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SEPARATION EFFICIENCY OF HOLPACK PACKING FOR ETHANOL – WATER RECTIFICATION IN DIFFERENT CONCENTRATION RANGES

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Abstract

In this study the mass transfer efficiency of HOLPACK packing is investigated in ethanol-water rectification in terms of high ethanol concentrations. The results are compared with previous study of this packing for low ethanol concentrations. The comparison shows variation of the efficiency depending on the investigated composition area. The reason is sought in highly variable physico-chemical properties of the system, its vapor-liquid equilibrium character and the different influence of some effects on mass transfer. The most part of the rectification column operates in the range of the highest concentrations, due to the specific type of this system equilibrium. Therefore it is important to study the process in these conditions.

Key words: *Distillation, rectification, packed columns, HOLPACK packing, separation efficiency*.

1. INTRODUCTION

Rectification is one of the most commonly used methods for separating mixtures in the chemical, petrochemical, food, pharmaceutical and other industries. A major problem in studying the process is determining the height of the rectification column. These difficulties come from the complexity of many phenomena accompanying the process.

In packed columns hydrodynamic pattern is too complex and diverse. It is characterized by axial mixing, radial maldistribution, bypass flows, stagnant areas and others. Because of insufficient theoretical knowledge, the impact of these phenomena is ignored or is attempted to be treated with various hydrodynamic models.

Despite the great diversity of methods and equations for calculating packed columns, it can be argued that the kinetic dependences for rectification process are not completely explored. As an example may be given a comparison of results (Stichlmair and Fair, 1998) from the calculation of rectification column filled with Pall rings packing for separation of ethylbenzene – styrene by different models. Quite large differences in the values of column height, calculated in each model have been found. The reason is the different conditions under which the dependencies are derived. Of great significance is also the model system, with which the experiments were performed.

Therefore we can say that despite the intensive research activity, rectification columns cannot be designed developed and operates absolutely reliable only through calculations, without experiment.

The structured metal packing HOLPACK is largely used in ethanol production. There is a model for design of columns with HOLPACK packing, based on results from experiments, carried out at low and medium ethanol concentrations (Kolev and Semkov, 1991). It is important to study the packing at high concentrations, because the prevailing part of the column operates in such a range.

Ethyl alcohol is widely used in chemical and food industry. Its industrial importance has recently grown related to the production of bio-ethanol for fuel. It is therefore important to investigate the efficiency of modern packings applicable to its production. Additional difficulties in design of rectification columns create highly variable physico-chemical properties of ethanol-water system and the nature of its equilibrium.

The aim of the present work is to study the efficiency of the structured HOLPACK packing in rectification under conditions similar to industrial. For this purpose, experiments were carried out at high and medium concentrations, since because of the characteristics of the phase equilibrium major part of the column operates in this concentration range. The results are compared with old data from experiments carried out at low concentrations.

2. EXPERIMENTAL INSTALLATION

HOLPACK packing consists of expanded metal sheets, located horizontally in the column at a distance from one another (fig. 1). To improve the uniform gas and liquid distribution at the column cross-section is only necessary to alternate the direction of the slits from sheet to sheet, as each sheet is rotated in the same direction as the previous to 90°.

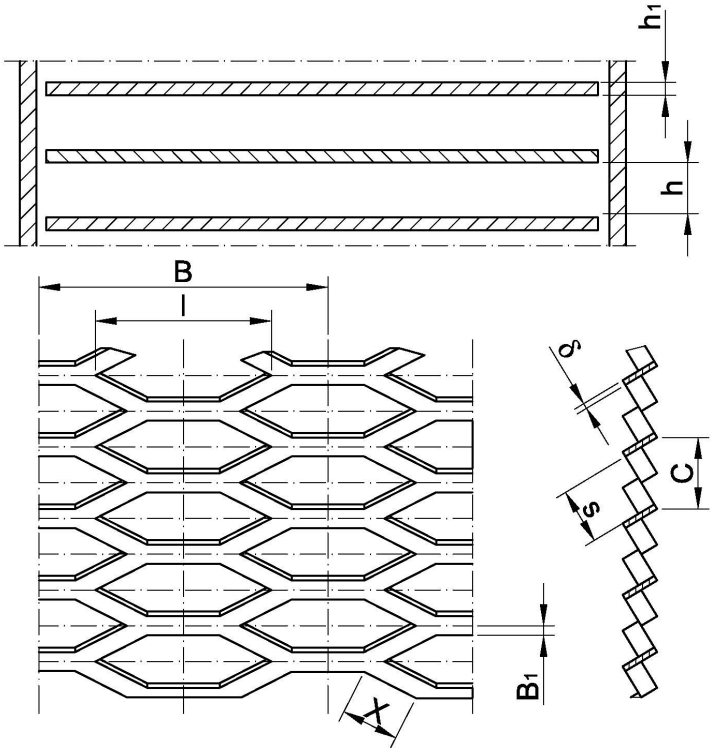


Fig.1. Scheme of HOLPACK packing

Two types of HOLPACK packing are investigated, which geometrical characteristics are given in Table 1. For comparison are used the results for packings with similar sizes (Semkov, 1989).

Table 1. Geometric characteristics of HOLPACK packing

Packing	Hydraulic diameter of the openings - d_e, mm	Distance between the sheets - h, mm	Sheet thickness – h_1, mm	Specific area – a, mm	Free volume – □
HOLPACK 1	6.8	20	3.5	56.15	0.92
HOLPACK 2	25.6	20	13.2	41.7	0.96

The experimental installation consists of a column, condenser, connecting pipelines, devices for monitoring and measurement and control panel. At the lower part of the column body is located reboiler with volume of about 80 l, into which are incorporated electric resistance heaters with total power of 45 kW. The column is made of stainless steel and has inner diameter of 0.213 m. It is built of separate sections connected by flanges. The packing layer is situated on support grid and has a height of 2.8 m for packing No. 1 and 1.54 m for packing No. 2 respectively. The scheme of the experimental installation and its detailed description are given in (Darakchiev and Semkov, 2008).

3. EXPERIMENTAL RESULTS

The results for packing efficiency are presented as height of transfer units HTU.

According to the method of transfer units, the column height *H* is determined by the relation:

$$H = HTU.NTU \tag{1}$$

where *HTU* is the height of transfer unit and *NTU* is the number of transfer units

$$HTU = \frac{H}{NTU} = \frac{w}{K_G a} \tag{2}$$

$$NTU = \int_{y_W}^{y_D} \frac{dy}{y^* - y}. \tag{3}$$

The overall mass transfer coefficient is determined by integrating the mass transfer equation for the plug flow model

$$w.dy = K_G a(y^* - y)dh \tag{4}$$

$$K_G a = \frac{w}{H} \int_{y_W}^{y_D} \frac{dy}{y^* - y} \quad . \tag{5}$$

On figure 2. are shown the results for packing HOLPACK 1 at two concentration ranges – low concentrations (average concentration for the studied range x_{av} = 0.0824 mol/mol) and high (average concentration x_{av} = 0.7343 mol/mol). We see that the packing shows higher efficiency at high concentrations, as this difference reaches 33 % at high vapor velocities.

For the larger packing size, the difference is even greater (figure 3). Here the results are for three different concentration ranges. The packing shows the highest efficiency at medium concentrations.

The obtained results are a visual illustration of the complexity of the rectification process. The combined influence of various effects that are superimposed on the mass transfer process, which are

strongly dependent on the physicochemical properties of the mixture, led to a strong change in the packing efficiency. Moreover, ethanol – water mixture is characterized by the fact that it change greatly as its properties and its phase equilibrium depending on concentration.

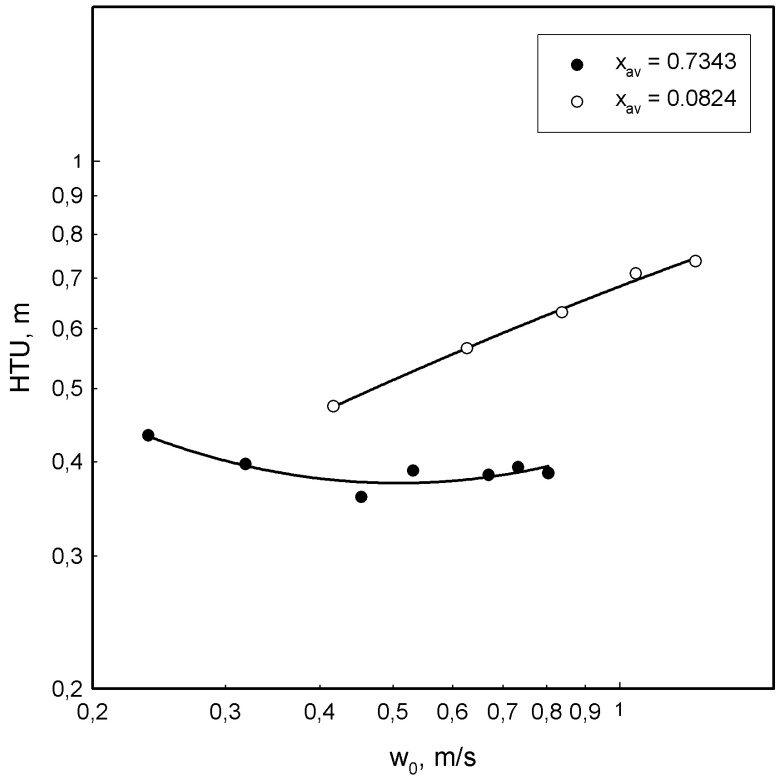


Fig.2. HTU vs. vapor velocity for HOLPACK 1 at low and high concentrations

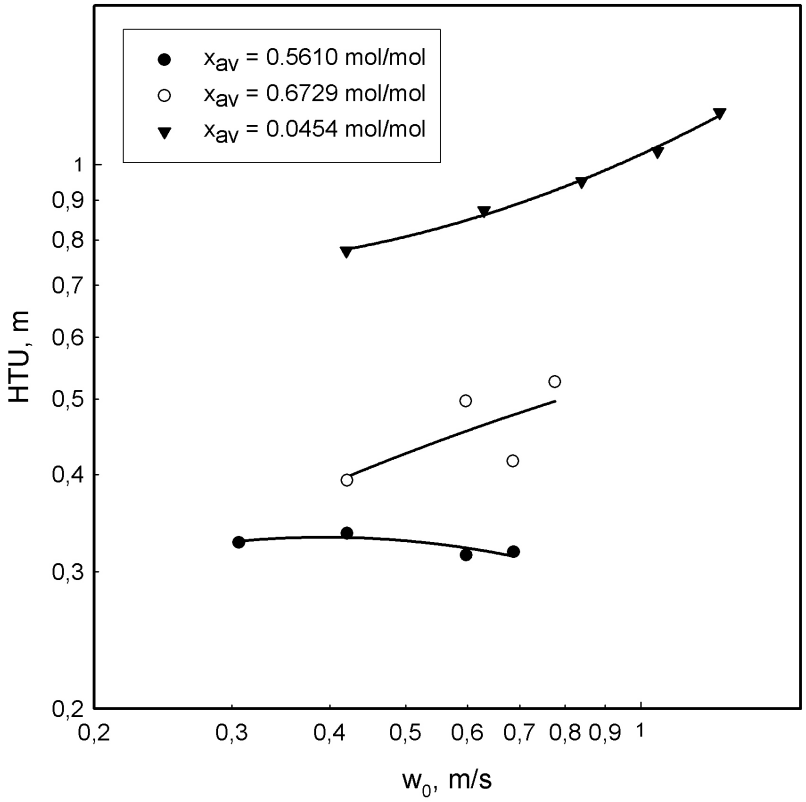


Fig. 3. HTU vs. vapor velocity for HOLPACK 2 at low, medium and high concentrations

To analyze the situation will look in more detail at the variation of the properties and the system equilibrium at concentration change. Physicochemical properties of pure ethanol and water components strongly differ, which leads to their great difference at different mole fractions. The physicochemical properties for each series of carried out experiments are given in Table 2. There can be seen that the difference between the vapor density of the mixture at low and high concentrations is about 49%, and the difference in the density of the liquid is 18%. Respectively the viscosity of the liquid mixture at low concentrations differs by 33 % than in high. Diffusivity coefficient of the mixture is almost constant at various concentrations, while the greatest influence has the surface tension – it differs by 63 %.

Table 2. Physical properties of ethanol – water system for each experimental series

Packing	x_{av}	ρ_V	ρ_L	$\mu_V.10^5$	$\mu_L.10^4$	$D_V.10^3$	$D_L.10^9$	σ
HOLPACK 1	0,0826	0,69	925,38	1.13	4,18	2,42	2,03	0,0498
HOLPACK 1	0,7343	1,34	754,97	1.04	5,22	2,31	1,94	0,0195
HOLPACK 2	0,0454	0,65	936,67	1.15	3,77	2,45	2,06	0,0541
HOLPACK 2	0,5610	1,17	790,25	1.05	5,63	2,32	1,95	0,0215
HOLPACK 2	0,6729	1,28	764,42	1.04	5,35	2,32	1,94	0,0199

Figure 4 shows the variation of the surface tension in the entire concentration range. We see the unsteady character of this variation – there is strong change from low to medium concentrations and permanent character from medium to high concentrations.

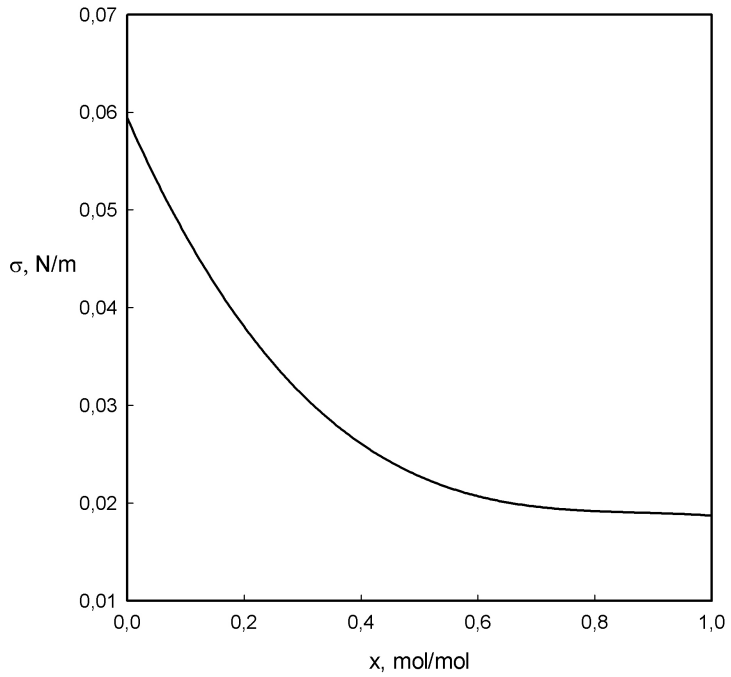


Fig.4. Surface tension vs. ethanol concentration in liquid phase

With the surface tension is connected a common effect in rectification process – the so called Marangoni effect. It is known that in most cases the concentration differences lead to surface tension gradients at the interface. In some cases, these gradients cause interfacial turbulence, which affects the mass transfer. As a result spontaneous circulation fluxes may be induced in the liquid phase, near the interface which, in fact, enforces liquid exchange in volumes of various concentrations between the interface and the phase bulk. This mechanism and the experimental study show, that interfacial turbulence causes mass transfer enhancement only in the liquid boundary layer.

To evaluate this effect, mass transfer enhancement can be correlated with the difference between the interfacial surface tension and the surface tension of the bulk phase ($\Delta\sigma = \sigma_i - \sigma$). There are various ways to determine the influence of Marangoni effect on mass transfer. A method based on a large amount of experimental data is used in this study, which is applicable to various apparatuses, systems and conditions of process release (Kolev and Semkov, 1991). Figure 5 shows this effect for each series of the experiments. It can be seen that at low concentrations the effect is relatively strong, at medium concentrations its influence is quite variable and in practice there is no such effect at high concentrations.

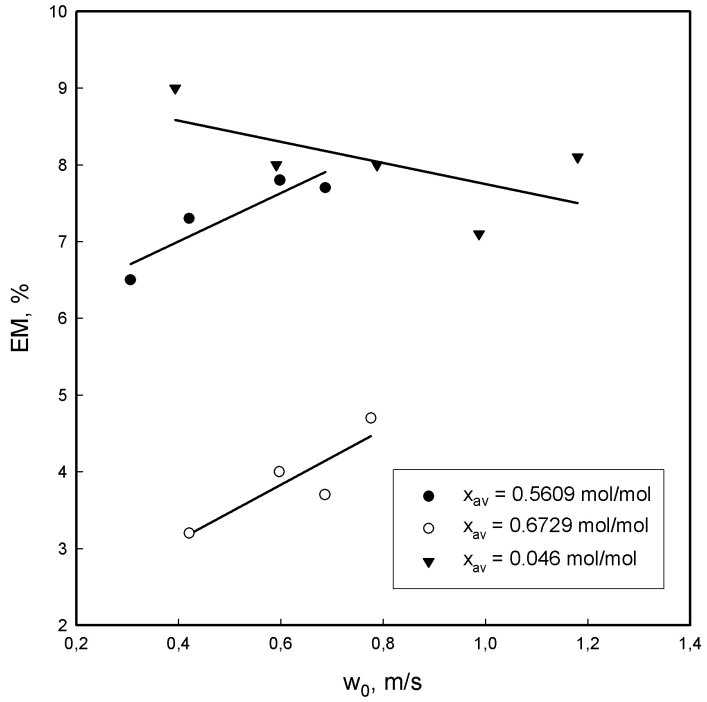


Fig. 5. Influence of the Marangoni effect on mass transfer efficiency for HOLPACK 2 at three different concentration ranges.

Ethanol – water system is known by its highly variable equilibrium. Equilibrium curve is characterized by a steep section in the zone of low concentrations of ethanol, the slope becomes flat at high concentrations, and there is azeotrope at a concentration of 96.2 % vol.

The equilibrium concentration is calculated by the well-known equation

$$y^* = \frac{\alpha x}{(\alpha - 1)x + 1} \tag{6}$$

with coefficient of relative volatility α

$$\alpha = \frac{P_A \gamma_A}{P_B \gamma_B}. \tag{7}$$

The pure component vapor pressure is determined by the Antoine equation (Stabnikov, 1962)

$$\log P = A - \frac{B}{C + t} \quad . \tag{8}$$

The values of the constants are obtained from literature data for saturated vapor pressure for three temperatures: 70, 90, 100^{0}C (Stabnikov, 1962):

for ethanol - A = 10,021; B = 1459,6; C = 212,69

for water - A = 10,156; B = 1705,9; C = 231,22

The activity coefficients are determined by the relations:

$$\ln \gamma_A = 1{,}58(1-x)^3; \tag{9}$$

$$\ln \gamma_B = 0{,}79(3-2x)x^2. \tag{10}$$

The equations used for equilibrium calculations in ethanol – water system correspond to experiments with error lower than 0,5 %.

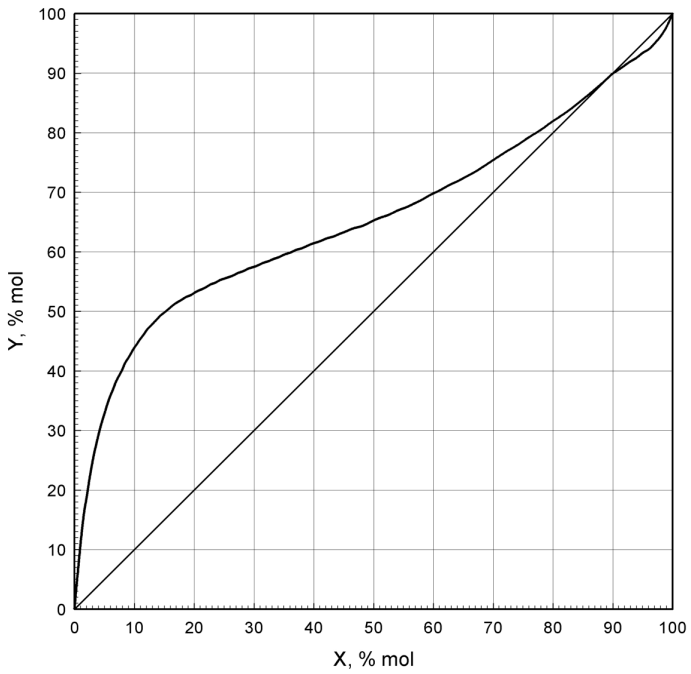


Fig. 6. Phase equilibrium of ethanol – water system

Figure 7 shows the dependence of the relative volatility and the slope of the equilibrium curve *m* on the concentration. Here again is seen significant difference in these characteristics, depending on the composition. It is known that in many model systems, used for process study, the relative volatility is constant. Moreover the liquid and gas phase resistances depend on the slope of the equilibrium curve and thus it directly affects the performance.

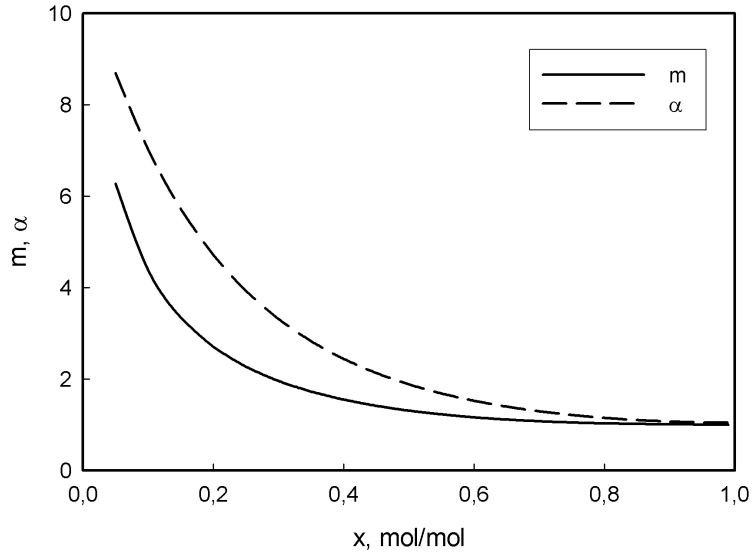


Fig. 7. Relative volatility and slope of equilibrium line vs. ethanol concentration in liquid phase.

Another important factor, which affects the mass transfer efficiency, is the non-uniformity in the distribution of the liquid and gas flows. Very strong influence on HOLPACK packing performance has the effect of axial mixing. Its account can be done by using one-parameter diffusion model, which is described in details in (Kolev and Semkov, 1991). The axial mixing effect, evaluated in this way for the experiments with HOLPACK 2 is shown on Figure 8. It is evident that the influence of the axial mixing is most intense at the lowest concentrations – up to 60 %, while at medium concentrations it is between 30 and 40 %. Calculated for HOLPACK 1 packing, at the highest concentrations, the influence of this effect increase again to 60 %.

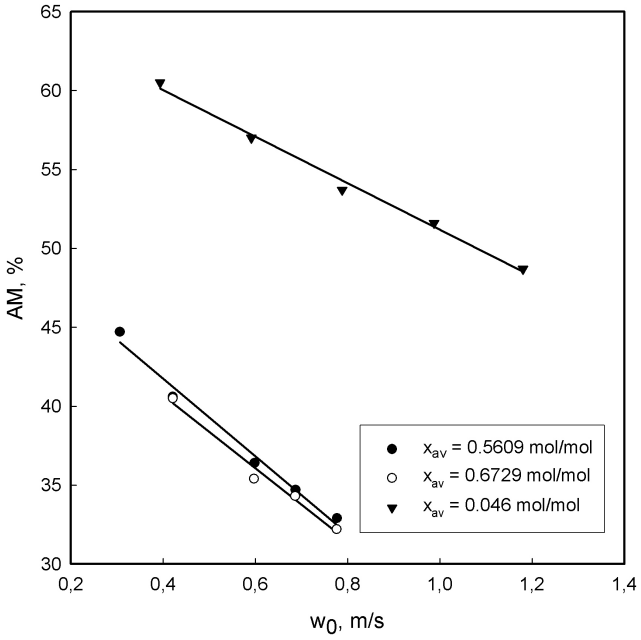


Fig. 8. Influence of the axial mixing on mass transfer efficiency for HOLPACK 2 at three different concentration ranges.

It is known that a basic problem for the packed columns is the non-uniform flow distribution over the column cross section. It changes the flow velocity and mass transfer intensity and results in variable concentrations in radial direction and efficiency reduction. It is shown (Mullin, 1957) that the negative effect depends not only on the rate of radial non-uniformity, but also on the deviation between operational and equilibrium concentrations. If in some column sections these concentrations are rather close, the non-uniformity can provoke concentration "pinch" and significantly reduction of mass transfer efficiency (Mullin, 1957, Billingham and Lockett, 2002). For this reason, strong effects are expected for rectification systems in a concentration zone of low relative volatility and large number of necessary theoretical stages.

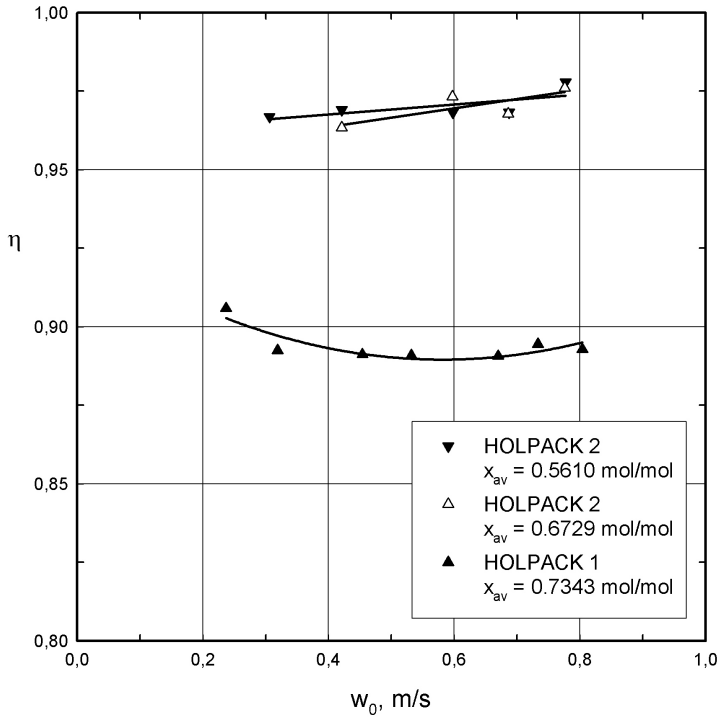


Fig. 9. Influence of vapor phase maldistribution on packing efficiency

A stochastic model for determination of the influence of vapor phase maldistribution on mass transfer efficiency was proposed recently (Semkov and Darakchiev, 2010), for which is of importance the equilibrium of the system. At high concentrations, the equilibrium curve is closer to the operating and small changes in the vapor / liquid ratio (as a result of maldistribution) lead to efficiency reduction, while at low concentrations the distance between the equilibrium curve and the operating line is bigger and the maldistribution does not has such effect. The results are shown on figure 9 as efficiency coefficient, which represents the ratio between the numbers of transfer units with and without considering radial vapor maldistribution. It is found that radial maldistribution of vapor phase decrease packing efficiency for HOLPACK 1 at average operating concentration x = 0.7343 mol/mol with 10 – 12 %. For HOLPACK 2 packing at lower concentrations this decrease is only 3-4 %.

4. CONCLUSION

The analysis, carried out in this work shows that determination of packed column efficiency for rectification is a complex task and depends on many factors. This should be taken into account when modeling the process and consider the specific conditions for its realization. Specifically for ethanol – water system is important to carry out experiments at high concentrations, as in this range operates about 80 % of the column. It is shown that the efficiency of the apparatus is lower when operates at lower concentrations. At medium concentrations the efficiency is highest and decreases again at high. The reason for the uneven packing efficiency is highly variable physicochemical properties and phase equilibrium of the ethanol – water system, as well as the different influence of some phenomena on mass transfer process.

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SYMBOLS USED:

D	-	coefficient of molecular diffusivity, m^2/s;
H	–	column height, m;
HTU	–	height of transfer unit, m;
$K_G a$	–	overall mass transfer coefficient, 1/s;
l	-	current coordinate (height), m;
$m=dy^*/dx$	-	slope of the equilibrium curve;
NTU	–	number of transfer units;
P_A	-	saturated vapor pressure of pure ethanol, Pa;
P_B	-	saturated vapor pressure of pure water, Pa;
y	-	ethanol molar concentration of vapors, mol/mol;
y^*	-	equilibrium vapor concentration, mol/mol;
y_W	-	initial vapor concentration, mol/mol
y_D	-	final vapor concentration, mol/mol,
w	-	mean vapor velocity based on column cross-section, m/s;
x	-	concentration in the liquid phase, mol/mol;
x_{av}	-	average concentration in the liquid phase, mol/mol;
α	-	relative volatility coefficient;
γ_A	-	activity coefficients of ethanol;

γ_B – activity coefficients of water;

μ - dynamic viscosity, Pa.s;

ρ - density, kg/m^3;

σ - surface tension, N/m.

Subscripts:

V - vapor phase;

L - liquid phase.

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