Features of liquid mixtures separation in large-scale distillation columns with structured packing. New ideas and approaches

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Abstract. Negative vapor stratification along the height of distillation column caused by different density of vapor mixture components and higher temperature at the column bottom, leads to formation of large-scale maldistribution of temperature and mixture composition over the column cross-section even at uniform irrigation of the structured packing. Experimental results concerning the dynamic effect of packing irrigation on separation efficiency of the two-component mixture of R-21 and R-114 are presented in this paper. The structured packing Zulser 350Y was installed in the distillation column with the diameter of 0.9 m. Experiments were carried out on the 10- and 19-layer packing with an overall height of 2.1 and 4 m, respectively. The liquid distributor with independently controlled 126 valves for each irrigation point, developed by the authors, was used for packing irrigation. The experiments showed that the periodic impact of the irrigation system on the large-scale non-uniformity of mixture composition, formed in the packing, could significantly affect the distribution of flow parameters over the cross-section and height of the mass transfer unit. Essentially non-uniform periodic irrigation of the packing can improve the separation efficiency of the column within 20%, if the switching periods are comparable with the times of formation of large-scale non-uniformity.

1. Introduction
Distillation columns are widely used by petroleum, chemical and food industries to separate the mixtures of various liquids. The basic requirement for the effective operation of distillation column is uniform distribution of the liquid film over the packing and the counter-current vapor flow over the column cross-section. To achieve this, the packing of metal sheets with deposited structures of different scales and highly permeable porous packing were designed [1]. In recent years, in order to increase the production capacity and improve the separation efficiency at distillation, the scholars have performed a lot of study on the structured packing [2–10]. These researches were mainly focused on the improvement of packing structure, special treatment of packing surface and organization of the optimal structure of irrigation points in the liquid distributor. The use of the modern structured packing allows more uniform distribution of a liquid film over the packing surface, but it does not completely solve the problem of uniform distribution of the flows along the entire height of the column.

As it is mentioned in [4], negative vapor stratification along the packing height caused by different densities of vapor mixture components and higher temperature at the column bottom can lead to formation of large-scale maldistribution of temperature and mixture compositions over the column cross-section even at uniform packing irrigation by liquid. In our experiments, we have implemented the idea of non-stationary use of the liquid distributor for destruction of such stationary large-scale
non-uniformities of local liquid and vapor flow rates, and mixture composition over the column cross-section.

2. Set-up and method description

The experiments were carried out on the large-scale research setup “Large Freon Column”, described in detail in [2-4]. The structured packing Zulser 350Y was installed in the distillation column with the diameter of 0.9 m. The experiments were performed on the 10- and 19-layer packing with the overall height of 2.1 and 4.0 m, respectively. The mixture of Freon R-21 and R-114 was used as the working liquid, chosen by its characteristics to model the separation of cryogenic mixtures. The liquid distributor specially designed by the authors was used for packing irrigation. This distributor included 126 electromagnetic valves overlapping the holes of 5-mm diameter. Each valve was controlled independently by a PC using specially developed software allowing manual control of each valve, control of an arbitrary group of valves and programmed valve switching by any predetermined algorithm with time discretization of 1 s. A response of the column to the influence of liquid distributor was observed in real time by indications of three groups of thermometers mounted in 3 different cross-sections of the column along the packing height. Each group consisted of 16 thermometers uniformly distributed over the packing cross-section. Their readings were displayed on the computer monitor as the topograms. The groups of thermometers were installed in the bottom (two layers from the packing bottom), middle and upper (two layers from the packing top) packing cross-sections. The on-screen information allowed evaluation of both the structure of large-scale temperature non-uniformity and the value of this non-uniformity.

The experiments were carried out for different irrigation point patterns of the liquid distributor, keeping the total number of irrigation points. The maximal number of experimental data was obtained for two patterns of irrigation points, namely: Pulse pattern and Half_Column (HC) pattern (Fig.1). Pulse pattern provides packing irrigation through the holes distributed uniformly over the cross-section during time $t_1$, then all holes are closed for period $t_2$ (Fig.1a). Then, the cycle is repeated. HC pattern provides irrigation of a half of the packing cross-section during time $t_1$ (Fig.1b), the second half of cross-section stays closed. Then, the valves are switched, and the irrigated and dry halves of cross-section change for period $t_2$ (Fig.1c). Then, the cycle repeated periodically. The main data array for the HC pattern is obtained for equal values of $t_1$ and $t_2$ within 5 – 160 s. The pictures of packing irrigation by HC pattern are shown in Fig. 2.

![Figure 1. Irrigation point patterns in different experiments. a) uniform irrigation by Pulse pattern; b) first semi-period of irrigation by HC pattern; c) second semi-period of irrigation by HC pattern.](image-url)
3. Results and discussions

Dependences of the height of transfer unit (HTU) on the period of valve switching for the HC pattern, determined as a sum of times $t_1$ and $t_2$, are shown in Fig. 3. The experiments were carried out on the packing with the heights of 2.1 and 4.0 m at the same superficial velocity of vapor $K_v = 0.024$ m/s. The zero period corresponds to the conditions of uniform packing irrigation.

As it can be seen from the diagram, the efficiency of mixture separation in a short packing (10 layers, $H = 2.1$ m) is significantly higher than in the packing of 4 m (19 layers). This fact is well known and it is explained by formation of large-scale maldistribution of local flow parameters along the packing height [4]. In engineering practice, the liquid redistributors are usually installed in 10 packing layers to align these maldistributions. In our experiments, at periodic packing irrigation by the HC pattern, we observed the improvement of mixture separation efficiency in the range of switching periods of up to 60 s for the packing height of 4 m and up to 20 s for the height of 2.1 m.

The following increase in the switching period led to deterioration in the efficiency of mixture separation. When switching with a short period, separation efficiency was close to separation efficiency at the zero period.
efficiency at uniform irrigation because during irrigation of a half of the packing cross-section liquid did not have time to penetrate through a significant part of the packing height. Switching irrigation to another half of the column cross-section of the column, in fact, created the conditions of uniform irrigation. An increase in the period of irrigation led to the fact that during one semi-period of irrigation liquid spread over the packing height, and the large-scale non-uniformity, corresponding to non-uniform irrigation of the first semi-period, started forming in the packing. Switching at this moment to the second semi-period of irrigation, whose structure is diametrically different to the first semi-period led to rearrangement of the forming large-scale non-uniformity in the opposite direction. As a result, at switching period $t_{\text{min}}$, the maximal separation efficiency was achieved; it was 20% higher than separation efficiency at uniform irrigation. The value of optimal period $t_{\text{min}}$ is determined by the time of liquid flow along the packing height, and it depends on both the packing height and superficial velocity of liquid and vapor flows.

The experiments in the regime of periodic irrigation (Pulse pattern) were carried out on the packing with the height of 2.1 m at superficial vapor velocity $K_v = 0.024$ and 0.035 m/s. Dependence of the relative height of transfer unit is shown in Fig. 4 at different ratios of irrigation time ($t_1$) and time of irrigation stop ($t_2$).

According to the diagrams, short-term (within the time of liquid flow on the packing) complete periodical stop of the liquid flow irrigating the packing does not lead to deterioration of mixture separation; in some cases, we can even observe the improvement of separation efficiency within 5 - 10%.

4. Conclusions
According to the experiments, the influence of the periodic irrigation on the large-scale non-uniformity of mixture composition, formed in the packing, can affect significantly the structure of flow parameter distribution over the cross-section and height of the mass transfer surface. Essentially non-uniform periodic irrigation of the packing can improve separation efficiency of the column within 20%, if the switching periods are comparable with the times of formation of large-scale non-uniformity. Short-term (within the time of liquid flow on the packing) complete periodical stop of the liquid flow irrigating the packing does not lead to deterioration of mixture separation; in some cases, we can even observe the improvement of separation efficiency within 5 – 10%.

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