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Gas-Solid Fluidized Bioreactor

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Clarke et al.: Treatment of VOC Emissions in a Gas-Solid Fluidized Bioreactor

TREATMENT OF VOC EMISSIONS IN A GAS-SOLID FLUIDIZED BIOREACTOR

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ABSTRACT

A fluidized bioreactor, with a bed of moist sawdust and glass spheres, successfully treated ethanol-contaminated air. The maximum elimination capacity was $75 \text{ g m}^{-3} \text{ sawdust h}^{-1}$ for the fluidized bed compared to $225 \text{ g m}^{-3} \text{ sawdust h}^{-1}$ for a packed bed (essentially a biofilter).

INTRODUCTION

Biological techniques, such as biofiltration, are highly effective for the treatment of waste gases with low contaminant concentrations (1). Biotreatment is often less expensive and more environmentally-friendly than physical and chemical treatment methods which include incineration, adsorption and scrubbing (2). For contaminated air treatment, biofilters (packed beds where micro-organisms coat the packing) are already used in several industrial situations. Biofilters are normally designed to treat superficial gas velocities from 0.014 to 0.042 m s^{-1} , with contaminant concentrations less than 5 g m^{-3} (1). Le Cloirec et al. (1) investigated a biofilter of moist wood chips to treat ethanol in air. They reported a decrease in biofilter performance with decreasing residence time of the polluted air. At a superficial gas velocity of 0.06 m s^{-1} , the removal efficiency was 98% at an ethanol loading of $143 \text{ g m}^{-3} \text{ h}^{-1}$, while at 0.43 m s^{-1} the removal efficiency was 98% but the loading was limited to $101 \text{ g m}^{-3} \text{ h}^{-1}$. Removal efficiency is defined as the difference between the inlet and outlet concentration as a percentage of the inlet concentration, and ethanol loading is calculated as the ethanol feed rate per total volume of sawdust in the bioreactor.

Although biofilters have been used to treat waste gases such as volatile organic compounds (VOCs), they have several operating problems. In particular, it is difficult to maintain uniform humidity, pH and cell growth across the bed, resulting in gas channelling and lower biodegradation efficiencies (3). A fluidized bed bioreactor may overcome the difficulties associated with biofilters. The advantages of fluidization include homogeneous conditions in the bed due to the rapid and uniform mixing of particles and high rates of heat and mass transfer between the fluid and particles (4).

The use of gas-solid fluidized bioreactors for treating air pollution is relatively new. In gas-solid fluidized bioreactors, micro-organisms are immobilized on solid particles

and fluidized with a gas stream. Wright and Raper (5) used a three-phase spouted bed to treat ammonia-contaminated air. Mineral solution was continuously dripped onto the bed, and the liquid caused high wall adhesion and aggregation of particles which interfered with spouting characteristics. The spouted bed achieved removal efficiencies from 0 to 40%, treating ammonia loadings of 500 to 650 g m⁻³ h⁻¹. The performance of the spouted bed exceeded that of a packed bed (5). Leslous et al. (3, 6) treated ethanol and toluene-contaminated air using a gas-solid fluidized bioreactor. The fluidized bed consisted of moist, scrap-wood particles with a Sauter mean diameter of 0.54 mm. The bioreactor achieved removal efficiencies of 100% at ethanol loadings less than 200 g m⁻³ h⁻¹ and 80% at a loading of 1150 g m⁻³ h⁻¹.

The scope of the present investigation is to measure biotreatment of ethanol, a candidate VOC, in a gas-solid fluidized bioreactor. The bioreactor packing consists of moist sawdust and glass spheres. Moisture is mandatory for biotreatment. Previous studies of fluidized biotreatment found that moist biomass particles are difficult to fluidize because the particles tend to agglomerate (5, 6). In this study, glass spheres are added to improve the fluidization properties. Although Leslous (3, 6) investigated a fluidized bioreactor for treating ethanol, this study will also compare fluidized bed performance to that of a packed bed (essentially a biofilter).

EXPERIMENTAL

A cylindrical, bench-scale bioreactor that may be operated as either a fluidized or a packed bed was used for this study (Figure 1). The vessel inner diameter is 0.139 m and the perforated-plate distributor has a 6.4% open area. At superficial gas velocities greater than 0.0024 m s⁻¹, ambient air was supplied with a blower and the air was saturated by periodically injecting low pressure steam. The inlet air was cooled to 19 to 21 °C with a heat exchanger. At superficial gas velocities of 0.0024 m s⁻¹, compressed laboratory air was used and the air was humidified in two bubblers containing deionized water. Humidity and temperature of the inlet air were measured with a HMP 230 series probe by Vaisala (Helsinki, Finland). Ethanol concentrations in the gas stream entering and exiting the bioreactor were determined using a Hewlett Packard 5890 series gas chromatograph with an FID. The samples were collected in glass bulbs from ports on the vessel and sub-samples from the bulbs were injected into the GC. Moisture content in the sawdust was measured with a Mettler Toledo (Columbus, OH) HB43 halogen moisture analyzer.

The bioreactor bed material consisted of 26 vol.% moist sawdust and 74 vol.% glass spheres. Sawdust with a Sauter mean diameter of 0.625 mm was produced by grinding waste spruce wood. The moisture content of the sawdust was adjusted between 67 and 233 wt% (dry basis). A-070 specification glass spheres with a Sauter mean diameter of 0.516 mm, were obtained from Potters Canada (Moose Jaw, SK, Canada). More detailed particle characterization is presented in Clarke et al. (7). Bed pressure drop profiles were obtained for sawdust (67 wt% moisture) and sphere mixtures, in order to determine fluidization regimes in the bed. The pressure profiles were measured in beds with heights of 14 cm, at increasing velocity, by the method described in Clarke et al. (7).

Transient mass transfer experiments were conducted in the bioreactor vessel in the absence of biodegradation, with fresh glass spheres and sawdust particles (67 wt%

moisture) which were initially free of ethanol. Ethanol was introduced into the inlet air at time zero. Ethanol breakthrough curves were obtained for a packed and fluidized bed by measuring inlet and outlet concentrations of ethanol until steady state was reached. Superficial gas velocities were 0.0024 and 0.155 m s^{-1} in the packed bed trials and 0.7 m s^{-1} in the fluidized bed trials.

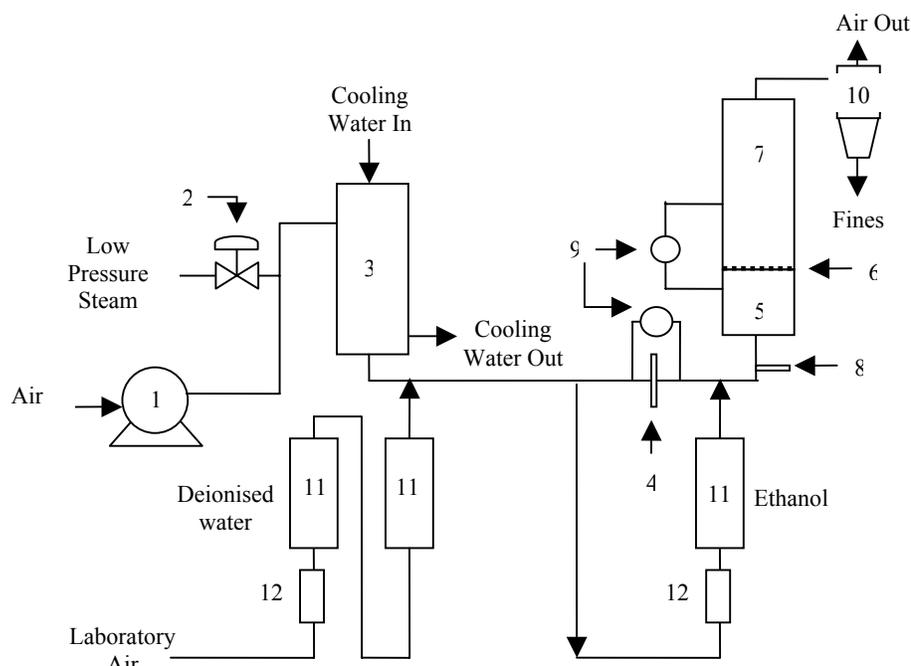


Fig. 1 - Experimental apparatus: (1) blower; (2) steam control valve; (3) heat exchanger; (4) orifice plate; (5) windbox; (6) distributor and wire screen; (7) bioreactor; (8) humidity and temperature probe; (9) differential pressure transducers; (10) cyclone; (11) bubbler; (12) rotameter

In the biodegradation experiments, total packed bed volume was 3.1 L, resulting in a packed bed height of 20 cm. Ethanol-contaminated air was continuously fed into the vessel. Conditions in the system were not sterile, and a mixed culture of microorganisms developed that could thrive on ethanol. Thus, at an industrial scale, this process does not require costly sterilization equipment. A nutrient solution was periodically mixed into the packing in 50 to 125 mL batch additions every two to four days. Biodegradation was measured in packed bed experiments as sawdust moisture content, inlet ethanol concentration, and superficial gas velocity were varied. Biodegradation was also measured in fluidized bed trials at various ethanol concentrations with a superficial velocity of 0.7 m s^{-1} . For both the packed and fluidized bed experiments, the bioreactor was allowed to reach steady state before biodegradation data was recorded. The bed was operated either in a packed or fluidized bed state continuously for seven months.

RESULTS AND DISCUSSION

Bed pressure drop profiles of two different compositions of sawdust and spheres are presented in Figure 2. At minimum fluidization there is the formation of a plug causing a peak in the pressure drop, which is followed by a decrease in pressure drop due to channelling. At higher gas velocities, this channelling gives way to a

regime of bubbling/slugging fluidization that was deemed acceptable for these experiments. The slugging action causes particles to fly upwards and to settle down through the bed in a step-wise motion. Note that in fluidization pressure drop is 7.7 kPa/m bed, contributing to higher air compression costs than for a packed bed.

A bioreactor with a mixed culture of micro-organisms was successfully operated in both a fluidized and packed bed mode to treat ethanol-contaminated air. Figure 3 presents packed bed biodegradation at varying sawdust moisture concentrations, at a superficial gas velocity of 0.0024 m s^{-1} . Each data point represents the ethanol removal efficiency of the bioreactor in a steady state experiment at constant inlet ethanol concentration. It is observed that removal efficiency is 100% at ethanol loadings up to $73 \text{ g m}^{-3}\text{sawdust h}^{-1}$, after which removal efficiency declines. Furthermore, Figure 3 illustrates that biodegradation is independent of sawdust moisture for the moisture range studied (67 to 233 wt%, dry basis). A sawdust moisture content of 67 to 75 wt% was used in the remaining experiments, because sawdust-sphere mixtures with lower moisture fluidize better (Z). Although moisture is required for biodegradation, increasing sawdust moisture above 67 wt% in packed bed operation does not affect the biodegradation efficiency or capacity. Perhaps either the additional moisture above 67 wt% does not increase the available moisture to the microbial culture or the amount of moisture which is required by the culture for maximum growth is less than 67 wt%.

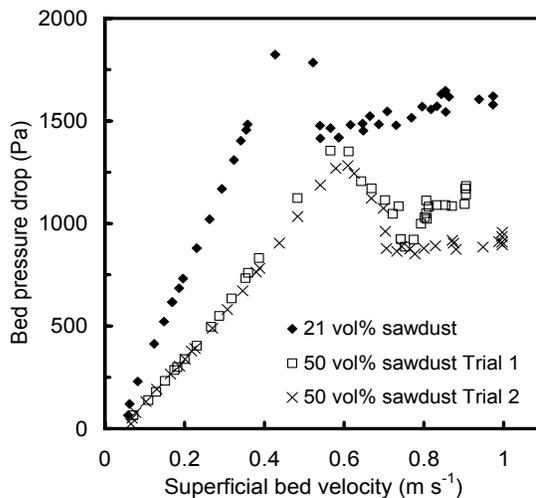


Fig. 2 – Pressure profiles of glass spheres and sawdust at increasing gas velocity (sawdust moisture = 67 to 75 wt%).

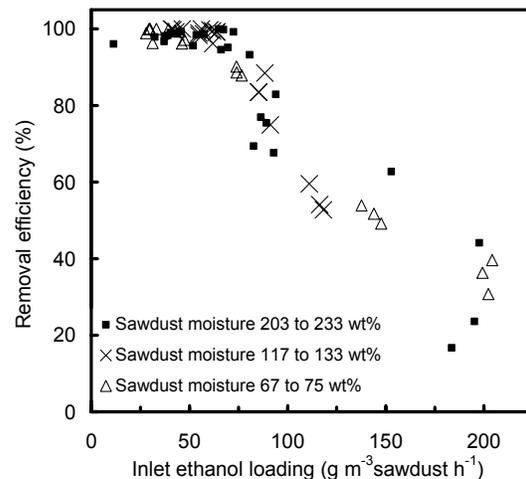


Fig. 3 – Effect of moisture on removal efficiency in a packed bed bioreactor (gas velocity of 0.0024 m s^{-1}).

Figure 4 illustrates the effect of superficial gas velocity on biodegradation in a packed bed. The results are shown again in Figure 5 in terms of elimination capacity (EC), which is defined as the mass of ethanol consumed per sawdust volume per unit time. EC reaches a maximum value of $225 \text{ g m}^{-3}\text{sawdust h}^{-1}$ at velocities of 0.155 and 0.25 m s^{-1} , while the maximum is only $73 \text{ g m}^{-3}\text{sawdust h}^{-1}$ at 0.0024 m s^{-1} . Biodegradation in the fluidized bed bioreactor is illustrated in Figures 6 and 7 where it is seen that the fluidized bed performance is comparable to that of the packed bed operated at a velocity of 0.0024 m s^{-1} . The maximum EC for the fluidized bed is approximately $75 \text{ g m}^{-3}\text{sawdust h}^{-1}$. It was observed that over several months the sawdust particles appeared to age, as they changed color from white to dark brown.

However, reproducibility experiments taken at different times demonstrated similar biodegradation performance.

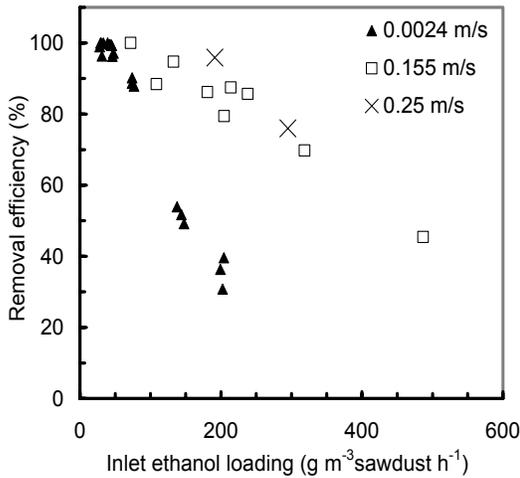


Fig. 4 – Effect of superficial gas velocity on removal efficiency in a packed bed (sawdust moisture = 67 to 75 wt%).

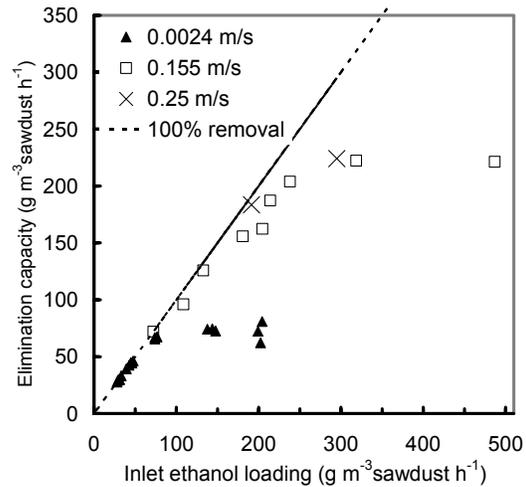


Fig. 5 – Effect of superficial gas velocity on EC in a packed bed (sawdust moisture = 67 to 75 wt%).

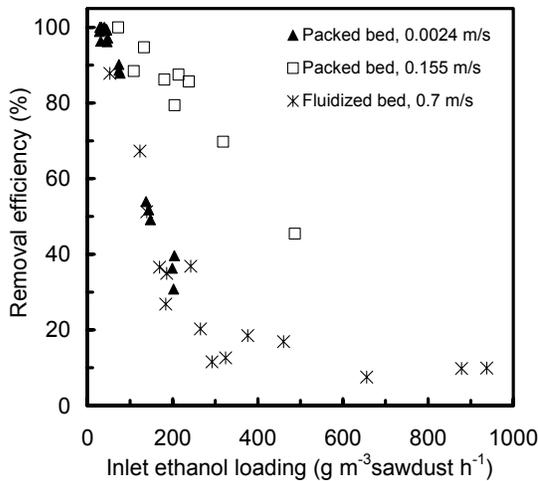


Fig. 6 – Fluidized versus packed bed bioreactor performance (sawdust moisture = 67 to 75 wt%).

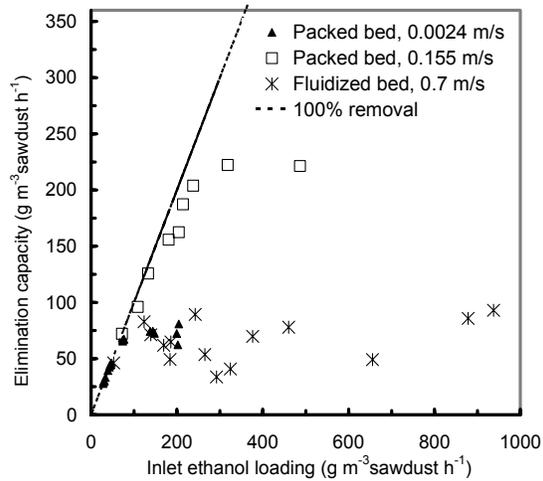


Fig. 7 – Fluidized versus packed bed bioreactor performance (sawdust moisture = 67 to 75 wt%).

As loading to biofilters (packed beds) increases from zero, the removal efficiency is 100%, until EC reaches a maximum (8), and at higher loadings EC may decline (9). The shape of the EC curve is due to a shift in the overall rate-limiting mechanism. Delhomenie and Heitz (9) and Ottengraf et al. (10) suggest that below the maximum EC, diffusion in the biolayer limits the overall biodegradation rate, and above the maximum EC, microbial growth kinetics are limiting. Microbial growth rate can often be described by Monod (11) kinetics, where it is assumed that only one substrate limits growth. Substrate inhibition is ignored because ethanol concentrations are low in this study (12):

$$-\left[\frac{dS}{dt}\right]_{\text{Biodegradation}} = \left(\frac{\mu_m SX}{K_s + S}\right) \frac{1}{V_{xs}} \tag{1}$$

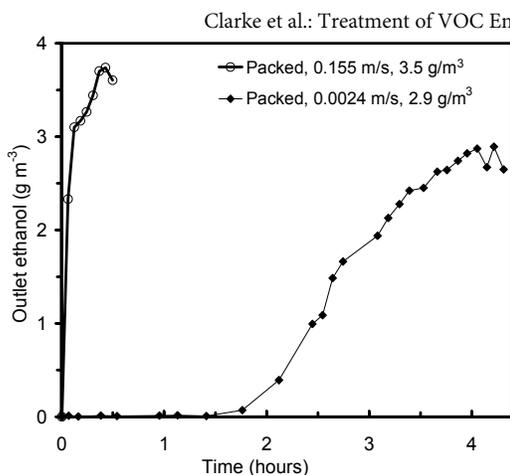


Fig. 8 – Breakthrough curves in packed beds (sawdust moisture content 67 to 75 wt%).

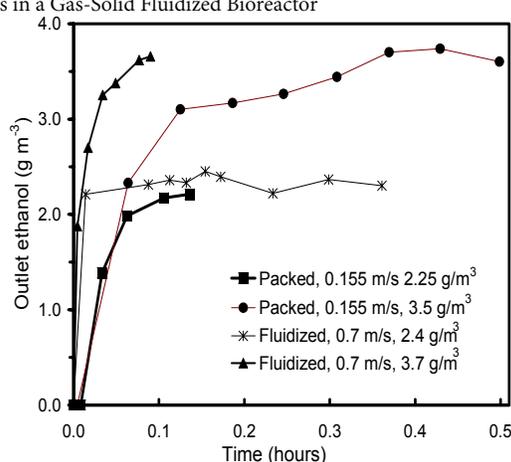


Fig. 9 – Breakthrough curves in fluidized and packed beds (sawdust moisture content 67 to 75 wt%).

Breakthrough curves for the packed and fluidized beds are shown in Figures 8 and 9, respectively, while the external mass transfer coefficients determined from these experiments are summarized in Table 1. The mass transfer coefficient for the packed bed is calculated by modelling the gas phase as non-dispersed plug flow (13). Two values are presented for the coefficient in the fluidized bed, corresponding to the two ideal limits of non-dispersed plug flow and perfect mixing of the gas (14).

Table 1 – Mass transfer in packed and fluidized beds

Bioreactor	Superficial gas velocity, m s^{-1}	Mass transfer coefficient, h^{-1}
Packed	0.0024	13
Packed	0.155	32
Fluidized	0.70	81 (plug flow); 49 (mixed)

It is observed that the mass transfer coefficient increases with velocity in a packed bed, and is highest in a fluidized bed. Note that these experiments were conducted without biodegradation. Thus the mass transfer coefficient is controlled by the external resistance across the gas and moist particle interface. The observation that the mass transfer coefficient increases with gas velocity is consistent with the Ranz correlation for external mass transfer coefficient in a gas-solid fixed bed (4):

$$Sh = 2 + 1.8(\text{Re})^{1/2} Sc^{1/3} \quad (2)$$

Applying Equation (2), the external mass transfer coefficient of the 0.7 m s^{-1} fluidized bed would be 2.1 times higher than that of the 0.155 m s^{-1} packed bed. However, the higher mass transfer is of no value for bioremediation purposes once the overall rate in the bioreactor is controlled by the kinetics of growth.

Deshusses and Johnson (8) propose that biofilter performance generally depends on contaminant loading and not concentration, such that the EC curves will be the same for different combinations of inlet gas velocity and concentration. On the other hand,

if substrate concentration (S) is very low, it is likely much less than K_s (8). Then according to Equation (1), the reaction is first order with respect to S . At higher S , growth kinetics become zero order. Concentrations used in the fluidized and high velocity packed (superficial gas velocity 0.155 m s^{-1}) bed experiments were 0.001 to 0.02 g m^{-3} , and 0.007 to 0.045 g m^{-3} respectively. Because of these low concentrations, if the kinetic of both the fluidized and high velocity packed bed were first order with respect to ethanol, then the microbial growth rate at maximum EC would be greater in the high velocity packed bed, which has higher ethanol concentrations. If microbial kinetics were first order in the fluidized bed and zero order in the high velocity packed bed, the high velocity packed bed would again be operating at higher microbial growth rate as per Equation 1. Furthermore, the amount of microbial cells available in a fluidized bed may be less than that of a packed bed. The fluidization motion may reduce the steady state concentration of cells on the packing. According to Equation 1, the biodegradation rate would be reduced if cell concentration (X) was reduced.

In the low velocity (0.0024 m s^{-1}) packed bed biodegradation trials, inlet ethanol was in the range of 0.07 to 1.2 g m^{-3} . Growth kinetics were likely zero order as per Equation 1, and the growth rate should be faster than that of the high velocity packed bed. However, Figure 5 shows that the maximum EC of the low velocity packed bed was much lower than that of the high velocity bed. Possibly, a process other than microbial kinetics limits the maximum EC of the low velocity bed. Kim and Deshusses (15) suggest that above the maximum EC of a biotrickling filter, biodegradation is limited by microbial growth kinetics, transport in the liquid, or diffusion in the biolayer. In addition, mass transfer of oxygen to the biomass was not measured. In the low velocity packed bed, the low external mass transfer coefficient for ethanol from the gas phase to the particles suggests that the oxygen mass transfer coefficient may also be low, which may limit the maximum EC. Another explanation is that in the low velocity packed bed, the gas stream bypasses sections of the bed, a well-known phenomenon in biofilter operations. As a result, a smaller portion of the micro-organisms come into contact with the contaminants, and X in Equation 1 is actually smaller in the low velocity versus the high velocity packed bed. Thus biodegradation rate and maximum EC are lower in the low velocity bed.

CONCLUSIONS

A binary mixture of moist sawdust and glass spheres has potential as a packing for a fluidized bioreactor. In this study, classic biodegradation EC curves are obtained in both packed and fluidized modes which confirms that the bed is capable of supporting micro-organisms, even when fluidized. The maximum EC is greater in the high velocity packed bed (0.155 m s^{-1}) than the fluidized bed, even though the fluidized bed has a higher external mass transfer coefficient. At the maximum EC, if the overall biotreatment rate is controlled by growth kinetics, then the high velocity packed bed, which has a higher ethanol concentration, may have a higher microbial growth rate.

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NOTATION

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K_s	Saturation constant (g m^{-3})
Re_p	Particle Reynolds number
S	Concentration of growth limiting substrate in liquid phase (g m^{-3})
Sc	Schmidt number
Sh	Sherwood number
t	Time (h)
X	Biomass (cell) concentration in liquid phase (g m^{-3})
Y_{xs}	Microbial yield: mass of biomass (cells) produced per mass of substrate consumed
μ_m	Maximum growth rate achievable (h^{-1})

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