



Prediction of acid hydrolysis of lignocellulosic materials in batch and plug flow reactors



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HIGHLIGHTS

- We proposed a model to predict hydrolysis in batch and plug flow reactors.
- The model considers the volume expansion in the hydrolysis suspension.
- The model predicts experimental data within a 5% error.
- The model unifies experimental results for batch and plug flow reactors.

ARTICLE INFO

Article history:

Received 5 March 2013

Received in revised form 15 May 2013

Accepted 17 May 2013

Available online 24 May 2013

Keywords:

Cellulose hydrolysis
Hemicellulose hydrolysis
Liquid–solid ratio
Batch reactor
Plug flow reactor
Sugar yield

ABSTRACT

This study unifies contradictory conclusions reported in literature on acid hydrolysis of lignocellulosic materials, using batch and plug flow reactors, regarding the influence of the initial liquid ratio of acid aqueous solution to solid lignocellulosic material on sugar yield and concentration. The proposed model takes into account the volume change of the reaction media during the hydrolysis process. An error lower than 8% was found between predictions, using a single set of kinetic parameters for several liquid to solid ratios, and reported experimental data for batch and plug flow reactors. For low liquid–solid ratios, the poor wetting and the acid neutralization, due to the ash presented in the solid, will both reduce the sugar yield. Also, this study shows that both reactors are basically equivalent in terms of the influence of the liquid to solid ratio on xylose and glucose yield.

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1. Introduction

Bioethanol production from lignocellulosic material is receiving broad attention in literature because it results promising from an energetic and sustainable point of view (Sun and Cheng, 2002). The lignocellulosic material requires a pretreatment in order to open the biomass structure, expose the cellulose and produce glucose (Kumar et al., 2009). Acid hydrolysis of several kind of feedstock has been largely reported in the literature and it is used at industry as a pretreatment method (Panagiotopoulos et al., 2012; Wei et al., 2012) for obtaining glucose (Taherzadeh and Karimi, 2007; Saeman, 1945) and xylose (Rodríguez-Chong et al., 2004; Aguilar et al., 2002). This kind of hydrolysis has been performed in batch (Alves-Gurgel, 2010; Rodríguez-Chong et al., 2004; Aguilar et al., 2002; Madelaine et al., 1990; Saeman, 1945) and continuous reactors (Converse et al., 1989; Mcparland et al., 1982; Church and

Wooldridge, 1981) and one of the conclusions from those studies is that the liquid to solid ratio affects the xylose and glucose yield. For instance, xylose yield from hydrolysis of hemicellulose using batch reactors increases as the liquid to solid ratio (ϕ_{LSO}) rises (López-arenas et al., 2010; Vargas Betancur and Pereira, 2010; Neureiter et al., 2002; Lavarack et al., 2002, 2000). In contrast, Madelaine et al. (1990) have shown no influence of the liquid to solid ratio on sugar yield in hydrolysis of wood in batch reactors. On the other hand, using plug flow reactors, hydrolysis of cellulose has not shown influence of the liquid to solid ratio on glucose yield (Church and Wooldridge, 1981; Thompson and Grethlein, 1979). These conclusions are counter-intuitive considering that batch and plug flow reactors are in principle similar on reaction time basis.

It has been shown that for low liquid to solid ratios: (i) the effect on sugar yield is negative (Vargas Betancur and Pereira, 2010), (ii) The sugar decomposition rate increases, also reducing the sugar yield (Lavarack et al., 2002, 2000).

A model that includes the effect of the liquid–solid ratio on sugar yield has been used for several authors (López-arenas et al.,

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Nomenclature

| | | | |
|----------------------------------|---|-------------------------------------|--|
| Batch reactor; Plug flow reactor | R | universal gas constant, [J/(mol K)] | |
| A | reactor transversal area, [m ²] | R_M | = C_M/C_{M0} , concentration ratio |
| C_A | acid concentration, wt%/v or wt%/wt | t | reaction time, [min]; = $ALZ/(v_0q)$, residence time in [min] |
| C_M | = m_M/v_L , sugar concentration, in [kg/m ³ liquid] | t_{cr} | = $1/t_{k_i(T_{ref}, C_{A, ref})}$ characteristic reaction time, [min] |
| C_{MD} | = m_{MD}/v_L , concentration of monomer degradation products, [kg/m ³ liquid] | T | temperature, [K] |
| \bar{C}_G | = m_G/v , glucose concentration based on suspension volume, [kg/m ³ suspension] | v | suspension volume, [m ³ suspension]; suspension volumetric flow, [m ³ suspension/min] |
| C_P | = m_P/v , polymer concentration, [kg/m ³ suspension] | v_L | liquid phase volume, [m ³ liquid]; liquid volumetric flow, [m ³ liquid/min] |
| C_S | = m_S/v , solid concentration, [kg/m ³ suspension] | v_{LO} | initial liquid phase volume, [m ³ liquid]; feed liquid volumetric flow, [m ³ liquid/min] |
| C_{sol} | = $100/\phi_{LS}$, solid concentration, [%] | v_0 | initial suspension volume, [m ³ suspension]; feed suspension volumetric flow, [m ³ suspension/min] |
| CF | conversion factor, 0.88 for xylose and 0.90 for glucose | x_i | mass fraction of component i |
| E_i | activation energy of reaction i , [J/mol] | Y_M | = m_M/m_{M0} , sugar yield |
| F | fraction of polymer of fast reaction rate | Y_{MD} | = m_{MD}/m_{M0} |
| f_P | fraction of soluble compounds | δ | longitudinal coordinate, [m] |
| k_i | reaction rate constant, [min ⁻¹] | Z | = t/t_{cr} ; = δ/L |
| k_{i0} | pre-exponential factor, [min ⁻¹ (wt%) ^{-γ_i}] | Z | = $(\psi + \Omega Z)$ |
| L | reactor length, [m] | α_P | = x_P/FC |
| M_L | = m_L/m_{L0} | β_i | = $k_i t_{cr}$; = $k_i AL/v_0$ |
| M_P | = m_P/m_{M0} | γ_i | acid concentration exponent |
| M_S | = m_S/m_{S0} | ε | = v_L/v , void fraction |
| m_L | mass of liquid, [kg]; mass flow of liquid, [kg/h] or [kg/min] | ε_R | reactor void fraction |
| m_{L0} | initial mass of liquid, [kg]; feed mass flow of liquid, [kg/h] or [kg/min] | θ | solid porosity |
| m_M | mass of sugar, [kg]; mass flow of sugar, [kg/h] | λ | = $1/(f_P \alpha_P)$ |
| m_{M0} | initial mass of potential sugar, [kg]; feed mass flow of potential sugar, [kg/h] or [kg/min] | ξ | = $\psi/\Lambda e^{-Z}$ |
| m_{MD} | mass of monomer degradation products, [kg]; mass flow of monomer degradation products, [kg/h] or [kg/min] | ρ_L | liquid density, [kg/m ³] |
| m_P | mass of polymer, [kg]; mass flow of polymer, [kg/h] or [kg/min] | ρ_S | solid density, [kg/m ³] |
| m_S | mass of solid, [kg]; mass flow of solid, [kg/h] or [kg/min] | ϕ_{LS} | = m_L/m_S , liquid–solid ratio |
| m_{S0} | initial mass of solid, [kg]; feed mass flow of solid, [kg/h] or [kg/min] | ϕ_{SL} | = $1/\phi_{LS}$, solid–liquid ratio |
| NC | neutralization capacity, g_{acid}/g_{solid} | σ | = 1; = $\varepsilon/\varepsilon_0$ |
| q | = 1; = v/v_0 | ψ | = $1 - \rho_L/\rho_S$ |
| q_L | = 1; = v_L/v_{L0} | ω | = $1 + \phi_{LS0}$ |
| | | Λ | = $\lambda(\omega - \psi) + \psi$ |
| | | Ω | = $\lambda(\omega - \psi)\beta_P$ |
| | | \mathcal{W} | Lambert function |

2010; Lavarack et al., 2002, 2000). This model shows that sugar yield is reduced as the liquid–solid ratio decreases as follows:

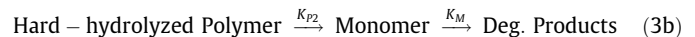
$$\bar{Y} = \frac{k_H(e^{-k_H t} - e^{-\phi_{LS0} k_X t})}{\phi_{LS0} k_X - k_H} = \frac{\phi_{LS0} \beta_H}{\beta_X - \phi_{LS0} \beta_H} \left(e^{-\beta_H \tau} - e^{-\frac{\beta_X \tau}{\phi_{LS0}}} \right) \quad (1)$$

where ϕ_{LS0} is the liquid–solid ratio, the inverse of the solid–liquid ratio (ϕ_{SLO}), and \bar{Y} is the sugar yield. Furthermore in order to describe the sugar production, the kinetic parameters used are function of the solid to liquid ratio (Alves-Gurgel, 2010).

This study unifies the contradictory conclusions presented in literature about the acid hydrolysis of lignocellulosic material, using batch and plug flow reactors, regarding the influence of the liquid–solid ratio on glucose and xylose yield. Thus, mass balances are developed for these reactors and, based on these results, the influence of the liquid to solid ratio, and the void fraction on xylose and glucose yield and concentration are theoretically studied. The theoretical predictions are compared with experimental data reported in the literature.

2. Methods

Two hydrolysis reactions are used in literature to describe the acid hydrolysis of cellulose or hemicellulose and the degradation of monomers (Jacobsen and Wyman, 2000):



The reaction rate expressions in Eq. (2) are linear function of the reactant concentration, with the following reaction rate constant of Arrhenius type:

$$k_i = k_{i0} C_A^{\gamma_i} e^{-\frac{E_i}{RT}} \quad (4)$$

The reaction rate expressions in Eqs. (3a) and (3b) are reduced to the one in Eq. (2) by Lee et al. (2000):

$$k_P = Fk_{P1} + (1 - F)k_{P2} \quad (5)$$

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