

APPLICATION OF NUMERICAL MODELLING TO SCALING-UP OF ELECTRICALLY INDUCED EXTRACTION FROM AN ORGANIC MIXTURE USING AN IONIC LIQUID

Kamil Kamiński¹, Laurence R. Weatherley², Jerzy Petera^{*1}

¹Lodz University of Technology, Faculty of Process and Environmental Engineering, Wólczajska 213, 90-924 Łódź, Poland

²Department of Chemical and Petroleum Engineering, University of Kansas, Lawrence KS66045, USA

Dedicated to Prof. Ryszard Pohorecki on the occasion of his 80th birthday

Liquid-liquid extraction provides an environmentally friendly process as an alternative to azeotropic distillation, pervaporation and reverse osmosis because these techniques require the use of large amounts of energy, may involve volatile organic compounds, and operation at high pressure.

Ionic liquids (ILs) continue to gain wide recognition as potential environmentally friendly solvents due to their unique properties. However due to their current high cost, their use in industry is seriously limited without an efficient methodology for recovery and recycle.

In this paper we describe an innovative methodology for a liquid-liquid extraction process based on an electrically induced emulsion of an ionic liquid as the extracting solvent dispersed in an organic mixture. This offers a most efficient exploitation of the solvent. On the other hand we present our own design of a pilot (semi-industrial) scale extractor based on this methodology and which demonstrates effective recovery of the ionic liquid. In order to achieve this goal we used a numerical modelling tool implemented using our own simulation software based on the finite element method. We also used our original previous experience with generating and investigating liquid-liquid electrosprays using phase Doppler anemometry. Finally we present recommendations for contactor geometry and for the preferred operating conditions for the extractor.

Keywords: scaling-up, finite element analysis, liquid-liquid extraction, ionic liquids, electrospray

1. INTRODUCTION

Extraction in a liquid-liquid system is a unit operation which may be applied for product separation in the chemical and petrochemical industries. In many industrial processes the separation of close-boiling or azeotropic mixtures may be necessary. It is an important problem in chemicals production, since conventional distillation cannot be used to obtain pure products. Liquid-liquid extraction may be considered an alternative method for the separation, in particular enabling azeotrope breakage. The most common methods to separate azeotropic mixtures include extractive distillation, membrane extraction, pervaporation and reversed osmosis. However there are major drawbacks associated with these methods including high-energy requirements, use of volatile organic compounds, operation at either high or very low pressures. In light of such drawbacks, liquid-liquid extraction may be considered a more environmentally friendly separation process. The modern variant of classical liquid-

*Corresponding author, e-mail: jerzy.petera@p.lodz.pl

cpe.czasopisma.pan.pl; degruyter.com/view/j/cpe

liquid extraction presented here is separation of organic mixtures with the use of ionic liquids in the presence of an electric field.

Ionic liquids are known as “designer solvents” because their structure can be molecularly engineered for specific tasks through the appropriate choice of the cation/anion combination. These molten salts might be also considered as attractive substitutes for conventional organic solvents because of their physico-chemical properties, such as good thermal stability, low vapour pressure at room temperature and selective miscibility with water and organic solvents (Li Zaijun et al., 2011). The main disadvantage of ionic liquids is high viscosity, which may be detrimental to the attainment of high interfacial area and also can be a cause of slow heat and mass transfer. One of the methods for developing interfacial area and enhancing the mass transfer in a liquid-liquid system is application of an electric field. Using electrical forces for efficient dispersion of ionic liquids is a low energy method proposed as a substitute for conventional mechanical agitation.

Ionic liquids can act as effective extractants for azeotropic, organic mixture separation processes. Application of ionic liquids as solvents for the petrochemical extraction process for the removal of alkane from its azeotropic mixture with alcohol (Pereiro and Rodriguez, 2008; Pereiro et al. 2006) and separation of the ester-alkanol system (Xuesheng Hu et al., 2008) may serve as examples. Ionic liquids were also successfully tested in extraction of aromatic hydrocarbons from their mixtures with aliphatic compounds (Garcia et al., 2012). At present, aromatic extraction at industrial scale is usually performed using sulfolane. However, ionic liquids may be considered as an attractive substitute for the organic solvent in the sulfolane process if the concentration of aromatics is low (Larriba et al., 2014). Ionic liquids have also been studied as novel extraction agents for the desulfurization of fuel-oils (Rodriguez-Cabo et al., 2013).

Generally, imidazolium based ionic liquids (IL) are characterised by high chemical and thermal stability and comparatively low viscosity. The literature provides strong evidence of the ionic liquid, 1-butyl-3-methylimidazolium methylsulfate [BMIM] [MeSO₄], to act effectively as an extraction solvent in petrochemical process for the removal of heptane from its azeotropic mixture with ethanol (Pereiro, Rodriguez, 2008). Particularly, the suitability of this IL for the ternary system heptane + ethanol + [BMIM] [MeSO₄] was proved. The solute distribution ratio, the selectivity and the extraction factor assessed at 298.15K by means of the experimental liquid-liquid equilibrium confirmed the solvent efficiency. A comparison with other IL in terms of solvent capacity, 1-Hexyl-3-methylimidazolium hexafluorophosphate [HMIM][PF₆] (also considered as a good candidate for this extraction process, Pereiro et al. (2006)) proved superiority of the former one.

Most of the research to date associated with ionic liquids has been conducted in the laboratory scale. This was mainly due to current high costs of ionic liquids and thus practical use of these solvents must be accompanied with efficient regeneration (recovery) process. Here in this research an attempt is made to design the extraction process to be conducted at pilot scale (semi-industrial scale) preferably in the continuous regime. The main objective was to define a preferred geometry and operational conditions on the basis of our significant experience gained during previous research over the last decade. Particularly we show significant intensification of the extraction process by use of an electric field to produce electrosprays of ionic liquids which are injected into a continuous organic phase. This method develops sprays of high interfacial area, comprising drops which can be close to being monodispersed. In addition, the presence of electrical charge may also create phenomena at the interface which further intensify mass transport. One of the tools used here is numerical modelling methodology based on the finite element method implemented in our own simulation software developed over last two decades.

2. LIQUID-LIQUID EXTRACTION AT LABORATORY SCALE

As mentioned above, the ultimate goal of the research presented here is development of a pilot scale extraction system for potential application in industry (e.g. pharmaceutical). In order to design an appropriate contactor expertise gained during the small scale laboratory experiments was used in parallel with our own original numerical modelling program. The proposed numerical model was validated with experimental results obtained at laboratory scale described in prior work (Kamiński et al., 2014).

In the experiments, ethanol was extracted from a mixture with n-heptane using an ionic liquid, [bmim][MeSO₄] in the presence of an electric field. In all our experiments we used 1-butyl-3-methylimidazolium methylsulfate (CAS Number: 401788-98-5) produced by BASF for Sigma-Aldrich, with a purity of more than 95%. It should also be noted that the ionic liquid was reused after every series of experiment after purification using a rotary evaporator with a vacuum pump. Purification of the extract was relatively easy because of negligible volatility of the ionic liquid. This fact is important in view of potential application of the method in the industrial scale process. The ionic liquid can be reused without significant decrease of its quality, thus resulting in manufacturing cost decrease. The relevant physical properties of the mixture components are tabulated below in Table 1. These data were taken from the literature (Beigi et al., 2013; Pereiro et al., 2007; Zeberg-Mikkelsen et al., 2006).

Table 1. Physical properties of the three components in the system

At temperature 25°C	n-heptane	ethanol	[bmim][MeSO ₄]
Viscosity [mPa·s]	0.3715	1.01	213.2
Density [kg/m ³]	679.8	785.5	1212.2
Electric conductivity [μS/cm]	0	0.0013	20.0
Electric permittivity [-]	1.9	24.3	-

The contactor comprised a cylindrical glass column with a diameter of 0.1 m and with a volume of about 2 dm³. The distance between the nozzle (earthed electrode) and the high voltage electrode was set to 0.2 m. At the beginning of the experiment the contactor was filled with 1.75 dm³ of feed containing 32.6% by mass of ethanol. The ionic liquid was pumped into the organic mixture through a dispersion nozzle using a diaphragm pump at a constant flowrate of 90 ml/h. During the experiment samples of raffinate phase were gathered after 30 minutes from the beginning and periodically at one hour intervals. The collected samples were analysed by gas chromatography and as a result concentrations of ethanol in raffinate phase were determined. Additionally another experiment without application of the electric field was performed as a control against which the experiments conducted in the presence of electrical field could be compared, see Figure 1. In the control experiment, the volumetric flowrates of ionic liquid, the initial concentration of ethanol, the distance between electrodes and the nozzle diameter were exactly the same as in the experiment conducted in the presence of the electric field.

Fig. 1 shows that the measured concentrations of ethanol in the continuous liquid phase in the column as time progressed were considerably lower when the electric field was applied. The final amount of the remaining alcohol obtained in electrically driven extraction was about two-and-a-half times lower in comparison to the experiment without the electric field, which proves significant enhancement of mass transfer by electrical forces in the studied liquid-liquid system. It can be observed that for the first three hours of the experiment, the current stabilization system significantly reduced the electric potential difference applied between the electrodes in order to keep the current at a constant level see Fig. 2 and Fig. 3. As a result of current stabilisation, poor dispersion of the ionic liquid in the extractor was observed, since the size of produced droplets depends on the voltage between the nozzle and the

electrode (Kamiński et al., 2014). However, even during that time, separation of the organic compounds was more effective when the electric field was applied. After three hours of the experiment the applied voltage increased to 4kV, as the conductivity of the organic mixture in the extractor dropped, and the ionic liquid was dispersed to fine droplets. Results presented in Fig. 1 shows that the reduction in ethanol concentration was the most significant during the last hour of the experiment, in the electrostatic regime.

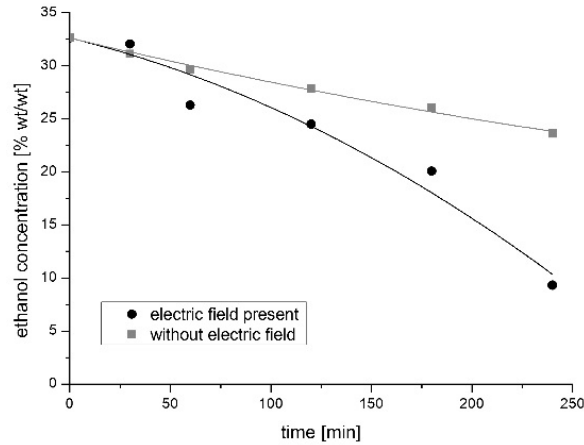


Fig. 1. Changes of ethanol concentration in time for the experiment with (black circles) and without (gray squares) electric field, presented as a function of time (for 32.6% initial concentration of alcohol - EtOH)

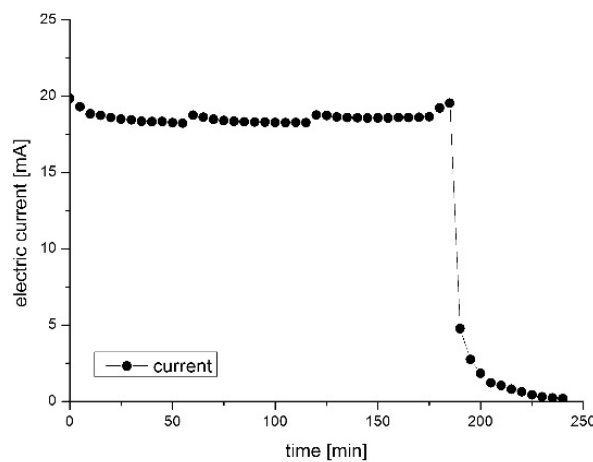


Fig. 2. Electric current presented as a function of time (for 32.6% initial concentration of EtOH)

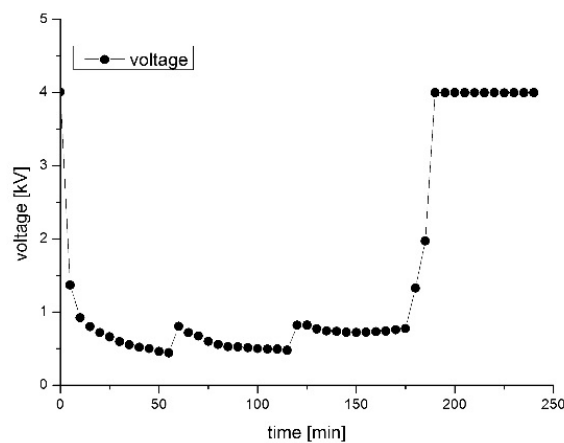


Fig. 3. Voltage presented as a function of time (for 32.6% initial concentration of EtOH)

3. NUMERICAL MODEL OF THE MASS TRANSFER BETWEEN DISPERSED AND CONTINUOUS PHASES

The numerical simulation described below aimed to reflect all the phenomena occurring during the subsequent experimental program. Particularly we wanted to investigate important features of the electrostatically induced extraction with the use of ionic liquids as follows:

- enhanced mixing, caused by the clouds of droplets and their motion inside the contactor,
- increase of mass transfer rate at higher voltages between the electrodes,
- investigating of the electric current characteristics which may dramatically influence the extraction process to be applied in a pilot scale experimental procedure.

The theoretical approach adopted here was based upon the fundamental relationships governing:

- the electrical field and potential distribution,
- the equations of motion for both continuous and dispersed phases (taking into account mutual coupling in their hydrodynamics), the mass transfer between the phases, and the electric current distribution.

The solution of the field equations was obtained rigorously using our own simulation software based on the finite element method (FEM). Most of the details of our numerical algorithms have been described elsewhere (Kamiński et al., 2014; Petera et al., 2009).

The solution of the momentum and continuity equations computationally is by far the most expensive fragment of the general algorithm. We applied a state of the art numerical solver developed in project "PARDISO" (Schenk and Gartner, 2002; Schenk and Gartner, 2004) specially optimised for Intel processor architecture available in the Fortran compiler contained in Intel Parallel Studio. We adapted that direct solver for our own iterative procedure in which the full solution was done only once and the system matrix was kept in memory while the right hand side was updated at every time step. The modification of the right hand side resulted from variation of the local viscosity (due to change of the concentration) and variation of the discrete phase distribution.

The mass transport coefficient was calculated on the basis of known formulae given for example in our previous papers (Kamiński et al., 2014; Petera et al., 2009). Some basic formulae are cited below to facilitate the main assumptions and the numerical procedure. The mass flux r_A between droplet surface and the continuous phase may be expressed as:

$$r_A = \rho_c k_c (x_{Et} - x_{Et,i}) \pi d^2 \quad (1)$$

where $x_{Et} - x_{Et,i}$ is the difference between ethanol mass fraction in the raffinate and its value at the interface.

For very small drops in a liquid-liquid system as in the present case, the mass transport coefficient can be calculated on the basis of rigid spherical droplets, from the very well known Newman formula.

We use the standard procedure for deriving the total mass transfer rate to obtain

$$r_M = K_{tot} a_d (x_{Et} - x_{Et,e}), \quad r_{Ma} = \frac{r_M}{a_d} = K_{tot} (x_{Et} - x_{Et,e}) \quad [\text{kg}/(\text{m}^2 \cdot \text{s})] \quad (2)$$

where

$$K_{tot} = \frac{1}{\frac{1}{k_c \rho_c} + \frac{1}{k_e k_d \rho_d}} \quad (3)$$

and $a_d = n_d \pi d^2$ (can be understood as the specific interface area a_v), calculated locally on the basis of the droplet number density distribution n_d , calculated independently from the clouds characteristics; $x_{Et} - x_{Et,e}$ is the total driving force.

Then, the distribution coefficient $k_e = y^*/x$ defining the liquid-liquid equilibrium was obtained as described later, and of course $x_{Et,e} = y_{Et}/k_e$. We have used a simple correlation for k_e justified by numerical procedure, which calculates the distribution coefficient on the basis of actual concentration in discrete phase to calculate the corresponding equilibrium value in the continuous phase. The driving force $x_{Et} - x_{Et,e}$ is then obtained.

$$k_e = A - B y, \text{ hence } y^* = \frac{A}{B + \frac{1}{x}} \quad (4)$$

For the purpose of our computations, we used data taken from literature (Pereiro and Rodriguez, 2008) for which we estimated $A = 27.9$ and $B = 46$. For the mass transfer in the discrete phase we have

$$\frac{d(y_{Et} m_d)}{dt} = \pi d^2 r_{Ma} \quad (5)$$

where in the particular case of a single droplet the expression m_d is the mass of a droplet that represents a cloud of droplets of the same diameter.

The last equation is in principle a first order differential equation and was solved numerically within the overall algorithm giving the concentration evolution inside a droplet as the result of mass transfer. The resultant mass transported to/from continuous phase is calculated as the sum of local contributions from different clouds. This gives evolution of the concentration of the components involved in the reaction.

4. NUMERICAL MODELLING FOR THE PURPOSE OF THE SCALING UP PROCEDURE

Using this algorithm the numerical simulation of the real extraction as described above was performed. The computational mesh for the laboratory scale contactor (generated by the commercial software ANSYS™), consisted of 198241 tetrahedral elements, and 36721 nodal points and was presented in our previous paper (Kamiński et al., 2014). Based on comparison with experiment at this laboratory scale some model parameters such as dispersion coefficient in the continuous phase were calibrated for future use in case of similar process simulation (e.g. the subsequent pilot scale investigation).

The computational mesh for the pilot scale contactor (also generated by the commercial software ANSYS™), was prepared for several variants of geometry and anticipated operating conditions. These included the variants with a partially and fully filled container and particularly important, different thickness of the high voltage electrode which resulted in different levels of electric field strength. The variant presented consistently throughout the subsequent analysis consisted of 436240 tetrahedral elements, and 74626 nodal points. The initial and boundary conditions (analogous to the case of laboratory scale) are shown in Fig. 4.

First we investigated the flow patterns in the contactor resulting from the flow imposition forced by the pump between inlet and the main outlet 1, on one hand, and the secondary flows generated by the dispersed phase motion under external electric field combined with the drag and gravity, on the other. The distance between the upper (earthed) electrodes and their space configuration should not be detrimental to the free circulation of the continuous phase. As seen from Fig. 5 this aim was achieved.

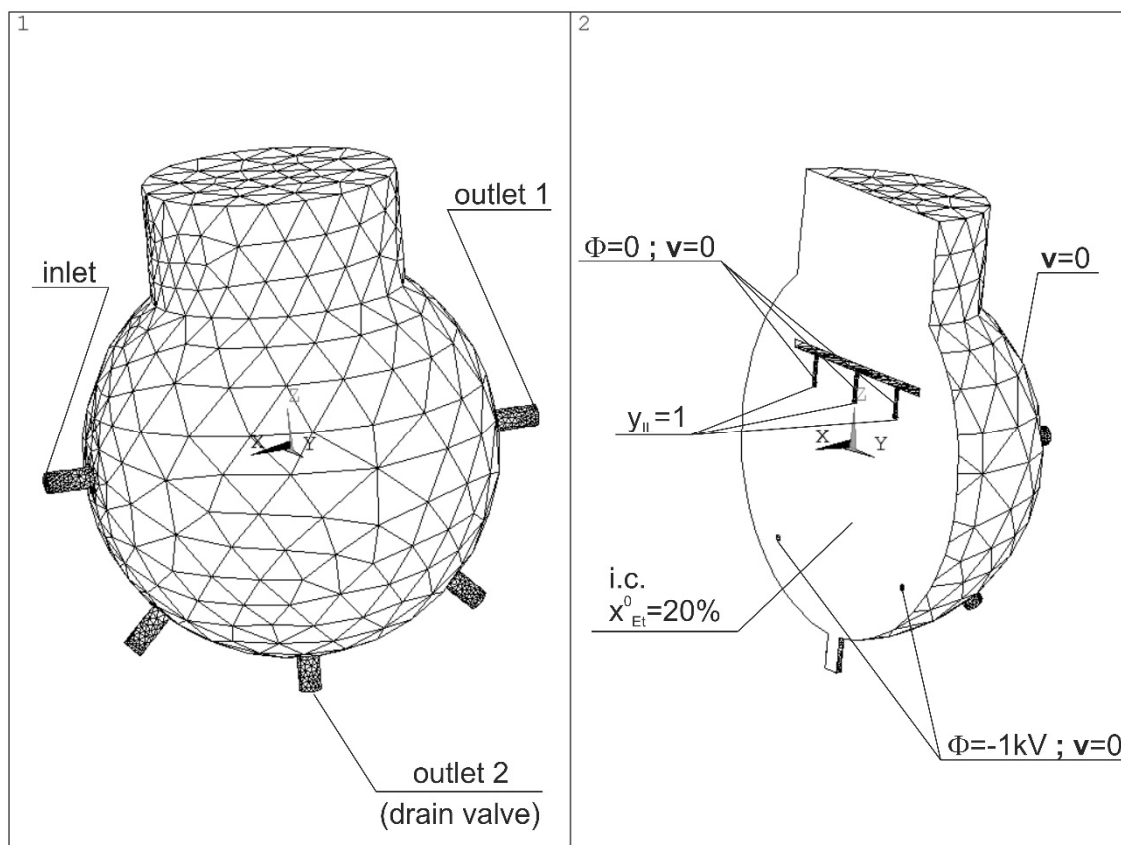


Fig. 4. Computational mesh and the initial and boundary conditions for the spherical extractor

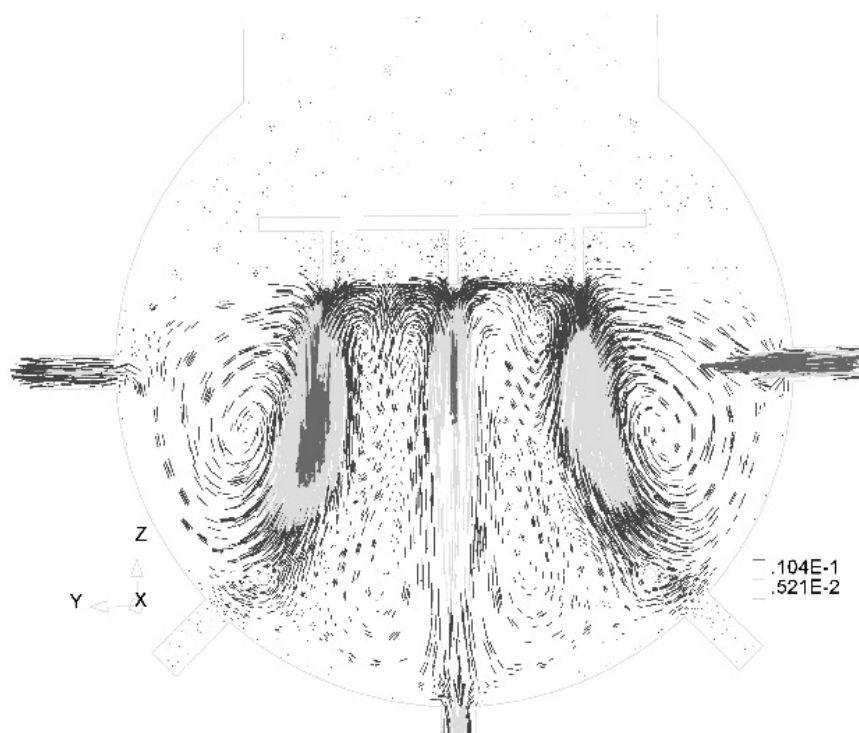


Fig. 5. Development of the secondary flow in the continuous organic phase generated by motion of the electrically dispersed ionic liquid droplets

The predicted electrical potential field in the contactor is shown in Fig. 6. The upper (nine) earthed electrodes serve as the nozzles producing the electro spray while the lower one is the high voltage ring

(HV-electrode) under negative electric potential ranging between 0 to -5kV. One can see that the electric field is distributed fairly uniformly due to the correct space configuration of the electrodes.

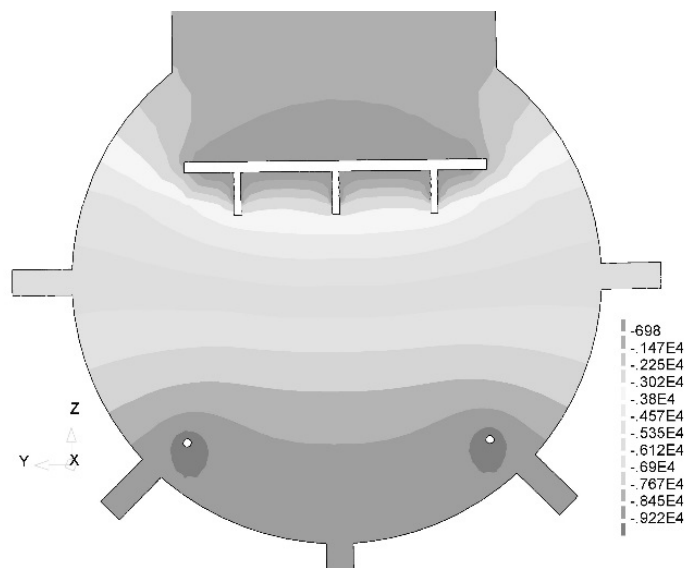


Fig. 6. The electric potential contours in the spherical contactor centerline 2D section

Figure 7 gives an example of the droplet dispersion (holdup) simulated by means of the cloud model, and a 3D picture of clouds is presented in the form of the droplet volumetric concentration contours. The centerlines of each monodispersed cloud are also visible. As mentioned, all monodispersed clouds are assembled statistically to form a poly-dispersed discrete phase. They reflect quite well the shape and distribution of the real clouds presented in the photographs later in this paper.



Fig. 7. The droplet dispersion (holdup) simulated by means of the cloud model, 3D picture of clouds

Then for the continuous phase the concentration field for the ethanol after 2 hours of extraction process is shown in the Fig. 8. There is no doubt that due to the higher number of droplets and the bigger interfacial area near the center of the contactor the efficiency is higher in this region. This results in the lower concentration zone of ethanol.

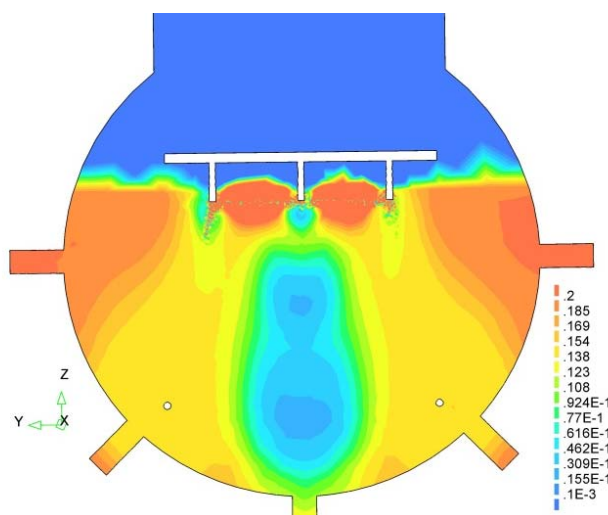


Fig. 8. Calculated ethanol concentration profiles in the contactor at applied electrical potential -1 kV after 120 min

The simulations were repeated for all variants of geometry and operating conditions investigated in this research. Visually and thus qualitatively, all the results were similar and not presented here for the other variants. However, quantitative comparison expressed in the form of average final concentration after completion of each experiment exhibited significant differences. The average final concentration is a measure of effectiveness of the apparatus equivalent to the amount of the extracted component per unit volume at the defined operating conditions (electric potential, volumetric flow rate, amount of organic phase, etc.).

Figure 9 shows the comparison of the average concentration evolution in time for three selected variants, defined as follows: 1) fully filled, thick HV-electrode, 2) fully filled, thin HV-electrode, 3) partially filled, thick HV-electrode.

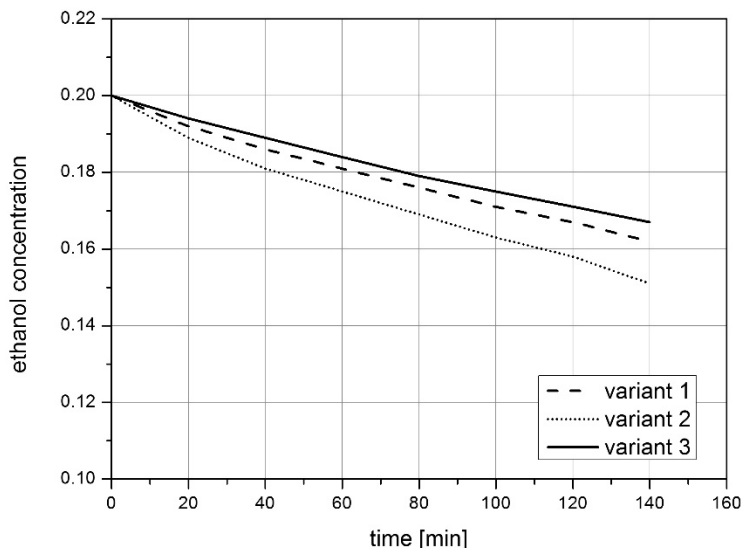


Fig. 9. Comparison of the simulated ethanol concentration evolution in time for the three variants of the contactor design

The advantage of the thin HV-electrode is evident. It produces much higher electric field strength and thus more intensive mixing generated by the dispersed phase due to higher electrophoresis forces. The next set of comparisons, Fig. 10, confirms the conjecture showing much higher concentration of arrows of the electric field strength near the HV-electrode.

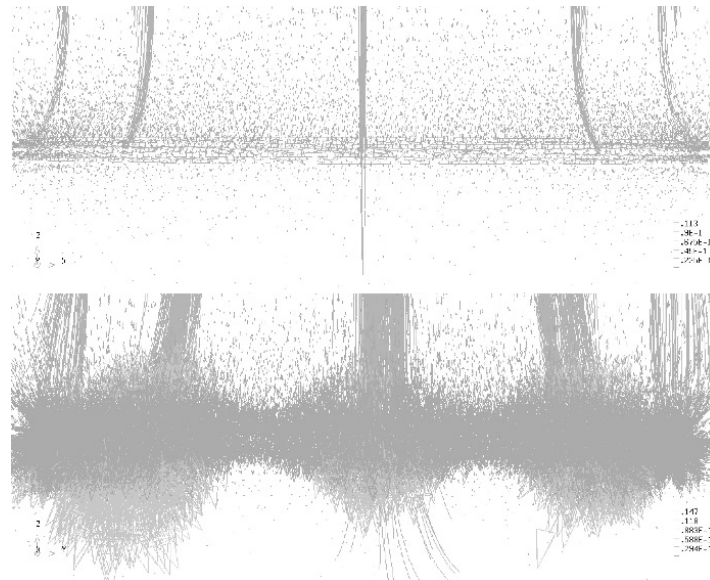


Fig. 10. Comparison of the electric field strength at the HV-electrodes;
Upper picture: thick electrode; Bottom picture: thin electrode

Apart from this observation we conclude that the longer travel distance covered by the droplets due to a fully filled contactor also creates congenial conditions for longer mass transfer process experienced by each droplet.

The last issue which has to be taken into account is the limitation that must be imposed on the electric current. Although an automatic control of the current is inherently implemented in our electric power generator, in some circumstances the variation of the electric conductivity during the process (due to change of mixture composition) may inhibit the spray stability or at least deteriorate the quality of the spray. In order to limit the current, a greater separation distance between the electrodes is thus recommended. This was confirmed by our simulations as can be seen in Figure 11 where full filling of the contactor gave a lower range of current level.

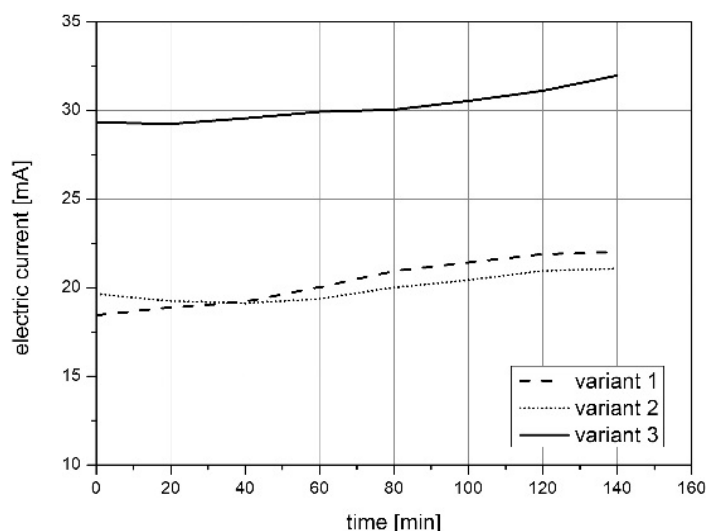


Fig. 11. Comparison of the simulated electric current evolution in time for the three variants of the contactor design

This conclusion was in agreement with subsequent experimental observation when conducting the real process where a stable spray was produced as described below.

5. LIQUID-LIQUID EXTRACTOR AT PILOT SCALE

The most important task for scaling-up was to test the possibility of electrically enhanced extraction of alcohol from an organic mixture with the use of an ionic liquid at pilot scale. Based on conclusions drawn from the numerical modelling within the constraints resulting from availability of appropriate equipment on the market a new spherical borosilicate glass contactor with a maximum volume of about 50 dm³ and a diameter of 0.49 m was designed and built (Fig. 12).

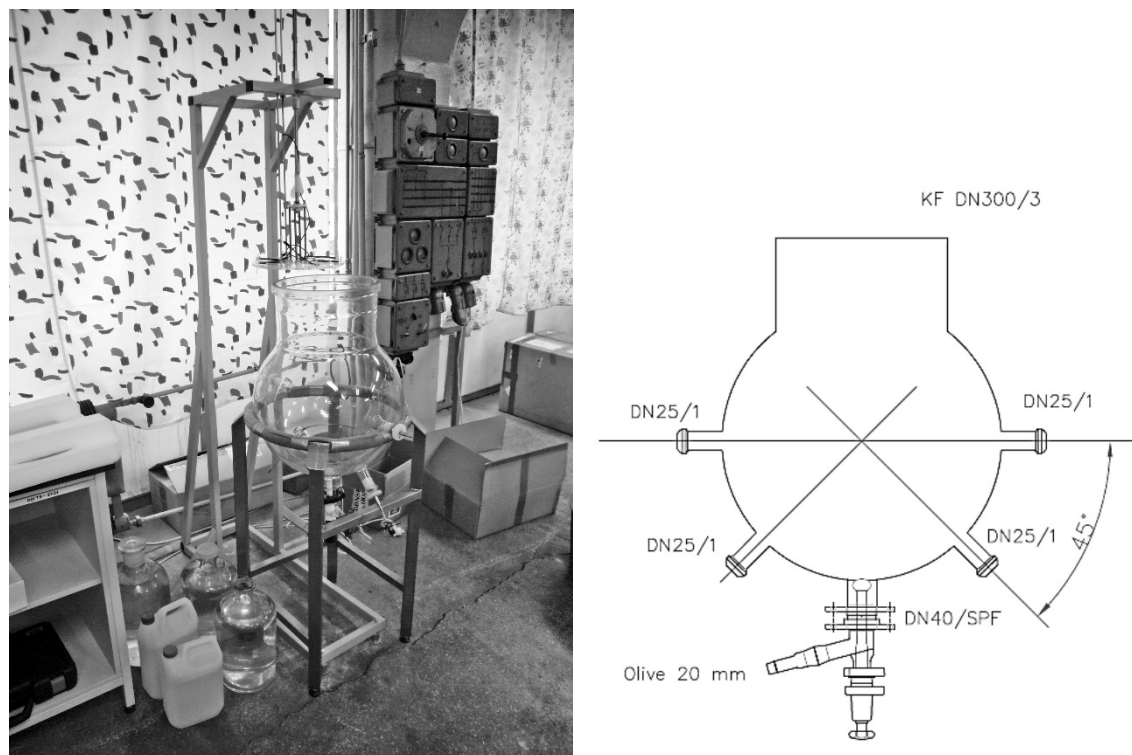


Fig. 12. Pilot scale spherical glass contactor for ethanol extraction (overall view on the left and a draft with dimensions on the right)

Since the weight of the contactor filled with a feed can be significant a special stainless steel support was constructed on which the contactor was located. On a longitudinal symmetry plane of the contactor four cylindrical ports with diameters of 0.025 m were located. Two of the ports near the top of the contactor were used as inlet for the extraction feed and for outlet of the raffinate respectively. One of the ports located close to the base of the contactor enabled connection of a high voltage electrode. The remaining port was blanked off. Because all the ports have slightly different profiles, four different polyamide plugs were designed and produced individually. The plugs together with rubber seals and fitting systems ensured avoidance of leakage of chemicals from the contactor. Near the bottom part of the extractor a port with a diameter of 0.04 m was located. The port was connected to a PTFE drain valve which enabled removal of the extract, as a heavier phase, from the contactor.

The high voltage circular electrode, made of stainless steel rod of a diameter of 0.004 m (the same as in the recommended numerical variant), was placed near the bottom of the contactor. The electrode was connected to a 100 W high voltage power source capable of delivering electric potential up to 5 kV at a maximum current of 20 mA. A specially designed multi-nozzle system for feeding the ionic liquid into the organic continuous phase was located at the top of the contactor. The system consisted of an ionic liquid distributor connected through silicon pipes with nine nozzles made of medical needles. One nozzle was placed at the centre of the acrylic glass cover which had a diameter corresponding to the diameter of the high voltage electrode. The remaining eight nozzles were arranged circumferentially

around the cover. All the nozzles and the distributor were earthed. The distance between the high voltage electrode and the nozzles was considered to an important parameter for effective dispersion of the ionic liquid. Therefore the multi-nozzle system was designed to enable variation of the vertical distance between the nozzle assembly and lower electrode. The position of the nozzles could be adjusted manually using a long bolt with a bearing connected to the cover of the contactor and to the overall supporting steelwork as shown in Fig. 13.

Two diaphragm pumps were used for introducing both the organic continuous phase and the ionic liquid dispersed phase, see Fig. 13. The heads of the pumps were made of stainless steel and included PTFE diaphragms in order to ensure compatibility with the liquid phases. The delivery pressure rating of the pumps was selected to allow accurate and stable pumping of the relatively viscous ionic liquid through the nozzles. Raffinate was received from one of the upper ports gravitationally and afterwards was delivered to a separator made of two coaxial polycarbonate tubes. At the end of the inner tube, a filter made of small ceramic spherules was installed. The main function of the filter was to remove traces of entrained ionic liquid from the raffinate. After the filter, in the space between the inner and the outer tube, the remaining dispersed ionic liquid could settle gravitationally. Finally, the clear raffinate was received at the top of the outer tube by use of another pump. The raffinate could be either recirculated to the extractor, or treated as a product, depending on the concentration of the remaining alcohol. Using the separator was necessary because the electric field can produce very fine droplets of ionic liquid which do not easily settle gravitationally in the contactor.



Fig. 13. The experimental setup for electrically induced extraction in a pilot scale contactor

The proposed experimental setup for separation of organic mixtures in the presence of an electric field, as described in the modelling section of this paper, concerned extraction of ethanol from a mixture with n-heptane using 1-butyl-3-methylimidazolium methylsulfate ([bmim][MeSO₄]). A general concept of the experiment and the arrangement of fluid flows are shown in Fig. 14. At the beginning of the experiment the contactor was filled with the mixture of ethanol and n-heptane. The mixture initially included 20% (wt/wt) of alcohol, the same as in the modelling part above. The tips of the nozzles were immersed in the organic mixture and the electric field was applied across the space between the nozzle assembly and the high voltage electrode.

The experiment demonstrated that electrically induced extraction of ethanol at pilot scale is possible. At the tips of the nine nozzles the ionic liquid was satisfactorily dispersed to fine droplets (Fig. 15) which generally is not obvious. As proposed in the simulation, from visible observation, the multi-nozzle

system enabled satisfactory distribution of the dispersed phase over the whole volume of the extractor which should give a satisfactory concentration distribution (see the simulation results, Fig. 8).

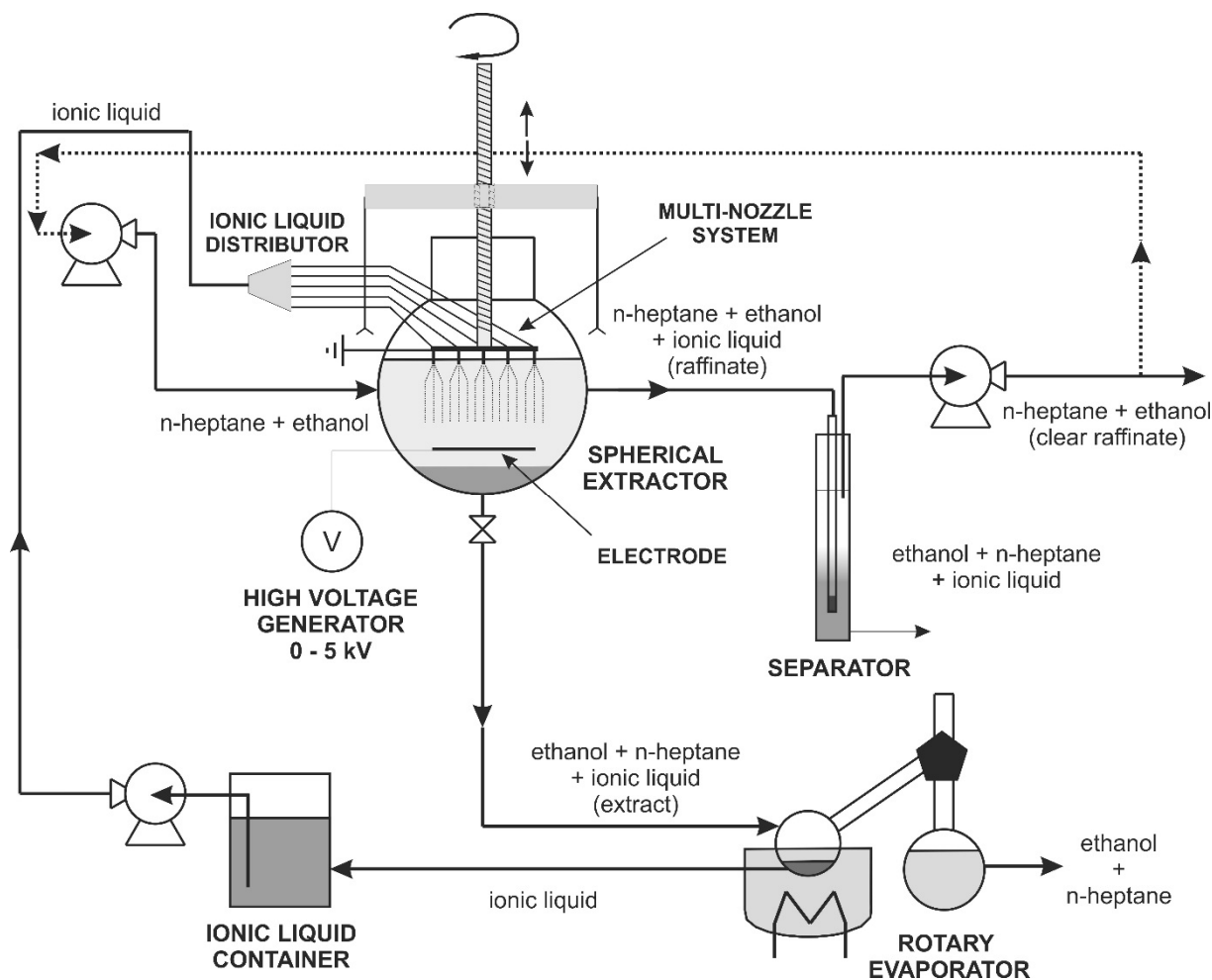


Fig. 14. Scheme of the general experimental arrangement



Fig. 15. Electro-spray produced in the extractor

The extract phase (mainly ethanol and the ionic liquid) settled gravitationally at the bottom of the contactor. However, after a few minutes the continuous phase became cloudy (Fig. 16) which confirmed the necessity of the filter-separator for obtaining a clear raffinate. It was observed that increasing the distance between the electrode and the nozzles resulted in an increase of applied electric

potential applied across the contactor in comparison to the laboratory scale extraction column (with the electric current stabilized at the level of 20 mA). It was also observed that dispersion under the electric field was “current limited”. Due to progressive mass transfer between the phases in the contactor the conductivity of the continuous phase increased. When the electric current in the extractor reached the maximum level (20 mA) the stabilization system of the high voltage generator limited the electric potential and as a result larger discrete droplets of the ionic liquid were produced.

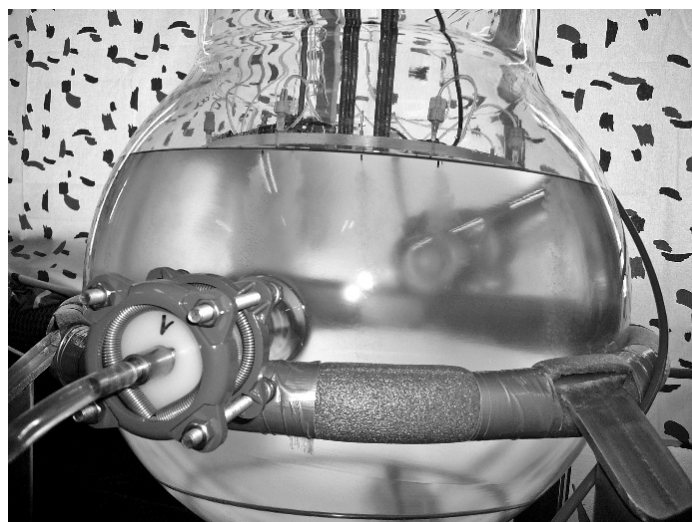


Fig. 16. The organic mixture became cloudy in the course of time; on the left: one of ports with a polyamide plug and a sealing system

6. CONCLUSIONS

In our opinion a consistent method for the new extractor design was proposed. Based on our preliminary experiments conducted in a semi-industrial pilot scale contactor we demonstrated stable behaviour of the spray once the dispersed (electrostatic) regime was attained. Generally, this is a crucial requirement. This was achieved owing to electronic control of the electric current level and by using the correct distance between the electrodes. The geometry proved to be satisfactory in terms of creation of secondary flows which were observable visually. Also our recovery system for the ionic liquid performed well.

Performance of additional experiments at the pilot scale was placed into abeyance pending review of additional relevant laboratory procedures necessary for pilot scale studies. For the pilot scale case therefore, the study focused principally on numerical simulation with validation demonstrated using the photographically captured images of the spraying. Future work is needed to verify all the CFD results obtained for the pilot-scale extractor with experimental measurements.

SYMBOLS

A	total membrane area, m^2
a_d	specific interface area, m^2/m^3
d	droplet diameter, m
K_{tot}	total mass transfer coefficient, $kg/(m^2s)$
k_c	mass transport coefficient in continuous phase, m/s

k_d	mass transport coefficient in dispersed phase, m/s
k_e	liquid-liquid distribution coefficient at the equilibrium, -
m_d	mass of droplet that represents a cloud of droplets of the same diameter, kg
n_d	droplet number density distribution, 1/m ³
r_M	volumetric mass transfer rate, kg/(m ³ s)
r_{Ma}	mass transfer rate, kg/(m ² s)
t	time, s
x_{Et}	mass fraction of ethanol in the raffinate, -
$x_{Et,i}$	mass fraction of ethanol in the raffinate at the interface, -
$x_{Et,e}$	equilibrium value of mass fraction of ethanol in the raffinate corresponding to y_{Et} , -
y_{Et}	mass fraction of ethanol in the extract, -

Greek symbols

$\alpha=k/(c\rho)$	thermal diffusivity, m ² /s
ρ_c	continuous phase density (kg/m ³),
ρ_d	continuous phase density (kg/m ³)

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