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Large-scale maldistributions of local flow parameters at distillation on a structured packing

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The results of research and operation of industrial distillation columns with the most widely used regular packing have shown that the separation efficiency with increasing diameter can significantly decrease due to formation of large-scale nonuniformity of liquid flows, as well as concentrations along the column section, which significantly affects the performance and purity of the finished product. The experiments were carried out on a large-scale experimental setup, designed to study the integral and local characteristics of mixture separation by distillation on a structured packing. To simulate the process of separation of liquefied air, a mixture of *R*114/*R*21 freons was used. The separation of mixture was studied on a structured package Mellapack 350.Y with a diameter of 0.9 m and height of 2100 mm. We studied the efficiency of mixture separation, dynamics of formation of large-scale temperature field non-uniformity over the column cross-section during mixture separation, and distribution of the density of local liquid flow rates at the packing outlet. The experiments were carried out at a pressure of 0.3 MPa and initial concentration of the *R*114 / *R*21 mixture of 12%. The conducted experimental studies showed that large-scale temperature maldistribution over the cross-section and height of the distillation column is formed. The conditions leading to formation of this nonuniformity are considered. The obtained experimental data will be used in the development of new approaches for numerical simulation of mass transfer and mixture separation efficiency in distillation columns with structured packing in the framework of a joint project.

Keywords: distillation columns, structured packing, numerical simulation, large-scale nonuniformity

INTRODUCTION

Distillation columns are widely used to separate two- and multicomponent mixtures in the chemical industry [1-4]. Mixtures are separated during heat and mass transfer under conditions of countercurrent motion of vapor and liquid phases using contact devices of various types [5, 6]. In accordance with the laws of thermodynamic vapor-liquid equilibrium, the vapor phase is enriched with low-boiling and the liquid phase is enriched with high-boiling components. The main requirement for contact devices is to ensure uniform distribution of the liquid film and countercurrent vapor flow over the column cross-section, as well as the maximum development of the liquid-vapor contact surface with a minimum pressure drop on the contact devices. Therefore, contact devices determine to a large extent the total efficiency of the separation process. The heat and mass transfer plates, all kinds of filling elements, as well as regular structured packing are used as contact devices [7]. Moreover, the columns with a structured packing have higher efficiency, wide range of stable operation, relatively low cost and simplicity of design, as well as low hydraulic resistance [8]. Currently, the diameter of widely used industrial distillation columns with regular packing can reach 7 m or more. However, the efficiency of mixture separation with increasing diameter can be significantly reduced. One of the reasons for this is the formation of large-scale maldistribution of liquid and vapor flows, as well as both phase concentrations over the column cross-section, which significantly affects the productivity and purity of the finished product.

The experimental results on the formation dynamics of large-scale temperature field maldistribution over the cross-section and height of a distillation column with a regular packing under conditions of negative stratification of the countercurrent vapor flow are presented in this paper.

EXPERIMENTAL

The experiments were carried out at a large-scale research setup [9], made to study hydrodynamics and heat and mass transfer in large-scale separation columns during distillation on structured packing. Separation of liquefied air to obtain pure cryogenic products (oxygen, nitrogen, argon, etc.) is a very significant aspect of industrial distillation. However, the studies in large-scale cryogenic columns are extremely complex and expensive.

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Therefore, it is justified to use modeling mixtures for a comprehensive study of the processes of hydrodynamics and heat and mass transfer on large-scale elements of complex geometry composing the packing of modern columns. To perform the studies, we used a mixture of freon R114/R21, selected to simulate the process of liquefied air separation. The mixture was separated on a structured Mellapack 350.Y packing with a diameter of 0.9 m and height of 2100 mm (10 layers of the packing with a height of 210 mm) under conditions of full reflux. The mixture poured into the bottom of the column (bottom space) evaporated, vapor passed through a structured packing and turned into the liquid phase in the condenser completely. The packing was irrigated by the jets from a liquid distributor located in the column head. The circulation pump fed liquid from the condenser to the distributor, in the bottom of which there were 83 holes with installed nozzles for packing irrigation. The flow rate of irrigating liquid changed within 0.8 – 2.0 l/s. To study the dynamics of formation of large-scale temperature field non-uniformity during mixture separation, the miniature temperature sensors were installed in layers No. 2, 5, and 8, counting from the lower layer, in three layers of the packing cross-section. In each cross-section, 16 sensors were installed on the upper edge of the layer. To study the distribution of local liquid flow density over the packing of a distillation column, a flow meter with a diameter of the receiving collector of 28 mm was used. The flow meter was mounted on a two-coordinate device located directly below the bottom layer of the structured packing.

RESULTS AND DISCUSSION

The experiments were carried out at a pressure of 0.3 MPa and initial concentration of the R114/R21 mixture of 12%. The vapor velocity, averaged over the column cross-section, was 0.15 – 0.3 m/s (F-factor 0.63 – 1.26 $(Pa)^{0.5}$). The data on temperature changes in the middle cross-section of the packing in the process of column transition to the stationary regime of mixture separation are shown in Fig. 1.

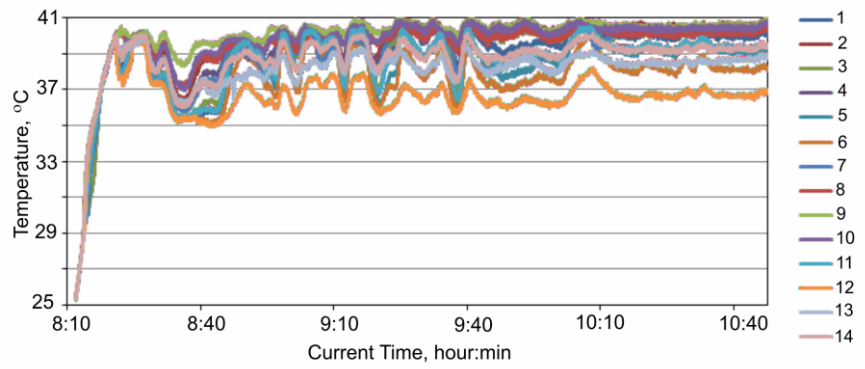

Fig. 1. The temperature change in the middle cross-section of the packing in the process of column transition to the stationary regime. F-factor = 0.63 $(Pa)^{0.5}$; liquid flow rate = 0.85 l/s.

As it can be seen in the diagram that at the initial stage, the temperature field over the packing cross-section is quite uniform. In the process of mixture separation, when the temperature in the packing cross-section is about 39-40°C, the readings of thermometers within one packing cross-section start stratifying substantially. Within one and a half hour, the operating parameters of the column become stable, and the column enters the stationary operation regime. The diagram shows that in the process of establishing a stationary regime, the temperature field in the column cross-section undergoes significant changes, but in general the character of non-uniformity is kept. In the stationary regime of column operation, the temperature field in the column cross-section remains almost unchanged. The diagrams of spatial distribution of the temperature field in the middle (Fig. 2a) and lower (Fig. 2b) cross-sections of the column are presented in Fig. 2.

This diagram illustrates the large-scale temperature maldistribution formed in the packing cross-section. The volatile component of the mixture (*R*114) has a higher density than the high-boiling component (*R*21). As a result, during mixture separation in the upper part of the column, the vapor density in the upward flow becomes higher than its density in the lower part of the column. Negative stratification of the vapor density in the upward flow is formed.

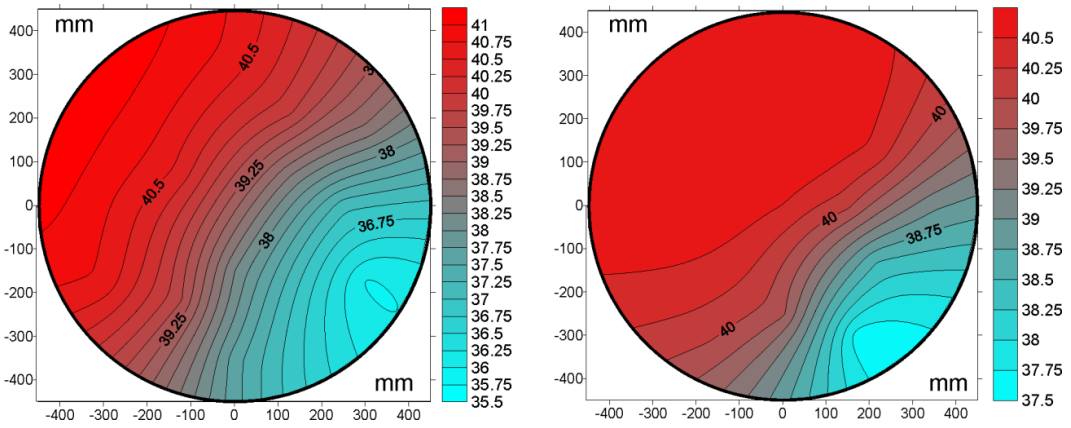

Fig. 2. Diagram of spatial distribution of the temperature fields in the column cross-sections: a) middle cross-section, layer 5; b) lower cross-section, layer 2. F-factor = 0.63 $(Pa)^{0.5}$; liquid flow rate = 0.85 l/s.

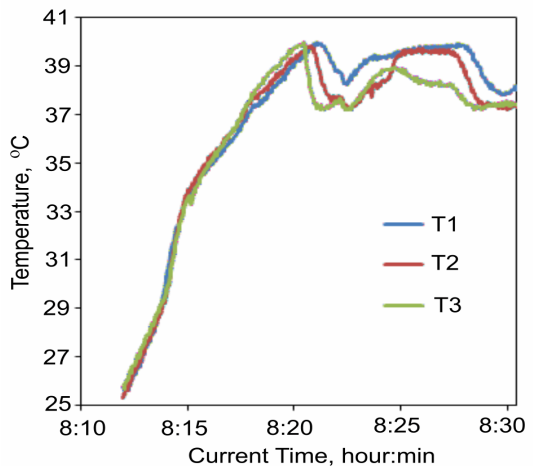

Fig. 3. Temperature changes in different packing layers during stabilization of column operation. T1 – layer 2, T2 – layer 5, T3 – layer 8. F-factor = 0.63 $(Pa)^{0.5}$; liquid flow rate = 0.85 l/s.

As a result, when reaching the critical values of the vapor density gradient, a loss of stability of the uniform countercurrent flows can occur and a large-scale maldistribution of the local parameters of mixture over the column cross-section may form. The scale of the perturbing effect, which can lead to a loss of stability in the upward vapor flow and formation of a large-scale maldistribution of local flow parameters in the column cross-section, is estimated in [10]. As it can be seen in the diagram, when a temperature close to 40°C is reached, a rapid decrease in temperature to 37°C is observed in the upper part of the packing (T3 thermometer). Relative to the top part of the packing (T3), the temperature drop in the middle part (T2) is delayed for 30 s. At the packing bottom (T1), the temperature drop is delayed for 72 s. The perturbation spread along the packing height at a velocity of 0.015 - 0.021 m/s. At that, the average velocity of the vapor flow over the column cross-section was 0.15 m/s.

Such a low velocity of perturbation propagation may indicate in favor of another formation mechanism of large-scale non-uniformity. It is observed in the packed columns that a significant portion of liquid flows down the column wall (up to ~ 8% or more of the total flow rate in the column, [11]), but not along the elements of the structured packing. Hydrodynamics of a two-phase mixture in a structured packing (a falling liquid film in contact with an upward vapor flow) depends significantly on the stress tensor [12], i.e., the tangential components of the stress tensors in two phases should be equal at the phase boundary:

$$\mu_G \frac{\partial u_G}{\partial y} = \mu_L \frac{\partial u_L}{\partial y}, \qquad (1)$$

where u_G, u_L, μ_G, μ_L are velocities and viscosities in the vapor and liquid phases, and the coordinate is directed along the normal to the interface. It can be derived from (1):

$$\frac{\mu_G \bar{u}_G}{\mu_L \bar{u}_L} \frac{\partial U_G}{\partial y} = \frac{\partial U_L}{\partial y}, \qquad (2)$$

where $\bar{u}_G, \bar{u}_L, U_G, U_L$ are average and dimensionless velocities in the vapor and liquid phases. For the industrial columns, the ratios of values in (2) are of the following order:

$$\frac{\mu_G}{\mu_L} \sim 10^{-4}, \frac{\bar{u}_G}{\bar{u}_L} \sim 10, \frac{\mu_G \bar{u}_G}{\mu_L \bar{u}_L} \sim 10^{-3}, \qquad (3)$$

i.e., under these conditions (below the loading regime, which develops at higher vapor velocities), the motion of the vapor phase does not significantly affect the motion of the liquid phase, but the liquid film flow affects the motion of the vapor phase.

When modeling hydrodynamic and heat and mass transfer processes during distillation in packed columns, a problem arises because of the need to take into account the presence of flowing liquid on the column wall, when there is a main flow of liquid in the volume of the structured packing and a significant liquid flow on the column wall. There are a number of design solutions that help to reduce the probability of liquid flow on the wall and increase the probability of liquid return from the wall to the nozzle (the character of initial irrigation, specific channel shapes, the presence of wipers and distance spacers), but these measures cannot eliminate the liquid flow on the column wall completely.

Liquid flows over the packing surface, and when it reaches the column wall, its most part flows on this surface since this part of liquid cannot return completely to the structured packing due to the limited contact surface between the column wall and the packing. With an increase in the thickness of the flowing liquid film, conditions for more intensive reverse flow of liquid from the wall to the volume of the packing are created, when these two reciprocal processes can be equalized (strictly speaking, these two processes depend on a gap between the column wall and the structured packing, features of connection and arrangement of sheets in the structured packing, molar flow rates of liquid and vapor, geometry and material of the used wipers and clips fixing the packing, number of packing layers in the column, etc.). In the process of such repeated mutual liquid flows between the packing and the column wall, the liquid film can ultimately reach its constant maximum thickness with which it flows down the column bottom. The amount of liquid entering the film flowing down the column wall causes a decrease in the total amount of liquid in the volume of the structured packing. Thus, the considered factor of the transverse liquid flow creates a significant radial non-uniformity of the axial component of liquid velocity (and non-uniform distribution of the local vapor flow over the packing cross-section, respectively) in the column and finally, can significantly reduce the mass transfer intensity in the liquid and vapor phases and, as a result, efficiency of mixture separation.

The surface of the falling liquid film is the interface between the vapor and liquid phases. For large values of the liquid film thickness (and the average velocity in it, respectively), the velocity of the vapor phase at the interface with the falling liquid can be directed downward, i.e., against the velocity of the bulk of the vapor phase. As a result, the direction of velocity in the vapor phase in a local zone near the boundary of liquid flowing along the column wall can change; as a consequence, this can lead to creation of a large-scale circulation zone. Thus, if the conditions of assembly and installation of the structured packing in the distillation column create the possibility of organizing a significant liquid flow on the column wall, this can lead to the presence of a large-scale zone of vapor circulation, which is observed in the performed experiments.

The diagram of distribution of the local density of liquid flow from the structured packing, averaged over the azimuthal coordinate, along the column radius is shown in Fig. 4.

As it can be seen in the diagram, a slight decrease in the density of liquid flow from the packing center to its edges is observed along the column cross-section.

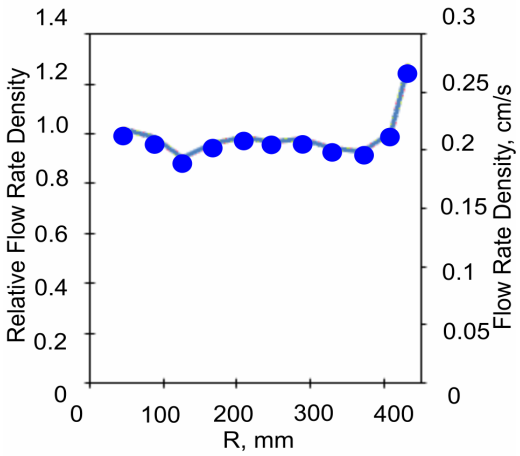

Fig. 4. The diagram of distribution of the local flow density of liquid from the structured packing, averaged over the azimuthal coordinate, along the column radius. F-factor = 1.27 $(Pa)^{0.5}$; liquid flow rate = 1.4 l/s.

However, when approaching the packing periphery, we can see a fairly significant increase in the density of liquid flow rate to approximately ~ 1.25 of the average density of the liquid flow rate in the structured packing. The diagram of distribution of the local liquid flow density in an annular cross-section, bounded by a radius 417 < R < 445 mm, is shown in Fig. 5. The average density of the liquid flow rate at the outlet of the nozzle is 0.22 cm/s.

According to the diagram, the distribution of the liquid flow density at the packing periphery is substantially uneven. The density of the liquid flow is mainly in the range from 0.15 to 0.4 cm/s. The areas of increased flow rate density are observed with a periodicity of 90°. With rotation at such an angle, the layers in the column packing alternate. There is no direct correlation of the position of

large-scale areas of the maximum and minimum temperatures with the flow density at the packing periphery, but this periodicity of the liquid flow density may be the initiator of a large-scale convective process.

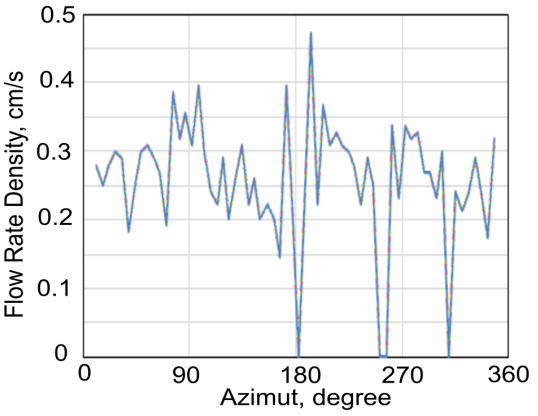

Fig. 5. The diagram of distribution of the local liquid flow density in a near-wall annular cross-section, bounded by a radius 417 < R < 445 mm. F-factor = 1.27 $(Pa)^{0.5}$; liquid flow rate = 1.4 l/s.

CONCLUSIONS

Experimental studies have shown that in the distillation column of a 0.9-m diameter a large-scale temperature maldistribution is formed over the cross-section and height of the column. The velocity of maldistribution propagation along the column height is 0.015 - 0.021 m/s at a countercurrent vapor flow velocity of 0.15 m/s.

At the outlet of the structured packing, a decrease in the local density of liquid flow from the packing center to its edges is observed over the column cross-section. However, when approaching the packing periphery then, a significant increase is observed in the density of the liquid flow rate to approximately ~ 1.25 of the average density of the liquid flow rate in the structured packing.

The results of processing the obtained array of experimental data will serve as the basis for the construction and verification of models aimed at developing methods for calculating the efficiency of mixture separation during distillation on the structured packing in industrial columns, allowing the quantitative consideration of the scale factor influence associated with large-scale maldistribution of local parameters of liquid and vapor flows.

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REFERENCES

1. J. Fang, X. Cheng, Z. Li, H. Li, C. Li, *Chin. J. Chem. Eng.*, **27 (6)**, 1272 (2019).
2. M. Markowski, K. Urbaniec, *Proc. Syst. Eng.: Energy Systems Eng.*, **5**, 301 (2008).
3. M. Elhelw, A. Abdurrahman, A. Alsanousie, A. Attia, *Alexandria Eng. J.*, **59 (2)**, 613 (2020).
4. J. Fang, Zh. Li, G. Huang, H. Li, Ch. Li, *Ind. Eng. Chem. Res.*, **59 (4)**, 1668 (2020).
5. S. K. Churakova. *Bashkir Chem. J.*, **18 (2)**, 39 (2011).
6. V. Bessoua, D. Rouzineau, M. Prévost, F. Abbé, Ch. Dumont, J. - P. Maumus, M. Meyer, *Chem. Eng. Science*, **65 (16)**, 4855 (2010).
7. R. Battisti, R.A.F. Machado, C. Marangoni, *Chem. Eng. & Proc. – Process Intens.*, **150** (2020).
8. A. R. Muzafarova, E. A. Emelyanycheva, *University of Technology Herald*, **19 (2),** 63 (2016).
9. A. N. Pavlenko, V. E. Zhukov, N. I. Pecherkin, V. Yu. Chekhovich, O. A. Volodin, A. Shilkin, C. Grossmann, *AIChE*, **60 (2),** 690 (2014)
10. A. N. Pavlenko, V. E. Zhukov, N. I. Pecherkin, E. Yu. Slesareva, *JET*, **(2)**, 1 (2020).
11. A. N. Pavlenko, N. I. Pecherkin, V. Yu. Chekhovich, V. E. Zhukov, A. F. Serov, A. D. Nazarov, S. Sunder, P. Houghton, *JET*, **13 (1),** 1 (2005).
12. Chr. Boyadjiev, Theoretical Chemical Engineering. Modeling and simulation, *Springer-Verlag, Berlin Heidelberg*, 2010, p. 594.