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DOWNSTREAM PROCESSING OF BIOREFINED LACTATE FROM LAKE BOTTOM ZERO FIBER DEPOSIT - A TECHNO-ECONOMIC STUDY ON ENERGY EFFICIENT PRODUCTION OF GREEN CHEMICALS

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ABSTRACT

This paper is built on a study performed to investigate the possibilities to take care of fiber sludge from a former pulp mill in the city Tampere, Finland. A pilot plant biorefinery process has been developed by Finnoflag and tested during 2018-2019 for the conversion of the cellulosic material into useful chemical products by the help of a microbial process. The results from this are used as input to a study on downstream processing performed through lab and pilot plant tests of different technologies, as well as system simulations. An economic analysis is also performed with respect to installation costs for the different alternatives of downstream processing – ion exchange, distillation and membrane filtration. It was found that all three processes would successfully concentrate the main product (lactic acid), however distillation would likely be energy intensive, and ion-exchange costly for materials. Therefore, the recommendation from this study is to focus on reverse osmosis after ultrafiltration, with the addition of evaporation if necessary. Besides the resulting chemicals fraction the biorefinery produced hydrogen, and the residues could be converted effectively into methane. These gases provided energy for the industrial unit and could be converted into energy, too. On the other hand, the feasible production of the chemicals gave an economic justification for the energy producing units.

Keywords: mixed fermentation, membrane filtration, distillation, ion exchange, energy recovery, biogas

1. INTRODUCTION

In Tampere city, Finland was a pulp mill operating for almost a century from the early 1900. This operation left fiber sludge of the so called zero fiber in the range of 1.5 million ton at the bottom of the bay outside the plant. The city wants to remove this sludge to avoid smell and other negative impact on those living around the bay, where the city plans for 25 000 new apartments. Previously experiments have been made with bio-refinery using mixed fermentation plant to produce organic acids in the EU-project ABOWE (Hakalehto, 2015), and different methods for separation of organic acids have been presented for ion-exhangers by Bishai et al (2015). John et al (2008) have tested Amberlite IRA 67, which is a weak anion exchanger resin, and received recovery of lactic acid to 92.5- 98.7 %, which shows that a good recovery can be achieved under suitable conditions.

Kumar et al (2006) have described a method for reactive distillation. Daengpradab and Rattanaphanee (2015, 2018) have investigated how to optimize the production. Lee (2015) has studied how the process can be integrated in a thermomechanical pulp (TMP) plant, while Komesu et al (2017) have investigated “non-conventional” distillation methods. There are many reports on membrane filtration of fibrous material using microfilter membranes. This is normally the necessary pre-filtration to removed particles, before recovering the acids by using RO (reverse osmosis). Lim and Bai (2003) tested how to keep a high flux with hollow fiber MF membranes when used for activated sludge processes with different fiber size. They concluded that a combination of clean water backwashing, sonication and chemical cleaning with alkali and acid could achieve almost complete flux recovery. Héran M. and Elmaleh (2001) have tested ceramic membranes with tubular alumina membranes (0.02, 0.05 or 0.2 µm), with internal skin for similar application. They found that cross flow flux was as good as back flushing. Xu et al (2012) has studied separation of organic acids from condensate from pharmaceutic production and have concluded that pH of feed water had a strong effect on the desalination rate and total organic carbon (TOC) removal rate.

In this study, a bio-refinery pilot was operated with samples from the lake and evaluated different downstream processing methods for the fermentation products. In this case the bio-refinery process produced a metabolic mixture, and the application is directly towards recovery of lactic acid from fermentation broth with as little as possible of other substances. Lactic acid can be converted to polylactic acid, which is a base chemical for production of bio-degradable plastics. However, the focus in this paper is on the techno-economic evaluation of the down-streaming process comparing reverse osmosis, ion exchange and distillation.

2. MATERIALS AND METHODS

2.1 SYSTEM DESIGN

Various challenges of the mixed fermentations could provide means for economically feasible applications, if the valuable products could be separated in a cost-effective way (Hakalehto and Jääskeläinen, 2017). In the current work, the hydrolyzed zero fiber was used as a substrate for the fermentation process. The energy demand of the bio-refinery and chemical plant could be satisfied by the production of methane biogas and possibly bio-hydrogen, as well as by the electricity from these processes. The pilot plant was based on a batch reactor design, and the data was extrapolated to show the design of continuous reactors with different capacity. Figure 1 demonstrates the balances for 2 ton, 20 ton and 200 ton 10% dry solids/h the alternative downstream processes options and how the system can be integrated into a combined heat and power system.

Due to the high concentration of lactate compared with the other organic acids in the fermentation broth, focus on refinement of lactate and further conversion to polylactic acid was deemed the primary goal of these processes.

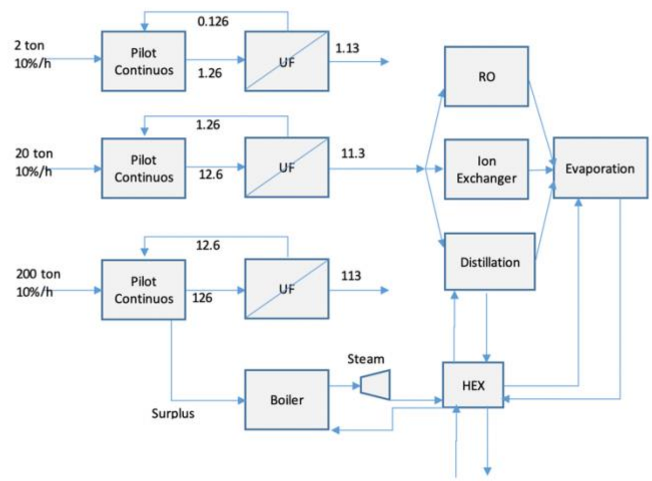

Figure 1. Pilot plant process design. UF = ultrafiltration, RO = reverse osmosis, HEX = heat exchanger.

2.2 EXPERIMENTS

The batch bio-refinery pilot was used for eight different tests runs, though they were found to produce similar product compositions. Fermentation broth from batch test #3 was used for the experimental work on downstream, with the following composition: glucose 2.3 g/L, acetate 2.5 g/L, lactate 22.1 g/L, ethanol 0.13 g/L and butyrate 1.1 g/L. The product concentrations were confirmed using NMR (Nucleic Magnetic Resonance) spectroscopy as described elsewhere (Laatikainen et al., 2016).

The first step in the proposed process was ultra-filtration to separate all suspended solids from the fermentation broth. The ultra-filtration was performed using ceramic tubular membranes in a pilot plant, with 4 m/s cross flow at the membrane surface. The flow rate through the membrane flux in the beginning was 150 L/m^2 x h.   The membrane flux decreased with time to 100 L/m^2 x h to produce a concentrated liquid free of suspended particles. From earlier experience we know that the flux can be kept at these levels long term with washing frequently to avoid severe fouling.

After the ultra-filtration, three processes for purification of lactic acid and other potential organic acids were proposed: ion-exchange, distillation and reverse osmosis. The first downstream process explored was ion exchange with Amberlite IRA 67. As feedstock fermentation broth from test run 3 after 40 h was used. The main acids of interest for this project were lactate, acetate, propionate and n-butyrate. Amberlite IRA67 can be regenerated using 2M NaOH, to remove the absorbed organic acids in a concentrated form.

The second downstream process that was proposed was distillation. To perform distillation experiments ASPEN plus simulation software was used, which gave reasonable design dimensions for a broth with the following composition: glucose 2.3 g/L, acetate 2.5 g/L, lactate 22.1 g/L, ethanol 0.13 g/L and butyrate 1.1 g/L.

3. RESULTS AND DISCUSSION

3.1 EXPERIMENTAL RESULTS

The results of the Amberlite IRA67 tests are shown in Figure 2. Lactate, acetate, propionate and n-butyrate concentrations were initially 14.2, 2.2, 0.5 and 0.4 g/L, respectively. The figure shows effluent concentrations of each of the acids at one bed volume intervals of 15 mL. The pH decreased from 7 to 4.7 as the ion exchange media became saturated and more acids passed through the column into the effluent.

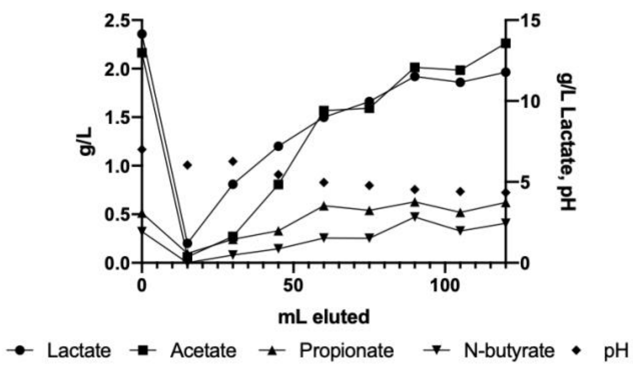

Figure 2. Experiments with Amberlite IRA67

This test showed that the absorption of acids was high for the first bed volume but then decreased with time. Compared with literature tests using the same adsorbent, the feedstock in this case is a complex mixture which includes substances that may influence the ion-exchange (i.e. humic acid).

After absorption, the ion exchange column was washed with NaOH to release the bound organic acids. Figure 3 presents the resulting concentrations of the organic acids during elution with the regeneration solution. Complete regeneration occurred after 4-5 bed volumes. The process was considered inefficient for this feedstock due to the other substances that impeded the absorption.

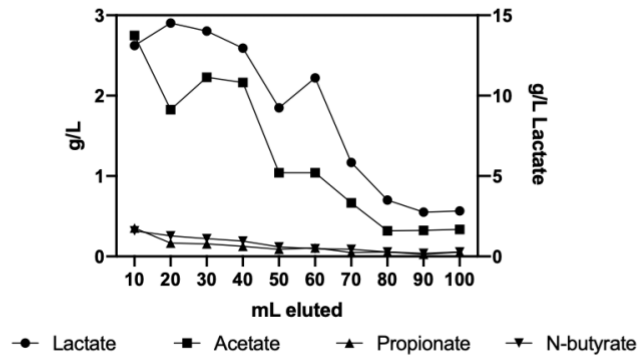

Figure 3. Elution of the absorbed organic acids

One of the main considerations for the viability of distillation in this application are the boiling points for the organic acids: acetic acid 118°C, lactic acid (2-hydroxipropionic acid) 122°C, butyric acid 164°C. Acetic and lactic acid are very close in boiling point making it more difficult to separate using distillation. The actual vessel for a 2 ton liquid/h would be a 14 m tower with 0.6 m diameter with 20 trays. Glucose will be in the bottom fraction, ethanol in the top fraction, although very low concentrations can be expected in a real plant. If necessary, an evaporator could be used to concentrate the acids further.

Finally, reverse osmosis was considered as an alternative for concentration of the product acids. It would not provide separation of different organic acids from each other but would be a simple process that could remove water to provide a better solution that can be separated further with distillation.

3.2 ECONOMIC CONSIDERATIONS

To determine the economic viability of each downstream process, a feedstock stream of 20 ton/h was used. Investment cost was reported in k€ and energy use in kW in Figure 4. The energy for the process could be obtained from the residual fraction of the fermentation broth, which consists of less than 50% of the original biomass (Hakalehto, 2015). The combustible gases and electricity produced from the side stream of the biorefinery process could provide most of the energy needed for the unit.

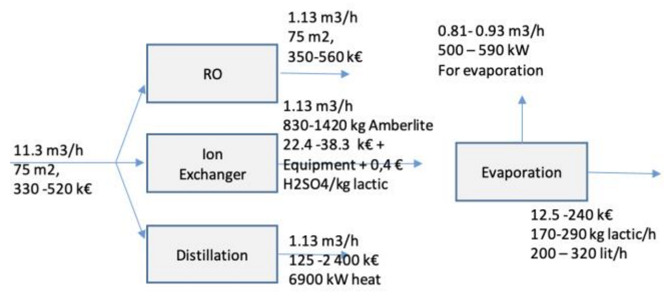

Figure 4. Detailed downstream processing analysis

The detailed downstream processing analysis begins with an ultrafiltration step thus the first set of costs are associated with membrane surface area and investment costs for the ultrafiltration system (far to the left). The figure then splits to evaluate each of the downstream alternatives: RO (reverse osmosis), ion exchange (Amberlite IRA67), and distillation. Reverse osmosis costs were primarily for electric power, cleaning chemicals, and filter replacement (assuming 3-5 year lifespan). The ion exchange process includes cost for the actual ion-exchange material, the column, and cost for regeneration with NaOH. Sulfuric acid can be produced from exhaust gas and was considered per kg of lactic acid. For the distillation the investment cost as well as the kW heat demanded for the operations were estimated. After either of these three alternatives there is a final step to concentrate the product using evaporation. The investment cost as well as the heat demand for evaporation is also given.

4. CONCLUSION

From the evaluation we can see that we can reach reasonable concentration of products, especially lactic acid. This can be refined through downstream processing based on the energy from the biorefinery residues. From estimates reverse osmosis after ultra-filtration and possible further concentration by evaporation is most economic. Distillation as main step is costly with respect to energy but could be interesting as a final step for separating fractions in the concentrate to produce concentrated lactic acid.

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