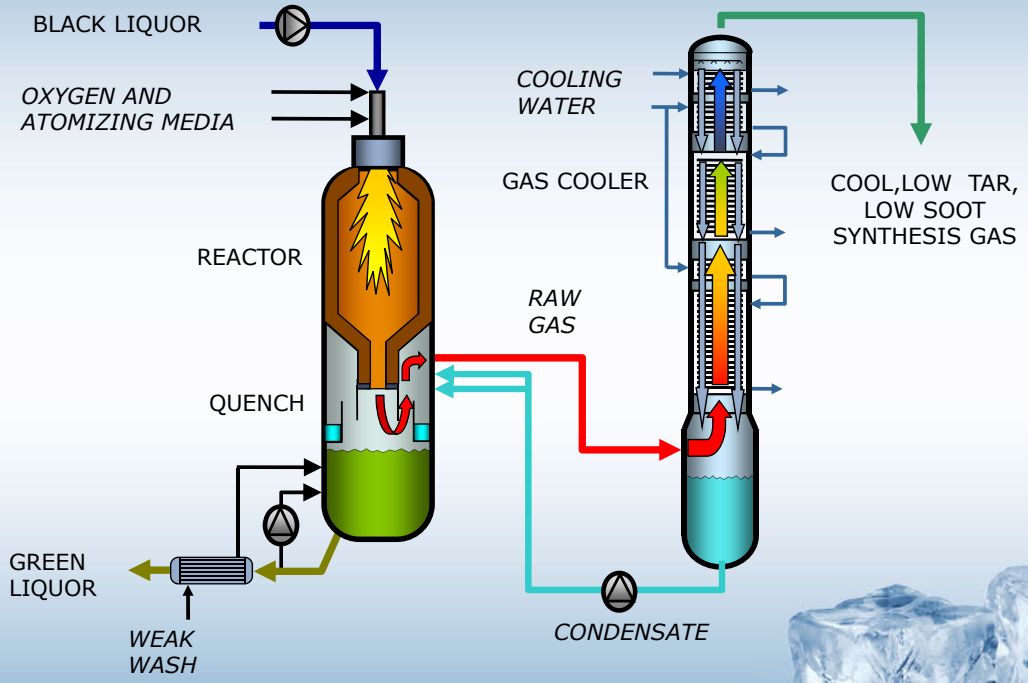
Black liquor gasification to motor fuels



- The recovery boiler can be boosted or replaced with a gasifier
- Need to increase supply of low grade biomass to maintain energy balance
- Small increase in energy use in lime kiln for regeneration of cooking chemicals

The Chemrec Black Liquor Gasifier



Carlsson, P., "Large scale experiments and modeling of black liquor gasification", PhD thesis, Luleå University of Technology, 2011

Jafri, Y., "Entrained flow gasification of black liquor and pyrolysis oil", LicEng thesis, Luleå University of Technology, 2016

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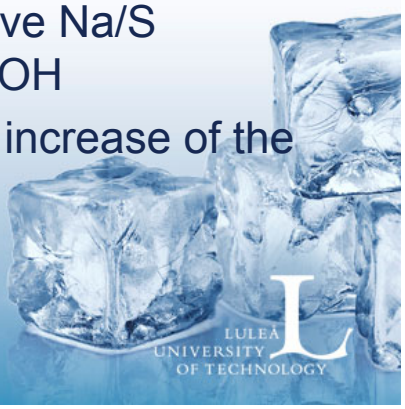
Excellent syngas and green liquor from BLG

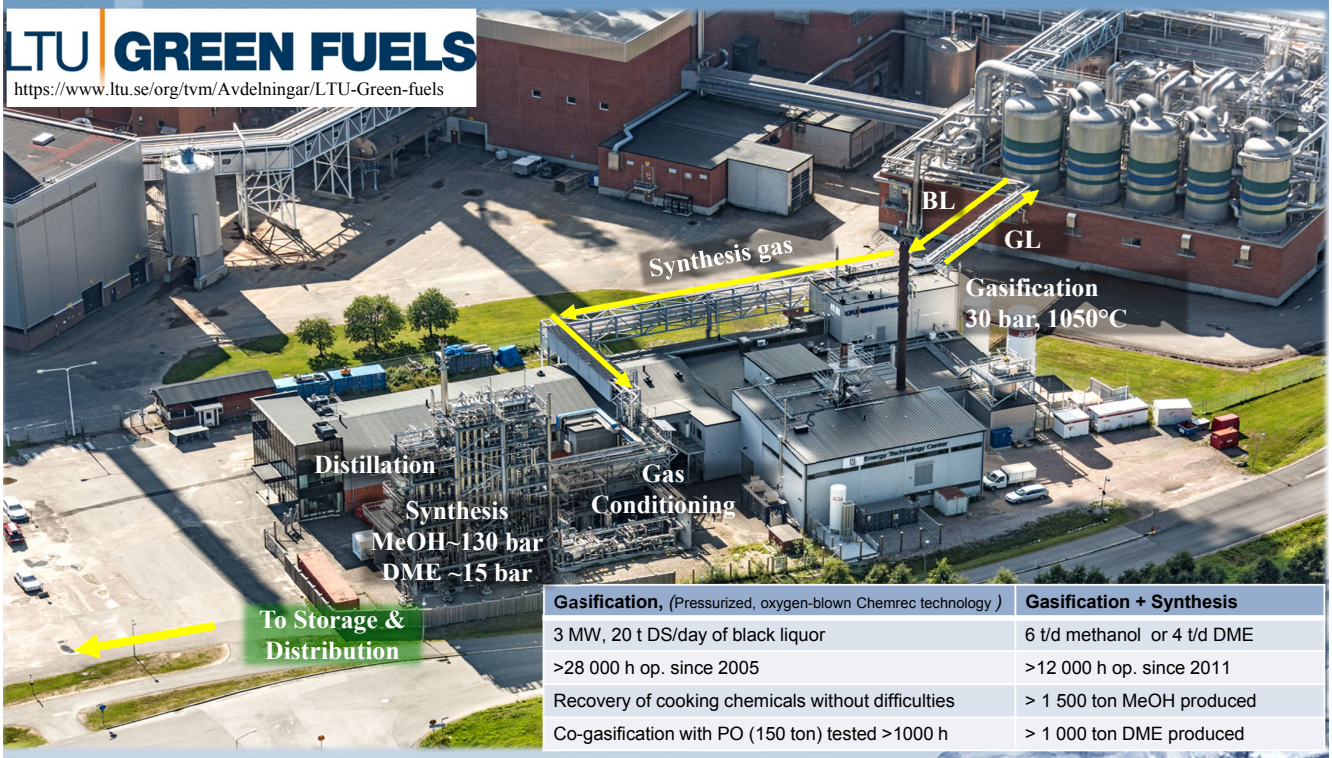
- Syngas
  - H2/CO ratio 1.2-1.4
  - 28.7% CO
  - 34.3% H2
  - 33.9% CO2
  - 1.35% CH4
  - 1.65% H2S
  - Practically tar free
  - Low soot
- Green liquor
  - Equal to recovery boiler GL in many respects
  - Low amount of sludge
  - Slightly higher causticising need due to formation of carbonates in the smelt

Sulphur split

- Approximately 30% of the sulphur will go to the gas phase as H_2S
- The sulphur will be recovered in the gas cleaning as H_2S or as elemental sulphur
- This opens up for:
  - Selective bleeding of sulphur to improve Na/S balance without need for make-up NaOH
  - Polysulphide cooking with about 10% increase of the pulp yield



LTU GREEN FUELS
https://www.ltu.se/org/tvm/Avdelningar/LTU-Green-fuels



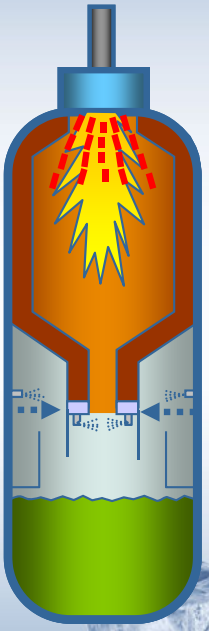
Gasification, (Pressurized, oxygen-blown Chemrec technology)	Gasification + Synthesis
3 MW, 20 t DS/day of black liquor	6 t/d methanol or 4 t/d DME
>28 000 h op. since 2005	>12 000 h op. since 2011
Recovery of cooking chemicals without difficulties	> 1 500 ton MeOH produced
Co-gasification with PO (150 ton) tested >1000 h	> 1 000 ton DME produced

- The plant was built by Chemrec in several collaborative research projects since 2004
- In 2013 the plant and staff was transferred to Luleå University of Technology

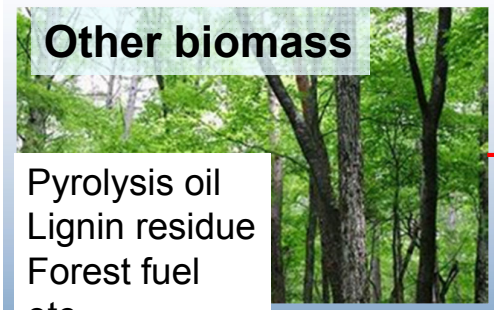
Catalytic co-gasification – mixing BL with pyrolysis oil

Low heating value
High reactivity

Mixing



Efficient gasification?



Other biomass

Pyrolysis oil
Lignin residue
Forest fuel
etc.

High heating value
Low reactivity

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Validation of co-gasification concept

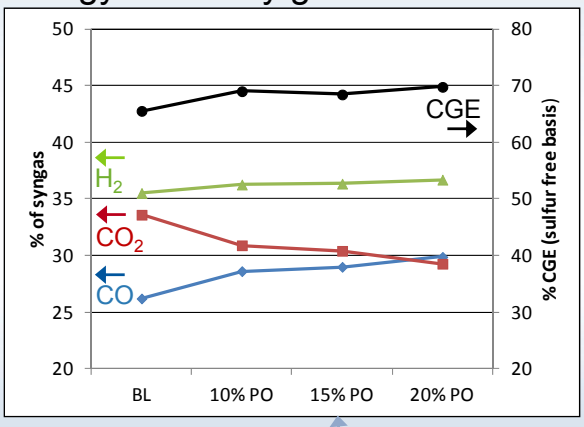
Small modification of the pilot

Summary of co-gasification

- First attempt spring 2015
- Operation 1100 h, app. 170 ton PO
- Operation 900 h with metanol/DME
- Up to 20% PO; 3,5 MW_{th}
- No difference in green liquor or syngas quality
- Robust operation, no technical difficulties

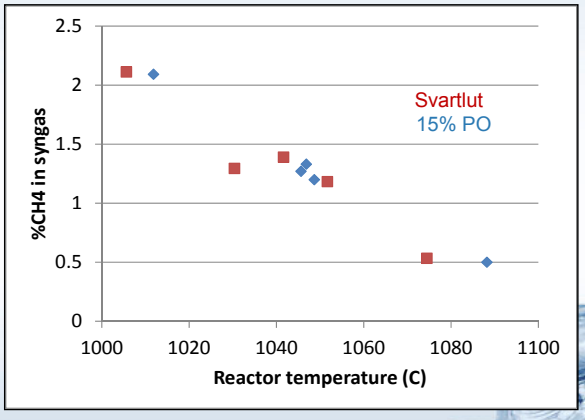
Co-gasification in pilot scale

Energy efficiency gasification

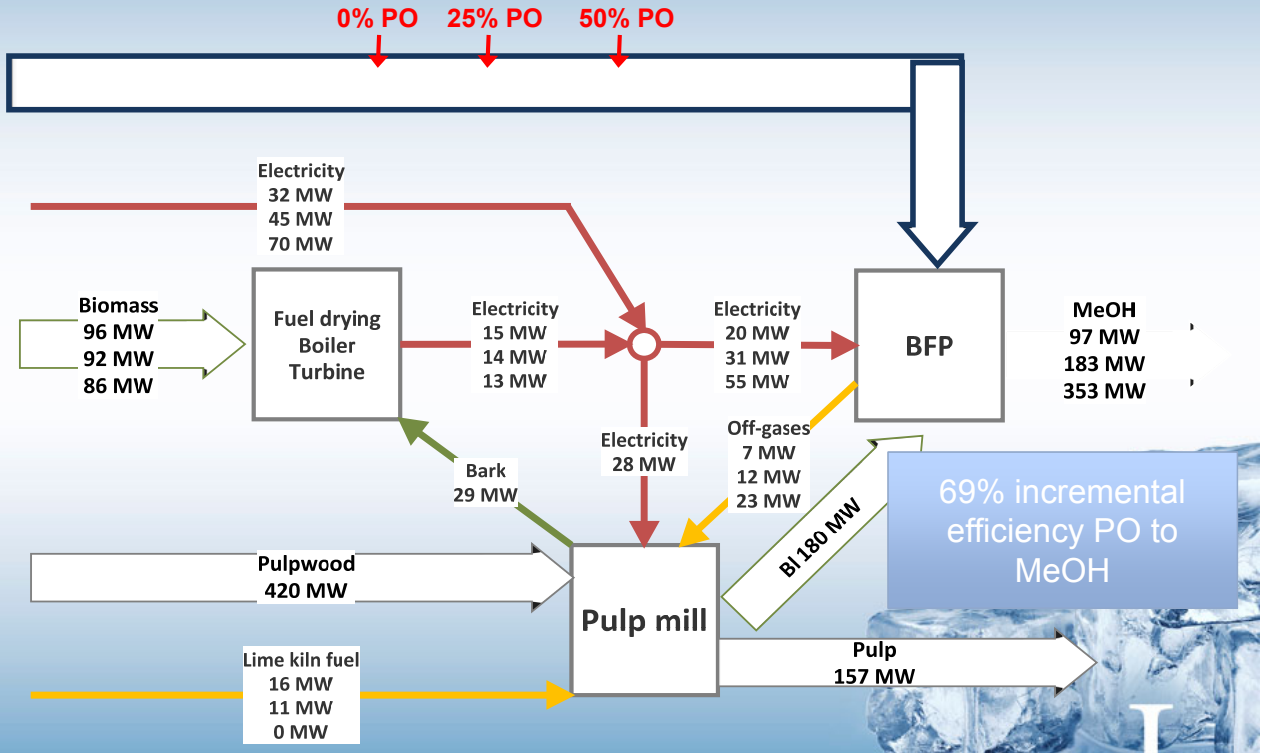


1.3% CH_4
1.5% N_2
1.4% H_2S
114 ppm C_6H_6
11 ppm C_2-C_3

Temperature sensitivity

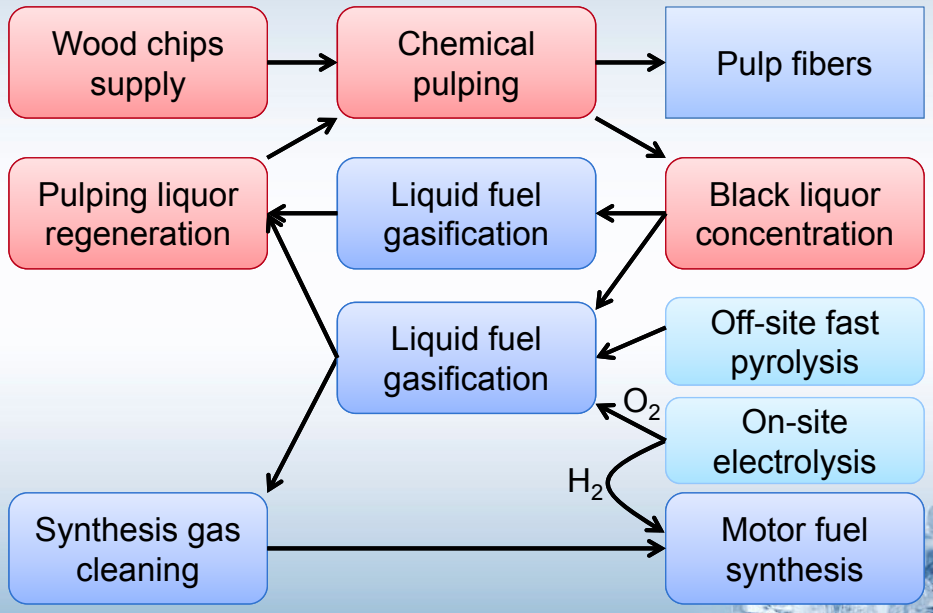


Co-gasification: energy balance



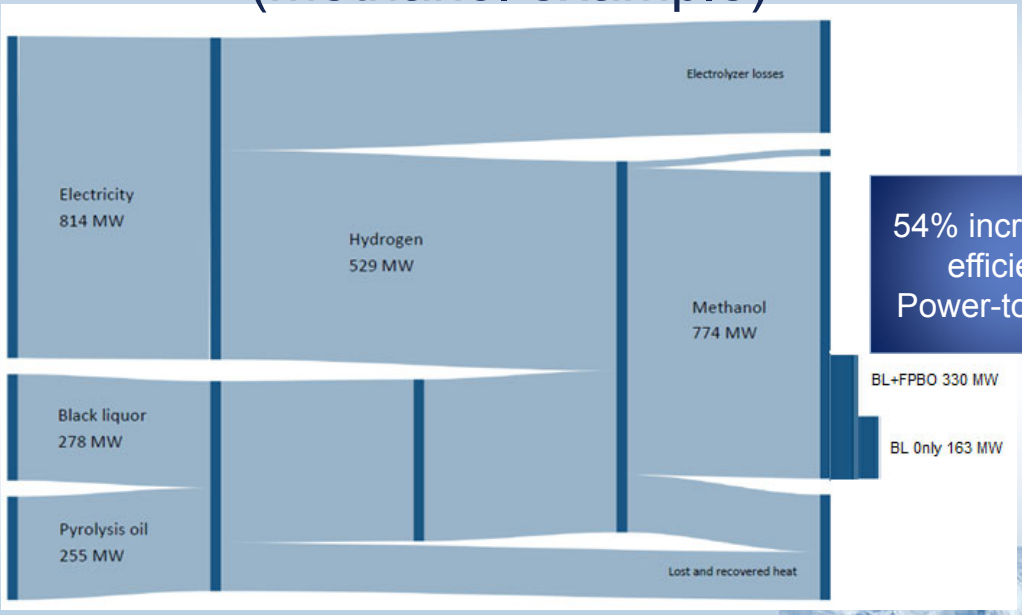
Andersson, J., Lundgren, J., Furusjö, E., & Landälv, I. (2015). Co-gasification of pyrolysis oil and black liquor for methanol production. *Fuel*, *158*, 451–459. https://doi.org/10.1016/j.fuel.2015.05.044

Next improvement – combine with electrolysis



- Elimination of CO-shift and ASU, lower CAPEX and lower specific OPEX
- Push synthesis to convert all CO_2
- Large capacity even with partial stream of black liquor

Efficiency for the new electrofuel process (methanol example)

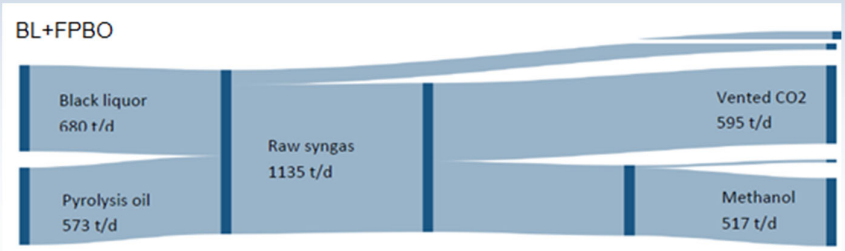


54% incremental efficiency Power-to-MeOH

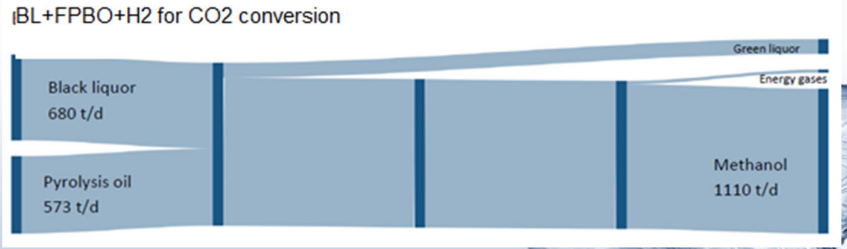
- More than a doubling of capacity compared to gasification only
- Needs a lot of renewable power (will be stored as chemical energy)
- Same production cost as gasification only (with current power price)

Very high carbon efficiency

Base case



New process

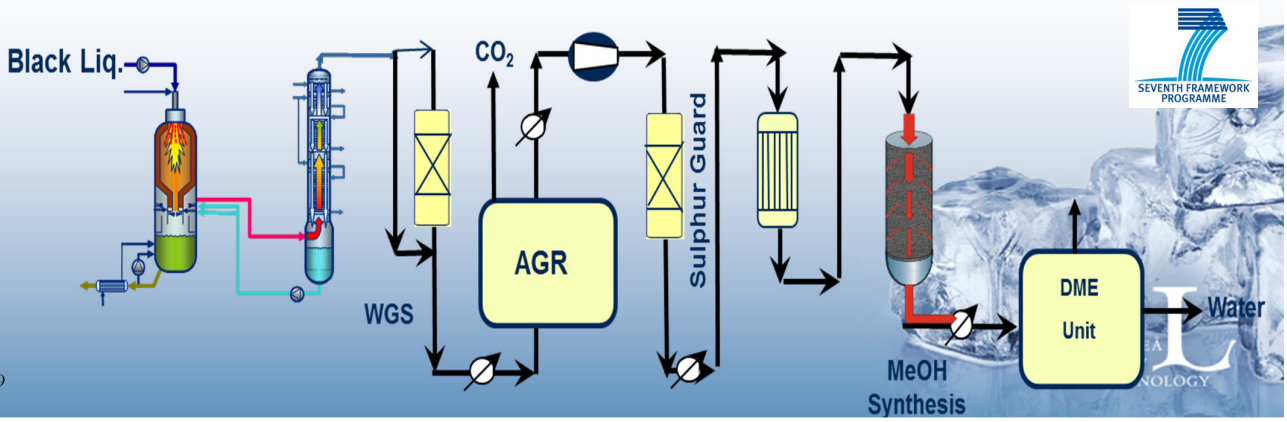


- Doubling of carbon efficiency compared to gasification only
- Almost all carbon is bound in methanol, the rest is recycled

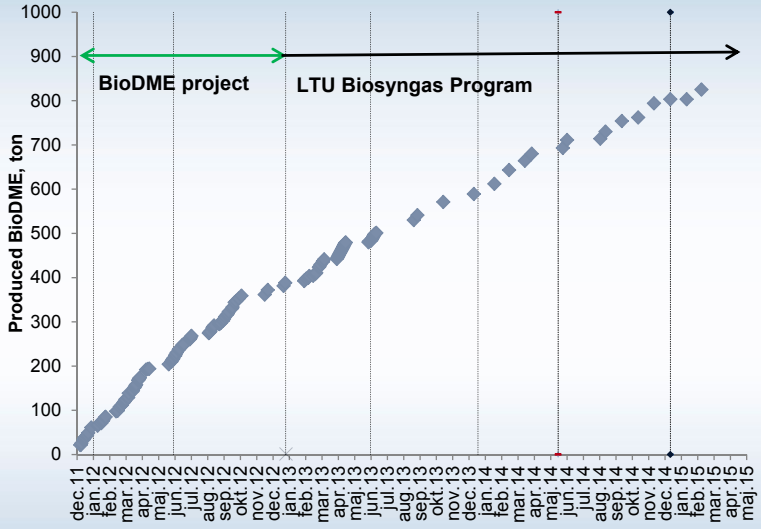
The BioDME project

- Demonstration of new once-through methanol technology
- Demonstration of integrated fuel production from black liquor at pulp mill
- BioDME demonstrated as diesel substitute
- BioDME project partners:

CHEMREC HALDOR TOPSØE ETC TOTAL preem DELPHI VOLVO



Cumulative BioDME production



More than 1000 ton DME + MeOH was produced until May 2016

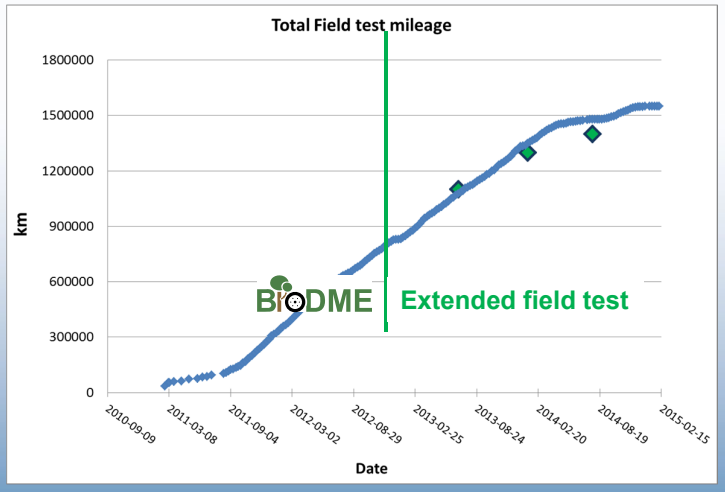
(1) Landalv, I.; Gebart, R.; et al., "Two years experience of the BioDME project - A complete wood to wheel concept", *Environ. Prog. Sustain. Energy,* 2014, 33 (3), 744–750.

Development of filling stations

Field test with HD trucks in commercial operation

Volvo field test

(1000 Km)	Status 2015-01-31	Target June 2014
Total mileage	1 550	1 400
1 truck	328	250



Process economics for BioDME

- Estimated necessary sales price with 15% IRR: 70-90 €/MWh*
- Typical plant size: 150 000 ton DME per year
- Flexible output of methanol or DME
- Methanol can be upgraded with MTG to gasoline that satisfies the current gasoline standard

* Carvalo, L., ”Opportunities to broaden biomass feedstocks in thermochemical conversion technologies”, PhD thesis, Luleå University of Technology, 2018
Andersson, J., ”Systems analysis of chemicals production via integrated entrained flow biomass gasification”, PhD thesis, Luleå University of Technology, 2016

Conclusions

- Black liquor is an important biomass resource and an excellent gasification fuel
- Black liquor gasification is a mature process after more than 28 000 hours of operation
- Combination of gasification and electrolysis
  - Eliminates the need for an ASU
  - More than doubles the potential of black liquor and bio-oil
- Co-gasification of black liquor and bio-oil
  - benefits from the catalytic effect of the black liquor
  - Increases the potential of black liquor with 50-100%
- BioDME has been demonstrated in heavy duty trucks in commercial operation for more than 1 500 000 km
  - Low soot, Nox and GHG performance
  - Excellent well-to-wheel efficiency

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Thank you for your attention!

Field testing of bio-methanol

- Passenger car > 20 000 km with M56
- Speedway motorcycle with M100
- Drag racing car with M100
- Pilot boat (converted diesel engine)
- 50 ton bio-MeOH supplied as catalyst to bio-diesel production

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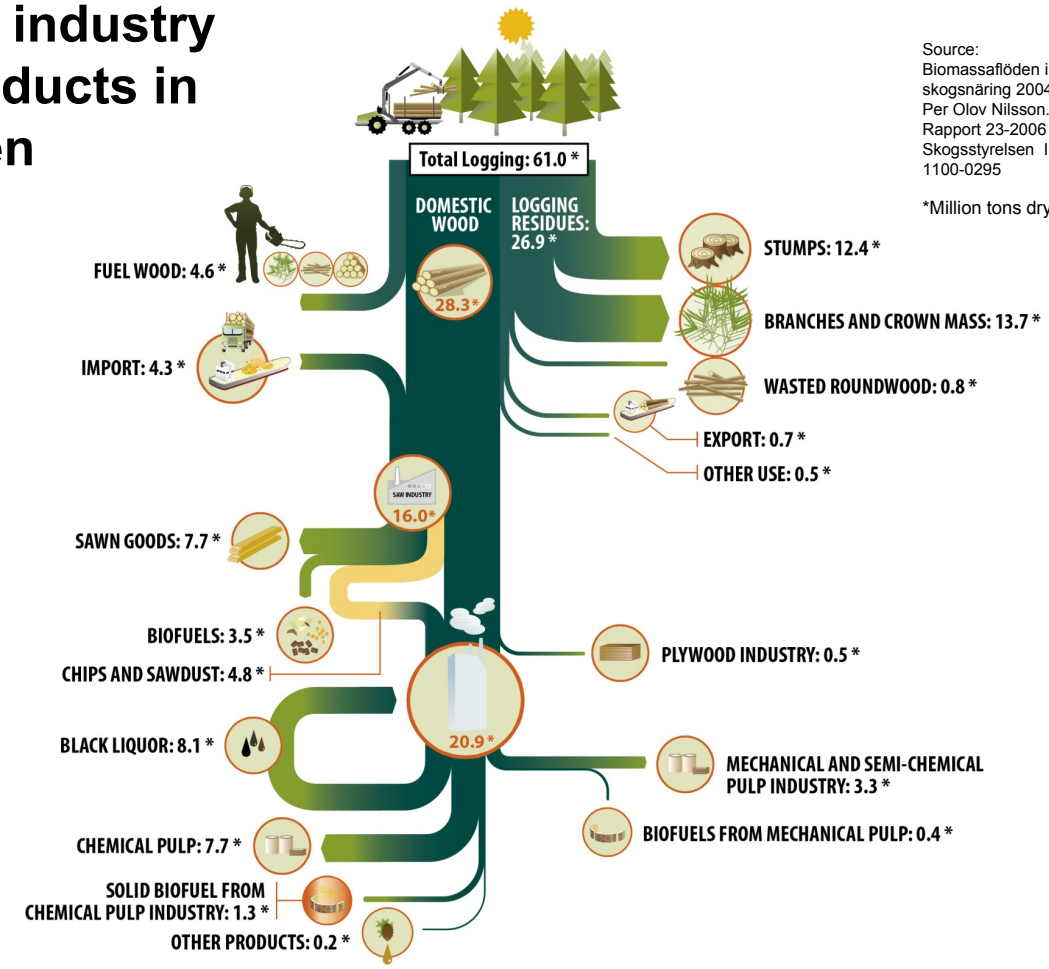
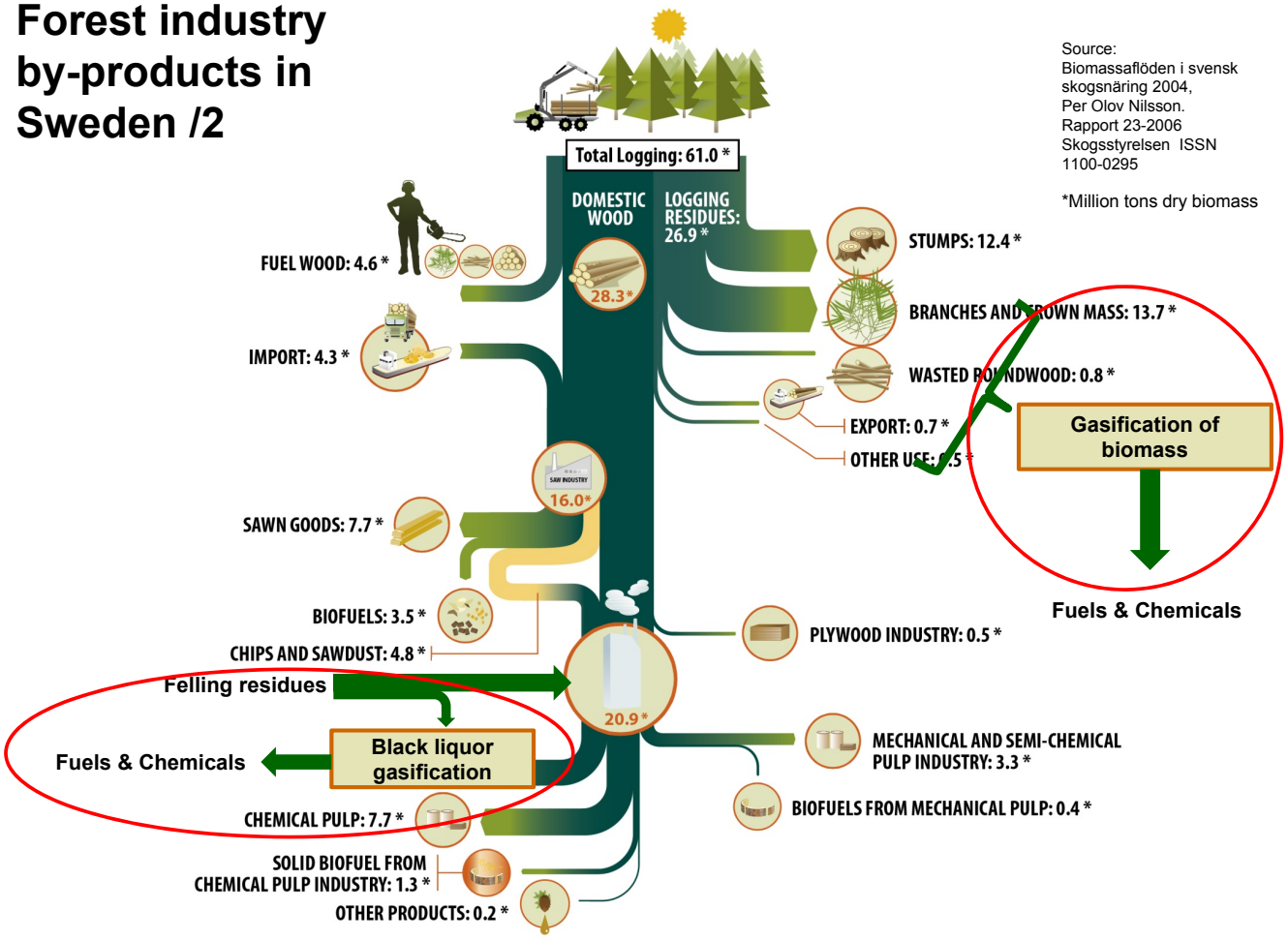
Forest industry by-products in Sweden



Source:
Biomassaflöden i svensk skogsnäring 2004,
Per Olov Nilsson.
Rapport 23-2006
Skogsstyrelsen ISSN 1100-0295

*Million tons dry biomass

Forest industry by-products in Sweden /2



Source:
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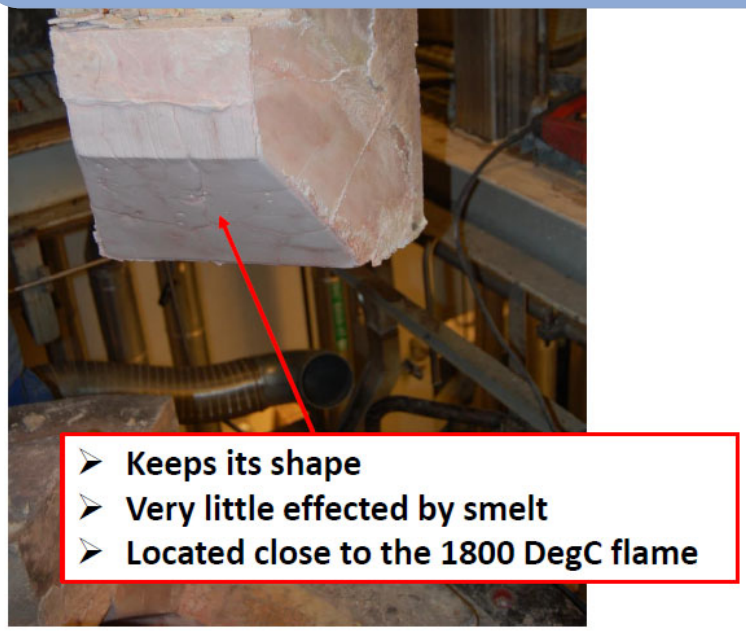
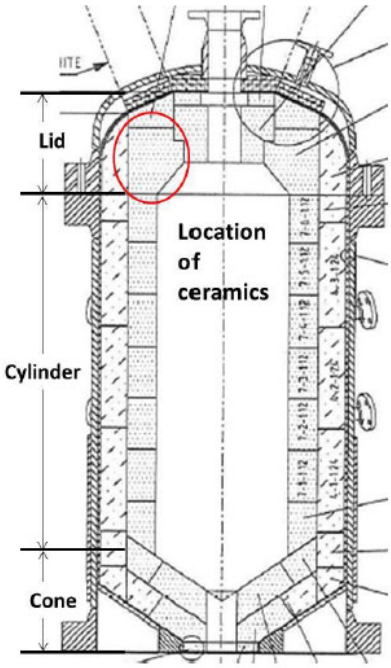
*Million tons dry biomass

Durable containment materials: The key to gasification of high alkali fuels





Source: Ingvar Landälv, LTU