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**Fractionation of Lignocellulosic Biomass using the OrganoCat Process**

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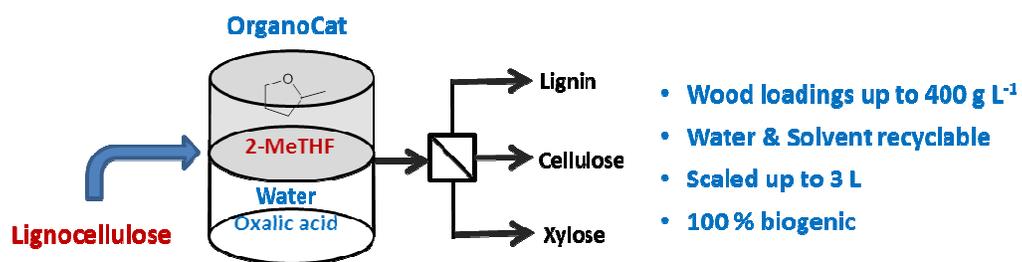
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## GRAPHICAL ABSTRACT



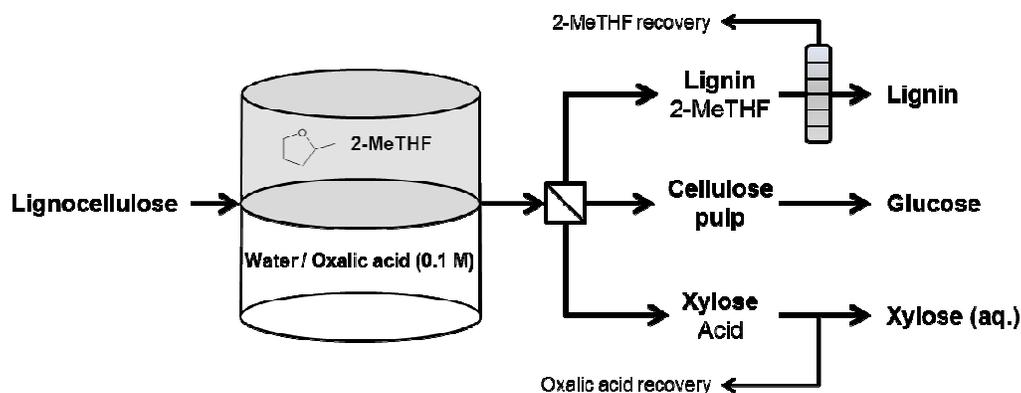
## Summary

The fractionation of lignocellulose in its three main components, hemicellulose, lignin and cellulose pulp can be achieved in a biphasic system comprising water and bio-based 2-methyltetrahydrofuran (2-MeTHF) as solvents and oxalic acid as catalyst at mild temperatures (up to 140 °C). This so-called OrganoCat concept relies on selective hemicellulose depolymerization to form an aqueous stream of the corresponding carbohydrates, whereas solid cellulose pulp remains suspended and the disentangled lignin is to a large extent extracted *in situ* with the organic phase. In the present paper, it is demonstrated that biomass loadings of 100 g L<sup>-1</sup> can be efficiently fractionated within 3 h whereby the mild conditions assure that no significant amounts of by-products (*e.g.* furans) are formed. Removing the solid pulp by filtration allows to re-use the water and organic phase without product separation in repetitive batch mode. In this way, (at least) 400 g L<sup>-1</sup> biomass can be processed in 4 cycles, leading to greatly improved biomass-to-catalyst and biomass-to-solvent ratios. Economic analysis of the process reveals that the improved biomass loading significantly reduces capital and energy costs in the solvent recycle, indicating the importance of process integration for potential implementation. The procedure was successfully scaled-up from the screening on bench scale to 3 L reactor. The feedstock flexibility was assessed for biomasses containing moderate-to-high hemicellulose content.

## 1. Introduction

Lignocellulosic biomass represents a major source for biogenic feedstocks that may allow for the production of chemicals and fuels without compromising the food supply.<sup>1-4</sup> The fractionation of the raw material into the main lignocellulosic components – hemicellulose, cellulose and lignin – provides access to valuable feedstock streams for subsequent (bio)chemical transformation into the desired products.<sup>1-7</sup> In particular, this opens the possibility to explore novel processes and products by analysis of the transformation pathways and target properties on a molecular basis. A number of fractionation methods have been developed, mainly with the aim to provide carbohydrate fractions for further processing *via* fermentation.<sup>1,2</sup> The Organosolv process has been investigated as part of the so-called integrated lignocellulosic biorefinery to provide all three components in a processable manner.<sup>6,7</sup> The protocols normally rely on the use of a homogeneous solvent system comprising water and miscible organic solvents (typically ethanol or acetone). The hemicellulose fraction is hydrolyzed, the lignin is dissolved in the organosolv liquor and purified cellulose is produced as a pulp (after which the lignin can be recovered by precipitation or solvent evaporation).<sup>7</sup> Although the separation steps are efficient, the economics of the process are challenging for practical implementation due to the number of processing steps and the formation of by-products from degradation.<sup>8</sup>

Very recently we disclosed the so-called “OrganoCat process” that delivers hemicellulose sugars (mainly xylose), cellulose pulp, and lignin directly as separate feedstock streams in a single processing step. The concept relies on selective acid-catalyzed depolymerization in a biphasic water-organic solvent system to separate the three components directly into the individual phases aqueous, organic, and solid (Scheme 1).<sup>5</sup>



**Scheme 1.** The concept of the OrganoCat process for the integrated fractionation of lignocellulose to provide directly processable streams of xylose, cellulose pulp, and lignin.<sup>5</sup>

The selective depolymerization of hemicellulose is achieved with a bio-based organic-acid (typically oxalic acid) at mild temperatures not exceeding 140 °C.<sup>5</sup> Under these conditions the less-structured regions (mainly hemicelluloses) are exclusively depolymerized to liberate the monomeric sugars (mostly xylose in case of beech wood) into the aqueous phase.<sup>9</sup> The low temperatures minimize sugar-degrading paths (e.g. formation of furans) that are known to occur in a significant extent only at temperatures higher than 150 °C.<sup>13-16</sup> The more crystalline polysaccharide fractions (celluloses) are not hydrolyzed under these conditions, thus retaining a high molecular weight and remaining insoluble as pulp in the reaction mixture. The disentangled lignin is *in situ* extracted in the organic phase for which the biogenic solvent 2-methyltetrahydrofuran (2-MeTHF) provides a preferred choice.<sup>10</sup> The lignin can be recovered from the 2-MeTHF solution by distillation and the

solvent can be directly recycled,<sup>5</sup> representing a crucial step for the economics of the process.<sup>8</sup> The obtained cellulose pulp can be hydrolyzed by cellulases to render fermentable glucose, demonstrating that a large delignification occurs along the process. The catalyst, oxalic acid, can be recycled from the aqueous effluent by crystallization. Xylose, which remains in solution, can be directly converted into furfural,<sup>11</sup> or fermented to produce itaconic acid<sup>12</sup> without the need for further purification or isolation.

According to the economic assessment reported by some of us, the OrganoCat process was found to provide the potential for a competitive approach as compared to other Organosolv-like approaches even under the initial non-optimized conditions.<sup>8</sup> The raw material loading and the solvent recycling were identified as important factors affecting capital investment and operating costs. The present paper therefore focusses on further optimization of these parameters and the resulting effect on the economic evaluation. It also reports on the scale-up from bench scale to a 3 L reactor and provides a first evaluation of the feedstock flexibility of the OrganoCat approach.

## 2. Results and Discussion.

### 2.1 Experimental Study.

The OrganoCat process is a recently published process for the fractionation of the three main components on lignocellulosic biomass.<sup>5</sup> In a reactor (metal-based and glass-made high-pressure reactors) 100 g L<sup>-1</sup> (referring to the aqueous phase) beech wood lignocellulose is suspended in water with oxalic acid (0.1 M referring to the aqueous phase). For the *in situ* extraction of lignin, pure 2-MeTHF is added to form

an organic phase and the reaction is stirred at 125-140 °C for 3-6 h. After cooling of the reactor, the organic phase is separated by decantation, and 2-MeTHF is evaporated to obtain the lignin fraction. The aqueous phase is filtered to isolate the cellulose-pulp. The solid residue is washed with distilled water until neutral pH and dried until constant weight. Starting from those process parameters of the OrganoCat process,<sup>5</sup> a preliminary optimization reduced the reaction time to 3 h and 0.1 M oxalic acid loadings (instead of 0.2 M) (Table 1). Under the biphasic conditions studied, beech wood loadings higher than 100-150 g L<sup>-1</sup> (related to the aqueous volume) resulted in highly viscous mixtures which are hard to mix. This resulted in significantly lower xylose yields, most likely at least in part due to mass transfer limitations.

**Table 1.** Preliminary optimization of the production of xylose, glucose and lignin in the OrganoCat process.

Catalyst loadings [mol L <sup>-1</sup> ]	water: 2-MeTHF	Reaction time   Reaction temperature	Wood loadings [g L <sup>-1</sup> ]	Aqueous phase		Organic phase
				Xylose yield [wt%] <sup>a</sup>	Glucose yield [wt%] <sup>a</sup>	Lignin yield [wt%] <sup>a</sup>
0.2	1:1	3 h   140 °C	100	18	1.2	13
0.1	1:1	6 h   125 °C	100	19	0.5	13
0.1	1:1	3 h   140 °C	100	17	1.2	14
0.2	1:1	3 h   140 °C	200	15	2.0	9
0.1	1:0.5	3 h   140 °C	100	18	1.2	10

<sup>a</sup>: wt% related to total beech wood loadings.

Depending on the availability of local resources, different sources of biomass may be envisaged for valorization. In order to evaluate the compatibility of the OrganoCat approach with different biomass sources, several raw materials were

fractionated under the optimized processing conditions. Results are summarized in Table 2.

**Table 2.** Production of xylose, glucose, and lignin with different lignocellulosic materials *via* OrganoCat in a glass-made high-pressure reactor. Conditions: Biphasic system water/2-MeTHF (1:1 v/v, 40 mL total volume), oxalic acid, 0.1 M in the aqueous phase, lignocellulose, 100 g L<sup>-1</sup> in the aqueous phase, 140 °C, 3 h. Xylose and glucose production were analyzed by HPLC (see Experimental). Lignin production was determined gravimetrically after removing 2-MeTHF *via* distillation.

Raw material	Aqueous phase		Organic phase
	Xylose [wt%] <sup>a</sup>	Glucose [wt%] <sup>a</sup>	Lignin [wt%] <sup>a</sup>
Beech wood ( <i>Fagus sylvatica</i> )	17.0	2.3	11.0
Theoretical content (lit)		(ca. 21) <sup>17</sup>	(ca. 26) <sup>17</sup>
Mate tea ( <i>Ilex paraguariensis</i> )	5.9	2.1	10.0
Theoretical content (lit)		(ca. 7) <sup>21</sup>	(ca. 28) <sup>21</sup>
Reed ( <i>Phragmites australis</i> )	19.0	1.2	14.0
Theoretical content (lit)		(ca. 20) <sup>18</sup>	(ca. 17) <sup>18</sup>
Spruce ( <i>Picea</i> sp.)	2.6	0.3	0.0
Theoretical content (lit)		(ca. 9) <sup>22</sup>	(ca. 27) <sup>22</sup>

<sup>a</sup>: wt% related to total biomass loadings.

In addition to beech wood, another successful substrate was reed (*Phragmites australis*), a plant that displays an efficient cultivation profile, and therefore can be envisaged for further growth and (bio)chemical valorization.<sup>[18,19]</sup> Yields in xylose (aqueous phase) and lignin (organic phase) were in the same range as values obtained for beech wood, and the process could be applied without any modification. Similarly, the post-brewed Mate tea leaves could be used directly after drying under the standard conditions. Mate tea (*Ilex paraguariensis*) represents a largely consumed and popular tea infusion in South America. Up to 280.000 ton of mate tea leaves are consumed annually in some countries (e.g. Argentina), and then discarded.<sup>20</sup> Assuming an average content in hemicellulose of ca. 7 wt% in *I. paraguariensis*,<sup>21</sup> it can be concluded that the xylose production was nearly quantitative and lignin recoveries were also in the same range as those obtained for beech wood. Spruce (*Picea* sp.), however, proved less suitable for the OrganoCat process due to its low content in hemicellulose and high lignin content (in particular guaiacyl lignin).<sup>22</sup> Under the mild conditions of the standard procedure, 1 mm spruce particles were not disintegrated and neither carbohydrates nor lignin was brought into solution effectively. Thus, a high content of hemicellulose is highly beneficial for the OrganoCat approach (e.g. for hardwoods). The processing of other biomasses would eventually demand more severe conditions – at the cost of degradation products formation –, leading to more Organosolv-type approaches.

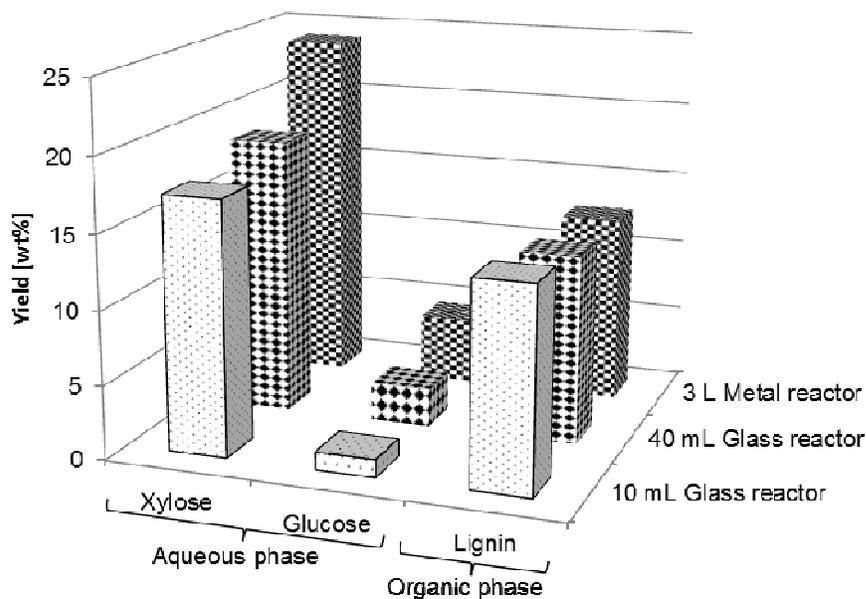
Using beech wood as prototypical feedstock with large potential for the region of this study, the fractionation system was scaled up to 3 L (total volume) in a stainless-steel high-pressure reactor (Figure 1).



**Figure 1.** Reaction-setup at 3 L scale. Left: Filled glass inlay with substrate, catalyst and solvents; Right: Closed high pressure reactor.

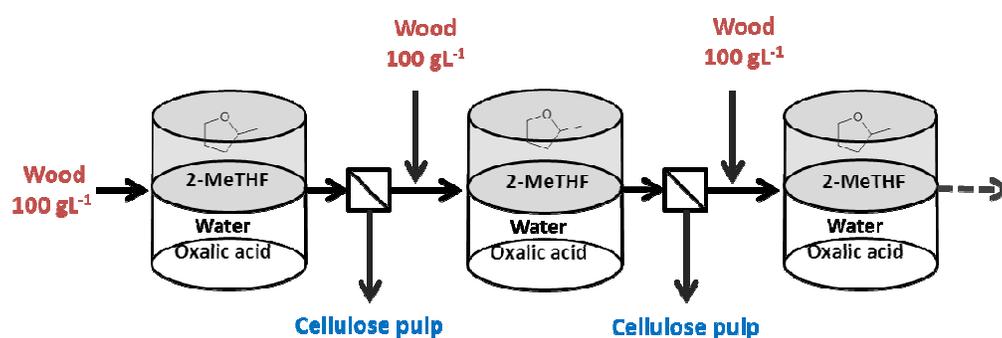
Beech wood chips (150 g, accounting for loadings of  $100 \text{ g L}^{-1}$  in the aqueous phase) were suspended in a solution of 1.5 L of water and oxalic acid (0.1 M), and 1.5 L 2-MeTHF was subsequently added in a glass insert (Figure 1). Processing temperature was kept at  $140 \text{ }^\circ\text{C}$  during 3 h and the mixture stirred by a sealed agitator shaft. Under these conditions, the pressure within the reactor increased up to ca. 10 bar at  $140 \text{ }^\circ\text{C}$  without the need of applying external gas pressure. After cooling and filtration, 79.6 g of cellulose pulp (53 wt% of the initial biomass loading) was achieved, fully consistent with cellulose amounts expected in beech wood lignocellulose,<sup>17</sup> and also consistent with the quantities obtained on smaller scale in the glass-made reactors (Figure 2). Similarly, the lignin fraction obtained from the organic phase was largely identical to that obtained in bench-scale experiments. The amounts of carbohydrates liberated into the aqueous phase at larger scale were slightly higher both for glucose (1.2% and 4.5% respectively) and xylose (ca. 18 to ca.

22 wt%, Figure 2). This may at least in part reflect better mass transfer and homogeneity of the mixture due to improved mixing in the 3 L reactor setup.



**Figure 2.** Carbohydrate yields from the aqueous phase (xylose and glucose) and lignin yield from the organic phase 2-MeTHF obtained at different scales and in different high-pressure reactors. Conditions: Biphasic system water/2-MeTHF (1:1 v/v), oxalic acid, 0.1 M in the aqueous phase, beech wood 100 g L<sup>-1</sup> in the aqueous phase, 140 °C, 3 h. Xylose and glucose production were analyzed by HPLC (see Experimental). Lignin production was determined gravimetrically after removing 2-MeTHF *via* distillation.

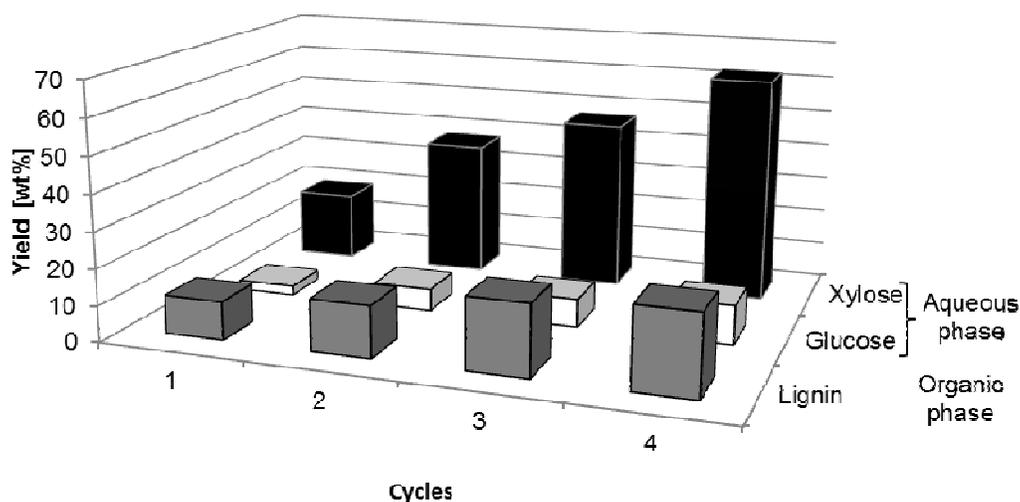
A key parameter for the efficacy of the process is the biomass loadings. To further optimize the biomass-to-solvent ratio, a repetitive batch approach was envisaged: Beech wood loadings of  $100 \text{ g L}^{-1}$  were processed during 3 h, the solid suspended cellulose pulp was then filtered, and both organic and aqueous effluents (together with xylose and oxalic acid in aqueous solution and with lignin dissolved in 2-MeTHF) were directly recycled by adding another beech wood loading of  $100 \text{ g L}^{-1}$  (Scheme 2).



**Scheme 2.** Repetitive-batch mode for fractionation without isolation of the product streams from the water and organic phase to increase to biomass-to-solvent ratio.

This repetitive-batch mode of operation could be successfully applied in four consecutive cycles of lignocellulose processing. This corresponds to overall beech wood loadings of  $400 \text{ g L}^{-1}$  in respect to the initially charged amounts of water, catalyst and organic phase. Consequently, significantly larger product concentrations of carbohydrates (xylose and some glucose) in the aqueous phase and lignin in the organic phase are obtained with this procedure (Figure 3). Remarkably, formation of

solid humins or furfural – at the cost of xylose –, was not observed in the aqueous phase under the mild reaction conditions applied.<sup>23</sup> Xylose concentrations of up to ca. 55-70 g L<sup>-1</sup> were reached after four processing cycles. Overall, largely optimized biomass-to-liquid performances are reached, thus contributing significantly to the economics of biorefineries, as well as to their ecological footprints in solvent utilization and energy demand (see below).



**Figure 3.** Amounts of xylose, lignin and glucose obtained with the OrganoCat process in repetitive-batch mode reusing water and 2-MeTHF during four cycles (see Scheme 2). Reaction conditions: 1<sup>st</sup> cycle: Biphasic system water/2-MeTHF (1:1 v/v, 10 mL total volume), oxalic acid 0.1 M in the aqueous phase, beech wood (6 mm), 100 g L<sup>-1</sup> in the aqueous phase, 140 °C, 3 h. Following cycles: Filtration (paper filter, MN615), addition of new beech wood (6 mm) loadings, 100 g L<sup>-1</sup> in the aqueous phase, 140 °C, 3 h. Xylose and glucose analyzed in the aqueous solution by HPLC (see Experimental). Lignin determined gravimetrically upon evaporation of 2-MeTHF.

With regard to the lignin extraction efficiency – and the subsequent pulp accessibility to cellulases –, some noticeable aspects are observed after the 3<sup>rd</sup> cycle of the OrganoCat process. The efficiency of the lignin extraction decreases during the 3<sup>rd</sup> and the 4<sup>th</sup> cycle (Figure 3), presumably due to saturation of the 2-MeTHF phase with lignin (what could be circumvented by additional lignin extractions among cycles). This aspect leads to higher amounts of lignin remaining at the cellulosic pulp. Thus, a lower efficiency during the enzymatic digestion may be expected. Actually, enzymatic hydrolytic rates of the pulp obtained during the first cycle ( $\sim 0.9 \text{ g glucose L}^{-1} \text{ h}^{-1}$ ) are  $\sim 4$ -fold higher than those rates observed for the pulp obtained after the 4<sup>th</sup> cycle ( $\sim 0.25 \text{ g glucose L}^{-1} \text{ h}^{-1}$ ). Although the conceptual approach in aqueous and organic phase recycling (proposed in this work) is valid and successful, obviously fine-tuning adjustments are needed to enable high quality fractions for further valorization. In this respect, each biomass type will certainly need specific conditions for a proper pretreatment.

## 2.2 Energetic and economic assessment.

The experimental study demonstrated the potential for scale-up and addressed three process parameters to gain higher efficiency: the amount of oxalic acid catalyst was reduced by 50%, the residence time was shortened from 6 h to 3 h, and the biomass-to-liquid ratio was increased from  $100 \text{ g L}^{-1}$  to  $400 \text{ g L}^{-1}$  in repetitive batch mode. The effect of those modifications on the energetic and economic analysis was assessed based on an updated version of the process model reported by Viell *et al.*<sup>8</sup>

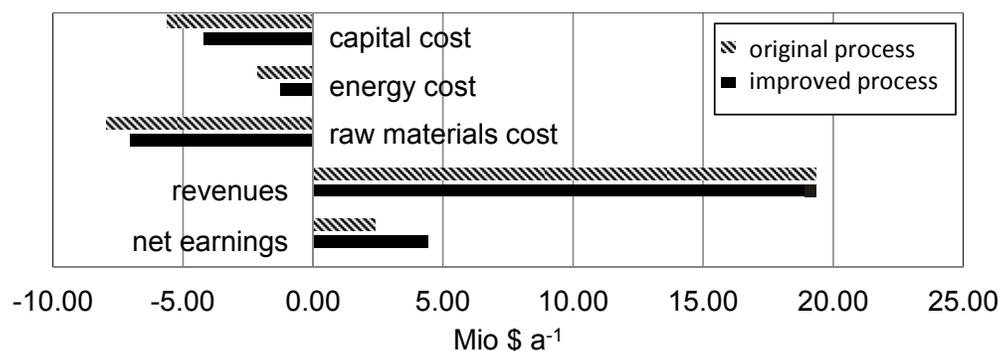
The smaller concentration of oxalic acid in the process is clearly favorable, but the effect on economics is relatively small. With the assumption that the loss of acid is proportional to the concentration of acid in the fractionation reactor, the need for fresh oxalic acid will decrease from 76 kg/h to 13 kg/h. However, the materials costs are still dominated by the feedstock costs and cellulases in the downstream processing. Furthermore, the oxalic acid recovery is still uncertain and would have to be investigated for a more thorough assessment of the oxalic acid consumption. Therefore, the savings on oxalic acid alone do not significantly increase the economic balance of the process concept.

A more significant effect results from the reduction of the residence time to fractionate the biomass, as this enables the utilization of a much smaller reactor. The existing conceptual design<sup>8</sup> assumes a simple stainless steel pressure reactor with countercurrent flow for in-reactor washing of the pretreated biomass. A residence time of 3 h instead of 6 h requires only half the volume at the same productivity. The price of the reactor, which was originally estimated to be 2.4 Mio \$, is then decreased to 1.6 Mio \$. Although the design of the individual unit operations is not yet very detailed and the solids handling involves a certain degree of uncertainty of the price estimation, a total investment in the range of 30 Mio \$ can be estimated for the overall process. Thus, the savings in the residence time translate into savings on CAPEX in the range of 2-3%.

The biomass-to-liquid is the third parameter, which is increased in this study from 100 g L<sup>-1</sup> to 400 g L<sup>-1</sup> due to consecutive solvent utilization. The higher loading with products in the two liquid phases results in smaller solvent streams in the solvent recycling at the same productivity. The original process analysis<sup>8</sup> already pointed out that the solvent recycling is one of the most expensive process steps due to high

energy consumption and costly compressors in the heat integration scheme. The model-based simulation has been updated accordingly to assess the impact of this parameter.

The result is a primary energy demand of the heat-integrated solvent recovery. The equipment for vapor recompression, heat exchange and solvent evaporation can be designed smaller and requires less energy. The improved OrganoCat process therefore consumes only 15% of the energy demand of wood instead of 24% in the original design, which is clearly a significant increase in energetic efficiency.



**Figure 4.** Comparison off the costs of the original OrganoCat design (dashed bars) with those of the improved processing (solid bars).

The positive effect on the energy balance translates directly into improved process economics as displayed in Figure 4. It compares the costs and revenues of the original design with those of the improved process. While the revenues for products and wood costs are identical, differences are obvious in the annual energy and capital cost and total raw material cost. The latter is due to a smaller oxalic acid make-up due to fewer recovery cycles. It only results in 10% savings in raw material costs. The

smaller capacity of the solvent recycle results in lower investment cost thus result smaller annual capital cost of 4.17 instead of 5.64 Mio \$ a<sup>-1</sup>. The energy costs even show a reduction of 42% due to the reduction of expensive electricity demand to run the compressors in the heat integration scheme. Additionally, less energy is required to heat the smaller solvent stream returning to the pretreatment section.

Altogether, the lower costs at the same productivity lead to higher net earnings of 4.4 Mio US\$ a<sup>-1</sup>, which is an increase by 87% in comparison to the original design with 2.35 Mio US\$ a<sup>-1</sup>. The corresponding payout time (POT) of the investment is 2.3 years and a return on investment (ROI) of 17% is computed. Hence, the improved process parameters result in approximately twice the economic attractiveness in comparison to the previous design (POT=3.7, ROI=7%). The most effective impact on the improvement among the studied parameters results from the increased capacity of the solvent and hence lower recycling effort.

An important factor in the economic analysis of biorefinery concepts is the value associated with the lignin stream. In accord with general practice,<sup>2</sup> the lignin has been treated in the present analysis as a high-value product at 1000 US\$ t<sup>-1</sup>. Consequently, 35% of the overall revenue is related to the lignin fraction, representing a major risk factor in the analysis. However, we note that the improved OrganoCat process described here exhibits net earnings for the process even at a lignin price of 90 US\$ t<sup>-1</sup>, which would correspond to the energetic value of lignin. This is an important fact, as even if lignin were degraded or condensed along subsequent processing steps, the *simple* energetic value would still provide promising economic figures.

The cost distribution in Figure 4 also shows that the raw materials are a dominant factor determining 57% of the overall costs. This value is close to what is experienced with mature chemical processes, i.e., 60-75%.<sup>24,25</sup> Therefore, further optimization potential will be hard to achieve by discrete improvements of the fractionation process. Rather, the integration of the fractionation process with the downstream processing likely enables further enhancement in efficiency and economic attractiveness. The integrated conversion of the solutions of carbohydrates and lignin without isolation of the products from the process streams offers such possibilities to further reduce the energy demand. For example, the aqueous xylose stream derived from OrganoCat has been successfully used without isolation for FeCl<sub>3</sub>-catalyzed conversion to furfural<sup>11</sup> and for the fermentation to itaconic acid.<sup>12</sup> Further studies in this direction are currently under way in our laboratories.

### 3. Experimental Section

*3.1. Materials.* Oxalic acid, 2-methyltetrahydrofuran (2-MeTHF), D-glucose and D-xylose were obtained from Sigma-Aldrich and used without further purification. Beech wood, reed and spruce were obtained from local suppliers, the particle size was reduced with a cutting mill with a 10 mm sieve and dried at 50 °C until weight constancy (ca. 24 h). Mate tea leaves were obtained in supermarket, after tea consumption and drying, the leaves were used for fractionation systems.

#### *3.2. Exemplary procedure for lignocellulose fractionation, OrganoCat.*

In a glass-made high pressure reactor 2 g beech wood lignocellulose and 0.1804 g (0.002 mol) of oxalic acid were suspended in 20 mL distilled water. 20 mL

2-MeTHF was added to form an organic phase and the reactor was closed. The reactor was heated to 140 °C for 3 h. After cooling of the reactor, the organic phase was separated by decantation. The aqueous phase was filtered to isolate the cellulose-pulp, and xylose and glucose concentrations were determined in the aqueous phase *via* HPLC. The solid residue was washed with distilled water until neutral pH and then dried until constant weight. 2-MeTHF was evaporated to obtain the lignin fraction.

*3.3. Procedure for lignocellulose fractionation at 3 L scale.* In a glass inlay 150 g beech wood and 13.5 g (0.15 mol) of oxalic acid were suspended in 1.5 L distilled water. 1.5 L 2-MeTHF was added to form an organic phase, the glass inlay was inserted into the metal-based high-pressure reactor and the reactor was closed. The reactor was heated to 140 °C for 3 h. After cooling of the reactor, the organic phase was separated by decantation. The aqueous phase was filtered to isolate the cellulose-pulp, and xylose and glucose concentrations were determined in the aqueous phase *via* HPLC. The solid residue was washed with distilled water until neutral pH, dried until constant weight, extracted with 2-MeTHF (1.5 L) at room temperature for 24 h and then filtrated and the solid residue dried again until constant weight. The organic phase was united with the other, and 2-MeTHF was evaporated to obtain the lignin fraction.

*3.4. Xylose and glucose determination (HPLC).* HPLC analysis was conducted on a Jasco HPLC equipped with a SUGARSH1011 column with a 0.01 wt% aqueous acid solution as eluent. The flow rate was set to 0.6 mL min<sup>-1</sup> and samples of 30 µL were injected. Amounts of xylose and glucose present in the hemicellulose fraction were determined based on calibration curves built using commercially available authentic substrates.

#### 4. Conclusions.

This paper has successfully explored several practical aspects of the one-step oxalic-acid catalyzed biphasic lignocellulose fractionation system (OrganoCat). The process was experimentally found to be compatible with different biomass sources as long as the hemicellulose fraction is accessible to the mild depolymerization conditions. The concept proved to be scalable, because the results from bench-scale screening and optimization could be successfully transferred to the 3 L scale. Biomass loadings of up to 100-150 g L<sup>-1</sup> (based on the aqueous phase) were found to present practical limits for efficient agitation of the reaction mixture. However, recirculation of the water and the organic phase after filtration of the cellulose pulp and adding new lignocellulose loadings (repetitive-batch mode) was demonstrated, corresponding to (at least) 400 g L<sup>-1</sup> of biomass loadings with a largely improved biomass-to-catalyst and biomass-to-solvent ratios. Model-based process analysis and economical assessment shows reduced capital costs and energy demand in the solvent recovery section at the same productivity. Thus, the annual earnings increase from 2.4 to 3.7 Mio US\$ y<sup>-1</sup> and the economical break-even point of the process is reduced to a lignin price of 140 US\$ t<sup>-1</sup>. Further improvements in reactor design and integration with downstream processing can be envisaged to provide even more optimized (economic) outcomes with a practical perspective for biorefineries.

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