Multiscale structure of gas-liquid flow and interfacial heat and mass transfer in complex channel systems

V V Kuznetsov$^{1,2}$
$^1$Kutateladze Institute of Thermophysics, Siberian Branch of the Russian Academy of Sciences
1, Academician Lavrentyev Av., Novosibirsk, 630090, Russian Federation
$^2$Novosibirsk State University, 2, Pirogova Str., Novosibirsk, 630090, Russian Federation
E-mail: vladkuz@itp.nsc.ru

Abstract. The paper considers the mechanisms of the development of large-scale instability in counter-current gas-liquid flow in complex channel systems of the structured packing, contributing to deterioration of the separation efficiency for a gas mixture. The paper discusses the contribution of the gravity-induced convective flows and capillary-gravity flooding to the development of the maldistribution of the fluid velocity and concentration fields in the distillation columns filled with Koch 1Y and Mellapak 500Y structured packings. The correlation for prediction of the gas and liquid capacity factors corresponding to the packing flooding is proposed based on the experimental data.

1. Introduction

Recently, the interfacial heat and mass transfer during gas-liquid flow in complex channel systems have been intensively studied. This is due to important applications of these systems in the energy, chemical, and cryogenic industries. The equipment that uses complex channel systems is characterized by the high surface area and enhanced heat and mass transfer. One of the important examples of this equipment is packed column operated in a counter-current gas-liquid mode. Being introduced early in the 20th century, this concept has been developed towards more effective packing design to increase the packing efficiency as well as to reduce the operating costs [1]. Currently, packed columns with regular structured packing are widely used in distillation and absorption processes since they offer the excellent mass transfer performance at a small pressure drop. For the structured packing of various types, the characteristics of the packing efficiency for different operation conditions were studied experimentally, and mass transfer correlations were proposed, for example [2]. However, the application of these correlations is limited by the operation conditions where they were obtained.

The separation of the multicomponent mixture becomes effective if the difference in the components concentration for liquid and gas phases is well organized. Therefore, the occurrence of large-scale concentration maldistribution causes the deterioration in separation efficiency [3]. Experimental studies of counter-current gas-liquid flow in the structured packing [4, 5] showed that liquid accumulates near the gap between packing elements at gas loads above the loading point. This effect causes the limitation for capacity of the distillation column. The liquid accumulation around the zone where two packing elements touch each other was confirmed in [5] measuring the liquid hold-up by a capacitance probe. This results in an increase in pressure drop when the flooding occurs.
Despite a number of published papers, there is currently limited information on the mechanisms of maldistributions appearance for the velocity and concentration fields during gas mixture separation in complex channel systems. In this paper, the mechanisms of the generation of large-scale instability during counter-current gas-liquid flow in complex channel systems of Koch 1Y and Mellapak 500Y structured packing are considered. The conditions when the maldistribution of flow and concentration fields may occur, contributing to the deterioration of the separation efficiency for binary gas mixtures, are discussed.

2. Gravity-induced convection in complex channel systems

The most efficient equipment for cryogenic separation of air is the distillation column with structured packing. The experimental observations [2] show that the limitations on effective gas mixture separation occur both for low and high load factors for gas (vapor) phase. The limitation on separation effectiveness at low load factor for gas usually referred to the appearance of the dry spots on the packing surface. Nevertheless, another mechanism of mass transfer deterioration can be essential if the ratio of liquid and gas fluxes is near the unity. This mechanism is related to the nature of the separation, when the ascending gas flow is enriched by light-boiling component due to counter-current mass transfer between the gas and liquid. Therefore, the negative density gradient in the column with respect to the direction of gravity arises, and the gas density increases towards the top of the column. In this case, the most plausible mechanism for the formation of large-scale concentration maldistribution is the gravity-induced convection.

Let us consider counter-current gas-liquid flow in the column with Koch 1Y structured packing when the separation of a binary mixture of Ar/O$_2$ occurs. The liquid flows downward on the surface of the packing under the action of gravity and the gas rises upwards under intense mass exchange on the vapor-liquid interface. The numerical calculations of the Ar/O$_2$ separation in the column filled with Koch 1Y structure packing, based on one-dimensional non-equilibrium separation model [6], showed that the largest gas density gradient is located near the bottom of the column. The variation of the argon concentration along the column is shown in Fig. 1 (a) for the ratio of liquid and gas mole fluxes $L/V$ equal to unity. Here $z$ is the distance from the column top and $H$ is the height of the column. As seen, the gas density in an upper part of the column is higher than that near the bottom. Therefore the origin of convective flow in the column is possible.

Assuming small variation in the argon concentration $Y$ and gas density along the column it follows:

$$
\rho_{gas}(Y) = \rho_{gas0} + d\rho_{gas}/dY \big|_0 Y',
$$

where $Y = Y_0 + Y'$ is the argon concentration, $\rho(Y)$ is the gas density, index 0 corresponds to the non-disturbed state. Taking into account the notation $\beta_Y = -\partial\rho_{gas}/\partial Y \big|_0 \rho_{gas0}^{-1}$, the steady dimensionless momentum equation with Boussinesq approximation is:

$$
(\mathbf{u}_{gas} \nabla)\mathbf{u}_{gas} = -\frac{\nabla'\rho_{gas}}{\rho_{gas0}} \frac{N_{conv} Y' g}{\operatorname{Re}} + \frac{1}{\operatorname{Re}} \Delta\mathbf{u}_{gas}
$$

Here the dimensionless variables are introduced using the superficial gas velocity $U_{gas}$, the characteristic transverse size $l$ and mole concentration $Y' = l \cdot dY_0/dz$, the argon density $\rho_{gas}$, the gravity acceleration $g$, $\operatorname{Re}$ is the Reynolds number, and $\mathbf{g}$ is the gravity vector. The dimensionless convection number $N_{conv}$ defines the ratio of the gravitational and inertial forces in convective flow as follows:

$$
N_{conv} = \frac{l^2 g \beta_Y dY_0/dz}{U_{gas}^2}
$$

(3)
For development of the convective flow in a column, convection number $N_{\text{conv}}$ should be much higher than unity. The variation of the convection number along the gravity vector for $L/V=1$ is shown in Fig. 1(b). In the calculation, we consider perturbations with the characteristic size $l$ equal to the column diameter $D$. As seen, the convection number considerably exceeds unity if the column diameter exceeds 0.2 meter. Under these conditions, one can expect the appearance of large-scale convective flow and significant maldistribution in the concentration of gas species in the column cross-section that was observed in [2] for the mixture of the refrigerants R21/R114.

3. Capillary-gravitational flooding in complex channel systems

The flooding capacity consists in a determination of the amount of fluid in the packing at which the liquid starts to overflow. In structured packing columns, packing layers usually have thickness equal to 200 mm. Therefore, the reason for the formation of large-scale structures in the counter-current gas-liquid flow is the capillary-gravitational flooding near the gaps between the layers of the packing for high gas loading factors [4].

The conditions for the appearance of capillary-gravity instability were determined experimentally for the multilayer packing of Mellapak 500Y within the range of flow parameters that are practically realized during the distillation. The experiments were performed using the rectangular column with transverse dimensions of 800 and 400 mm, and the packing element thickness of 100 mm. The column is filled with nine layers of the packing and each layer contains nine sheets of the packing. The standard structured packing with a corrugation angle of 45° results in 180° change in the gas flow direction at the transitions between packing elements. The packing layers are moved away from the column walls and fixed through a system of the vipers to prevent the liquid flowing down the walls. During the experiments, distilled water with the additives of surfactant and antifoam is pumped through the heater and feed to the top of the column where the liquid distributor is located. In heater the working fluid archives the temperature of 25-27 °C, the surface tension for this temperature equals to 0.03 N/m. Air from the high pressure system enters the lower part of the column, where the gas distributor is located. The gas distributor includes sieve tray that has 1650 holes with a diameter of 2.2 mm. A vortex flow meter is used to measure the air flow rate. The liquid flow rate is measured by a turbine flow meter, the liquid and gas temperatures are measured at the bottom of the column by insulated thermocouples.

To determine the gas loading factor $C_{\text{gas}}=U_{\text{gas}}/\rho_{\text{gas}}/\left(\rho_{\text{liq}}-\rho_{\text{gas}}\right)$ that corresponds to the formation of capillary-gravity instability, pressure drop measurements were made for uniform irrigation of the upper part of the structured packing at different liquid and gas flow rates. It was obtained that for gas loading factor $C_{\text{gas}}$ higher than 0.066, the pressure drop is stratified depending on the ratio of the liquid and gas flow rates. The high-speed video shows that just before loading point, the progressive flooding of the

![Figure 1. Variation of argon concentration (a) and convective number (b) along the column for $L/V=1$.](image)
lower layer of the packing occurs. When loading point is achieved, the flooded zone extends over the entire column when the gas or liquid loading factor is increased. To measure the gas loading factor at the flooding, friction factor for the gas is used. The dependence of the friction factor on the gas Reynolds number is shown in Fig. 2 for dry gas flow and gas-liquid flow with \( \frac{G_{\text{liq}}}{G_{\text{gas}}} = 0.76 \). The friction factor and Reynolds numbers were calculated as follows: 
\[
f_{\text{fp}} = 2A(p_d/d)_{\text{fp}} \rho_{\text{gas}}/G_{\text{gas}}^2 P, \\
\text{Re}_{\text{gas}} = G_{\text{gas}}D_h/\mu_{\text{gas}}. 
\]
Here \( G \) is the mass flux, \( A \) is the flow open cross-section, \( P \) is the packing perimeter, \( D_h \) is the hydraulic diameter of packing, \( \mu_{\text{gas}} \) is the gas viscosity. As seen, the friction factor shows drastic growth when the flooding arises and the loading point can be easy determined.

To generalize the obtained experimental data, the physical model of the flooding based on the approach proposed in [7] for round tube was used. Taking into account the geometric characteristics of the gas and liquid flows through the channels of the packing and gravity vector direction, one can obtain the Kutateladze-Sorokin coordinates of \( k \) and \( N \):
\[
k = \frac{C_{\text{gas}}}{\varepsilon(\cos \varphi)^{25}(\sigma_{\text{liq}}/(\rho_{\text{liq}} - \rho_{\text{gas}}))^{25}}, \\
N = \frac{\text{We}_{\text{liq}}^{0.5}}{4 \text{Bo}_{\text{liq}}(\cos \varphi)^{0.55}} \left(1 + \frac{31}{\text{Ga}_{\text{liq}}^{0.55}(\cos \varphi)^{0.55}}\right) 
\]
(4)

to find the conditions that provide the weighting of liquid layer by ascending gas flow. Here \( \varphi = \theta/2 \), \( \theta \) is the angle between the channels directions, \( \sigma \) is the surface tension, \( \text{We}_{\text{liq}} = \rho_{\text{liq}}C_{\text{liq}}^2D_h/\sigma \), \( \text{Bo}_{\text{liq}} = \rho_{\text{liq}}8D_h^2/\sigma \), \( C_{\text{liq}} = U_{\text{liq}}/(\rho_{\text{liq}} - \rho_{\text{gas}}) \), \( \text{Ga}_{\text{liq}} \) is the Galilean number, \( \varepsilon \) is the porosity.

The experimental data on the dependence of \( k_{\text{flood}} \), determined by the point of inflection in the friction factor, on \( N \) number is shown as rhombs in Fig. 3 for the columns with width of 800 mm and 400 mm. The water-surfactant solution was used as working fluid in the experiments with Mellapak 500Y structured packing. The experimental data [5, 8] on the beginning of liquid accumulation in water-air experiments with Mellapak 250Y structured packing is also shown in Fig. 3. The solid line shows the result of the calculations according to the equation:
\[
k_{\text{flood}} = 0.168 \cdot N^{-0.26} \tag{5}
\]

**Figure 2.** Dependence of friction factor on Reynolds number for air and air-water+ surfactant flows for \( \frac{G_{\text{liq}}}{G_{\text{gas}}} = 0.76 \).

**Figure 3.** Dependence of \( k_{\text{flood}} \) on \( N \) number for Mellapak 500Y and 250Y. Lines are the prediction according to Eqs. (5) and (6).
As seen, the calculations agree well with the experimental data if the exponent in Eq. (5) is somewhat different from original exponent for round tube [7]. The equation with original exponent can be presented as follows:

$$k_{\text{flood}} = 0.095 \cdot N^{-0.347}$$  \hspace{1cm} (6)

One can see that the calculations according to Eq. (6) also can be used for the flooding prediction.

The processing of the experimental data showed that the loading point can be predicted by the following equation, accounting that the Galilean number is much smaller than unity:

$$C_{\text{gas,flood}} = 0.168 \cdot \varepsilon \left( \cos \phi \right)^{1.25} \sqrt{\frac{\sigma_g}{\rho_{\text{liq}} - \rho_{\text{gas}}}} \left( \frac{We_{\text{liq}}^{0.5}}{4Bo_{\text{liq}} \cos \phi} \right)^{-0.26}$$  \hspace{1cm} (7)

It is important to note that proposed equation contains physical properties of the gas and liquid, and can be used to predict the capillary-gravitational flooding for different fluids.

4. Conclusions
The presented results make it possible to quantify the necessary conditions for the development of large-scale instability in counter-current gas-liquid flow in complex channel systems of the structured packing, contributing to deterioration of the separation efficiency for distillation columns. It was shown that the considerable maldistribution of the velocity and concentration fields within the range of flow parameters practically realized in distillation arises due to appearance of the gravity-driven convective flows and capillary-gravity flooding for Koch 1Y and Mellapak 500Y structured packings. The equation for prediction of the gas and liquid capacity factors at flooding in the channels of structured packing is proposed based on the experimental data. It is important that proposed equation contains physical properties of the gas and liquid and can be used to predict the capillary-gravitational flooding for different fluids. The results presented in this paper could be applied to predict the mass transfer deterioration in the packed columns.

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