

## The influence of small scale maldistribution in the vapor phase on the efficiency of the rectification in packed columns

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Received November 2, 2009, Revised March 29, 2010

An analysis is made of the possibilities for accounting for small and large scale maldistribution in gas and liquid phases in packed columns. A stochastic parallel model is proposed for mathematical modeling of small scale maldistribution in gas (vapor) phase. It is assumed that the non-uniformity is normally distributed with standard deviation, equal to the maldistribution factor  $M_f$ . The reality of this assumption is confirmed by an analysis of previous experimental data for gas phase small scale maldistribution using modern random IMTP and RSRM packing. The model is tested for ethanol–water rectification using structured HOLPACK packing. Experimental data for seven types of modern packing (IMTP, Raschig Super-Ring (RSR) Metal and Plastic, and Ralu Flow Plastic) at high ethanol concentration are used to estimate the impact of vapor phase small scale maldistribution. It is found that its influence on mass transfer efficiency is quite different, and for various packing it is between 14 and 38 %. For comparison, this value for structured HOLPACK packing is 10-12 %. It is shown that a preliminary analysis of the efficiency reduction due to small scale maldistribution can provide information about the expedience of using a particular type of packing at particular operational conditions.

**Keywords:** Gas maldistribution, distillation, efficiency, packed columns

### INTRODUCTION

In the last decades random packing of third and fourth generation have been developed and applied for industrial use. The examples are Nutter rings, Ralu Flow, IMTP, Raschig Super Rings (RSR), etc. Such packings are mainly made of metal using wasteless technology but there are also plastic varieties. They ensure high efficiency at low pressure drop and are very prospective for industrial heat and mass transfer processes. For this reason, it is necessary to develop reliable methods for determination of column efficiency when operating with these packings.

Up to now some basic parameters of these packings are investigated, such as pressure drop, effective surface, and mass transfer coefficients for processes, controlled by liquid or by gas phase. Also, for the reliable forecast of the efficiency, required is information about other substantial parameters which characterize important events in the packed columns, related to the hydrodynamic and mass transfer mechanisms, and are used in mathematical models. Important events are for example the axial mixing, the liquid phase

distribution, the redistribution of gas (vapor) phase, the influence of packing discrete structure. There are studies aiming at determination of liquid flow distribution coefficient for the considered packings [1], which coefficient is important for irrigation devices design. Also, the gas phase distribution is experimentally studied, and information is collected about the maldistribution due to discrete packing structure [2, 3].

It is known that a basic problem for the packed columns is the non-uniform flow distribution over the 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 [4] 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 [4,7]. For this reason, strong effects are expected for rectification systems in a concentration zone of low relative volatility, as well as large number of theoretical stages.

The published studies on rectification with modern random packings are limited to a couple of systems, for example isobutene/n-butane, cyclo-

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hexane/n-heptane [8] and i-octane/toluene [9]. Recently thorough study has been published [6] dealing with an important industrial system, ethanol–water at different phase ratio in the high concentration zone, where equilibrium and operation line are very close.

The considered packings have low pressure drop due to their open structure. Hence, it is to expect higher minimal irregularity of the gas (vapor) phase in comparison with the traditional and especially with the structured packings. This conclusion is supported by experience. Therefore, more significant impact on the column efficiency has to be expected.

The aim of this paper is to study the influence of non-uniform vapor phase distribution and to develop a method for determination of this effect. Data are used from a case study using IMTP, Raschig Super Rings, and Ralu Flow packings, and the method is illustrated by the experimental results obtained from ethanol-water rectification.

#### PARALLEL COLUMN MODEL AND ITS APPLICATION

The parallel column model is often used for description of flow non-uniformity. A packed column is represented by a number of columns connected in parallel. Due to flow non-uniformity, the ratio of gas (vapor)/liquid [mol/mol] in each column is different.

According to the first model applications in rectification [4], the column is divided in two geometrical zones, wall and central zones, presented by two parallel columns. More often this concept is applied dividing a column in velocity zones instead of geometrical zones. For example, applying the two-column model, two zones with maximum deviation of gas-liquid ratio are formed, which provokes concentration pinch in the column ends [7]. Also, a scheme with three columns is proposed with two extreme and one mean value of the phase velocity ratio [10]. Models with even more columns are known [11]. Although some attempts to account on radial mixing by a two-column model [5], almost all parallel models neglect the radial mixing between the zones.

Generally, the parallel models treat liquid phase non-uniformity while the gas phase is assumed to be homogeneously distributed between the columns. The main shortcomings of these models are [7]: i) neglecting of radial mixing; ii) undefined number of parallel columns; iii) insufficient information about the real non-uniformity. For this reason the analyses are phenomenological, they show in general the

influence of process parameters on the efficiency, also they show that the efficiency should be reduced, plus the impact of the non-uniformity rate which results in efficiency reduction. However, the results cannot be used for apparatus design.

An additional disadvantage of the parallel model is the lack of differentiation, in terms of global treatment, between the large- and small scale maldistribution, although some comments are known about the eventual different influences of radial mixing on these two types of maldistribution [7]. On the other hand, the model is relatively simple and is well ground physically. A profound analysis of the conditions, at which it should be applied, can lead to improvement of its adequacy and extend its practical application.

Large-scale maldistribution is due to the initial phase distribution and formation of local flows as wall flow or bypass gas flows. They can be reduced or eliminated by technical devices. The initial distribution can be ameliorated by use of better inlet devices. Quick reduction of large-scale maldistribution is obtained after passing through relatively short packing layer due to its redistribution ability. Large-scale maldistribution is successfully modelled by radial dispersion model applying as main parameter spreading coefficient (for the liquid), and distribution coefficient (for the gas phase). We propose a relation to determine the redistribution layer height by multipoint irrigation devices with multiple orifices [12]. Also, we develop a method [14] for evaluation of gas distribution devices and for determination of penetration depth. The latter is defined as the height of packing layer necessary to attain minimal value of gas maldistribution factor [15, 16]. The wall flow can be limited with simple technical devices, i.e. wall flow deflecting rings [17] having less width than the characteristic width of a packing element. They are mounted horizontally on the column wall at relatively small distance and change the flow distribution in the wall zone. In case of rectification, a method is developed [18] for determination of the optimal distance between the rings, resulting in equal volume integral mean superficial velocity in wall zone and in the column bulk. It is mentioned [19] that these rings stop also the wall bypass gas (vapors) flow. It is to conclude that large-scale maldistribution can be eliminated except in short zones after the distribution devices.

Small scale maldistribution is due to the packing discrete structure and cannot be eliminated. Expressed by the value of maldistribution factor, it is manifested as a ‘noise’ which has specific value for each kind of packing [20, 25].

Small scale maldistribution provokes axial mixing in both phases which can be represented by the axial dispersion model. The packing is considered homogeneous in structure, and maldistribution is reflected by the axial mixing coefficients, i.e. by Peclet number which have to be determined experimentally. It is also necessary to identify the values of volume mass transfer coefficients that fit the dispersion model [21, 22, 24]. Regarding the liquid (dispersed) phase, the small scale maldistribution affects also the packing effective surface area but it is taken into account in the experimental determination of this surface. Significant impact of the liquid phase axial mixing on the mass transfer efficiency is registered, and it is rising at decreasing of the superficial velocity [22-24].

Considering the gas phase, the investigation of axial mixing goes through a relatively difficult and imprecise experiment due to faster transient process. However, relatively small axial mixing impact is found in the case of traditional random packings, about 19 %, which allows for using of relatively imprecise relations for its determination [21]. For some structured packings it is even admitted to neglect the gas phase axial mixing. There is no information about studies on the axial mixing in gas or liquid phase in columns with new highly efficient random packings. However, significant values of maldistribution factor after the penetration depth are experimentally determined which should be attributed to their open structure and existence of more than one characteristic geometrical dimension.

It can be concluded, resuming the existing information, that:

- Large-scale maldistribution can be limited to small zones in the column and can be successfully modeled by the dispersion model with radial mixing. Parallel model is not applicable to large-scale maldistribution because of large radial gradients and significant radial mixing.
- Small scale maldistribution in the liquid phase can be modeled with acceptable precision by the dispersion model with axial mixing. The packing can be regarded as homogeneous medium with uniform radial distribution. For this reason, it is not expedient to apply parallel model at these conditions.
- The experimental study of the gas phase axial mixing in the packing is difficult and relatively imprecise. There is no data on the modern random packings. However, detailed information is available about the maldistribution factor after

the penetration depth where the large-scale maldistribution is already eliminated. At these conditions radial velocity gradients are small and radial mixing is low. The parallel model can be applied.

#### THE STOCHASTIC PARALLEL MODEL

According to the above considerations, we propose to tighten the zone of parallel model application in order to ameliorate its adequacy. The model will be applied for evaluation of the impact of the gas (vapor) phase small scale maldistribution in packed columns. The following assumptions were taken:

- The liquid phase is uniformly distributed over the column cross-section;
- The radial mixing in the gas phase is negligible;
- Gas (vapors) velocity profile over the cross-section is described by normal random distribution law.

The last assumption eliminates the need to introduce undefined number of parallel columns. It corresponds well to the experiments, as it will be shown later.

The density of normal distribution is given by the expression

$$\varphi(U) = \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{1}{2}U^2\right), \quad (1)$$

where  $U = \frac{z - \xi}{\sigma}$  is a standard normal variable.

The model solution is based on an approximate presentation of the Standard normal distribution with a mean 0 and standard deviation of 1 in nine specific intervals, when the center interval is within the quarter of a standard deviation of a mean, and each of the other intervals are a half standard deviation wide, except for the tails. This presentation is in terms of the so called stanines (standard nine intervals) [26], Fig. 1.

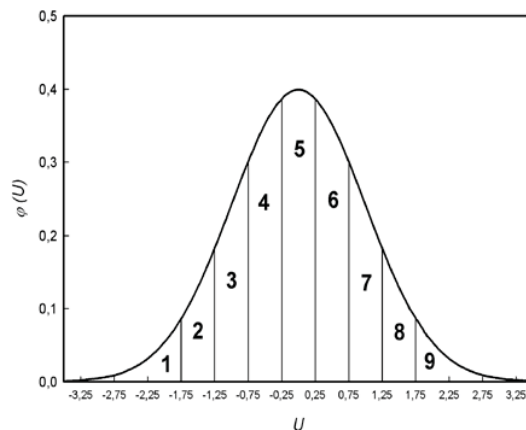


Fig. 1. Standard normal distribution in terms of “stanines” (standard nine intervals).

terms in Eq. (1) have the meaning

$$z \equiv G/G_0; \xi \equiv 1; \sigma \equiv M_f; U = \frac{G - G_0}{G_0 M_f} \quad (2)$$

For every stanine, the mean value of random variable is introduced

$$Sn_i \equiv (i - 5)0,5M_f \quad \text{for} \quad i = 2 \dots 8 \quad \text{and} \\ Sn_9 = -Sn_1 = 2,375M_f. \quad (3)$$

Then the mean density of the gas (vapor) molar flow in a particular stanine is

$$G_i = (1 + Sn_i)G_0, \quad i = 1, \dots, 9. \quad (4)$$

According to the first model assumption, the superficial velocity is the same for each stanine, i.e.  $L_i = L_0$ . Then the ratio of molar flows densities becomes

$$\frac{L_i}{G_i} = \frac{1}{(1 + Sn_i)} \frac{L_0}{G_0}, \quad i = 1, \dots, 9. \quad (5)$$

The partial flow for  $i^{th}$  stanine is given by the coefficient  $k_i$  (see Table 1).

**Table 1.** Part of the results for the stanines

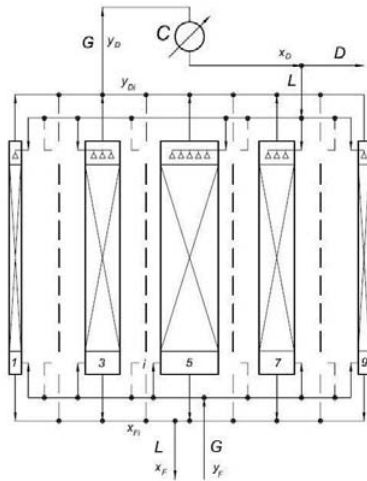
$i$	1;9	2;8	3;7	4;6	5
$k_i$	0,04	0,07	0,12	0,17	0,20

Additionally, by definition

$$\sum_{i=1}^9 k_i = 1, \quad (6)$$

taking into account the symmetry of (3)

$$\sum_{i=1}^9 Sn_i = 0 \quad \text{and} \quad \sum_{i=1}^9 k_i Sn_i = 0. \quad (7)$$



**Fig. 2.** Flowchart illustrating stochastic model with nine virtual parallel columns.

columns. In the present example the symbols, used in the model, are associated with the upper (enrichment) part of a rectification column with condenser C. In this case the model includes nine parallel columns, corresponding to nine stanines. The molar flow ratio is defined by (5) and the corresponding cross-sections are in the proportions, given in Table 1.

In the upper part of the parallel columns, a liquid flow (reflux) with concentration  $x_D$  is fed by the condenser C. In the lower part, vapor with concentration  $y_F$  comes from the column stripping part. As the ratio vapor/liquid is different for each parallel column, the corresponding outcome flows have different concentration –  $y_{Di}$  for vapors, and  $x_F$  for the liquid. The vapors are mixed to give a flow with concentration  $x_D$ . The liquid flows from the bottom of the columns are also mixed. The liquid with the resulting concentration  $x_N$  is used to feed the stripping part of the column. The material balance of the  $i^{th}$  column is

$$G_i(y_{Di} - y_F) = L_i(x_D - x_{Fi}), \quad (8)$$

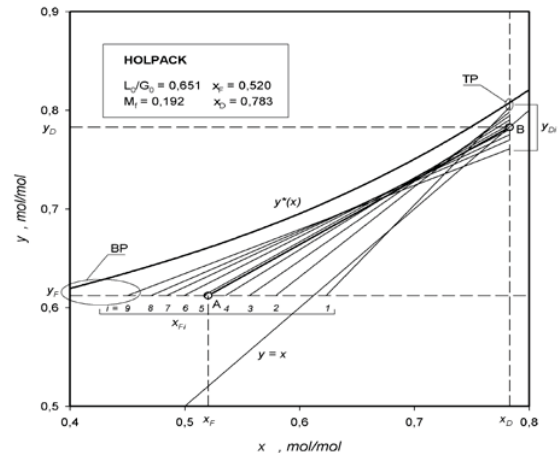
For a given cross-section with operational concentrations  $x_i$  and  $y_i$  it is

$$G_i(y_i - y_F) = L_i(x_i - x_{Fi}). \quad (9)$$

After rearrangement of (8) and (9), an expression for the operating line of the  $i^{th}$  column is obtained

$$y_i = \frac{L_i}{G_i} x_i + \left(1 - \frac{L_i}{G_i}\right) x_D + (y_{Di} - x_D). \quad (10)$$

This operating line differs from the traditional line for the column upper part. In our case  $x_D \neq y_{Di}$  for all stanines, including  $i = 5$  when  $L_5/G_5 = L_0/G_0$ . Fig. 3 illustrates the operating lines for the



**Fig.3.** Operating lines for a given condition of ethanol-water rectification.



particular case of ethanol–water rectification using the HOLPACK packing [6]. Considering for the condition  $L_i = L_0$  and from expression (4) the concentrations after mixing of outgoing flows become:

$$\text{-for the vapor phase} \\ y_D = \sum_i k_i (1 + Sn_i) y_{Di}, \quad (11)$$

$$\text{-and for the liquid phase} \\ x_N = \sum_i k_i x_{Fi}. \quad (12)$$

The correctness of Equations (11) and (12) can be easily confirmed as shown below. The concentration  $y_{Di}$  is obtained from Eq. (8) using (5):

$$y_{Di} = y_F + \frac{1}{1 + Sn_i} \frac{L_0}{G_0} (x_D - x_{Fi}) \quad (13)$$

Introducing Eq. (13) in (11) with regard to (6), (7) and (12), it is obtained

$$y_D = y_F \sum_i k_i (1 + Sn_i) + \frac{L_0}{G_0} \sum_i k_i (x_D - x_{Fi}) = \\ = y_F \sum_i k_i + \frac{L_0}{G_0} \left( x_D \sum_i k_i - \sum_i k_i x_{Fi} \right) = \\ = y_F + \frac{L_0}{G_0} (x_D - x_N) \quad (14)$$

As  $y_D = x_D$ , this expression represents the material balance of the column enriching part at mean vapor/liquid ratio, and the exit liquid concentration  $x_N = x_F$ .

The influence of the vapor phase maldistribution on the overall column efficiency can be evaluated by the number of transfer units, using HTU–NTU model. For a particular packing, defined operational conditions, including the  $L_0/G_0$  ratio, and known column height, the concentrations  $y_D = x_D$  and  $y_F$  can be measured. Then the number of transfer units in the vapor phase is

$$NTU^+ = \int_{y_F}^{y_D} \frac{dy}{y^* - y} \quad (15)$$

This expression can be interpreted as effective number of transfer units in the column at given degree of separation.

In our case the stochastic parallel model includes nine parallel columns with different vapor/liquid ratio, which depends on the model parameter,  $M_f$ . It is assumed for simplification that the mass transfer efficiency keeps constant value in the interval of vapor velocity changes due to small scale maldistribution. Hence, the same number of transfer units has to be placed in each column

$$NTU = NTU_i = \int_{y_F}^{y_{Di}} \frac{dy_i}{y^* - y_i} = const \quad (16)$$

where  $y_{Di}$  is determined via Eq. (13) which defines the relation between  $y_{Di}$  and  $x_{Fi}$ . Additionally, the conditions of the Equations (11) and (12) have to be satisfied. Eq. (16) represents the number of transfer units necessary to attain a preset separation in the presence of gas phase radial maldistribution. The impact on mass transfer efficiency can be expressed by the efficiency coefficient

$$\eta = \frac{NTU^+}{NTU} \leq 1 \quad (17)$$

This expression can also be regarded as the reduction rate of the driving force due to the vapor phase maldistribution.

## MODEL SOLUTION

At preset or experimentally measured concentrations  $x_F$  and  $x_D$  and the molar phase  $L_0/G_0$  ratio, the concentrations  $y_F$  and  $y_D = x_D$  can be determined from the operating line. Then  $NTU^+$  can be directly derived from Eq. (15). Equilibrium dependence is also needed, and the integration is generally made by a proper numerical method.

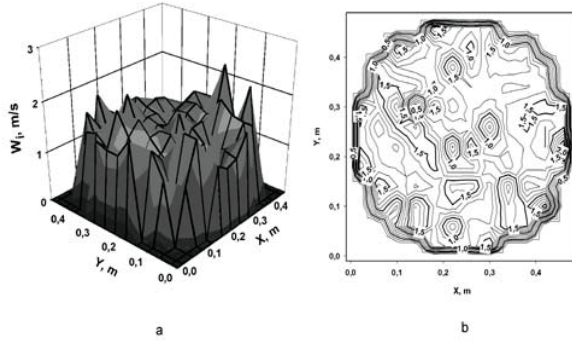
The term  $NTU$  in Eq. (16), together with the conditions (11, 12) has to be determined by iterative procedures. We shall only mention without detailed description that after testing of different methods, type of functions, solution sensitivity and convergence velocity of the solution, we have developed a particular two-level iterative procedure. At the first (low) level the iterations are made for  $y_{Di}$  for every stanine until a predefined value of  $NTU$  is attained. Then  $x_F$  is determined by Eq. (13). The values of  $y_D$  and  $x_N$  are obtained by summing the stanine concentrations, Equations (11 and 12). At the second (high) level the iterations are made for  $NTU$  until the condition  $x_N = x_F$  is satisfied. At both levels the new approximations are defined by the secant method, a numerical version of the Newton-Raphson method. The integration is made by the method of Simpson. The convergence velocity depends strongly on initial approximations of  $y_D$ , especially for  $i=1$  and  $i=9$ , where top (TP) and bottom pinch (BP) are obtained (see Fig. 3). For these cases the initial approximations have been chosen according to the pinch with small (0.5%) reserve in the safe side. In regards to the other stanines, the result for the previous stanine has been used as initial approximation for the next one. The software program developed is very stable and fast with relative deviation of  $1.10^{-4}$ , attained with 3-8 iterations for  $y_{Di}$  and 3-5 iterations for  $NTU$ .

### EXPERIMENTAL STUDY

The studies on gas phase distribution [2,3] have been carried out in a column of 0.47 m in diameter with random Raschig Super-Ring, IMTP and Ralu Flow packings, and a structured HOLPACK packing has been used for comparison [27, 16]. Thermoanemometry has been applied to measure the gas velocity profile (single phase with no countercurrent liquid flow) throughout the cross-section. The measuring device has been connected to a computer. A hundred values in each point have been taken and a mean value has been determined. The measurement points have been placed over two perpendicular diameters in 22 points on each of them. In some cases, when it has been technically possible (for example at the end of a column section with height 0.8 m), gas velocity has been measured in 112 points, regularly distributed throughout the cross-section. These results, considered to be more reliable, are used in this paper. Measurements for different packing height have been carried out, too. The results have been presented as maldistribution factor:

$$M_f = \frac{1}{w_0} \sqrt{\frac{\sum_{j=1}^n (w_j - w_0)^2}{n-1}}, n = 112 \quad (18)$$

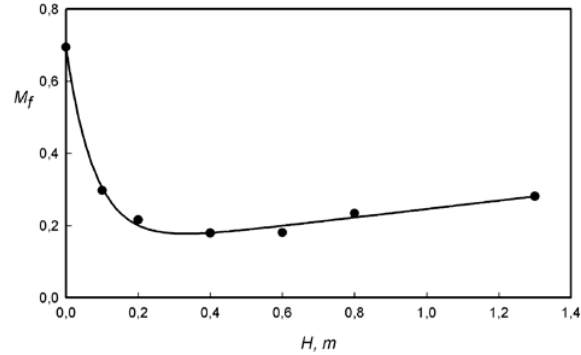
Fig. 4 represents the experimental gas velocity distribution of IMTP 25 at 0.8 m packing height.



**Fig. 4.** Experimental gas velocity distribution for IMTP 25 at 0.8 m packing height: a) 3D representation; b) contour plot.

Fig. 5 illustrates the results for the IMTP 25 packing at different packing height. This is a typical graph for most of the modern random packings. In the beginning the maldistribution factor descends sharply, passes through a minimum, and then begins slowly to rise. The first zone corresponds to the large-scale maldistribution, produced by the inlet gas distribution device. The minimum is attained at packing height of only 0.2 m. It is the so called

‘uniformity limit’ or the ‘noise’, which expresses the small scale maldistribution due to the packing discrete structure. The rising part of  $M_f$  represents a new development of relatively small large-scale maldistribution, which can be attributed to the formation of wall gas flow. It is typical for the random packings and can be attributed to the different geometrical structure of the wall and bulk zones. In the case of block or package packings with regular cell structure (Honeycomb [14] or HOLPACK [27, 16]), this effect is not pronounced.



**Fig. 5.** Maldistribution factor vs. packing height for IMTP 25-typical graph for most of the modern random packings.

The assumption for existence of normal standard distribution, Eq. (1) is verified by the test of Kolmogorov-Smirnov [28]. It is based on the determination of the maximal difference  $D_c$  between the cumulative frequencies of the empirical and theoretical distribution

$$D_c = \max |F_n(U) - F(U)|, \quad (19)$$

where  $F/U$  is a theoretical probability integral

$$F(U) = \Phi(U) = \frac{1}{\sqrt{2\pi}} \int_0^U \exp\left(-\frac{t^2}{2}\right) dt. \quad (20)$$

The value, derived by (19), is to be compared to a tabulated value. In case the latter is smaller, the hypothesis for normal distribution is not valid. When the number of points is larger than 35, the tabulated value at significance level of 0.05 is calculated by the expression [27]

$$D_{C|0.05} = \frac{1.36}{\sqrt{n}}. \quad (21)$$

For our experiments  $n = 112$  and  $D_{C|0.05} = 0.1285$ .

Fig. 6a shows a comparison between corresponding cumulative frequencies for 20 intervals (IMTP 25 packing, 1.3 m in height). Fig. 6b represents a comparison of theoretical and experimental distribution of nine stanines. The

values, derived from Eq. (19), are  $D_C = 0.055 < D_{C|0.05} = 0.1285$ , i.e. the hypothesis for normal distribution is approved. The same value of  $D_C$  is obtained for packing of 0.8 m height. For the RSRM 2, RSRM 3 and RSRP 2 packings at 0.8 m height, the corresponding values of  $D_C$  are respectively 0.073; 0.068 and 0.045. Hence, the hypothesis of normal distribution is applicable in these cases, too.

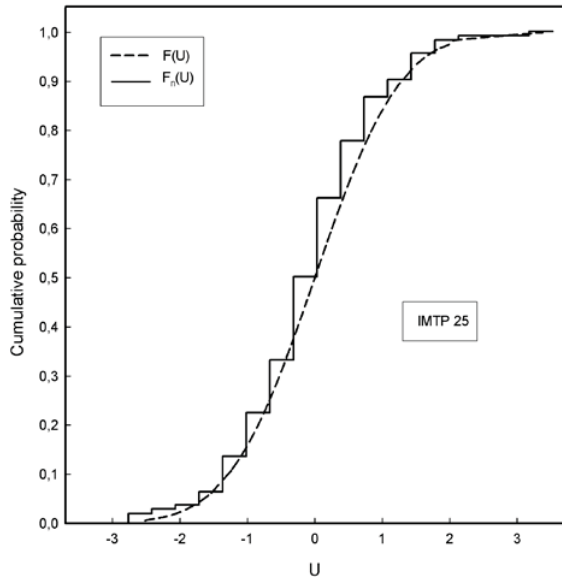


Fig. 6(a)

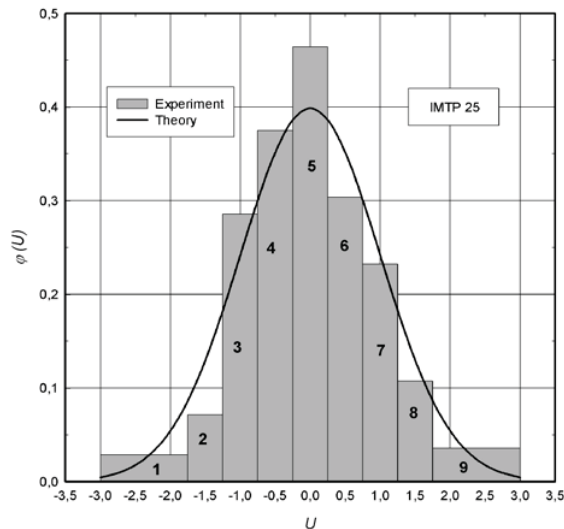
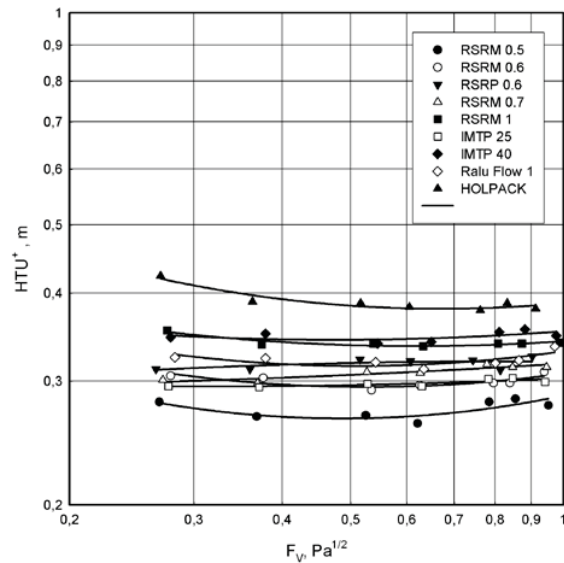


Fig. 6(b)

**Fig. 6.** Standard normal distribution representation of the gas velocities for IMTP 25 at 1.3 m packing height: a) comparison of corresponding cumulative frequencies for 20 intervals; b) comparison of theoretical and experimental distribution in terms of stanines.

The study on ethanol–water system rectification [6] has been carried out in the zone of high ethanol concentration, which corresponds to the common case for the main part of the industrial rectification columns. The experimental column is of 0.213 m diameter with packing layer of 2.8 m. Wall flow deflecting rings (WFDR) [17] are mounted on the inner wall surface at 200 mm distance. They serve to eliminate the liquid wall flow and its harmful impact [18]. Obviously, the rings also



**Fig. 7.** Experimental (effective) height of transfer units vs.  $F$ -factor at full reflux.

hamper the development of gas wall flow. The column can operate at constant or variable reflux.

Seven types of modern random packings are studied: four metal Raschig Super – Rings with characteristic sizes of 0.5; 0.6; 0.7 and 1”, one plastic – 0.6”, two types of IMTP, and one plastic Ralu Flow. For comparison, a structured metal HOLPACK packing, used currently in the ethanol production, is also tested under identical conditions. The experiments are carried out at vapor flow velocity of 0.253 – 0.936 m/s and liquid superficial velocity of  $4.44 \cdot 10^{-4} \text{ m}^3/(\text{m}^2\text{s}) - 1.63 \cdot 10^{-3} \text{ m}^3/(\text{m}^2\text{s})$  at full and variable reflux, attained at constant liquid superficial velocity and variable vapor velocity, as well as at constant vapor velocity and variable liquid velocity. Fig. 7 illustrates the results, obtained at full reflux.

Table 2 contains the values of the gas phase maldistribution factor for the relevant rectification column conditions, using the available experimental results from different authors. In all cases, the values refer to the zone beyond the penetration depth where mainly small scale maldistribution is pronounced.

**Table 2.** Gas maldistribution factor and packing specific surface area.

Packing	$M_f$ [-]	$a$ , $m^2/m^3$
RSRM 0.5	0.260 [2]	236 [29]
RSRP 0.6	0.270 [2]	206 [1]
RSRM 0.7	0.241 [2]	176 [29]
RSRM 1	0.245 [2]	156 [29]
IMTP 25	0.181 [3]	243 [30]
IMTP 40	0.263 [3]	172 [30]
Ralu Flow 1	0.197 [3]	177 [30]
HOLPACK	0.192 [27, 16]	56 [6]

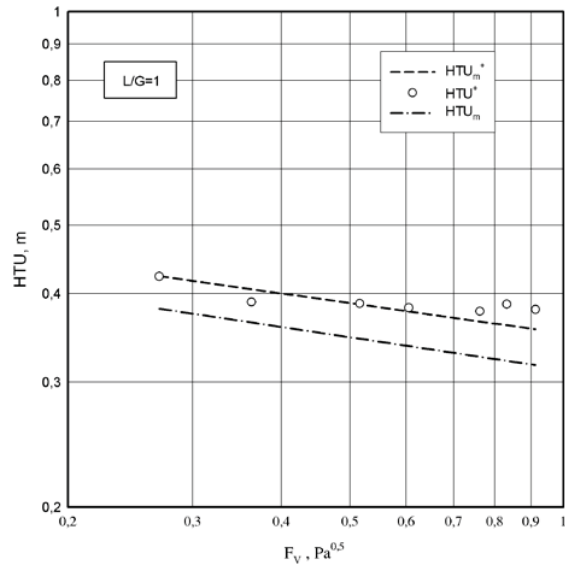
Table 2 includes also data on the packing specific surface area,  $a$ , in order to allow an approximate comparison of their efficiencies.

### RESULTS AND DISCUSSION

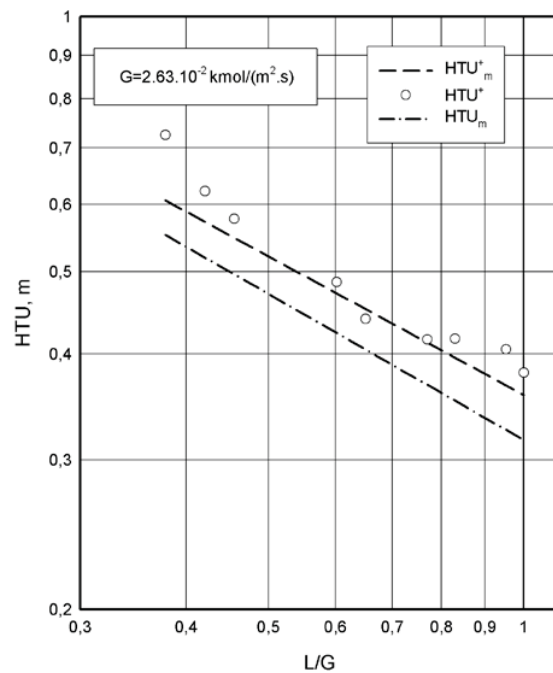
The stochastic model is applied to our previous experimental results for the ethanol–water rectification [6]. Preliminary check of the model performance was done using the rectification results for the structured HOLPACK packing. This packing is thoroughly investigated. The packing parameters are determined by unified method for modeling of heat and mass transfer processes [31, 32, 23] because of the lack of experimental data. These parameters are: mass transfer coefficients in both boundary layers [33, 34], effective surface area [35], liquid phase axial mixing (Peclet number) [36], and the impact of Marangoni effect [13, 23]. Only the influence of vapor phase axial mixing is not taken into account because of lack of experimental data. It is admitted [23] that the HOLPACK packing (perforated and extended metal sheets placed horizontally) ensures uniform distribution of vapors, and they have plug-flow structure.

Fig. 8 shows the results of the stochastic model application using data, obtained by a mathematical modeling. A comparison between the experimental results is also shown. The term  $HTU_m$  is the height of a transfer unit, determined by the abovementioned unified method, which neglects the vapor phase maldistribution. After applying the stochastic model and considering the vapor phase maldistribution, the effective height of transfer unit  $HTU_m^+$  is calculated. The corresponding experimental data for  $HTU_m^+$  are determined from Eq. (15). Obviously, the description is significantly improved when vapor phase small scale maldistribution is taken into account. The efficiency reduction for HOLPACK packing at the particular experimental conditions is about 10-11%.

Fig. 9 illustrates the results at full reflux, presented as influence of  $F$ -factor,  $F_V$ , on the efficiency coefficient,  $\eta$ , Eq. (17). The upper line



**Fig. 8a**



**Fig. 8b**

**Fig. 8.** Comparison between experiment ( $NTU^+$ ), and models without  $NTU_m$  and with ( $NTU_m^+$ ) taking into account vapor phase maldistribution for HOLPACK: a) at full reflux; b) at constant vapor load.

refers to the structured HOLPACK packing. Significant difference of efficiency coefficients is seen. It is to stress upon the strong impact of vapor phase maldistribution – up to 37% for the RSRM 0.5 packing. Nevertheless, the efficiency of this packing is the best, see Fig. 7.

Similar results are obtained at variable reflux. The result analysis of the results, as well as



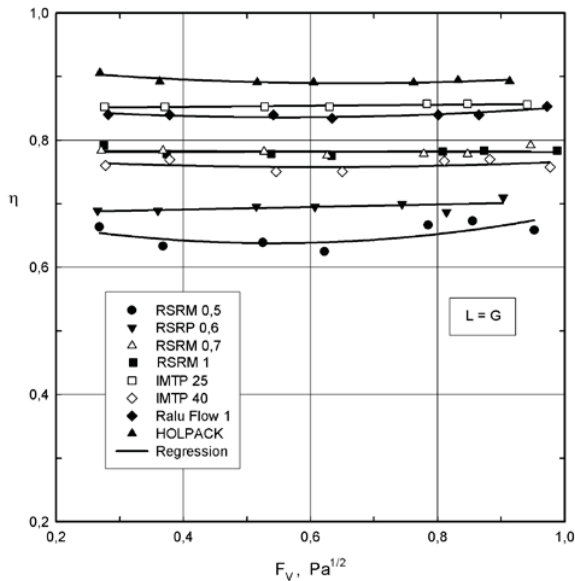


Fig. 9. Efficiency coefficient vs. F-factor at full reflux.

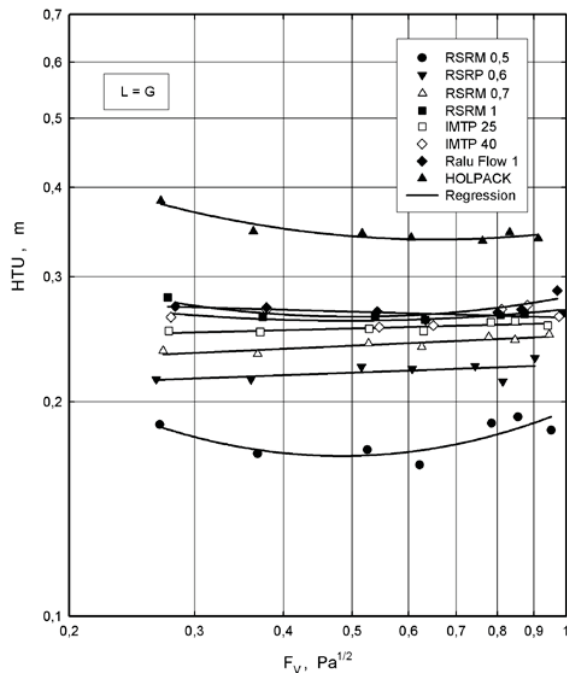


Figure 10. HTU neglecting the vapor phase maldistribution vs. F-factor at full reflux.

simulation results, show that the influence of the vapor phase maldistribution is stronger for more efficient packings and at a higher value of the maldistribution factor. The relative efficiency of random packings might be estimated in relation with their specific surface area, Table 2. Although it is a rough comparison, it might be done because the effective surface/specific surface area ratio does not practically depend on the specific surface [29]. This ratio values are between 0.72 and 0.89 in these particular experimental conditions. The same is true

for the IMTP and Ralu Flow packings [30] because of their similar structure. However, it is not applicable to the HOLPACK packing which has a completely different structure. Correspondingly, the value of the above mentioned ratio is about 1.5, i.e. the effective surface is significantly larger than the specific surface. The vapor phase maldistribution affects most strongly the packing with the largest specific surface and highest maldistribution factor, i.e. the RSRM 0.5. For IMTP 25 which has similar specific surface but small maldistribution factor, the impact is twice smaller, 15-16%. The smallest influence is registered in the case of the HOLPACK packing, which has small specific surface and small maldistribution factor.

Fig. 10 illustrates the height of transfer unit, neglecting vapor phase maldistribution. In this case more pronounced difference between the efficiency of different packages is observed. The ratio best/worst efficiency is about 2, while it is not greater than 1.4 in the case of experimental efficiency determination (Fig. 7).

#### CONCLUSION

A stochastic model is proposed which allows for quantitative evaluation of the influence of the vapor phase maldistribution on the packing efficiency based on data on the maldistribution factor. Using modern random packings, significant reduction of packing efficiency, between 16 -38 %, is found for the ethanol-water rectification in the zone of high ethanol concentration. Such a strong impact should not be neglected and has to be taken into account when selecting the type of packing. It is necessary to improve the accuracy of the experimental determination of the maldistribution factor because of its essential influence on the efficiency.

**Acknowledgements** This study has been carried out with the financial support of the Grant scheme № BG051PO001-3.3.04/30 /28.08.2009 under the Operational Program "Human Resources Development" 2007-2013, co-financed by European Social Fund of European Community.

#### NOMENCLATURE

Symbols used

- $a$  specific surface area of the packing,  $m^2/m^3$
- $F$  cumulative frequencies, [-]
- $F_V$  F-factor ( $F_V = w_0 \sqrt{\rho}$ ),  $Pa^{1/2}$
- $D_M$  molar flow of the distillate, mol/s
- $D_C$  term in Eq. (19), [-]

$G$	density of the gas (vapor) phase molar flow, mol/(m <sup>2</sup> s)
$G_M$	vapor molar flow, mol/s
$H$	height of the packing layer, m
$k$	weight coefficient, Table 1, [-]
$L$	density of the liquid phase molar flow, mol/(m <sup>2</sup> s)
$L_M$	molar flow of the reflux, mol/s
$M_f$	maldistribution factor, Eq. (18), [-]
$n$	number of experiments, [-]
$Sn$	mean value of the random variable $U$ , [-]
$w$	gas (vapor) flow velocity, m/s
$x$	liquid phase molar concentration, mol/mol
$y$	vapor phase molar concentration, mol/mol
$H$	height of the packing layer, m
$HTU$	height of the transfer unit, m
$NTU$	number of transfer units, [-]
$t$	current value, Eq. (20), [-]
$U$	standard normal random variable, [-]
$z$	random variable, [-]

#### Greek letters

$\eta$	efficiency coefficient, Eq. (17), [-]
$\rho$	gas (vapor) density, kg/m <sup>3</sup>
$\sigma$	standard deviation, [-]
$\xi$	mathematical expectation, [-]
$\Phi(U)$	probability integral, [-]
$\varphi(U)$	probability density, [-]

#### Subscripts

$i$	for $i^{\text{th}}$ stanine (parallel column)
$j$	for $j^{\text{th}}$ point
$m$	model
$n$	experimental for $n^{\text{th}}$ interval, Eq. (19)
$0$	mean value
$D$	in/out at column top
$F$	in/out at column bottom
$N$	summing value at the bottom exit

#### Superscripts

+	effective value including maldistribution impact
*	equilibrium value

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ОТЧИТАНЕ ВЛИЯНИЕТО НА ДРЕБНО-МАЩАБНАТА НЕРАВНОМЕРНОСТ В ПАРОВАТА ФАЗА ПРИ РЕКТИФИКАЦИЯ С МОДЕРНИ ВИСОКО-ЕФЕКТИВНИ НЕНАРЕДЕНИ ПЪЛНЕЖИ

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Постъпила на 2 ноември, 2009 г., преработена на 29 март 2010 г.

(Резюме)

Направен е анализ на възможностите за отчитане на дребно- и едромашабните неравномерности в двете фази при колони с пълнеж. Но тази основа е предложен стохастичен паралелен модел за математично моделиране на дребно-машабната неравномерност в газовата (паровата) фаза. При него се приема, че неравномерностите се подчиняват на нормално случайно разпределение със стандартно отклонение равно на фактора на неравномерност  $M_f$ . Правомерността на това допускане е показана на основата на анализ на получени по-рано експериментални данни за дребно-машабната неравномерност в газовата фаза при съвременни ненаредени пълнежи IMTP и Raschig Super-Ring (RSR). Възможностите на модела са проверени и доказани при ректификация на система етанол-вода със структурен пълнеж HOLPACK. Обработени са експериментални данни по ректификация на същата система при високи концентрации на етанола с осем вида съвременни пълнежи от сериите IMTP, Raschig Super-Ring (метални и пластмасови), и Ralu Flow. Показано е, че при конкретните условия влиянието на дребно-машабните неравномерности в паровата фаза върху ефективността на масообмена силно се различава за отделните пълнежи и варира между 14 и 38%. При използвания за сравнение структуриран пълнеж HOLPACK това влияние е 10-12%. Посочено е, че предварителният анализ на понижаването на ефективността вследствие на споменатата неравномерност може до голяма степен да реши целесъобразността на използването на даден вид пълнеж при конкретни условия на процеса.