Contents lists available at ScienceDirect

Food and Bioproducts Processing

journal homepage: www.elsevier.com/locate/fbp

Two industrial examples of coupling experiments and simulations for increasing quality and yield of distilled beverages

Martine Esteban-Decloux[a,b,c,*], *Sophie Deterre*[a,b,c], *Samir Kadir*[a,b,c], *Pierre Giampaoli*[a,b,c], *Joël Albet*[d,e], *Xavier Joulia*[f,g], *Olivier Baudouin*[h]

[a] *AgroParisTech, UMR 1145 GENIAL, F-91300 Massy, France*
[b] *INRA, UMR 1145 GENIAL, F-91300 Massy, France*
[c] *Cnam, UMR 1145 GENIAL, F-91300 Massy, France*
[d] *INRA, UMR 1010, F-31030 Toulouse Cedex 4, France*
[e] *Université de Toulouse, INPT-ENSIACET, Laboratoire de Chimie Agro-industrielle, F-31030 Toulouse, France*
[f] *Université de Toulouse INPT-UPS, Laboratoire de Génie Chimique, F-31030 Toulouse Cedex 4, France*
[g] *CNRS, Laboratoire de Génie Chimique, F-31062 Toulouse, France*
[h] *ProSim SA, Immeuble Stratège A, 51, rue Ampère, F-31670 Labège, France*

A B S T R A C T

The aim of this study was to check the ability of ProSim® software to model both continuous and batch distillations of two specific industrial units in order to obtain both a better understanding of the behaviour of aroma volatile components and a tool to optimise the still's operation. Simulations of multistage continuous distillation to produce neutral spirit from raw alcohol and of batch distillation to produce bitter orange distillate from bitter orange peels macerate were carried out with ProSimPlus and BatchColumn software. Simulations were compared with distillations performed in two industrial plants. For each case, the industrial plants were studied to determine all the operating parameters and the behaviour of certain compounds selected for their high concentration or quality impact. Then, the NRTL and Henry's law thermodynamic models were chosen. Simulation results of particular compositions of the selected compounds in the different extractions were analysed and compared with experimental measurements. Simulations represented faithfully the behaviour of compounds in the industrial plants. Therefore, it was possible for two totally separated cases to illustrate the interest of simulation software; for neutral spirit production to determine new operation set points in order to maximise productivity and improve quality for neutral spirit production and for bitter orange distillate production, to explain the choice of different cuts and the role of the presence of peels during distillation.

Keywords: Continuous distillation; Batch distillation; Simulation; Neutral spirit; Bitter orange; ProSim®

1. Introduction

Process simulators are very powerful tools that are scarcely used in food processing (Bon et al., 2009). The two main obstacles for their use in food processing are the lack of data on the different compounds involved and the complexity of the processes (Joulia, 2008).

Among the process simulators used to represent continuous distillation, ProSimPlus is a process engineering software package that performs rigorous mass and energy balance calculations for a wide range of industrial steady-state processes (ProSim, 2012). It was used by Decloux and Coustel (2005) to simulate a typical production plant of neutral spirit, which is a high purity ethanol used in the food, pharmaceutical and

* *Corresponding author at*: AgroParisTech, UMR 1145 GENIAL, F-91300 Massy, France. Tel.: +33 1 69 93 50 92; fax: +33 1 69 93 50 44.
E-mail address: martine.esteban-decloux@agroparistech.fr (M. Esteban-Decloux).
Received 27 December 2012; Received in revised form 29 September 2013; Accepted 3 October 2013
Available online 12 October 2013
0960-3085/$ – see front matter © 2013 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.fbp.2013.10.001

chemical industries. The entire distillation plant was comprised of a series of seven columns for concentrating and purifying, and six representative compounds plus ethanol and water were considered. Vapour–liquid equilibrium data were assessed using the UNIquac Functional-group Activity Coefficient (UNIFAC) modified Dortmund model (Gmehling et al., 1993), a group contribution predictive model available in Simulis Thermodynamics which is the thermodynamic properties and phase equilibria server for ProSim software. Their results allowed them to illustrate the specific role of each column in the sequence of purification steps performed during neutral spirit production. Nevertheless, they did not validate their results on an industrial scale. More recently, Batista and Meirelles (2011) simulated continuous Cachaça production with the Aspen Plus simulation software. They took into account ten compounds plus ethanol and water. The entire representation of the liquid–vapour equilibria by the Non Random Two Liquids (NRTL) model required the knowledge of three binary interaction parameters for each of the 66 associated binary mixtures. Binary parameters were adjusted for 43 binary mixtures from literature data. For the others, they used the NRTL parameters available in the Aspen databank or estimations from the UNIFAC predicted values. Simulation results were compared with data from the Santa Adélia Mill plant, which produces 300 m^3 of anhydrous ethanol on a daily basis. Good agreement between the simulated and experimental results made it possible to study the role of several process parameters and the degassing system. The same research group (Batista et al., 2012) validated the simulation of a typical bioethanol distillation process by considering an alcoholic wine with 19 components. More recently, Valderrama et al. (2012b) used the ChemCad process simulating software to simulate the behaviour of eight congeners in the production of neutral spirit from beer. They studied more particularly the variation of congeners' behaviour due to variation of the alcohol grade of the feed from 8.3 to 14.8% (v/v) using the NRTL thermodynamic model. Therefore, they were able to suggest a tool to optimise the positions and flows of lateral extractions.

Concerning the extraction of orange essential oil extraction, the recovery of aroma compounds from orange essential oil was simulated by Haypek et al. (2000) using the commercial steady state software Pro/II and the UNIFAC model for the calculation of vapour–liquid and liquid–liquid equilibria of 14 compounds plus ethanol and water. Since they obtained good agreement between the Cutrale industrial processing plant and simulated values for the first column used in the recovery of the aroma compounds from the aqueous stream during orange juice evaporation, they used the simulation to study the recovery of the aqueous essence phase.

Simulation of batch distillation is much more difficult than steady state simulation. Osorio et al. (2004) developed a mathematical model for simulating Pisco distillation as a multi-component reactive batch distillation process with reflux. In another study, Osorio et al. (2005) investigated, via process simulation, the operating recipes to obtain a distillate with a maximum of linalool and a minimum of octanoic acid. This recipe was validated with lab-scale experiments. Scanavini et al. (2010) studied Cachaça production in a lab-scale alembic. In that specific case, they modelled the alembic and compared the simulated and experimental results with seven components plus ethanol and water.

As the development of a simulation code is a time consuming task, both for setting up the problem and for solving it, Zamar et al. (2005) recommended the use of a simplified model even if it was less accurate. It is also possible to use commercial simulators. Claus and Berglund (2009) used the ChemCad batch distillation programme to model the batch multistage distillation still used to produce fruit spirits. They conducted experiments on the lab-scale (10 L and 150 L stills) and adjusted the simulation parameters to model the distillate flow rate, reflux ratio and ethanol concentration.

The objective of the present work was to check the ability of ProSim® software to model both continuous and batch distillations of two specific industrial units in order to obtain both a better understanding of the behaviour of aroma volatile components and a tool to optimise the stills' operation. The first part of the present work follows the study of Decloux and Coustel (2005) as it validated the modelling with ProSimPlus software of an industrial plant producing neutral spirit from raw alcohol. In this study, the UNIFAC predictive model was replaced by the NRTL with binary interaction parameters fitted on experimental data from literature. Once the plant was modelled, it first was possible to suggest several modifications of the circuits between the columns and then to use the optimisation capability of the software to search new control set points of the many fluxes between the columns with the view of maximising the productivity by reducing the ratio of low grade alcohol produced. The second part presents a batch distillation simulation with BatchColumn software in order to obtain bitter orange distillate. The aim was to develop a tool to explain the rationale behind the process control parameters, in particular, the choice of the separation of distillate fractions.

2. Simulation of the rectification plant of neutral spirit process

2.1. *Description of the neutral spirit continuous distillation plant*

Neutral spirit is produced by distillation from a must obtained by fermentation of diluted beet molasses or beet juice with *Saccharomyces cerevisiae* yeasts. This solution is commonly called wine, regardless of the raw material. The wine, composed mainly of water and ethanol (ethanol mass fraction around 0.08), contains volatile components at a lower concentration than ethanol that are referred to as congeners (esters, alcohols, aldehydes, acids or bases, amines or sulphur compounds). Production of neutral spirit from wine is achieved in two stages: (1) production of a raw alcohol with an ethanol concentration in volume around 93% (v/v) containing concentrations of volatile congeners that are greater than desired, followed by (2) purification of the raw alcohol by rectification to eliminate the remaining volatile congeners. This last separation unit requires at least four interconnected and energy integrated distillation columns (Decloux and Coustel, 2005). This study focused on the purification stage of raw alcohol. Fig. 1 presents the rectification unit of the industrial plant. It was composed of four columns named C30, C40, C50 and C60 and a liquid–liquid separator (decanter) S60.

Each column had a specific role. C30 (52 trays) was an extractive distillation column with water as the solvent. In this column, most of the congeners were removed into the head (H30). Water came partly from C40 bottom residue (B40) and partly from demineralised water (DW30). For good separation, Jacques et al. (2003) advised a ratio of nine parts water for one part alcohol. The effectiveness of this extractive column on each congener i is related to its relative volatility ($\alpha_{i,ethanol}$) to