

Direct tray and point efficiency measurements including weeping effects through a convenient add-on for air/water simulators

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S1. Analysis of the individual phase resistances

No reliable correlation is available for predicting the mass transfer coefficients pertaining to sieve trays. However, a rough estimate of the ratio of liquid-side and gas-side resistances can be obtained according to Lockett and Uddin¹. Assuming that both phases can be modelled according to the penetration theory and that the residence time of each fluid element at the interface is the same at both sides, the ratio between the liquid and gas transport coefficients is given by

$$\frac{k_L}{k_G} = \left(\frac{D_{IA,L}}{D_{IA,G}} \right)^{0.5}, \quad (S1.1)$$

where k_L and k_G are the mass transfer coefficients for the liquid and the gas phase, respectively. $D_{IA,L}$ and $D_{IA,G}$ are the diffusivities of isobutyl acetate in water and air, respectively. As reported in Section 4.1, the equilibrium of the system can be described using Henry's law. Using the two-film theory, the ratio of the liquid phase resistance to the mass transfer with respect to the total resistance is given as

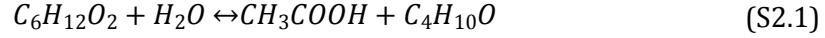
$$\frac{R_L}{R_{tot}} = \frac{1}{1 + \left(\frac{k_L}{k_G \cdot K^c} \right)}, \quad (S1.2)$$

where K^c is the dimensionless Henry's constant.

At 17 °C, the liquid phase resistance is approximately 70%. This shows that the system is neither fully liquid-phase controlled nor fully gas-phase controlled and thus, unsuitable for measuring the single-phase mass-transfer coefficient only.

S2. Assessment of hydrolysis reaction pertaining to species present in the aqueous solution

At acid pH values, the following acid-catalyzed hydrolysis reaction for isobutyl acetate occurs:



The formation reaction of isobutyl acetate from isobutanol and acetic acid has an equilibrium constant²

$K_{eq} = 4$, thus

$$\frac{[CH_3COOH][C_4H_{10}O]}{[C_6H_{12}O_2][H_2O]} = \frac{1}{K_{eq}} = 0.25. \quad (S2.2)$$

Since the reaction happens with an excess of water, the system reaches equilibrium at almost complete conversion of the isobutyl acetate.

The kinetics of the reaction depends on the concentration of H^+ ions acting as the acid catalyst. The pH value of deionized water decreases immediately if in contact with the atmosphere due to the dissolved carbon dioxide. In this study, pH = 5 was measured.

Rayne and Forest³ reported the kinetic constant (expressed per mol of catalyst) of the reaction at pH 4 and 25°C as

$$\log_{10} \chi_A = -4.02 \text{ mol}^{-1} \text{ s}^{-1}. \quad (S2.3)$$

Considering a first-order reaction, the isobutyl acetate concentration in the sample follows

$$c_{IA}(t) = c_{IA}|_{t=0} \cdot \exp(-10^{-pH} \chi_A t). \quad (S2.4)$$

It can be concluded that the change in the isobutyl acetate concentration in the sample as a result of the hydrolysis reaction is below 0.1%, provided that the sample is analyzed within two days after the batch preparation.

It should be noted that the neutral and basic hydrolyses have not been considered, since their effects in acid solutions are negligible.

S3. Established approaches for point efficiency calculation

Experimental data from the FRI database were analyzed by Zuiderweg⁴ using the slope and intercept method and the following correlations were derived for the froth regime:

$$\frac{k_G}{\rho'_G} = k'_G = \frac{0.13}{\rho_G} - \frac{0.065}{\rho_G^2} \quad (\text{S3.1})$$

$$\frac{k_L}{\rho'_L} = k'_L = 0.024 D_{IA,L}^{0.25} \quad (\text{S3.2})$$

$$ah_f = \frac{43}{A_f^{0.3}} \left(\frac{u_s^2 \rho_G h_{cl} FP}{\sigma} \right)^{0.53}, \quad (\text{S3.3})$$

where ρ'_G and ρ'_L are the molar gas and liquid densities, respectively, $D_{IA,L}$ is the diffusivity of isobutyl acetate in the liquid phase, A_f is the tray free fractional area, σ is the liquid surface tension, a is the contact surface between the gas and liquid phase per unit of froth volume, and FP is the flow parameter defined as $\frac{L_{vol}}{G_{vol}} \sqrt{\frac{\rho_L}{\rho_G}}$.

From the two-film theory, the number of the overall gas transfer units is given by

$$N_{OG} = \frac{\frac{k'_G ah_f}{u_s}}{1 + m \frac{k'_G \rho'_G}{k'_L \rho'_L}}, \quad (\text{S3.4})$$

whereas the point efficiency can be estimated as

$$E_{OG} = 1 - \exp(-N_{OG}). \quad (\text{S3.5})$$

Based on a separate experimental database concerning stripping and absorption systems, the following correlations for estimating the number of liquid and gas transfer units were developed by Gerster et al.⁵ and are known as the AIChE correlations:

$$N_G = \left(0.776 + 4.57h_w - 0.238F_s + 104.8 \frac{L_{vol}}{L_w} \right) Sc_G^{-0.5}, \quad (S3.6)$$

$$N_L = 1.97 \cdot 10^4 D_{iA,L}^{0.5} (0.40F_s + 0.17)\tau, \quad (S3.7)$$

where h_w is the height of the outlet weir, F_s is the f-factor, τ is the liquid residence time (that can be estimated as in Equation S4.8) and Sc_G is the gas Schmidt number defined as

$$Sc_G = \frac{\mu_G}{\rho_G D_{iA,G}}, \quad (S3.8)$$

where μ_G is the gas viscosity. Considering that, from the two-film theory

$$\frac{1}{N_{OG}} = \frac{1}{N_G} + \frac{\lambda}{N_L}, \quad (S3.9)$$

E_{OG} can be obtained using Equation S3.5.

S4. Tray efficiency models

The perfectly-mixed model describes the highest possible degree of axial liquid mixing. It assumes that the liquid on the tray as well as the incoming vapour are perfectly mixed, so that

$$E_{MV} = E_{OG}. \quad (S4.1)$$

In this case, the driving force for mass transfer on the tray is everywhere the lowest; thus, this value represents the lower limit for the tray efficiency value. Anyways, in extreme conditions $E_{MV} < E_{OG}$ is a possibility.

The plug flow model⁶ describes the lowest possible degree of axial liquid mixing. It assumes that both liquid and vapour traverse through the tray in plug flow, while the inlet vapour is perfectly mixed. For these conditions, the driving force for mass transfer is everywhere the highest; thus, this value represents the upper physical limit for the tray efficiency value, given by

$$E_{MV} = \frac{\exp(\lambda E_{OG}) - 1}{\lambda}. \quad (S4.2)$$

The AIChE model⁷ is the most applied model for tray efficiency prediction⁸. It is based on the assumptions of constant point efficiency and linear vapour-liquid equilibrium data, while representing partial liquid mixing axially. Considering the axial liquid mixing as a stochastic phenomenon, the molar mixing flux in the liquid phase can be described by a diffusion-like law according to

$$J_L = -D_\epsilon \rho_F \left(\frac{dc}{dz} \right), \quad (S4.3)$$

where D_ϵ is the eddy diffusion coefficient that can be estimated as

$$D_\epsilon = 3.0 u_s h_{cl} \left(\frac{\rho_G}{\rho_L} \right)^{0.5}, \quad (S4.4)$$

where u_s is the gas superficial velocity and ρ_G and ρ_L are the gas and liquid densities, respectively.

Performing a material balance between the gas and liquid phases, the following equation can be derived

$$\frac{E_{MV}}{E_{OG}} = \frac{1 - \exp[-(\eta + Pe)]}{(\eta + Pe) \left(1 + \frac{\eta + Pe}{\eta} \right)} + \frac{\exp(\eta) - 1}{\eta \left(1 + \frac{\eta}{\eta + Pe} \right)}, \quad (S4.5)$$

where

$$\eta = \frac{Pe}{2} \left(\sqrt{1 + \left(\frac{4\lambda E_{OG}}{Pe} \right)} - 1 \right), \quad (S4.6)$$

$$Pe = \frac{Z^2}{D_\epsilon \tau}, \quad (S4.7)$$

$$\tau = \frac{h_{cl} Z L_w}{L_{vol}}, \quad (S4.8)$$

where L_{vol} is the volumetric liquid flow rate, L_w is the weir length and Z is the flow path length.

Nomenclature

Symbol	Description	Unit
a	Interfacial area per unit of froth volume	m^{-1}
A_f	Tray free fractional area (hole area/bubbling area)	
c	Liquid concentration	kmol m^{-3}
D_{ij}	Diffusivity of component i in the phase j	$\text{m}^2 \text{s}^{-1}$
D_ϵ	Eddy diffusion coefficient	m s^{-1}
E_{MV}	Murphree vapour-side tray efficiency	
E_{OG}	Murphree vapour-side point efficiency	
F_s	Superficial factor $F_s = \rho_G^{0.5} u_s$	$\text{kg}^{0.5} \text{m}^{-0.5} \text{s}^{-1}$
FP	Flow parameter	
G_{vol}	Volumetric vapour flow rate	$\text{m}^3 \text{s}^{-1}$
J_L	Molar backmixing flux in the liquid phase	$\text{kmol m}^{-2} \text{s}^{-1}$
k	Mass transfer coefficient	m s^{-1}
k'	Mass transfer coefficient	$\text{kmol s}^{-1} \text{m}^{-2}$
K_{eq}	Reaction equilibrium constant	
K^c	Henry constant defined as gas/liquid concentration	
L_{vol}	Volumetric liquid flow rate	$\text{m}^3 \text{s}^{-1}$

L_w	Weir length	m
h_{cl}	Clear liquid height	m
h_f	Height of the froth	m
h_w	Weir height	m
m	Slope of the linearized vapour-liquid equilibrium data	
N	Number of transfer units	
Pe	Peclet number	
R	Resistance to mass transfer	
Sc_G	Schmidt number	
t	Time	s
u_s	Vapour superficial velocity referred to A_b	m s^{-1}
z	Axial coordinate on the tray	m
Z	Flow path length	m
η	Parameter in the AIChE tray efficiency model	
λ	Stripping factor	
μ	Viscosity	Pa s
ρ'	Molar density	kmol m^{-3}
ρ	Density	kg m^{-3}
ρ_F	Froth density (volume of liquid/volume of froth)	
σ	Surface tension	N m^{-1}

τ	Average liquid residence time	s
χ_A	Kinetic constant of acid-catalyzed hydrolysis	$\text{mol}^{-1}\text{s}^{-1}$

Subscripts

G	Gas
L	Liquid
OG	Overall gas
IA	Isobutyl acetate
tot	Total

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