

MATERIAL TRANSFER EFFICIENCY IN THE MULTISTAGE PACKED DISTILLATION AND ABSORPTION COLUMNS

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Abstract

Material transfer efficiency by height transfer units and height of equivalent theoretical plate was investigated. Several correlation models have been studied. The overall and individual height of transfer units were examined. Based on correlation models of the individual phase material transfer units for the gas and the liquid phases the overall material transfer units were determined. The models were tested with different systems. The comparative study of the different models which used for the prediction of height of transfer units and height of equivalent theoretical plate in packed columns. The obtained results show efficiency of material transfer and it using for column design and operation. These results can be used for packing efficiency determination.

Keywords: Packed column; substance transfer; separation; height of transfer units; height of equivalent theoretical plate; correlations.

1. Introduction

Substance transfer phenomena where diffusion occurs through the liquid and gas phases has been studied long period of time, in distillation columns. A different approach to the modelling of the packed column, particularly applicable to multicomponent distillation, was developed and applied in full-scale tests in papers [1-6]. They put forward the concept of substance transfer section and defined vaporization efficiencies to compensate for the deviation of each section from an ideal stage.

Correlations generally need to estimate is there any connection between two or more dependent or independent variables. If common probability distribution for two variables is known then can be calculated some measure linear dependence between them, which called correlation coefficient [7-8].

Substance transfer coefficients, HTU/NTU (Height of transfer units/Number of transfer units) and HETP (Height of equivalent theoretical plates) in distillation packed columns were investigated in papers [9-11].

Industrial separation processes continue to be of considerable interest. Mass transfer between the liquid and vapor phase of the systems with acetic acid association in a distillation column was studied [10-12]. Several correlation models for packed distillation columns were studied in literature [13-22].

In this paper correlation functions for the substance transfer in the packed distillation and absorption columns were examined. Several correlation models for HTU/NTU and HETP have been investigated. Correlation models for overall, individual and component transfer are examined.

2. Substance transfer in multistage and multiphase system

Substance transfer between the liquid and vapor phases in packed column can be interpreted in terms of two film theory if sensible heat exchanges throughout the column, heats of mixing and heat losses to the surroundings are negligible.

Flux through the column can be expressed:

$$d(Gy_i) = d(Lx_i) = N_i dN = N_i a S dz \quad (1)$$

For the gas phase, if assuming $G = \text{constant}$,

$$H = \int_0^H dz = \frac{G}{S} \int_{y_1}^{y_2} \frac{dy}{k_{OG} a (y - y_i)} = \frac{G}{S} \int_{y_1}^{y_2} \frac{dy}{k_{OG} a (y - y^*)} \quad (2)$$

For the dilution systems, $m = \text{constant}$, can be written:

$$H = \int_0^H dz = \frac{G}{k_{OG} a S} \int_{y_1}^{y_2} \frac{dy}{(y - y_i)} = \frac{G}{k_{OG} a S} \int_{y_1}^{y_2} \frac{dy}{(y - y^*)} \quad (3)$$

For the liquid phase,

$$H = \int_{x_1}^{x_2} dz = \frac{L}{k_{OL} a S} \int_{x_1}^{x_2} \frac{dx}{x_i - x_i} = \frac{L}{k_{OL} a S} \int_{x_1}^{x_2} \frac{dx}{(x - x^*)} \quad (4)$$

these integrals can be defined as the number of transfer units $(NTU)_{OG}$ or $(NTU)_{OL}$ and height of transfer unit $(HTU)_{OL}$ and $(HTU)_{OG}$:

$$(HTU)_{OG} = \frac{H}{(NTU)_{OG}} = \frac{H}{\int_{y_1}^{y_2} \frac{dy}{y - y^*}} \quad (5)$$

$$(HTU)_{OL} = \frac{H}{(NTU)_{OL}} = \frac{H}{\int_{x_1}^{x_2} \frac{dx}{x - x^*}} \quad (6)$$

Those expressions were used for overall $(HTU)_{OL}$, $(HTU)_{OG}$ or k_{OL} , k_{OG} determination by experimental measurement using method of numerical integration [10-12]. The relationship between the overall and individual HTU is given by the following relation [3-5]:

$$(HTU)_{OG} = (HTU)_G + \frac{mG}{L} (HTU)_L \quad (7)$$

$$(HTU)_{OL} = (HTU)_L + \frac{L}{mG} (HTU)_G \quad (8)$$

The relationship between the overall HTU and HETP (height of equivalent theoretical plate) is given by

$$(HETP) = (HTU)_{OG} + \frac{\left(\frac{mG}{L}\right) - 1}{\ln\left(\frac{mG}{L}\right)} \quad (9)$$

HETP is defined as

$$HETP = H / (N_t - 1) \quad (10)$$

where N_t is the required number of theoretical plate.

3. Packed tower correlated variables

Relation between correlation variables illustrates in Appendix A1. In separation processes need to define mixture variables and packed tower variables. The best fitting of those variables gives the optimal process condition [14].

$$\text{Substance transfer efficiency} = f(\text{column and packing properties, system properties}) \quad (11)$$

$$HTU = f_1(L, D_c, d_p, H) f_2(\mu, \rho, D, \sigma) \quad (12)$$

$$HETP = f_1(L, D_c, d_p, H) f_2(k, \alpha, \mu, \rho, D, \sigma) \quad (13)$$

The conflict resolution between f_1 and f_2 functions has given the optimal values for HTU and HETP.

Comparative study of different models for column efficiency determination over height of transfer units and height equivalent of theoretical plate were performed. The most adequate model of the real process efficiency make possible improving current and development a new separation technology.

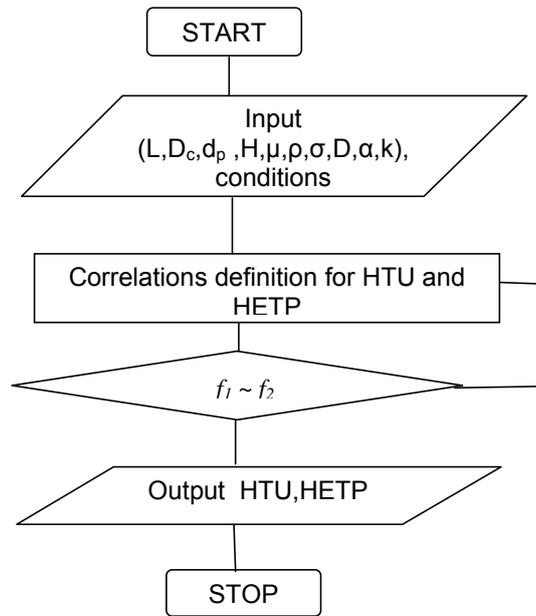


Fig.1 Correlation modelling

4. Modelling of the HTU and HETP

Many correlation models for HTU and HETP in packed columns are reported in the literature [15-22]. Many authors were correlated own experimental data. Some authors correlated experimental data from literature by different correlation models.

In this paper for HTU correlation models of Sherwood-Hollway, Cornell, Knapp and Fair, and Zuiderweg were examined.

Sherwood-Hollway [15]:

$$(HTU)_L = B_1(L / \mu_1)^n Sc_L^{0.5} \quad (14)$$

$$(HTU)_G = B_2 G^{0.31} / L^{0.33} \quad (15)$$

Cornell, Knapp and Fair [16-17]:

$$(HTU)_L = \Phi Sc_L^{0.5} (C')(H/10)^{0.5} \quad (16)$$

$$(HTU)_G = \psi Sc_G^{0.5} (L \varphi_1 \varphi_2 \varphi_3)^{-0.6} (d_c/12)^{1.24} (H/10)^{1/3} \quad (17)$$

Zuiderweg [18]:

$$(HTU)_L = C_1 w_L^{0.27} d_p^{0.33} D_L^{-0.5} g^{-0.17} \quad (18)$$

$$(HTU)_G = C_2 Re_G^{0.4} Sc_G^{0.67} w_L^{-0.56} \quad (19)$$

where $Re = (\rho d_c w / \mu)$, $Sc = (\mu_L / \rho D)$,

$$\varphi_1 = (\mu_L / 1.005)^{0.16}, \quad \varphi = (1 / \rho_L)^{1.25} \quad \varphi = (72.8 / \sigma)^{0.8},$$

D is diffusivity, μ is viscosity, and ρ is density for the liquid and gas phase were determined by equations for low pressure (see Appendix A2,A3 and A4), B_1 , B_2 , C_1 , C_2 are constants and Φ , ψ and C' are constants dependent of flow rate.

For HETP Murch, Savkovic-Stevanovic et.al. and Hands-Witte correlations were examined.

Murch [19]:

$$HETP = k_1 L^{k_2} D^{k_3} H^{1/3} (a \mu / \rho) \quad (20)$$

Savkovic-Stevanovic [11]:

$$HETP = L^{b_1} d_c^{b_2} H^{1/3} (k a \mu \rho)^{b_3} \quad (21)$$

Savkovic-Stevanovic -Simonovic-Popovic [20]:

$$HETP = \exp \{ a_1 + a_2 (\ln L) + a_3 (\ln L)^2 \} \quad (22)$$

Hands-Witt [21]

$$HETP = b(d_p \mu / L)^{0.5}, \quad b = \text{constant} \quad (23)$$

where μ is viscosity, and ρ is density for the liquid and gas phase were determined by equations for low pressure. α is relative volatility and k is specific chemical constant, k_1 , k_2 and k_3 and b_1 , b_2 and b_3 are constants.

Diffusivities D were calculated according to the equations Fuller et. al for the gas phase and Leffler Cullinan [23-24] and Savkovic-Stevanovic for the liquid phase [11,25].

Sherwood-Hollway models were tested in distillation packed column with the acetic acid-benzene, acetic acid-toluene and acetic acid-ethanol-ethyl-acetate-water systems. Also, Sherwood-Hollway correlations were tested in the packed absorber with Rashig rings with ammonia-water system. The fitted parameters are shown in Table 1.

Cornell, Knapp and Fair correlations were tested with acetic acid-benzene and acetic acid-toluene in distillation column which packed with metallic rings of 3mm diameter with inner column diameter 30mm. The fitted parameters for this systems are shown in Table1.

Zuiderweg correlations were tested in distillation packed column with the acetic acid-benzene, acetic acid-toluene and acetic acid-ethanol-ethyl-acetate-water systems. The fitted parameters are shown in Table 1.

Table 1 Parameters of correlation models for HTU

System	Sherwood-Hollway [14]	Zuiderweg [17]	Cornell-Knapp-Fair [15-16]
Acetic acid -benzene	$B_1=0.02$, $n=0.25$ $B_2=7.66$	$C_1=0.54E-4$ $C_2=0.49E-4$	$C'=f(\text{flow})$ $\Phi=f(\text{flow})$ $\Psi=f(\text{flow})$
Acetic acid-toluene	$B_1=0.38$ $n=0.25$ $B_2=17.61$	$C_1=0.20E-2$ $C_2=0.91E-6$	$C'=f(\text{flow})$ $\Phi=f(\text{flow})$ $\Psi=f(\text{flow})$
Ammonia -water	$B_1=0.0090$ $n=0.25$ $B_2=0.0772$	-	-
Acetic acid-ethanol-ethyl-acetate-water	$B_1=3.45$ $n=0.25$ $B_2=0.08$	$C_1=0.08$ $C_2=95.0$	-

5. Parameters determination

The parameters of the investigated correlation models were obtained by minimization method based on least squares method.

The objective functions were defined as:

$$F_G = [(HTU)_{OGexp.} - (HTU)_{OGcal.}]^2 = \min \quad (24)$$

$$F_L = [(HTU)_{OLexp.} - (HTU)_{OLcal.}]^2 = \min \quad (25)$$

$$F = [(HETP}]_{exp} - (HETP)_{cal} = \min \quad (26)$$

The obtained parameters have shown in Table 1 and Table 2.

6. Results and discussion

The comparison of experimentally determined and calculated values of HTU for the benzene -acetic acid and toluene -acetic acid was provided.

The correlation models of Cornell et. al. produced a good agreement between experimental and calculated values of the HTU with a mean deviation of 5.73 for acetic acid benzene and 13.05% for acetic acid- toluene system.

The comparison of experimental and the calculated values of $(HTU)_{OG}$ by the Sherwood-Hollway model showed a mean deviation of 18.84% for the acetic acid benzene and 22%

for the acetic acid- toluene system. The correlation model of Zuiderweg for overall HTU values gave a mean deviation of 19.12% for acetic acid-benzene and 26.63% for acetic acid- toluene system.

For the acetic acid-ethanol-ethyl-acetate-water system Sherwood-Holloway models gives goods agreement between experimentally determined and calculated values for $(HTU)_{OG}$ with an average deviation of 5.0% and $(HTU)_{OL}$ with an average deviation of 11.83%.

The correlation models of Zuiderweg gives goods agreement between experimentally determined and calculated values for $(HTU)_{OG}$ with an average deviation of 5.5% and $(HTU)_{OL}$ with an average deviation of 15.73%.

The correlation model of Murch gives good agreement between experimentally determined and calculated values for HETP with an average deviation of 7.01% for the acetic acid -benzene and 0.24% for the acetic acid -toluene.

The model suggested Savkovic-Stevanovic shows the best agreement between experimentally determined and calculated values of the HETP with an average deviation of 6.53% for the acetic acid-benzene and 0.11% for the acetic acid-toluene.

The correlation model of Hands-Whitt gives unsatisfied agreement between experimentally determined and calculated values for HETP with an average deviation 17.25% for the acetic acid -benzene and 7.13% for the acetic acid -toluene systems.

The correlation model of Savkovic-Stevanovic-Simonovic-Popovic has shown good agreement between experimentally determined and calculated values for HETP with an average deviation 7.25% for the acetic acid -benzene and 7.1% for the acetic acid - toluene systems.

For benzene-carbon-tetra-chloride system correlation model of Murch gives good agreement between experimentally determined and calculated values for HETP with an average deviation of 4.75%.

The correlation model of Hands-Whitt showed agreement between experimentally determined and calculated values for HETP with an average deviation of 18.5% for the benzene carbon -tetra-chloride system.

Table 2 Parameters of correlation models for HETP

Model	Acetic-acid benzene	Acetic acid - toluene	Benzene-carbon-tetra-chloride
Murch ^[18]	$k_1=0.0570$ $k_2=0.0570$ $k_3=0.0570$	$k_1=1.7085$ $k_2=-0.103$ $k_3=1.8235$	$k_1=2.6300$ $k_2=-0.0740$ $k_3=1.8235$
Hands-Witt ^[20]	-	-	$b=59.55$
Savkovic-Stevanovic-Simonovic-Popovic ^[19]	$a_1=0.3353E2$ $a_2=-0.1057E2$ $a_3=0.9150$	$a_1=0.4129E1$ $a_2=0.1030$ $a_3=0.0000$	$a_1=-0.29E2$ $a_2=0.03E2$ $a_3=-0.977$
Savkovic-Stevanovic ^[11]	$a_1=0.3172$ $a_2=0.0601$ $a_3=0.2217$	$a_1=-0.1030$ $a_2=0.6064$ $a_3=0.4478$	-

The correlation model of Savkovic-Stevanovic-Simonovic-Popovic gives good agreement between experimentally determined and calculated values for HETP with an average deviation of 3.6%.

Fig.2 shows the calculated overall height of transfer unit according to Sherwood-Holloway correlation model for the acetic acid-ethanol-ethyl-acetate-water system. Fig. 3 shows individual calculated values height of transfer units for the liquid and gas phase according to Sherwood-Holloway correlation model for the acetic acid-ethanol-ethyl-acetate-water

system. Deviations between calculated and experimental determined values $(HTU)_{OL}$ and $(HTU)_{OG}$ have shown in Fig. 4.

Fig.5 shows individual $(HTU)_L$ and $(HTU)_G$ according to Zuideweg correlation model for the acetic acid-ethanol-ethyl-acetate –water system.

In Fig.6 individual $(HTU)_L$ calculated according to Sherwood-Holloway correlation model have shown for the ammonia-water system.

Fig.7 shows $(HTU)_G$ calculated according to Sherwood-Holloway correlation model for the ammonia – water system.

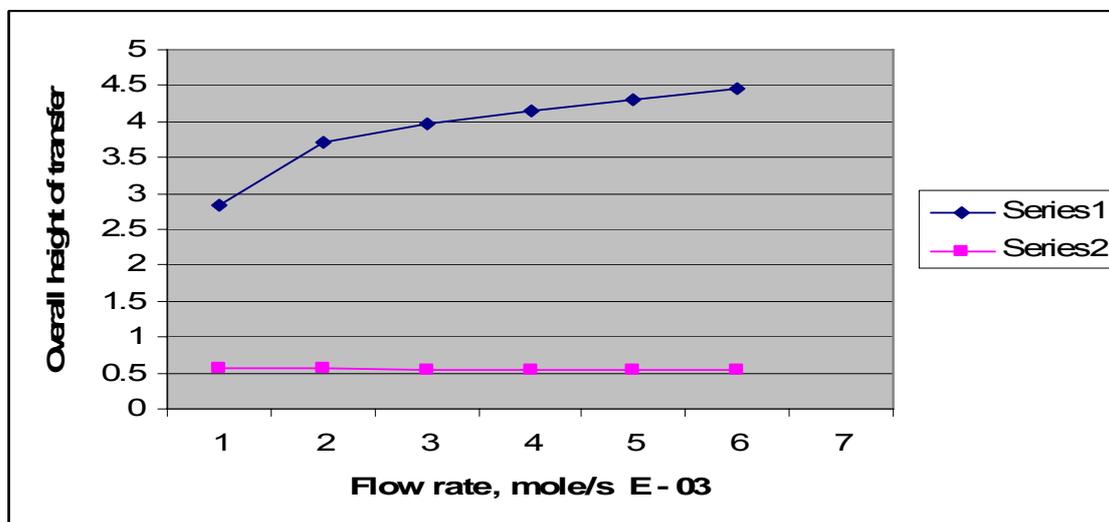


Fig. 2. $(HTU)_{OL}$ and $(HTU)_{OG}$ (cm) of the ethyl-acetate vs. flow rate for the acetic acid-ethanol-ethyl-acetate –water system according to Sherwood –Holloway correlation model (laboratory column with height of 1m and 33mm diameter packed with ion exchanger 0.8mm diameter) at the atmospheric pressure.

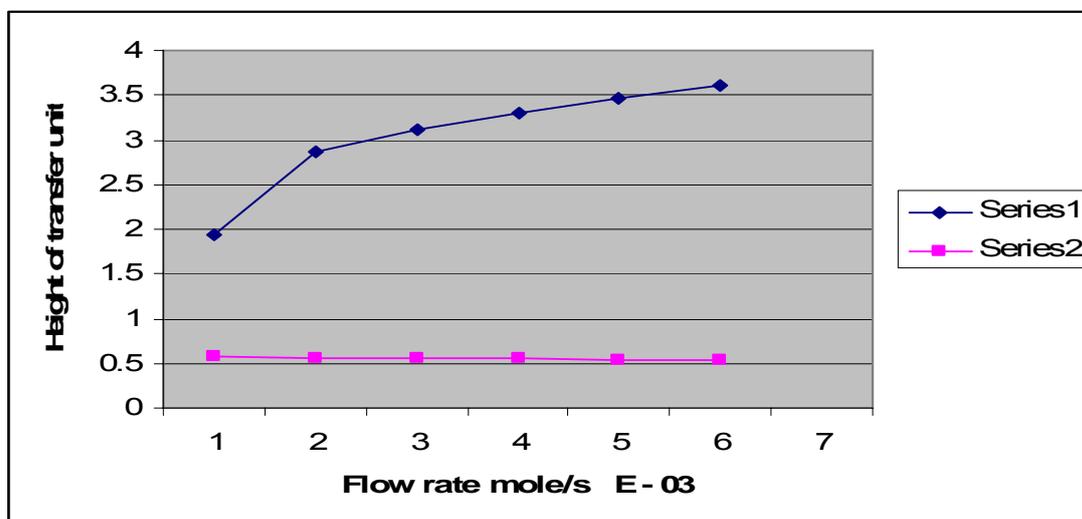


Fig. 3 $(HTU)_L$ and $(HTU)_G$ (cm) of the ethyl-acetate vs. flow rate for the acetic acid-ethanol-ethyl-acetate system according to Sherwood –Holloway correlation model (laboratory column with height of 1m and 33mm diameter packed with ion exchanger 0.8mm diameter) at the atmospheric pressure

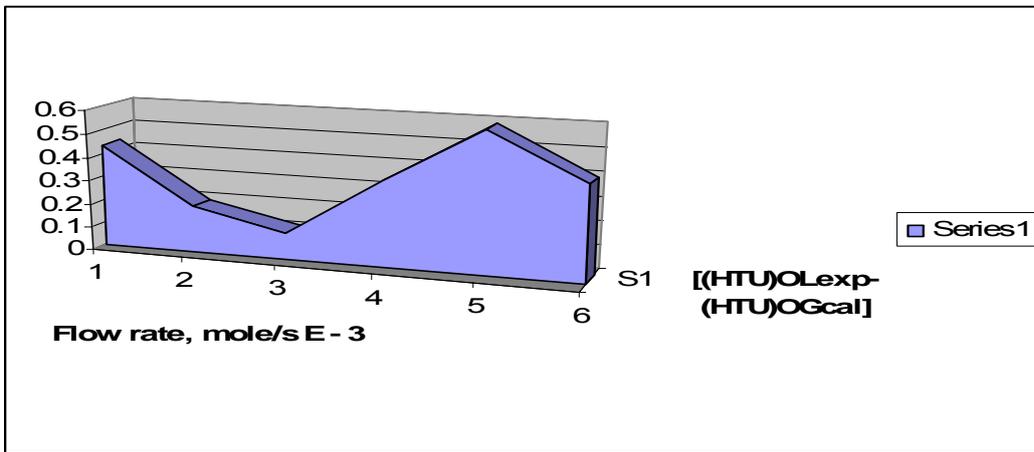


Fig.4. Deviation of the calculated by Sherwood -Holloway correlation model and experimental values of the $(HTU)_{OG}$ of the ethyl-acetate for the acetic acid-ethanol-ethyl-acetate -water system (laboratory column with height of 1m and 33mm diameter packed with ion exchanger 0.8mm diameter) at the atmospheric pressure.

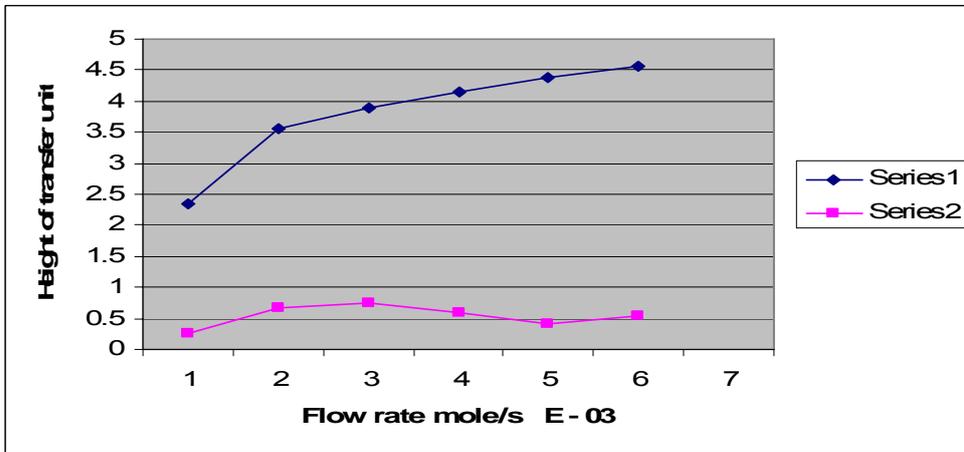


Fig.5 $(HTU)_L$ and $(HTU)_G$ (cm) of the ethyl-acetate vs. flow rate for the acetic acid-ethanol-ethyl-acetate system according to Zuiderweg correlation model (laboratory column with height of 1m and 33mm diameter packed with ion exchanger 0.8mm diameter) at the atmospheric pressure.

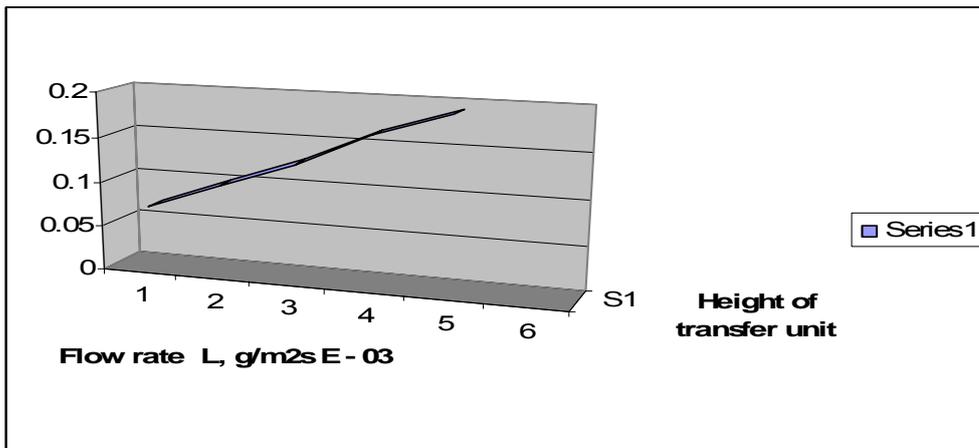


Fig.6 $(HTU)_L$ vs. (m) flow rate for the ammonia-water system calculated according to Sherwood -Holloway correlation model (industrial column with height $H=2308.8$ m and diameter 1827.8 mm with height of packed with metallic Rashig rings 49.0 x 49.0 x 5.0 mm) at the pressure $P=1.05$ bar.

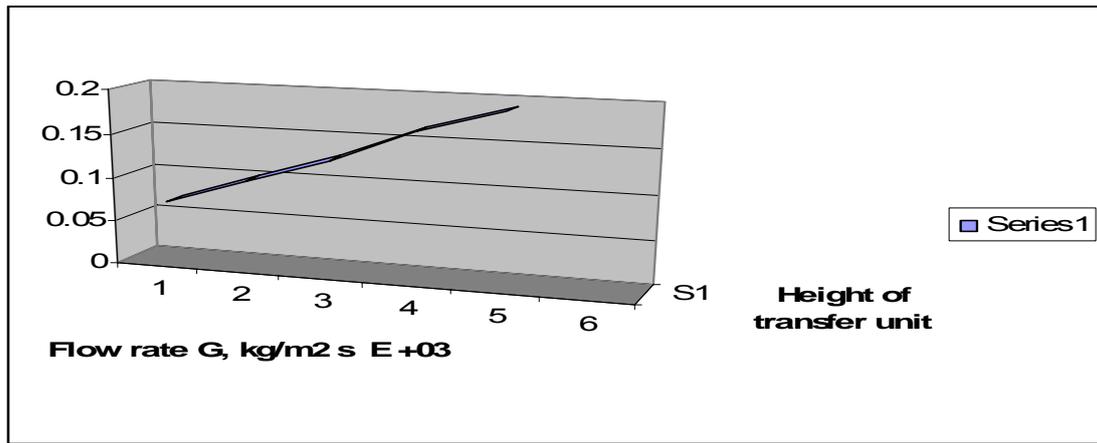


Fig. 7 $(HTU)_G$ vs. flow rate for the ammonia-water system calculated according to Sherwood Holloway correlation model (industrial column with height $H=2308.8$ m and diameter 1827.8 mm packed with metallic Rashig rings 49.0 x 49.0 x 5.0 mm) at the pressure $P=1.05$ bar

7. Conclusion

Several correlation models for HTU/ NTU have been examined. Also, several model for HETP were investigated. Correlation models for overall, individual and component transfer will be tested.

The correlation models of Cornell, Knapp and Fair gave the best agreement between experimental and calculated values of HTU for investigated systems the acetic acid-benzene and acetic acid-toluene.

The Zuiderweg correlations were proven inadequate for the both investigated systems acetic acid-benzene and acetic acid - toluene.

Sherwood-Holloway model have shown satisfied deviation from experimental data in packed column for the both investigated systems acetic acid-benzene and acetic acid-toluene. For ammonia-water this model has shown adequate.

For the acetic acid-ethanol-ethyl-acetate-water system Sherwood-Holloway model have shown better results that Zuiderweg models. The both of investigated models have given satisfied agreement with the experimental results.

The results of investigations showed that the HETP values determined from experiments are in a good agreement with the calculated ones.

The correlation model of Savkovic-Stevanovic showed the best agreement between experimental and calculated HETP values for the both investigated systems acetic acid-benzene and acetic acid-toluene.

The correlation model of Murch gives good agreement between experimentally determined and calculated values for HETP for the both acetic acid -benzene and acetic acid -toluene systems.

The correlation model of Hands-Whitt gives an unsatisfied average deviation for packed column for the all investigated systems the acetic acid -benzene, acetic acid-toluene and benzene -tetra-chloride systems.

The correlation model of Savkovic-Stevanovic-Simonovic-Popovic gives good results with satisfied average deviation for packed column.

The obtained results have shown efficiencies of substance transfer by HTU and HETP methods and their using for tower design. These results can be used for packing efficiency examination, too. The results can be applied in the others domain.

Acknowledgements

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Notation

a - interfacial surface per unit volume, cm^2/cm^3

a_t -total surface area of packing per unit volume, cm^2/cm^3

B_1, B_2 , b -constants

C_1, C', C_2 -constants

D -fluid diffusivity, cm^2/s

D_L, D_G -diffusivity of the liquid and gas phase, respectively, cm^2/s
 D_c -column diameter, mm
 d_p -diameter of packing, mm
 f_1, f_2 -functions
 G -gas flow rate, mole/s
 H -column height, m
 h - enthalpy of the vapour phase, J/mole
 HETP - Height equivalent of theoretical plate, cm
 HTU - Height of transfer unit, cm
 $(\text{HTU})_L, (\text{HTU})_G$ - height of transfer unit over the liquid and gas phase, respectively, cm
 K - equilibrium constant
 k -specific chemical reaction constant, $\text{l mol}^{-1} \text{s}^{-1}$
 k_L, k_G , individual transfer coefficient for the liquid and the gas phase, respectively, $\text{mol}/\text{cm}^3 \text{s}$
 k_{OG}, k_{OL} , overall substance transfer coefficient, $\text{mol}/\text{cm}^3 \text{s}$
 L -liquid flow rate, mole/s
 N_1 -total number of component
 N - total number of molecules
 N_t -number of theoretical plate
 n -constant
 NTU -Number of transfer unit
 R_G -gas constant
 Re - Reynolds number ($= \rho d_c w / \mu$)
 Sc - Schmidt number ($= \mu / rD$)
 Sh - Sherwood number ($= kd_c / D$)
 S -radial column surface, cm^2
 S_p - packing surface, cm^2
 T -temperature, K
 y - vapor composition
 y^* - equilibrium composition
 x -liquid phase composition
 V -volume, cm^3
 W -gas phase velocity, cm/s

Greek Symbols

α - relative volatility
 $\gamma_{XY}(\tau)$ - correlation coefficient
 γ - activity coefficient
 μ - fluid viscosity, Pas
 μ_X - mean values of quantity of X
 μ_Y - mean values of quantity of X
 μ_L - viscosity of the liquid phase, Pas
 μ_G - viscosity of the gas phase, Pas
 Φ -factor
 Ψ -factor
 Φ_1, Φ_2, Φ_3 - factors
 ρ -density, g/cm^3
 σ - surface tension, N/m

Subscript

c -column
 G -gas
 L -liquid
 OG -overall liquid phase
 OL -overall gas phase
 p -packing

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Appendixes

A1. Correlation

Scientists very often need to estimate is there any connection between two quantity, qualitative and if possible quantitative. If common probability distribution for two quantities is known then can be calculated some measure linear dependence between them, which called correlation coefficient. At this it is the same for dependent and independent variables. Covariation function of two quantities $X(t)$ and $Y(t)$ is called a function [7] :

$$\begin{aligned}\sigma_{XY}(t_1, t_2) &= \mathcal{E}\{[X(t_1) - \mu_X(t_1)][Y(t_2) - \mu_Y(t_2)]\} = \\ &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} (x - \mu_X)(y - \mu_Y) p(x, y; t_1, t_2) dx dy\end{aligned}\quad (\text{A1})$$

For steady state assembly covariation:

$$\sigma_{XY}(\tau) = r_{XY}(\tau) - \mu_X \mu_Y \quad (\text{A2})$$

Since, covariation quantity depends from quantities of X and Y two undimensional quantities can be introduced.

$$\frac{X - \mu_X}{\sigma_X(0)} \quad \text{i} \quad \frac{Y - \mu_Y}{\sigma_Y(0)} \quad (\text{A3})$$

where argument (0) denotes $t=0$. Correlation coefficients for steady state assembly is called covariation of two undimensional quantities:

$$\gamma_{XY}(\tau) = \frac{\sigma_{XY}(\tau)}{\sigma_X(0) \cdot \sigma_Y(0)} \quad (\text{A4})$$

For non-correlated X and Y covariation and correlation coefficient are equal zero. In oposite statement is not accurate. For example, two stochastic quantities which have Gauss normal distribution probability law can be uncorrelated but dependent from each other [7].

Correlation coefficients allow to estimate linear measure between two quantities by one number. Positive correlation means that σ_{XY} , mean square deviation allways positive, and negative value σ_{XY} means that large value of one quantity has connected with little value of the other.

A2. Diffusion coefficients in infinite dilution liquid

Diffusivities D_{L12}^0 and D_{L21}^0 were calculated by the equation of Lusi-Ratchlif [23-24].

$$\frac{D_{L12}^0 \mu_2}{T} = [8.52 \cdot 10^{-8} v_2^{(-1/3)} (1.40 \left(\frac{v_2}{v_1}\right)^{(1/3)} + \frac{v_1}{v_2})] \quad (\text{A5})$$

Diffusivities D_L were calculated by the equation Leffler- Cullinan [23-24].

$$D_{L,ij} = (D_{L,ij}^0 \mu_{ij})^{x_j} (D_{L,ji}^0 \mu_{ji})^{x_i} \left(1 + \frac{\partial \ln \gamma_i}{\partial x_i}\right) \quad (\text{A6})$$

and Savkovic-Stevanovic [25]:

$$\begin{aligned}D_{L,ij} &= (D_{L,ij}^0 \mu_{ij})^{x_j} (D_{L,ji}^0 \mu_{ji})^{x_i} \prod_{k \neq i, j, l \neq i, j, k}^{N_i} \\ &[(D_{L,ik} \mu_{ik} D_{L,jk} \mu_{jk})^{x_k/2}] \\ &[(D_{L,il} \mu_{il} D_{L,jl} \mu_{jl})^{x_l/2}]\left(1 + \frac{\partial \ln \gamma_i}{\partial x_i}\right)\end{aligned}\quad (\text{A7})$$

The activity coefficients γ for the liquid mixture was determined from experimental data.

A3. Diffusion coefficients of the gas phase

$$D_G = 0.001007^{7/4} P (v_i^{1/3} + v_j^{1/3}) \sqrt{\left(\frac{1}{M_i} + \frac{1}{M_j}\right)} \quad (\text{A2.1}) \quad (\text{A8})$$

where v is mole fraction and T is temperature.

A4. Viscosity and density

Viscosity μ of the binary mixtures at low pressure was determined by the following equation:

$$\mu_L (=) x_i \mu_i^{1/3} + x_j \mu_j^{1/3} \quad (\text{A9})$$

Density ρ for the binary mixtures at low pressure was calculated by the following equation:

$$\rho_L = \rho_1 (=) \frac{M_{mix}}{\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2}} \quad (\text{A10})$$

$$\rho_L (=) \rho_{ij} = \frac{M_{mix}}{\frac{x_i M_i}{\rho_i} + \frac{x_j M_j}{\rho_j}} \quad (\text{A11})$$