ASSESSMENT OF AN ORGANOSOLV LIGNOCELLULOSE BIOREFINERY CONCEPT BASED ON A MATERIAL FLOW ANALYSIS OF A PILOT PLANT

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The described work investigated ethanol-water fractionation of beech wood as the basic technology for a lignocellulose biorefinery. The main products were glucose, lignin and xylose. The fractionation technology was studied, scaled-up and successfully implemented on pilot scale. Based on an Aspen process simulation of an industrial production plant, the process was assessed economically and ecologically, pointing out the potential benefits of a "lignocellulose biorefinery" and the importance to valorize all the obtained fractions. The process assumptions of the assessment have been tested at the pilot plant and its feasibility was verified.

Keywords: biorefinery, Organosolv pulping, techno-economic assessment, Life Cycle Assessment

INTRODUCTION

In the "German Lignocellulose Feedstock Biorefinery Project",[1,2] it was intended to develop and scale-up a process for maximum material usage of the components of national wood resources. The second phase of the project ends by March 2014 with the successful operation of a pilot plant and the economic and sustainability assessment of the developed processes.[2] During the first phase of the project, beech wood was identified as the most available and suitable species for the conversion into platform components for the chemical industry. The organosolv process, i.e. pulping with ethanol-water, was selected as the most appropriate process for the fractionation of beech wood forest residues with the goal to achieve the highest material usage of the wood components.[1]

The main products of the fractionation process are glucose, lignin and xylose. Within the project, applications of the sugars for various fermentations and chemical conversions were tested. The lignin derived from the oganosolv pulping ("Organosolv lignin") was used in different thermoplastic and thermoset polymers or depolymerized to aromatics. Based on the findings of the first phase, during the second phase (2010-2014), a pilot plant for the fractionation of wood into glucose, lignin and xylose was developed, built and operated at the Fraunhofer CBP at the chemical site of Leuna, Germany. In the pilot plant, industrial wood chips are pulped with mixtures of ethanol and water. Glucose is obtained by enzymatic hydrolysis of the pulp. Lignin is precipitated from the pulping liquor via water dilution or thermal precipitation to receive Organosolv lignin. All liquid process streams are collected for solvent (ethanol) recovery. After ethanol recovery, a resulting fraction of C5-sugars (e.g. xylose) is obtained.

The assessment of the fractionation process was developed simultaneously during the project. Thus economic and ecological key factors could be identified and thus the development of the process supported. This assessment is based on an Aspen process simulation of an industrial scale production plant. The production costs of the fractionation products were determined and the ecological impacts were compared to (petrochemical) reference products using Life Cycle Assessment (LCA) methods.

In this paper, the set-up of the pilot plant and the results of its operation are shortly described followed by the most important results of the assessment period, which have been mainly verified by the first operation results of the pilot plant.

EXPERIMENTAL

Description of the pilot plant

The "lignocellulose biorefinery" pilot plant is designed to fractionate wood chips by ethanol-water-pulping using batch processes. The plant is divided into the following process units, which are shortly described:

- Pulping: The pulping section consists of a batch digester with forced circulation containing 400 L of industrial wood chips. Two additional temperature controlled pressure vessels and one atmospheric tank allow the preparation, preheating and storage of the cooking and washing liquors. The whole pulping section is built in Duplex steel and can be operated at maximum 200 °C and 36 bars. Wood chips can be pre-steamed prior to cooking and the pulping liquors can be displaced from bottom to top through the digester for washing with ethanol and water.
- Washing and dewatering: The pulped wood chips are discharged into a blow tank where the pulp is diluted to ~5% solids content and the fibers are disintegrated by an in-line disperser. The pulp is then dewatered by a screw press.
- Enzymatic hydrolysis: Two temperatures controlled stirred tanks with a volume of 1000 L are used for the enzymatic saccharification of the pulp. A chamber filter press is used to separate the hydrolysis residue from the liquid. Optionally, a fall film evaporator can be used to concentrate the glucose solution for stabilization.
- Lignin precipitation: Lignin is precipitated in a 1200 L stirred tank by the addition to water and filtered using a chamber filter press. Optionally, lignin can be precipitated by evaporating ethanol. The obtained organosolv-lignin can be washed on a membrane filter press and dried in a vacuum drier.
- Solvent recovery: Ethanol is recovered from the filtrates of lignin precipitation and from the washing liquors in a batch rectification column.

RESULTS AND DISCUSSION

Results from scale-up and operation

The pilot plant started its operation in April 2012. Since, the plant is running continuously in day-shift operation. The selection of the scalable unit operations could be confirmed and no major modifications of the plant have been deemed necessary so far. The scale-up of the processes from the laboratory experiments was successful and similar yields and product compositions to the laboratory-scale ones were obtained. Ethanol-water fractionation of 70 kg of debarked beech (*Fagus sylvatica*) wood chips at 170 °C for 100 minutes using 0.5% of sulphuric acid (based on o.d. wood) at a liquor-to-wood ratio of 3.2:1 resulted in the pulp, lignin, glucose syrup compositions described in Table 1.

The mass balances of the pilot plant could confirm the assumptions and basic data used for the process simulation and assessment of an industrial scale production process. Further work of the pilot plant focuses on the optimization of the single processes (e.g. pulping parameters, enzymatic hydrolysis, lignin precipitation and filtration, solvent recovery), the integration of the whole process (closing of heat and water cycles between the unit operations) and the quality of the products. The Fraunhofer CBP is involved with this "lignocellulose biorefinery pilot-plant" in various national and European projects (e.g. Leading-Edge BioEconomy-Cluster, Lignoplast, BioConSepT or Carboprec) in order to promote and develop the integration of the fractionation products into the value chains of the chemical industry and to prepare the industrial implementation of the Organosolv technology.

Assessment

The techno-economic and ecological assessments are based on material and energy flow balances obtained by modelling the whole value chain from forest production to plant gate (cradle to gate analysis). In the following, the most important results of the assessment will be shown (a research project report with more detailed descriptions and results will be published soon[2,3]).

Material and energy flows of industrial lignocellulosic biorefinery plant concept

During the research project a plant process for a capacity of 50 t/h dry wood was developed (flow sheet, Figure 1). The basic information for the process modelling and simulation are based on experimental results obtained in the lab and on pilot scale. The wood is pulped with a 1:1 ethanol/water mixture at 180 °C and 18 bar at a liquor to wood ratio of 4:1 with sulfuric acid as optional catalyst (<1% based on dry wood) followed by the phase separation into pulp and mother liquor.[2,3] 90% of the lignin dissolved in the liquor can be recovered after its precipitation and washing. The pulp is washed and converted to glucose via enzymatic hydrolysis at a conversion rate of cellulose to glucose of 86%.[2,3] The remaining hydrolysis lignin is recovered as additional product stream. All process water is collected for ethanol recovery and the distillation residue is considered as C5-sugar fraction, which contains xylose, oligopentoses and other decomposition products. By assuming this fraction as effluent, high costs for waste water treatment would arise and an economic operation of the biorefinery would not be possible.[2]

Table 1
Compositions of pulp and organosolv-lignin obtained from ethanol-water fractionation of beech wood and sugar syrup from enzymatic hydrolysis of the pulp[2]

Fraction/Components	Pulp	Sugar syrup	Organosolv-lignin
Glucose [% dry mass]	72.1	74.8	0.0
Xylose [% dry mass]	16.9	18.2	3.6
Lignin [% dry mass]	14.9	n.d.	88.5

However, laboratory tests and literature study indicate a possible recovery of the products or use as substrate. As further developments for the valorization of this fraction were not included in the project, we assume that the C5-sugar stream can be further processed. Therefore, revenues and emission credits for the C5-sugars were assumed for the economic and ecological assessment, respectively.[2,3] For the material and energy flow estimation, the process has been simulated in ASPEN PLUS® for pulping with and without sulfuric acid as catalyst. The simulation results for the product streams and energy consumption are shown in Table 2.[2,3]

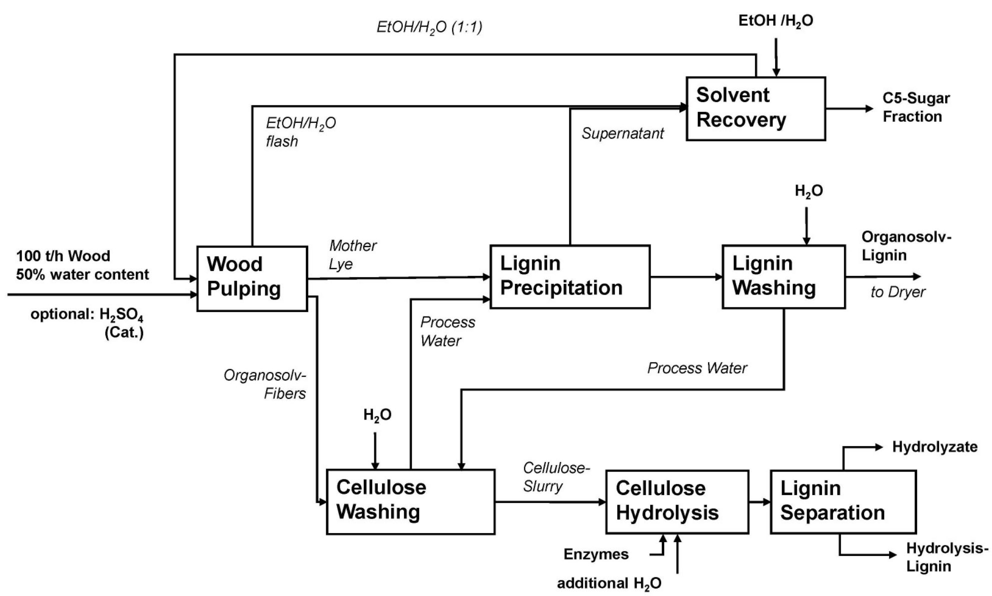

Figure 1: Process flow sheet of plant[2,3]

Table 2
Product flows and energy consumption[2]

		Pulping without H_2SO_4 4 h	Pulping with H_2SO_4 2 h
Products (t/h)	Organosolv-Lignin	7.1	7.3
	C5-Sugars (xylose+oligopentose)	2.1	4.0
	Glucose	19.0	19.0
	Hydrolysis-Lignin	4.5	4.2
Energy (MW)	Steam	32.2	29.2
	Electric energy	7.6	7.6

Economic assessment

For the economic assessment, investments in the biorefinery (inside battery limits) and glucose production costs are estimated for a capacity of 400,000 t dry wood per year and an operation of 8000 h/a. The total investment of the biorefinery for pulping with and without sulfuric acid as catalyst are estimated to 54.7 m€ and 61.0 m€, respectively, including 20% for planning. Glucose production costs are calculated by summarizing investment related costs (including depreciation with an expected life time of 10 years, taxes and insurances (2% of investment), maintenance (2.5%) and capital costs (4%)), costs for raw materials (e.g. dry wood (100 €/t)), operating materials and energy (variable costs), revenues for by products (phenol (630 €/t, 50% of mean phenol spot price EU), hydrolysis lignin (price for heating value, 118 €/t), C5-sugars (200 €/t)), direct and indirect labor costs (4 shifts à 5 employees) and costs for plant overhead (cf. Table 3).

Ecological assessment

All material and energy flows are modelled in Umberto® using the ecoinvent database[4] for the Life Cycle Impact Assessment (LCIA) for selected impact categories. In the LCIA the categories climate change, acidification, eutrophication, land use (competition) according to Guinée *et al.*[5] and the fossil cumulative energy demand (CED)[6] were considered as the most relevant and therefore assessed. The emissions of the biorefinery are mass allocated for its products and compared with reference processes (phenol for lignin, sugar solution from sugar beet for glucose and C5-sugars and heat from wood combustion for hydrolysis lignin).[2, 3]

All assessed impact categories showed 50-80 % lower emissions than references except for land use (competition) because of wood as a feedstock. However, as land use is compared to fossil feedstock, increased land use for forests has to be expected.[2, 3]

Table 3
Estimated glucose production costs (€/t glucose)[2]

Costs (€/t glucose)	Pulping without H_2SO_4 4 h	Pulping with H_2SO_4 2 h
Raw and operation materials	322	325
Energy	120	113
Investment related	73	65
Labor and other	27	27
Revenues (lignin, C5-sugars, hydrolysis lignin)	-685	-325
Glucose production costs	**257**	**218**

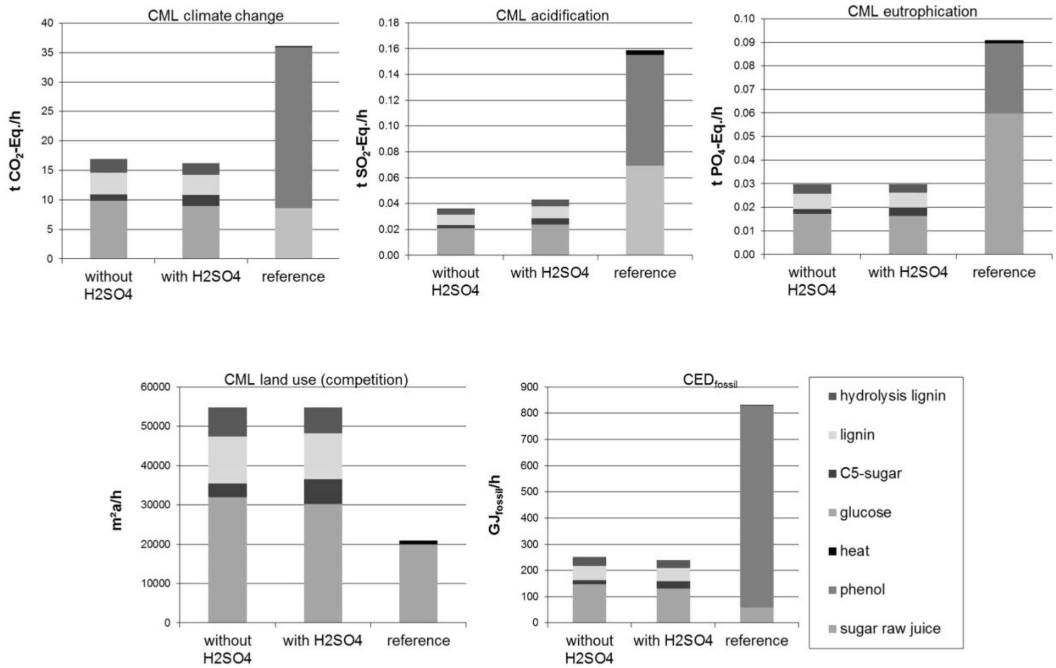

Figure 2: Comparison of ecological impact categories after mass allocation of emissions for biorefinery products (for two pulping variations) and reference products (emission-equivalents/h)[2,3]

CONCLUSION

In the "German Lignocellulose Feedstock Biorefinery Project", a lignocelluloses biorefinery based on an ethanol-water fractionation of beech wood was developed. The fractionation technology was studied, scaled-up and successfully implemented on the pilot scale, confirming the expected results from laboratory scale. The process optimization and integration at pilot plant scale is expected to further improve product yields and mass balances. The fractionation products were used for various promising product developments in the project. Moreover, the fractionation products, which are now available at the kg scale from the pilot plant, are used in different national and European joint projects to continue their integration into the value chains. The economic and ecological assessment of the process shows the potential competitiveness and benefits of a "lignocellulose biorefinery". Thus the developed process appears to be a feasible and promising, from an economic and ecological point of view, integrated material utilization concept for lignocellulosic feedstock. However, the economic assessment also pointed out the importance to valorize all the obtained fractions, including the C5 sugars, and to avoid effluent streams, which may burden the profitability.

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