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Alcohol recovery from fermentation broth with gas stripping: system experimental and optimisation

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Abstract. Effective liquid biofuel production from various lignocellulosic waste resources is dependant not only on pre–treatment and hydrolysis but also on effective removal of alcohols from the fermentation media. Distillation and rectification is not suitable in low alcohol content systems (butanol production with clostridia) or in cases when the fermentation is performed in a continuous mode. One of the technologies offering continuous, *in situ* removal of alcohol is gas stripping. Despite the recognition of this technology, it is still under evaluation and adjustment. Thus, the aim of this study was to evaluate if gas stripping technology at rapid flow conditions is efficient enough to recover ethanol from the fermentation media. The results showed that 60 l min^{-1} flow rate was optimal to recover more than 45% of the available ethanol in 8 hours of stripping with nitrogen gas. The technology was efficient if the ethanol content in the fermentation broth was 10 wt%. At lower concentrations the recovery showed to be inefficient. Application of CO_2 as the stripping gas was not suitable for ethanol recovery and should be tested prior use. In conclusion, the application of rapid N_2 flow rate for gas stripping of ethanol from fermentation media showed to be an efficient technology and could replace long time, low flow rate stripping.

Key words: lignocellulosic biomass, biofuel, gas stripping, alcohol recovery.

INTRODUCTION

Lignocellulosic biomass pre-treatment and hydrolysis is regarded as one of the most expensive steps in lignocellulosic biofuel production. To minimize the costs and increase production yields, various chemical, mechanical and biological technologies or their combinations have been introduced and tested over the last twenty years (Mood et al., 2013). At the same time, a significant source of cost and energy consumption in current alcohol production is the collection, dewatering and purification of the main product–alcohol (Taylor et al., 2010). Ethanol content in the culture broth usually varies from 2.5 to 10 wt%, however, to be used as fuel it has to have a 99.5% purity (Sanchez & Montoya, 2013), thus, purification costs can reach up to 40–60% of total plant energy consumption (Kumar et al., 2015). Similarly, butanol production via acetone-butanol-ethanol (ABE) fermentation is limited to 2 wt% (Huang et al., 2014) of alcohol in the broth. Above this concentration it becomes inhibitory to the fermenting bacteria.

Alcohol concentration and purification can be achieved with classical distillation and subsequent rectification (Sanchez & Montoya, 2013) or with *in situ* technologies

that can be run in a continuous mode and can even remove the toxic products selectively (de Vrije et al., 2013). These technologies are pervaporation, adsorption, vacuum fermentation, extraction with organic solvents or supercritical CO_2 extraction (Ritslaid et al., 2010; Sanchez & Montoya, 2013; Kumar et al., 2015). Pervaporation has been successfully applied *in situ* during ABE fermentation. Extractive fermentation can be coupled with simultaneous saccharification. Nevertheless, these technologies usually account for high energy and maintenance costs or, as for adsorption, the search for the most efficient and selective material is still ongoing (Sanchez & Montoya, 2013; de Vrije et al., 2013).

Another possibility for recovery of alcohols *in situ* is gas stripping, where oxygen free gas (N_2, CO_2 or H_2) circulates through the fermentation liquor. It is a relatively simple process that does not harm the fermenting culture and can be operated in a continuous mode even on an industrial scale (Quershi & Blaschek, 2001). Moreover, there is no need for an expensive equipment or reagents. Gas is sparged into the bioreactor through a sparger, which creates bubbles and these after breaking induce vibration of the liquid and subsequent volatile removal (Ezeji et al., 2005). Parameter sensitivity analysis has shown that the dominant variable in the process is gas flow rate (Liao et al., 2014). Unfortunately, rapid gas flow rate in the reactor removes large amount of water together with the ethanol, resulting in ethanol concentrations below 60% (Ponce et al., 2014), and can induce foaming in the bioreactor (Ezeji et al., 2005). Thus, the aim of this study was to evaluate if gas stripping technology at rapid flow conditions is efficient and suitable for recovery of ethanol from the fermentation media. A special attention to alcohol concentration in the bioreactor, overall energy consumption and overall product yields was made during the research.

MATERIALS AND METHODS

Experimental set-up.

To examine the potential of gas stripping, an experimental pilot scale unit was constructed (Fig. 1). It consisted of a glass bioreactor (Biotechnical Centre, Latvia) with a working volume of 2–4.5 l and equipped with one speed–controlled standard Rushton turbine type agitator with 6 blades and ring microsparger with ten 1 mm jets. Temperature and oxygen in the bioreactor were controlled with a programmable logic controller. To prevent the penetration of fermentation liquid into gas stripping system, a separator with adjustable working volume (0.5 5 l) and variable gas flow capacity was installed. Afterwards gaseous chemicals were condensed in a gas cooling system which consisted of a copper pipe inserted in a refrigerated water bath and collected into a collector.

Flow circulation of gas was performed with an air pump (Alita, USA). The flow rate was measured with a flow meter (Cole-Parmer, USA) with a measuring range of 0–150 l min^{-1} and accuracy of 1 l min^{-1}.

Energy consumption of the gas stripping system was measured by 3–phase indicator (Orno OR–WE–505, Poland).

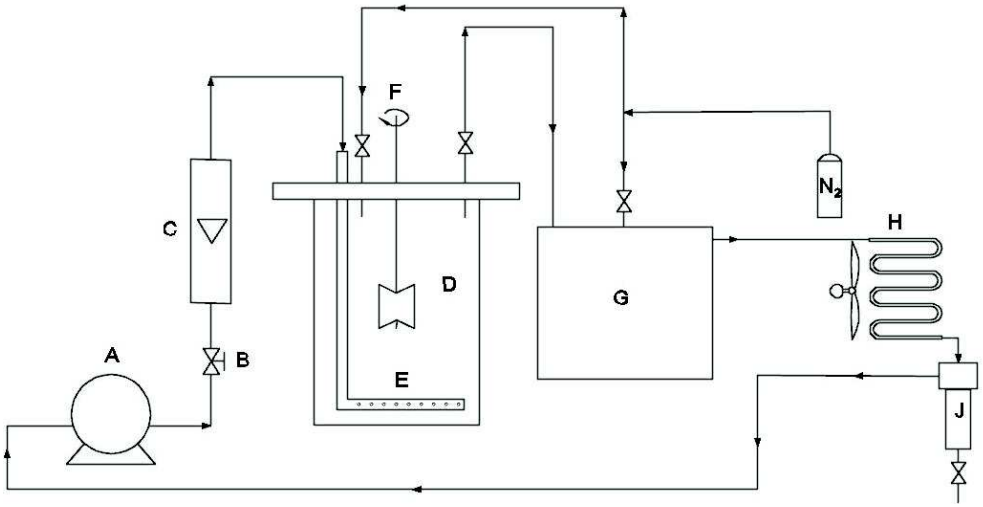

Figure 1. Experimental set–up of gas–stripping system: A–air pump; B–flow control valve; C–flow meter; D–reactor; E–gas sparger; F–agitator and motor; G–separator; H–cooler; J–product collector.

Gas stripping tests

To perform gas stripping tests a model fermentation broth consisting of 2; 10 or 22 wt% of ethanol in water was prepared. Total working volume was 3.5 l. The separation of alcohol from the fermentation broth was performed in a batch regime. Before all stripping experiments the cooling system and gas-circulation line was manually flushed with O_2–free–N_2 (> 99.95%) or CO_2 (99.99%) gases, to make it anaerobic. The temperature in the bioreactor was maintained at 36.7–37 °C, 50 rpm agitator speed and minus 4 to 2 °C in the cooling system. To estimate the efficiency of the gas stripping, 30; 50 and 60 l min^{-1} gas flow regimes were tested at various alcohol concentrations in the fermentation broth (Table 1). Each test giving a positive result was repeated at least once.

Sampling was performed at regular intervals from the product collector and bioreactor and measured with a hydrometer (Vinoferm, Belgium) to determine the final ethanol concentration. Energy consumption was read on an hourly basis all through the testing period. The overall testing time of each run was 8 hours.

Table 1. Experimental test regimes

No	Ethanol concentration wt %	Flow rate l min^{-1}	Gas type	Separator volume	No runs performed
1	22	60	N_2	Low	2
2	10	60	N_2	Low	3
3	10	60	N_2	High	2
4	10	50	N_2	Low	2
5	10	30	N_2	Low	1
6	10	60	CO_2	Low	1
7	2	60	N_2	Low	1

Statistical analyses

MS Excel 2007 *t–test* (two tailed distribution) and ANOVA single parameter tool (significance level ≤ 0.05) were used for analysis of variance on data from various sample setup`s.

RESULTS AND DISCUSSION

Despite the observation that gas stripping is directly related to the gas flow rate, the reported flow regimes are usually below 10 l min^{-1} (Qureshi & Blaschek, 2001; Abdehagh et al., 2014) and can be as low as 1.25 l min^{-1} (Lu et al., 2012). In these conditions gas stripping is usually performed for more than 24 hours. The rationale behind this is to control constant–low levels of the alcohol in the system, thus, preventing the accumulation of the inhibitors (Ezeji et al., 2005). At the same time, such treatment generates low concentration of alcohol in the recovered condensate. Lu et al. (2012) reported only 10–16% for the recovered butanol. Others had between 11 till 24% for the recovered ethanol (Taylor et al., 2010; Ponce et al., 2014). Within this study flow regimes of 30; 50 and 60 l min^{-1} were tested to evaluate the efficiency of alcohol extraction in a short period of time (in less than 8 hours). 10 wt% ethanol concentration in the reactor was selected to simulate yeast tolerance level. The results showed that at 30 l min^{-1} the overall ethanol recovery is low and it did not reached even 10% of the amount of absolute alcohol available in the reactor (Fig. 2), thus, corresponding to the previous observations. At the same time with 50 and 60 l min^{-1} flow rates it was possible to recover 37.8% and 45.4% of all available alcohol respectively, thus, decreasing the concentration of ethanol in the reactor to around 5 wt%. Only a slight difference ($p > 0.05$) was observed between the two fastest flow regimes. Despite the fact that 60 l min^{-1} allowed to collect from 2–24% more ethanol in all samplings, further increase in flow rate was omitted due to possible excess foaming (Ezeji et al., 2005) and potential damage to cells as a result of gas bubble breaking (Chisti, 2000).

The amount of water in the samples collected after 2 hours of gas stripping was below 40% (50 l min^{-1} and 60 l min^{-1} flow rate) and it had the tendency to increase with the stripping time irrespective of the flow rate. Correspondingly, after 8 hours of treatment more than 60% of the collected sample volume was represented by water. In distillation ethanol content after the first column is only around 50% (Sanchez & Montoya, 2013), thus, a second system for product treatment is necessary for both technologies.

After 8 hours of stripping the samples had only 25–30 wt% ethanol (50 l min^{-1} and 60 l min^{-1} flow regimes) (Fig. 2) and that represented less than 20% of all the collected amount of ethanol within previous 6 hours. Again no significant difference ($p > 0.05$) between 50 l min^{-1} and 60 l min^{-1} was observed. Moreover, no significant difference ($p > 0.05$) was observed for the overall energy consumption to perform gas stripping at 50 or 60 l min^{-1} flow rate. The average consumption rates in one hour work of the pilot scale system were around 0.94 MJ.

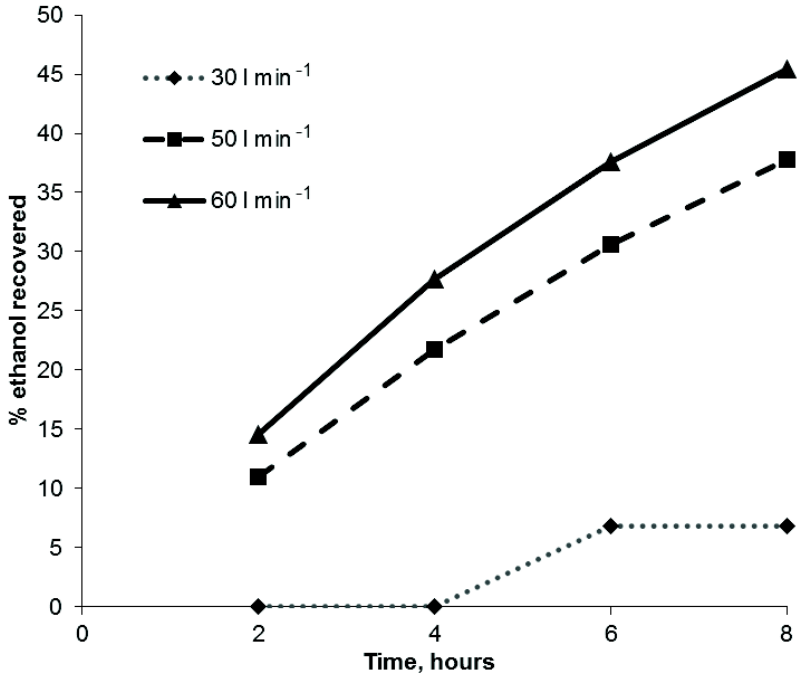

Figure 2. The percentage of total absolute ethanol (99.5%) recovered during 8 hours of gas stripping at 30, 50 and 60 l min^{-1} flow rate. Initial ethanol concentration in the reactor was 10 wt%. Values represent the average.

To decrease the amount of water in the collected liquid and limit water presence in the condensation system, an additional separator volume was installed (0.5 l min^{-1} was increased to 5 l). The function of the separator was to decrease liquid or foam entrance into the condensation system and increase separation intensity as such. This modification was tested only at 60 l min^{-1} flow. Test results showed no significant difference in the ethanol recovery with or without increased separator volume ($p > 0.05$) (Fig. 3). Like before, the amount of recovered ethanol decreased with gas stripping time. Thus, the increase in separator volume did not have any positive effect on the ethanol percentage in the liquid collected after gas stripping. At the same time it should be noted that the modification could be important in systems with high foaming intensity or in situations when prolonged gas stripping is performed.

Further, two additional ethanol concentrations – 2 and 22 wt% were tested to simulate the upper tolerance limit of ABE fermenting clostridia and a very high alcohol concentration, respectively. Both concentrations were tested at 60 l min^{-1} flow rate. It was impossible to collect enough samples after 2 and 4 hours with a reactor having 2 wt% alcohol. The overal amount of absolute ethanol recovered was only 2.4 mL(3.3% of all available ethanol). The amount of water in these samples was almost 90% (Fig. 3), thus, the selected approach showed to be unsuitable for the systems with low volatile content.

The increase in the reactor ethanol concentration to 22 wt% resulted in an increased amount of recovered ethanol. More than 120 mL(18.3% of available ethanol) were collected after 2 hours of gas stripping and after 6 hours almost 44% (317 ml) of available ethanol was recovered (Fig. 3). The increased volatile concentration resulted in higher process efficiency, however, the possibility that such concentrations will be regular in fermentation broth is low. Nevertheless, the results demonstrated that elevated concentrations (above 20 wt%) are suitable for gas stripping at 60 l min^{-1} gas flow regimes (Fig. 3).

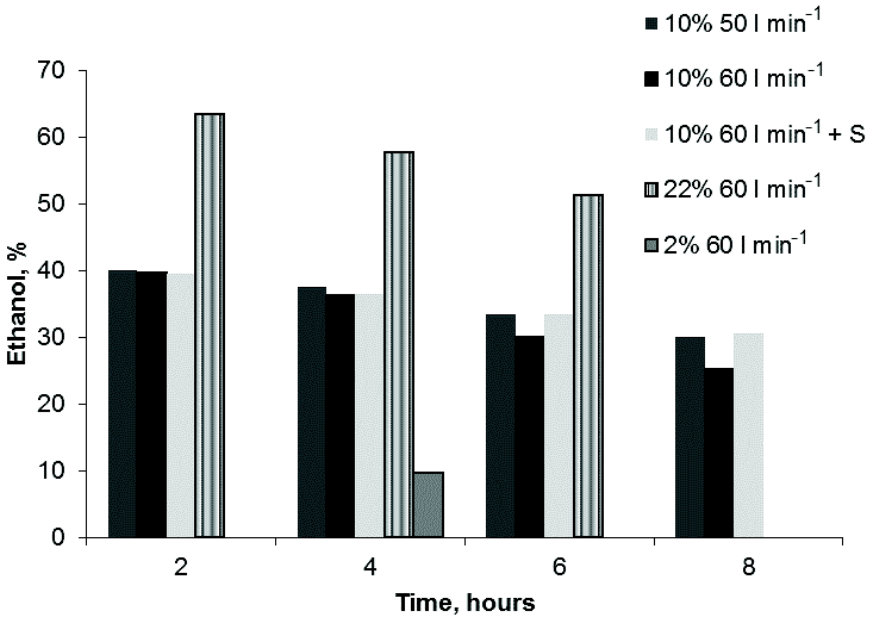

Figure 3. The percentage of absolute ethanol in samples collected after 2, 4, 6 and 8 hours of gas stripping at 50 or 60 l min^{-1} with or without extra separator. Ethanol concentration in the reactor was 2; 10 or 22 wt% depending on the run. Values represent the average from at least 2 runs.

To perform gas stripping, application of gases like CO_2, N_2 or even H_2 has been suggested and evaluated (Liao et al., 2014). In general, the application of CO_2 is advised because the gas is produced during the fermentation process itself, thus, no extra gas source is necessary (Xue et al., 2012). Moreover, it has been estimated that there is no economic reason to exchange CO_2 with any other gas (Taylor et al., 2010). However, it has been demonstrated that CO_2 can be inhibitory to yeast growth (Zhang et al., 2005) and nitrogen is a more suitable gas in both yeast ethanol production and ABE (Liao et al., 2014). To evaluate if other type of gas can increase the alcohol recovery at high gas flow regime, CO_2 was tested. The results showed that there is an increased consumption of CO_2 (at least 2 times more than N_2) to maintain anaerobic conditions in the whole system. Moreover, no increase in ethanol recovery was observed – even 13% lower recovery rate was observed with CO_2 than N_2. Also the same amount ($p > 0.05$) of water content was observed in the collected samples, thus, indicating on no superiority of CO_2. Thus, N_2 showed to be a more appropriate source of gas.

Efficient recovery of alcohol from the fermentation media is usually a struggle between the recovery efficiency, costs and system longevity. Introduction of gas stripping can be a good choice to maintain a continuous system and do not increase the production costs. Despite the problems with high water content, foaming or technological parameters, the resultant product yields can be on the same quality level as classical distillation. Introduction of a subsequent technology, like, membrane separation (Nigaz & Durmaz, 2016) will enable the production of alcohol above 99.5% that is a suitable fuel alcohol.

CONCLUSIONS

The study demonstrated that the efficiency of gas stripping is strongly dependent on the technological parameters, like, alcohol concentration in broth, gas flow rate and treatment time. High flow rate (60 l min^{-1}) is suitable for rapid extraction of ethanol from the reactor and does not introduce any excess foaming. Moreover, there was no significant ($p > 0.05$) increase in process energy consumption when the flow rate was increased from 30 to 60 l min^{-1}. The recovery rate of ethanol after 8 hours reach up to 45.4% of the available ethanol. CO_2 did not showed to be superior for ethanol recovery. The study showed that downstream process optimisation in lignocellulosic biofuel production can significantly decrease production costs.

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