HYDRODYNAMICS OF AN EXTERNAL–LOOP AIRLIFT REACTOR WITH INSERTED MEMBRANE

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Abstract – The objective of this study was to investigate the hydrodynamics of an external-loop airlift membrane reactor (ELAMR). The ELAMR was operated in two modes: without (mode A) and with bubbles in the downcomer (mode B), depending on the liquid level in the gas separator. The influence of the gas distributor's geometry and various alcohol solutions on the hydrodynamics of the ELAMR was studied. Results for the gas holdup and the downcomer liquid velocity are commented with respect to an external-loop airlift reactor of the same geometry but without the membrane in the downcomer (ELAR). Due to the presence of the membrane in the downcomer, acting as the local hydrodynamic resistance, the gas holdup in the riser of the ELAMR increases maximally by 16%, while the liquid velocity in the downcomer decreases up to 50%. The values of the gas holdup and liquid velocity predicted by the application of empirical power law correlations and a feed forward back propagation neural network (ANN) are in very good agreement with experimental values.

Keywords: membrane air lift reactor; hydrodynamics; dilute alcohol solution; geometry of distributor; multichannel membrane.

INTRODUCTION

Over the past few decades airlift reactors have been widely used in chemical and biotechnological processes, wastewater treatment and various fermentation systems. The airlift reactors have many advantages, such as the self-generated liquid circulation caused by hydrostatic pressure differences, simple construction, good heat transfer, good mixing, low costs, absence of moving parts, high capacity, relatively low energy consumption and minimal space requirements. Also, they easily enable fluidization of solid particles. There are many variations of these reactors, but the two major groups are: internal loop airlift reactors (ILAR) and external-loop airlift reactors (ELAR).

The progress in waste water treatment, chemical processes and biotechnology industries requires reactors with *in situ* product removal. *In situ* separation processes recover desired product or remove harmful components from the reactor. Therefore, airlift reactors were combined with separation processes such as: liquid−liquid extraction (Gianetto et al., 1988; Yuan et al., 2001) and adsorption or ion exchange (Sun et al., 1999). Nowadays, *in situ* membrane separation process coupled with reactor has received considerable attention (Carstensen et al., 2012). In this integrated process, both the biomass formation and liquid separation with soluble products take place in one membrane bioreactor (MBR). This process has many advantages, such as an excellent and stable effluent

quality, compact equipment, high volumetric load and less surplus sludge production (Liu et al., 2000). One of the newest types of MBRs is the airlift MBR (Futselaar et al., 2007). This reactor has lower energy consumption, compared to conventional cross-flow MBR, because it preferably uses air to control membrane fouling and to recirculate liquid (Futselaar et al., 2007). Namely, the liquid circulation is induced without a circulation pump. Therefore, in the past years different types of airlift MBR configurations have been designed and investigated. Liu et al. (2000) investigated hydrodynamics and its effect on transmembrane pressure in an internal airlift membrane bioreactor. They found that the cross flow velocities increase with an increase of the superficial gas velocity. Jajuee et al. (2006) conducted experiments in a 25 dm³ three-phase concentric airlift reactor with a semipermeable membrane. They observed that the mass transfer coefficient was strongly affected by p-xylene, naphthalene and solid loadings. The effect of hydrodynamic conditions and operating modes on the permeate flux in a submerged airlift hollow fiber membrane system was studied by Bérubé and Lei (2006). Li et al. (2008) developed an innovative single stage continuously aerated internal-loop membrane airlift bioreactor for simultaneous nitrification and denitrification of synthetic domestic wastewaters. Xu and Yu (2008) investigated the hydrodynamics of a novel multi-airlifting membrane bioreactor, constructed with four sintered stainless steel tubular filters serving as the risers and downcomers. In several studies the hydrodynamics in a draft tube airlift reactor with and without membrane have been simulated using computational fluid dynamics software (CFD) (Moraveji et al., 2012; Prieske et al., 2008). They concluded that the presence of membrane caused an increase in the gas holdup and decreased the driving force for liquid movement. Mihaľ et al. (2013) used a hybrid system consisting of an ILAR and membrane module immersed in the downcomer for *in situ* 2-phenylethanol removal from the fermentation medium.

In all mentioned airlift MBRs the membranes were submerged in the reactor. The key problem of this set-up is that the reactor requires small bubbles for high gas holdup and good gas-liquid mass transfer, while the membrane requires large bubbles to control membrane fouling (Cui et al., 2003; Futselaar et al., 2007). To overcome these problems, external-loop airlift membrane reactors (ELAMR) have been developed. In the ELAMR the riser acts as the reactor, while the separation takes place in the downcomer with an inserted membrane. One of the first configurations of this type of reactor was constructed by Norit for commercial application (Futselaar et al., 2007). Fan et al. (2006) successfully used an H-type recycling pipe external-loop airlift membrane bioreactor to treat and reuse municipal wastewater. Shariati et al. (2010) removed acetaminophen as the main pollutant of pharmaceutical wastewater in a rectangular ELAMR. The removal efficiency of acetaminophen was significantly higher for the ELAMR, in comparison to a conventional activated sludge laboratory system.

The hydrodynamics in an ELAMR has significance in the control of membrane fouling and maintenance of the steady operation (Liu et al., 2000). The gas holdup and the downcomer liquid velocity are the most important hydrodynamic parameters (Chisti, 1988). It is well known that gas holdup and downcomer liquid velocity in the ELAR depend on parameters such as superficial gas velocity, ratio of the downcomer to riser cross-sectional area, horizontal connector geometries, hydrodynamic resistance to the liquid flow, liquid height in the gas separator, type of gas distributor and physical properties of the liquid phase (Bello et al., 1984; Bentifraouine et al., 1997; Merchuk and Stein, 1981; Rujiruttanakul and Pavasant, 2011). A membrane module inserted in the downcomer represents hydrodynamic resistance to the liquid flow. Higher resistance in the ELAR leads to a decrease in the liquid velocity and an increase in the gas holdup (Bendjaballah et al., 1999; Cao et al., 2008; Merchuk and Stein, 1981; Pošarac, 1988; Verlaan et al., 1989; Vial et al., 2002).

The liquid height in the gas separator has an effect on the gas holdup and the downcomer liquid velocity (Al-Masry, 1999). The critical level of the liquid in the gas separator prevents the gas bubbles from entering into the downcomer. If the liquid level is below the critical level, the downcomer drags in gas slugs. This situation is preferable in the ELAMR, because of a bigger shear stress in the multiphase flow and minimization of membrane fouling (Bérubé and Lei, 2006; Böhm et al., 2012; Ratkovich et al., 2009).

Physical properties of liquids also influence both the gas holdup and the liquid velocity. Alcohol solutions were used as a model liquid phase of non-coalescing organic mixtures in coal liquefaction and bioreactors (Kelkar et al., 1983). Addition of 2-propanol to an air-water system induces a behavior similar to the fermentation media used in aerobic bioprocesses (McClure et al., 2014). In bubble columns and airlift bioreactors inorganic salts, sugars and metabolic products, such as alcohols and organic acids, were present in significant quantities in the culture medium (Jamialahmadi and Müller-Steinhagen, 1992; Schügerl et al., 1977). A small amount of alcohol (below 1%) remarkably decreased the surface tension of the aqueous solution, thus changing hydrodynamic properties of the airlift reactors. The surface tension of the dilute alcohol solution was the only physical property that differs from water (Freitas and Teixeira, 1998). In ELAR the presence of alcohols caused an increase in the gas holdup (Al-Masry and Dukkan, 1997; Gharib et al., 2013; Miyahara and Nagatani, 2009; Pošarac, 1988; Weiland and Onken, 1981). Also, Weiland and Onken (1981) and Pošarac (1988) reported that the addition of alcohol in an ELAR increased the liquid velocity in the downcomer. On the contrary, Al-

Masry and Dukkan (1997) and Miyahara and Nagatani (2009) found a marginal effect of alcohol solutions on the liquid velocity.

Gas distributor design has a major effect on the initial bubble size and, hence, on the hydrodynamics of the airlift reactor (Bendjaballah et al., 1999; Cao et al., 2008; Lin et al., 2004; Snape et al., 1995; Vial et al., 2000). Cao et al. (2008) performed the most comprehensive study about the influence of the gas distributor on the gas holdup and liquid velocity in the ELAR. They observed that the gas distributor had a noticeable effect on the gas holdup up to the superficial gas velocity of 0.25 m/s. At higher gas inputs, the distributor's influence was negligible. However, the effect of the gas distributor on the liquid velocity was evident in the range of gas velocities from 0.025 m/s to 0.045 m/s.

In this paper, the hydrodynamics of an ELAMR with a short multichannel ceramic membrane inserted at the bottom of the downcomer was investigated. Such a configuration enables high hydrostatic head pressure, which decreases the power necessary for permeate removal. Furthermore, by changing the liquid level in the gas separator, the set of experiments were done with gas slug entrainment in the downcomer. This operation mode is useful for preventing membrane fouling. Also, the influences of the gas distributor's geometry and the addition of aliphatic alcohols on the riser gas holdup and downcomer liquid velocity of the ELAMR were studied. Also, this reactor has been designed for possible use as a high efficiency equipment for removal of organic and inorganic pollutants from wastewater.

MATERIALS AND METHODS

Experimental setup

A schematic illustration of the setup is shown in Figure 1. The cylindrical external-loop airlift reactor made of Plexiglas consisted of a riser (54 mm i.d. and 2000 mm in height), downcomer (25 mm i.d. and 1950 mm in height) and rectangular gas separator (400×310×300 mm). The distance between the riser and the downcomer was 100 mm. When the ELAMR was operated without gas bubbles in the downcomer (mode A), the unaerated liquid level in the gas separator was 4 cm. Lowering the liquid level to 3 cm, gas bubbles were dragged into the downcomer (mode B). The air, sparged into the riser, was used as the gas phase. Three different gas distributors were tested: single orifice (4 mm i.d.), perforated plate (7 holes of 1 mm i.d., triangular pitch) and sinter plate (100-160 μm, average pore size 115 μm, porosity 8%). Porosity and average pore size of the sinter plate were obtained with a porosimeter (Porosimeter 2000 with Macropore Unit 120). The gas flow rates were controlled and measured by an electronic mass flow controller (model Bronkhorst High Tech F 201AV). The

superficial gas velocity, based on the riser cross-sectional area, was varied in the range 0.02 to 0.22 m/s for mode A. In mode B, the entrainment of gas bubbles in the downcomer started at $U_G = 0.15$ m/s, so the investigated range of U_G was 0.15 to 0.22 m/s. Two eDAQ (Australia) conductivity isoPods with miniature dip-in conductivity electrodes were used to determine downcomer liquid velocity. The tubular ceramic membrane (ZrO_2/TiO_2, Novasep, France) 20 cm in length and 2.3 cm in diameter, with 7 channels (6 mm i.d.), was installed in the downcomer. The filtration was disabled in the membrane module since our aim was to investigate only the influence of the membrane acting as additional resistance on the hydrodynamics in the reactor. Also, it was considered that the permeate flux would not affect the hydrodynamics (Böhm et al., 2012).

Gas-liquid systems

Tap water and dilute alcohol solutions (0.046 wt% ethanol, 0.011 wt% n-butanol and 0.0051 wt% n-hexanol) were used as the liquid phase. Added amounts of each alcohol correspond to their critical concentration reported by Keitel (1978). Increasing the alcohol concentration above the upper limiting concentration value, only enhances the liquid phase frothing and bubble coalescence (Camarasa et al., 1999; Freitas and Teixeira, 1998). Surface tensions of liquid phases and the surface tension gradient ($d\sigma/dC_A$) data were taken from Šijački et al. (2011).

Measurement of hydrodynamic characteristics

Gas holdup

The gas holdup values in the riser (ε_{GR}) and the downcomer (ε_{GD}) were obtained by measuring the pressures at the bottom and the top of the riser and downcomer using piezometric tubes, and calculated from the equation:

$$\varepsilon_G = \frac{\Delta z}{\Delta H} \qquad (1)$$

In order to reduce the liquid surface fluctuations in the piezometric tubes, capillaries (50 mm in length and 0.7 mm i.d.) were inserted at the entrance of the piezometric tubes. Therefore, the relative average error of these measurements was reduced to max ±2%. This experimental method was adopted from Zahradník et al. (1974). Also, it was visually observed that the gas bubbles did not enter into the piezometric tube due to frothing in the riser and the downcomer.

Liquid velocity in the downcomer

The liquid velocity in the downcomer (W_{LD}) was determined by the tracer response method. Two conductivity probes were placed in the downcomer section at a distance (L) of 1.4 m from each other. A volume of 25 cm³ of 4 M
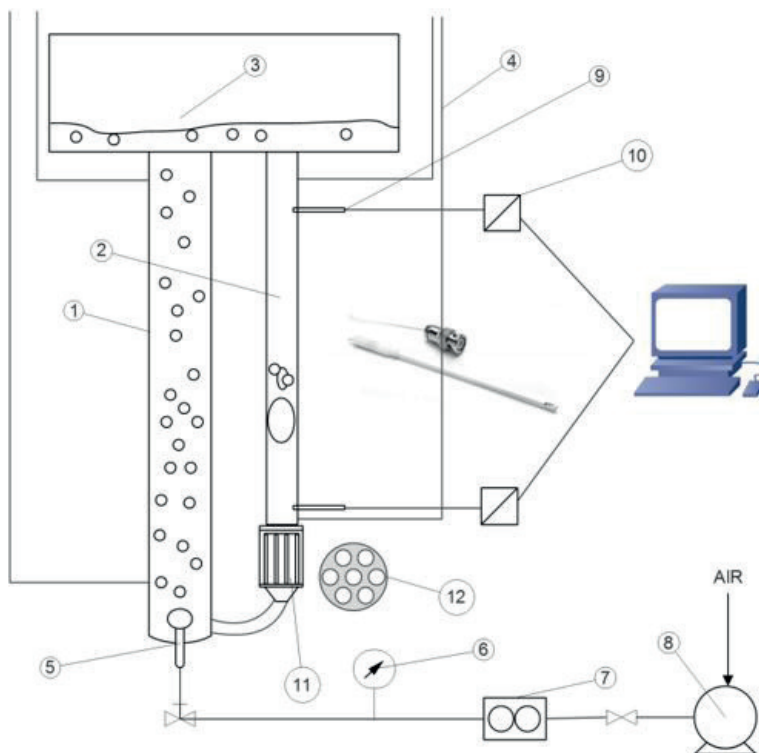
Figure 1. Experimental setup: 1-riser, 2-downcomer, 3-gas separator, 4-piezometric tubes, 5-gas distributor, 6-manometer, 7-mass flow controller, 8-air compressor, 9-conductivity electrode, 10-conductivity isopod, 11-membrane module, 12-membrane.

NaCl, used as a tracer, was instantaneously injected 15 cm above the top electrode. Liquid velocity in the downcomer was calculated from the measured time interval between the tracer peaks from the two conductivity probes and the known vertical distance between them, by the following equation:

$$W_{LD} = \frac{L}{t_2 - t_1} \qquad (2)$$

The cross flow velocity inside the membrane could be calculated using the continuity equations by knowing W_{LD}. Signals were recorded at a frequency of 0.1 s. For each value of a gas flow rate two measurements were performed and the average value of W_{LD} was calculated. The relative average error of this method was ±3%.

Friction coefficient

To quantify the hydrodynamic resistance of the membrane itself, it was necessary to calculate the overall friction coefficient (K_f) in both reactors. The K_f was derived according to Verlaan (1987) by plotting the square of the measured superficial liquid velocity as a function of the difference between gas holdups in the riser and the downcomer:

$$W_{LD}{}^2 = \frac{2gH}{K_f}(\varepsilon_{GR} - \varepsilon_{GD}) \qquad (3)$$

The commercial software pipe flow expert was used to calculate $K_{f(calc)}$. $K_{f(calc)}$ was estimated according to Garcia-Calvo (1992) and Milivojević (2011) as the sum of specific fittings in each reactor separately. In the estimation of $K_{f(calc)}$ the fitting that represented the resistance of the membrane was taken as a partially open valve for the ELAMR and as a fully open valve for the ELAR. The assumption was made that the type of distributor and alcohol do not influence $K_{f(calc)}$. A good agreement between calculated and experimental K_f values, with maximal relative error of 15%, was achieved.

RESULTS AND DISCUSSION

Hydrodynamic regimes

It is well known that the hydrodynamics of a bubble column (riser of the ELAR) in the gas-liquid co-current operation mode is characterized by different flow patterns depending on the gas flow rate: homogenous (bubble flow),

transition, heterogeneous (churn-turbulent flow) regimes and slug flow(Govier, 1972; Hatch, 1975; Wallis, 1969).

In both reactors, ELAR and ELAMR, for operating mode A the existence of all regimes was confirmed (Figures 2ac and 3a-c). The transition between regimes was identified by the change of the slope of the gas holdup vs. the gas velocity curves. A more detailed analysis of hydrodynamic regimes and transition superficial gas velocities was described in our earlier study (Kojić et al., 2015). The transition points in the ELAMR appeared at a slightly lower U_G compared to the ELAR. This is because the larger hydrodynamic resistance in the ELAMR caused lower W_{LD} that induced more intensive bubble coalescence. Joshi et al. (1990) and Bendjaballah et al. (1999) also noted that the resistance in the ELAR, and thereby W_{LD}, and the geometry of the downcomer influenced the regime transitions. On the other hand, Vial et al. (2002) reported that the hydrodynamic resistance had a minor influence on the regime transitions. As can be seen in Figures 2 and 3, added alcohol had a noticeable influence on the transition between regimes in both ELAR and ELAMR for the sinter plate and perforated plate. But, for a single orifice this transition was the same for all alcohol solutions. The presence of alcohols delayed the heterogeneous regime, due to their coalescence inhibiting nature (Pošarac, 1988; Weiland and Onken, 1981). The effect of the gas distributor type on the regime transition in the ELAR and ELAMR was also observed. When a single orifice was used, the absence of bubble flow was noted even at the lowest U_G. Both reactors operated instantly in the transition regime, that is in agreement with the observations of Bendjaballah et al. (1999) and Vial et al. (2001). In mode B, at $U_G \geq 0.15$ m/s, gas slugs started to be dragged into the downcomer. The length of the slugs was ~5 cm in water and ~ 3 cm in alcohol solutions. When the alcohols were added, smaller number of gas slugs were created in the downcomer, in comparison to water. Mode B is significant for the mitigation of membrane fouling. Large bubbles or slugs are more beneficial as they have larger wake regions, create stronger secondary flows and are more effective in promoting local mixing than smaller bubbles. Also, for a fixed bubble volume, an increase in bubbling frequency means more falling films and bubble wakes per unit time (Cui et al., 2003). According to Ratkovich et al. (2009) gas slugs increase the shear stress across the membrane and prevent membrane fouling.

Gas Holdup
Influence of the superficial gas velocity on the gas holdup

The influence of the superficial gas velocity on the riser gas holdup is presented in Figures 2 and 3 for the ELAR and ELAMR, respectively. For all the gas-liquid systems and both reactors (ELAR and ELAMR), the gas holdup increased with an increase in the superficial gas velocity,

and it was higher in the ELAMR than in the ELAR. The increase in ε_{GR} was the highest for the single orifice (about 16%), while for the perforated plate and the sinter plate it was lower (about 8%). When the ELAMR was operated in mode B, ε_{GD} was up to 0.03, while ε_{GR} was unchanged. ε_{GD} values were about 10 times lower, compared to ε_{GR} (Figure 3).

To quantify the influence of the membrane as the resistance to the flow in the ELAMR, the overall friction coefficient was calculated from equation 3 by fitting the experimental data. The insertion of the membrane in the downcomer of our ELAR increased the overall friction coefficient by 90% while ε_{GR} was slightly increased. Table 1 illustrates that increasing the resistance in the downcomer of the ELAR results in an increase in ε_{GR}. This effect is the most pronounced when the cross section of the downcomer is reduced by more than 50%. Furthermore, this contributed to an increase of K_f by more than 100%. Reduced liquid velocity decreased the rise velocity of the bubbles and consequently increased ε_{GR}.

Influence of added alcohol on the gas holdup

The effect of alcohols on the gas holdup in both reactors, without and with membrane, was the same (Figures 2 and 3). The addition of alcohols led to an increase in ε_{GR}, in comparison to water for all distributors, but more intensively for the sinter plate (SP) and perforated plate (PP). At $U_G \leq 0.05$ m/s only slight differences in ε_{GR} were observed, regardless of the geometry of the distributor and type of alcohol. In the bubble flow no coalescence was observed, even in the water. However, in the transition and churn-turbulent flow ε_{GR} was about 22, 17.5, and 10% higher for ethanol, n-butanol and n-hexanol solutions, respectively for SP and PP, in comparison to water. At high gas velocities, corresponding to the transition and heterogeneous flow, coalescence was much stronger with frequent bubble collisions. The inertial forces dominated over the surface forces and the effect of coalescence inhibition due to added alcohol was more pronounced. The effect of added alcohol on ε_{GR} for the single orifice (SO) (Figures 2 and 3) was lower than for the PP and the SP, so in the transition and the churn-turbulent regime alcohol solutions had only about a 7% higher ε_{GR} compared to water. The SO produced large bubbles and the gas-liquid system was instantly in the transition regime; therefore, it was difficult for alcohols to form monolayer around the bubbles and prevent coalescence. When the ELAMR was operated in mode B, added alcohols slightly decreased ε_{GD}, compared to water.

Influence of distributor type on the gas holdup

The effect of gas distributor: SO, PP and SP is shown in Figures 2 and 3. It can be clearly seen, that the SP for

Table 1. List of the influence of hydrodynamic resistance on the riser gas holdup and downcomer liquid velocity in the external-loop airlift reactors.

Ref., resistance and sparger type	Valve or orifice plate opening	Friction coefficient (K_f)	The increase of ε_{GR} relative to a fully open downcomer (%)	The decrease of W_{LD} relative to a fully open downcomer (%)
Verlaan (1987) (valve, single orifice)	lack of data	4.6	-	-
		8.9	7	24
		19.3	22	46
		61.5	42	68
		409	55	88
Pošarac (1988) (orifice plate)	100	15	-	-
	50	19	17	9
	30	35	38	26
	5	793	117	77
Bendjaballah et al. (1999) (valve, multiple orifice)	100	6.9	-	-
	77	7.2	2.2	5.9
	46	8.2	5.5	10.4
	23	15.3	23	30
	7	43.1	51	55
Vial et al. (2002) (valve, multiple orifice)	100	23	-	-
	70	25	2	12
	30	45	23	43
this paper (single orifice, perforated plate and sinter plate)	ELAR ELAMR	10.3-12.6 21.8-26.1 21.2-30.2(with slugs)	- 16	- 26

all used gas-liquid systems produced the highest ε_{GR}. It is obvious that the SP and PP are more efficient distributors than the SO. The membrane module reduced the effect of the SP and PP on ε_{GR} in comparison to SO (by 25% to 50% depending on the gas-liquid systems). The efficiency of both the SP and PP started to decrease at lower U_G in the ELAMR, because of the fact that the churn-turbulent flow appeared earlier than in the ELAR. The SP was more efficient than the PP ($U_G \leq 0.09$ m/s) until the inertial forces started to be dominant. Therefore, at higher U_G differences in ε_{GR} were less than 3%. Alcohols improved the efficiency of the SP and PP compared to SO (which remained the least efficient distributor), especially in churn-turbulent flow. For instance, in the region of surface forces domination ε_{GR} was higher up to 40%. However, at $U_G > 0.09$ m/s, when coalescence was intensive, ε_{GR} produced by the SP and PP was only 4% higher for water and about 20% for alcohol solutions. When the ELAMR was operated in mode B, the

SO produced slightly smaller ε_{GD} compared to the SP and PP.

Downcomer liquid velocity
Influence of superficial gas velocity on the downcomer liquid velocity

Figures 4 and 5 present the effect of U_G on the liquid velocity in the downcomer for both operating modes and all distributors in the ELAR and the ELAMR, respectively. For all the gas-liquid systems W_{LD} increased with increasing U_G. Insertion of a membrane module in the downcomer of the ELAR increased frictional energy losses and led to a decrease of W_{LD} in the range of 36-49% for both SP and PP, while for SO the decrease was in the range of 19-27%, depending on the gas-liquid system. In order to reduce membrane fouling, it is very important to know the liquid velocity through the channel of the membrane (cross-flow
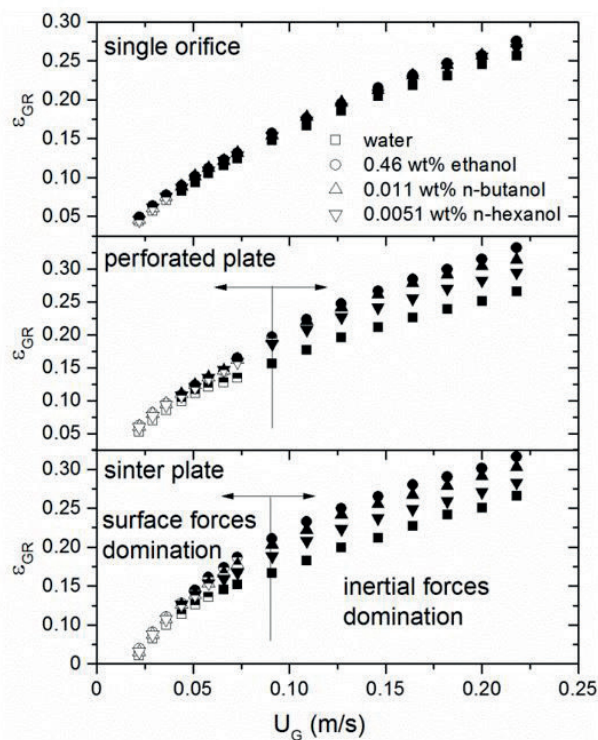
Figure 2. Influence of gas distributor type and added alcohol on the gas holdup in the ELAR. Hydrodynamic regime legend: open symbols, homogeneous regime; half solid symbols, transition regime; solid symbols, heterogeneous regime and slug flow.

Figure 3. Influence of gas distributor type, operation mode and added alcohol on the gas holdup in the ELAMR. Hydrodynamic regime legend: open symbols, homogeneous regime; half solid symbols, transition regime; solid symbols, heterogeneous regime and slug flow.

velocity). A continuity equation was used for its estimation. This velocity was about 2.5 times higher than W_{LD}. Our ELAMR at higher U_G can achieve the cross flow velocity for microfiltration and ultrafiltration (2-3 m/s), which was proposed by Rossignol et al. (1999) and Choi et al. (2005). When the ELAMR was operated in mode B, the slugs in the downcomer reduced W_{LD} by 5-15%, depending on the gas-liquid system and type of gas distributor. The velocities of the gas slugs in the downcomer were 35-46 cm/s, in the range of applied U_G.

Influence of added alcohol on the downcomer liquid velocity

The results in Figures 4 and 5 showed that the downcomer liquid velocity in the majority of the investigated gas-liquid systems and gas distributors changed with added alcohol. The reason for this is a higher driving force for the liquid circulation. Namely, the driving force is proportional to the hydrostatic pressure difference between the two vertical columns in the ELAR. Added alcohol increased the riser gas holdup, while the downcomer gas holdup remained unchanged because there
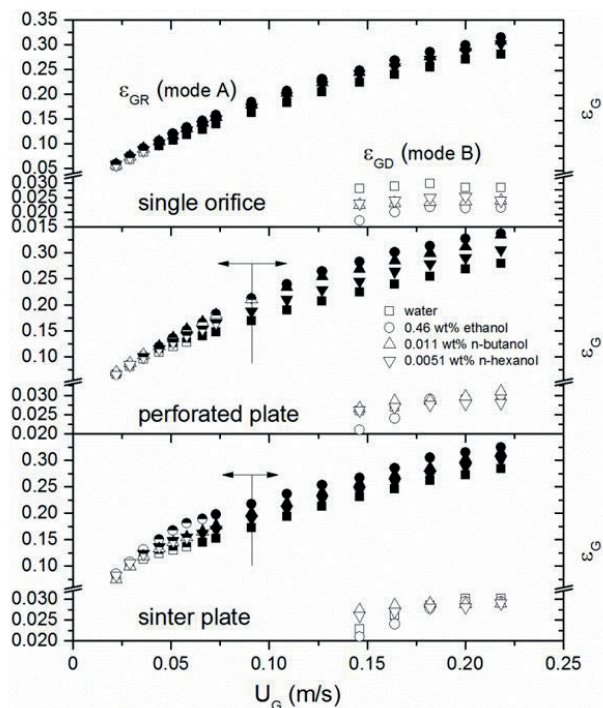
was no bubble entrainment in the downcomer (mode A). Therefore, W_{LD} was highest in the ethanol and n-butanol solutions for all distributors. The membrane, inserted in the downcomer of the ELAR, diminished the effect of alcohol on W_{LD}. For instance, added alcohols increased W_{LD} from 10% to 15.5% in comparison to water in the ELAR with SP, while in the ELAMR the increase was only 3.7% to 10%, depending on the alcohol solution. In both operating modes, the effect of added alcohols on W_{LD} was the same.

Influence of gas distributor on the downcomer liquid velocity

Figures 4 and 5 show the effect of U_G and gas distributor type on W_{LD}. In the ELAR, SP and PP were equally efficient. They gave higher W_{LD} (12-20% depending on the gas-liquid system) than SO. In the ELAMR the effect of gas distributor type was reduced because of more intensive coalescence that appeared at lower U_G compared to the ELAR. In the region of surface forces domination (bubble and transition flow) in the ELAMR the highest W_{LD} were achieved using SP, while the lowest ones were with SO. However, in churn-turbulent flow ($U_G > 0.09$ m/s), when
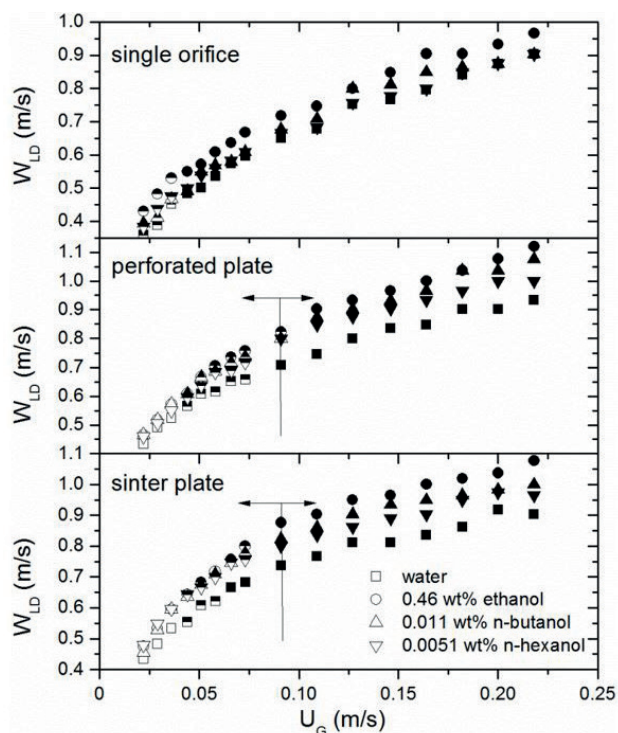
Figure 4. Influence of gas distributor type and added alcohol on the downcomer liquid velocity in the ELAR. Hydrodynamic regime legend: open symbols, homogeneous regime; half solid symbols, transition regime; solid symbols, heterogeneous regime and slug flow.
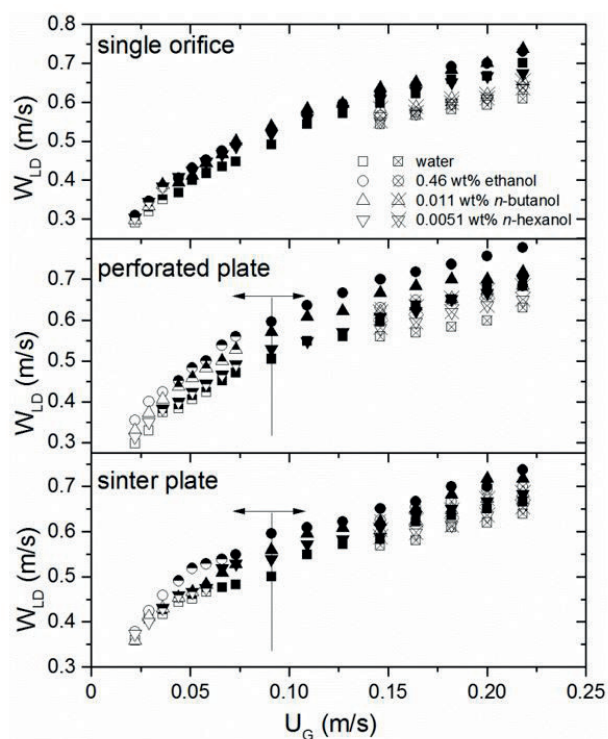

Figure 5. Influence of gas distributor type and added alcohol on the downcomer liquid velocity in the ELAMR. Hydrodynamic regime legend: open symbols, homogeneous regime; half solid symbols, transition regime; solid symbols, heterogeneous regime and slug flow; crossed open symbols, operation mode B.

inertial forces dominated, W_{LD} tended to be equal for all three distributors, as the differences in the W_{LD} between distributors were less than 3%. In both operating modes, the same influence of the gas distributor on W_{LD} was noticed.

CORRELATIONS

Empirical power law correlations

According to experimental results in both operating modes examined the general comment is that ε_{GR} and W_{LD} in the ELAR and ELAMR depend on the superficial gas velocity, distributor type, surface properties of the gas-liquid system and overall friction coefficient. Distributor type has a strong influence on the primary bubble dispersion and therefore on the global hydrodynamics. The main parameter that describes the gas distributor, i.e., initial bubble size, is the orifice diameter (d_o) (Šijački et al., 2011). The influence of alcohols on ε_{GR} and subsequently on W_{LD}, should be linked to changes in the surface tension gradient, i.e., to the coalescence suppression strength of

individual alcohols (Albijanić et al., 2007; Camarasa et al., 1999). The effect of hydrodynamic resistance on the liquid velocity in the downcomer depends on the friction coefficient (K_f) in the ELAR and ELAMR, in both operating modes. Therefore, the general form of the correlations we applied was:

$$(y)_{calc} = AU_G{}^B \left(1 + \left(-\frac{d\sigma}{dC_A}\right)\right)^C d_o{}^D K_{f(calc)}{}^E$$

$$(4)$$

The correlations were fitted for the following ranges of independent variables: $0.022 < U_G < 0.218$ m/s, $0.027 < (d\sigma/dC_A) < 1.985$ mNm²/mol, $0.115 < d_o < 4$ mm, $9 < K_{f(calc)} < 30$. The coefficients were determined by minimizing the least-square sums over nonlinear correlations. These calculations were performed in Mathcad software. Table 2 contains the values of estimated parameters in the proposed correlations (eq. 4) and the goodness of fit. The parity plot of calculated values versus experimental ones is presented in Figure 6.

Table 2. Values of correlations parameters for the riser gas holdup and downcomer liquid velocity and fit statistics

y_{cal} (eq. 4)	A	B	C	D	E
ε_{GR}	0.64	0.61	-0.01	-0.02	0.06
W_{LD}	3.85	0.34	-0.01	-0.03	-0.38

Fit statistics	Correlation		Artificial neural network	
	ε_{GR}	W_{LD}	ε_{GR}	W_{LD}
mean relative errors (%)	8.8	5.8	5	3.5
coefficient of determination	0.95	0.93	0.97	0.97
mean absolute errors	0.37	0.96	0.27	0.64
root mean square errors	0.018	0.045	0.013	0.03
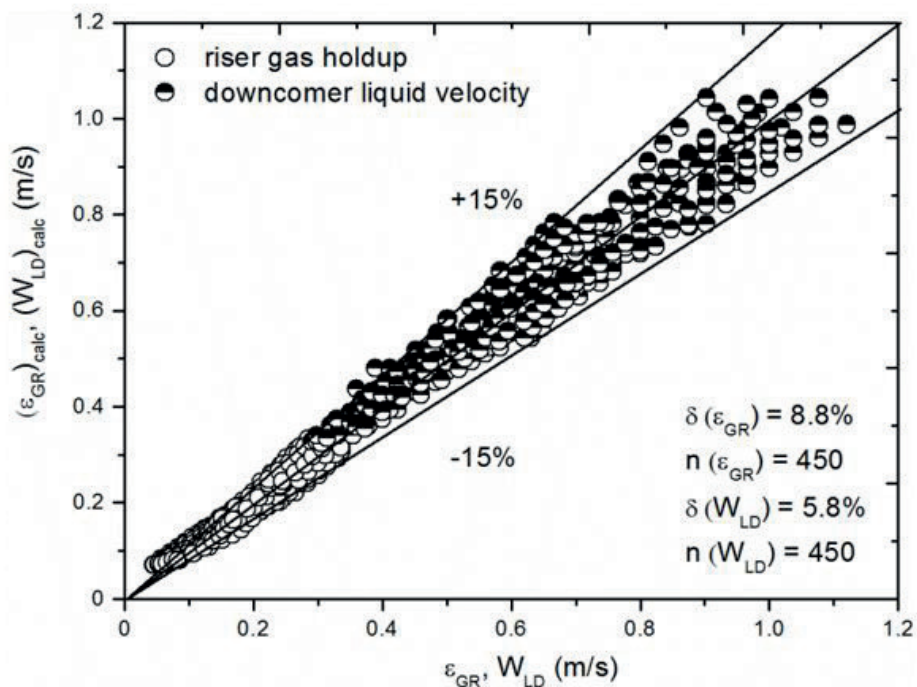
Figure 6. Comparison of experimental and predicted values for the riser gas holdup and downcomer liquid velocity using the proposed correlations (4).

Neural network simulations

In this study the feed-forward artificial neural network model (ANN) was used, beside the previously proposed empirical power law correlations, to predict ε_{GR} and W_{LD}. Al-Masry (2006), and Chen et al. (2013) pointed out that an ANN reasonably predicted experimental values for liquid velocities, gas holdup and volumetric gas-liquid mass transfer coefficient in the ELAR. Neural network inputs were U_G, $-d\sigma/dC_A$, d_o and $K_{f(calc)}$, the ranges of network inputs were the same as in the correlations (eq. 4) mentioned above. The number of neurons was 20 and 2 in the hidden and output layer, respectively. The network was trained with the Levenberg-Marquard back propagations algorithm. The transfer function was the logsigmoid. All 450 data points were used to train and develop the ANN: 70% of the data points for training, 15% of the data for validations and 15% of the data for the testing of the process. Figure 7 shows the parity plot of the experimental and predicted values for ε_{GR} and W_{LD} using the ANN on the whole database. The statistical analysis of prediction with the ANN is shown in Table 2. Based on Table 2, the ANN predicts the experimental results more accurately than power law correlations.
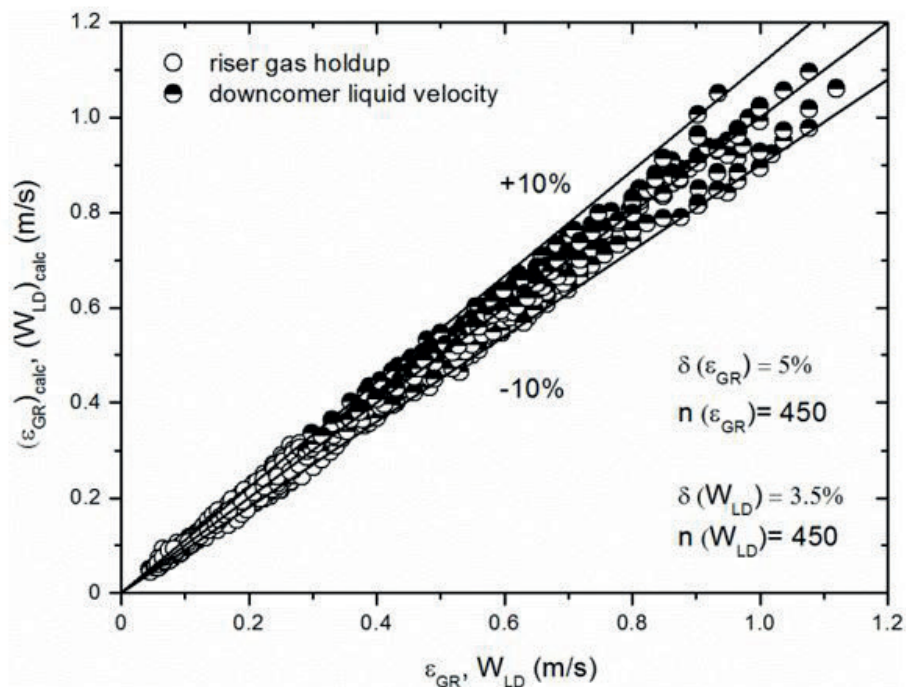
Figure 7. Comparison of experimental and predicted values for the riser gas holdup and downcomer liquid velocity using an artificial neural network.

CONCLUSION

The following conclusions could be drawn from the present study for both external-loop airlift reactors, with and without inserted membrane in the downcomer:

The insertion of the membrane in the downcomer caused a rise of the overall friction coefficient by 90% and a decrease of the downcomer liquid velocity up to 50%. However, the cross-flow velocity remained in the recommended range of velocities for ultrafiltration and microfiltration given in the literature. Decreased downcomer liquid velocity resulted in up to a 16% increase of the gas holdup, while the regime transitions shifted toward the lower superficial gas velocities. The presence of bubbles in the downcomer decreased the downcomer liquid velocity up to 15% and resulted in a downcomer gas holdup up to 0.03. An artificial neural network gave a good prediction of riser gas holdup and downcomer liquid velocity for both reactor configurations studied. Future research should be oriented toward enabling filtration within an external-loop airlift reactor with membrane in the downcomer.

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NOMENCLATURE

d_o	diameter of orifice (mm)
g	gravitational acceleration rate (m/s^2)
H	column height (m)
K_f	friction coefficient
L	distance between conductivity electrodes (m)
t	time (s)
U_G	superficial gas velocity (m/s)
W_{LD}	downcomer liquid velocity (m/s)

Greek letters

ΔH	distance between two measuring points (mm)
Δz	pressure drop between two measuring points (mm)
δ	mean relative error (%)
ε_{GR}	riser gas holdup
ε_{GD}	downcomer gas holdup
$-d\sigma/dC_A$	surface tension gradient (N m^2/mol)

Abbreviations

ANN	artificial neural network
ELAR	external-loop airlift reactor
ELAMR	external-loop airlift membrane reactor
MBR	membrane bioreactor

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