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Continuous dehydration of Ionic Liquids in a falling film evaporator

Niels Wellner, Karl Siebeneck, Stephan Scholl *

Technische Universität Braunschweig, Institute of Chemical and Thermal Process Engineering, Langer Kamp 7, 38106 Braunschweig, Germany

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ABSTRACT

Water is a commonly found impurity in Ionic Liquids (IL), which may be critical for many applications due to the changes of the ILs' physical properties. A falling film evaporator operated in continuous mode is used to separate water from IL/water solutions and determine favorable process parameters. The dehydration was investigated for two different IL at different feed rates and water concentrations at the inlet. Water contents from 0.0086 to 0.0121 g_{water}/g_{total} were achieved. Additionally, the thermal performance of the evaporator was assessed resulting in film heat transfer coefficients on the evaporation side ranging from ~1000 W m^{-2} K^{-1} to ~1800 W m^{-2} K^{-1} calculated according to VDI Wärmeatlas.

1. Introduction

Ionic Liquids (IL), also known as room temperature liquid salts, form a new class of solvents in today's science. Typically these solvents consist of two characteristic ions which may be inorganic or organic. IL have proven to be very useful as solvents for different purposes in extraction, absorption, extractive distillation, analytics or as process fluids [1,2]. Regarding ecological effects and economic reasons, IL may only compete with "traditional" fluids when their operational lifetime is extended or their application is more efficient. When used as process fluid in extraction, absorption or extractive distillation processes, separation, purification and recycling is mandatory to achieve economic as well as ecological competitiveness. Based on economical and ecological considerations a minimum number of recycles may be quantified which is necessary to achieve respective benefits for the use of IL [3]. The necessity of multiple recycles requires effective and gentle separation techniques in order to maintain solvent and process qualities. Therefore new strategies to purify and recycle Ionic Liquids need to be established.

Water is a commonly found impurity in Ionic Liquids, often resulting from synthesis or IL' hygroscopic properties [4]. To ensure constant product quality and a long operational lifetime, methods for dehydrating need to be elaborated and assessed. Investigations on batch-mode dehydration of IL in a falling film evaporator were found to be very promising, in terms of achieved water contents $<1 * 10^{-3}$ g_{water}/g_{total} at moderate process conditions, such as heating temperature of 120 °C, total pressure of 50 mbar, specific heat flux <15 kW m^{-2}, and short residence time. Furthermore a dependency of viscosity to water content and temperature showed the possibility to utilize the viscosity as a parameter to monitor water content online [5]. In industrial applications continuous processes often provide great potential savings on specific energy demand against batch processes for larger capacities. Therefore first experiments on the feasibility of continuous dehydration were conducted, in order to determine beneficial process parameters for increasing energy efficiency and economic profitability.

Falling film evaporators (FFE) are commonly used in the process industry for separating thermal sensitive products, e.g. fruit juices, milk or sugar syrup [6]. Driven by gravity, a thin liquid film is flowing down at the inner circumference of a vertical tube with the vapor phase in cocurrent or countercurrent flow in the central cross section. Due to the resulting low pressure drop and high film heat transfer coefficients moderate acting temperature differences are applicable [7]. A low liquid holdup results in short mean residence times on the heated surface and thus induces minimum thermal stress to the product.

The thermodynamic and fluidic behavior of the fluid in the evaporator tube have been assessed in several studies with different methods and aims. The experimental setups differ in heating (e.g. steam, ohmic), geometry (plate, tube), test fluid and for the tubes external or internal fluid flow [14–18]. Prost et al. [14] used an industrial-type, steam heated evaporator to determine the heat transfer coefficient of a sucrose-water solution whereas Mc Adams et al. [15] used pure water in copper tubes. On the other hand, Alhusseini [16] and Chun and Seban [17] used ohmic heating and outside wetted tubes with different pure components. Binary mixtures on outside wetted tubes were investigated by Palen et al. [18]. However, references in open literature for higher viscous

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* Corresponding author.
E-mail address: S.Scholl@tu-braunschweig.de (S. Scholl).

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Nomenclature

A	heat transfer area, m^2
α	film heat transfer coefficient, $W\ m^{-2}\ K^{-1}$
a	thermal diffusivity, $m^2\ s^{-1}$
c_P	heat capacity, $kJ\ kg^{-1}\ K^{-1}$
d	diameter, m
η	dynamic viscosity, $kg\ m^{-1}\ s^{-1}$
Δh_v	evaporation enthalpy, $kJ\ kg^{-1}$
k	overall heat transfer coefficient, $W\ m^{-2}\ K^{-1}$
l	length of evaporator tube, m
Nu	Nusselt number, –
λ	thermal conductivity, $W\ m^{-1}\ K^{-1}$
$\dot{m}$	mass flow, $kg\ s^{-1}$
Pr	Prandtl number, –
$\dot{Q}$	heat flux, kW
$\dot{q}$	heat flux density, $kW\ m^{-2}$
Re	film Reynolds number, –
s	wall thickness of tube, mm
T	temperature, K
ΔT	temperature difference, K
Γ	wetting rate, $kg\ m^{-1}\ s^{-1}$
v	kinematic viscosity, $m^2\ s^{-1}$

Indices

c	characteristic
Cond	heating steam condensate
Evap	evaporation
Film	film
I	inner
IL1	ionic liquid 1
IL2	ionic liquid 2
In	inlet
Lam	laminar
Log	logarithmic
Max	maximum
Min	minimum
O	outer
Out	outlet
SS	shell side
TS	tube side
Turb	turbulent
V	vapor

fluids and their thermal behavior in FFE are rare and in most cases based on simulations or modeling [11,20]. Experiments with IL in larger scale equipment as described have not been reported so far.

2. Experimental setup

The experiments were conducted in a single tube metal falling film evaporator with dimensions of $d_o \times s \times l = 32 \times 2 \times 2770$ mm, see Fig. 1. The heated section of the tube was 2700 mm as the distributor was not heated. The distribution of the film was realized using a flat weir. The liquid film on the inside of the inner tube was heated by condensing steam in the shell tube. All continuous experiments were carried out with a constant pump around flow of 150 L h^{-1}, regulated via a flow-meter controlled pump, resulting in a liquid load (wetting rate) per unit inner circumference at the tube inlet of $\Gamma_{IL1} = 0.568\ kg\ m^{-1}\ s^{-1}$ for IL1 and $\Gamma_{IL2} = 0.557\ kg\ m^{-1}\ s^{-1}$ for IL2. At this wetting rate a sufficient wetting of the tubes' inner wall and the formation of a fully developed liquid film could be ensured for all experiments. Depending on the viscosity, which changes with temperature and water content, the film Reynolds numbers were in a range of $14 < Re_{film} < 52$. The film Reynolds number was calculated according to Brauer [12]

$$Re_{film} = \frac{\Gamma}{\eta_{film}} \tag{1}$$

The pressure at product side was set to 50 mbar, the vapor pressure at service side was set to 2000 mbar which equals a condensation temperature of 120.26 °C [8]. These settings were chosen to exert the identical thermal stress as in batch experiments [5]. Pressure drop on both sides was neglected. The pump around flow of the IL/water mixture from the holding vessel D1 into the evaporator tube E1 and back was initiated via gear pump P1. The mixture was pre-heated using the heat exchangers E2 and E3. The steam condensate from E3 was collected in D5. Within the evaporator tube E1 the thin liquid film is partially evaporated with liquid and vapor phase in cocurrent flow. The vapor was separated from the liquid phase in the bottom part of the evaporator. The vapor was condensed in the cooler E5, collected in the product tank D3 and measured with the scale WIR 450. The concentrated IL/water mixture from the evaporator flowed back into the holding vessel D1 to be recirculated. The film temperature of the liquid was measured at the tube outlet via TIR 200, see Fig. 1. The total liquid product holdup of the FFE including tank D1 was about five liter.

Continuous dehydration of IL was operated at a constant feed rate which was controlled via pump P2 and valve V1 respectively. Local temperature, pressure and flow rates were constantly monitored and logged utilizing the process control program "LabVision" (HiTec Zang GmbH, Aachen/Germany). In this setup, the feed solution with high water load was pumped from the feed vessel D2 into the pump around circuit downstream of the bottom draw using the pump P2. The bottom draw was situated shortly behind the pump P1 and was collected in the bottom draw vessel D4. Feed drum D2, bottom draw tank D4, product condensate vessel D3 and steam condensate tank D7 were positioned on scales to determine each mass flow. The feed flow via pump P2 was varied in a range of 1–4 kg h^{-1} and the bottom draw was adjusted to ensure a constant level in the FFE concentrate tank D1. The distillate mass flux represents the evaporation capacity of the FFE under the given operating conditions. The distillate flux is obtained from the change of distillate mass over time, i.e. the slope of the product mass condensate per time, see Fig. 2. The bottom draw was controlled using valve V2, such that a constant liquid level was kept in tank D1.

Two streams of condensed water vapor from the service side were obtained, one from the outside of the inner tube, the other from the inside of the shell tube. The first condensate stream stands for the energy input to the evaporator, the second represents the energy loss to the environment. For energy balancing only the first stream was taken into account. This stream was cooled in the heat exchanger E4 and directed through the vessels D6 and D7 and into a bucket situated on a scale WIR440. The two vessels D6 and D7 enable a continuous removal of condensate from the service side while maintaining a constant service side pressure. This mass condensate was used to establish energy balances as basis for the calculation of heat transfer coefficients. The second stream, resulting from the energy loss to the environment was collected in D8 and was not taken into account for the energy balances.

The used IL in this work were dimethyl-dihydroxy-ethylammonium methylsulfonate [IL1] and 1-ethyl-3-methylimidazolium