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# CORRELATION BETWEEN HYSTERESIS OF GAS-LIQUID MASS TRANSFER AND LIQUID DISTRIBUTION IN A TRICKLE BED*

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**Abstract** The hysteresis of gas-liquid mass transfer rate and the corresponding radial liquid distribution in a trickle bed reactor are measured to provide evidence for the correlation between these two behaviors. Experimental results indicate that the hysteresis of gas-liquid mass transfer originates from the nonuniformity of the hydrodynamic state of gas-liquid flow and the radial maldistribution of local $k_{gl}a$ corresponds very well to the radial maldistribution of liquid flow in the bed. The local liquid flow rate is also found to be nonuniform in the azimuthal direction. In view of maldistributed liquid flow even in the pulsing flow regime, the conventional plug flow model seems oversimplified for describing the behavior of a trickle bed.

**Keywords** trickle-bed reactor, radial liquid distribution, gas-liquid mass transfer, hysteresis

## 1 INTRODUCTION

A trickle bed reactor involves a three-phase system in which liquid and gas flow cocurrently downward through a fixed catalyst bed and has been widely used in the process industry. It has been recently found that a trickle bed shows obvious hysteretic behavior when packed with small particles. Hysteresis indicates that there exist multiple hydrodynamic states, which depend not only on the system properties and operating conditions, but also on the operating mode or the path to reach certain operating conditions. So far, hysteresis in pressure drop, liquid holdup and liquid-solid mass transfer rate has been reported[1-3]. Hysteresis of gas-liquid mass transfer coefficient was also evidenced[4]. Christensen *et al.*[5] and Chu *et al.*[6] attributed hysteresis to the texture of liquid flow. Melli and Scriven[7] predicted hysteresis in terms of fluid distribution in the bed, but they presented no comparison with experimental results. The authors also anticipated that the different branches of a hysteresis loop corresponded to different hydrodynamic states of the gas and liquid flow in a trickle bed[4,8].

In this work, radial liquid distribution and local gas-liquid mass transfer rate were measured for a trickle bed operated in the Levec's mode[2], and experimental evidence is presented to support the proposition that hysteresis originates from the multiplicity of hydrodynamic states of trickling gas-liquid flow.

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## 2 EXPERIMENTAL SYSTEM

An air-water system was used in this study. The experimental trickle bed reactor was made of plexiglass tube of 70 mm I.D. and packed 1000 mm high with $\phi 2.7$ mm glass beads (Fig.1). The liquid was fed into the column through a liquid distributor consisting of 128 $\phi 1 \times 155$ mm stainless steel tubings. Underneath the supporting grid, a liquid diverter was attached to direct the localized gas-liquid streams to graduated cylinders for flow intensity measurement. The movable diverter consists of an array of 5 beveled $\phi 10$ mm tubes so that local flow intensity data may be obtained. The liquid flow distribution can be determined by taking measurements at various azimuthal positions. An oxygen meter (YSI Model 51B, Yellow Springs Instrument Co. USA) was used to monitor the inlet and outlet concentration of dissolved oxygen. The saturated concentration at the temperature of the experiments was measured in an aerated bottle before and after each experimental run. The gas-liquid mass transfer coefficient was calculated by using the simple plug flow model without backmixing in the trickle bed[4]. Experiments were conducted in the Levec's mode of operation. The procedure involved the following steps:(1) prewet the bed using pulsing gas-liquid flow; (2) stop the flow and allow the bed to drain for 20 minutes; (3) set the desired gas flow rate and (4) begin measurements with various liquid flow rates. The superficial mass velocity of air was 0.208 $\mathrm{kg \cdot m^{-2} \cdot s^{-1}}$ and the liquid flow rate ($L$) in the range of 0 to 15.0 $\mathrm{kg \cdot m^{-2} \cdot s^{-1}}$.

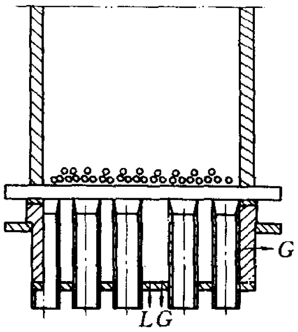
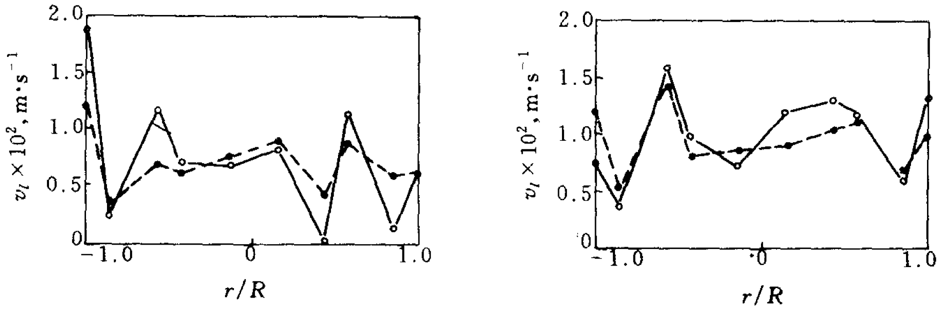


**Figure 1** Scheme of experimental trickle bed (lower part) and liquid diverter

## 3 RESULTS AND DISCUSSION

For the Levec's mode of operation, as stated in our previous papers, under the condition of nonzero gas flow rate, liquid distribution in the packing is likely to deviate from the uniform distribution as $L$ increases. When $L$ becomes large enough to induce flow pulsation, the difference between $L$-increasing and $L$-decreasing paths vanishes, because the phase distribution becomes quite uniform on a time-average basis and the solid packing is well wetted due to strong interaction between pulsing phases. On the $L$-decreasing path, the pulsation induced uniformity persists and good gas-liquid contact leads to higher value of $k_{gL}a$[4]. Experiments were conducted to verify the above anticipation.

Liquid distribution was determined along 4 diameters with azimuth of 0, 45, 90 and 135 degree respectively and Fig.2 (a) presents the local liquid flow rates along one diameter. Although $d_t/d_p$ is up to 25.9 in this work, the radial liquid distribution with $L$-increasing is less uniform than that of the $L$-decreasing path at the same liquid flow rate. A similar contrast is observed at higher liquid flow rate [Fig.2(b)].



(a) $L = 4.42\ \text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (b) $L = 7.37\ \text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$

**Figure 2 Liquid distributions across the diameter for operations along $L$-increasing and $L$-decreasing flow paths ($G = 0.208\ \text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$)**

—— $L$-increasing; ---- $L$-decreasing

The difference of liquid maldistribution for the $L$-increasing and $L$-decreasing gas-liquid flows was tested by the variance test for its significance. $F$ was the ratio of the variance of liquid flow intensity on the $L$-increasing path over that of $L$-decreasing path under the same superficial operation conditions. For an ensemble of 32 data on both the $L$-increasing and $L$-decreasing paths respectively, the variance ratio $F$ was 2.064. From the $F$ distribution table, $F_{(0.05)}(30, 30) = 1.840$ is below the measured $F$, indicating that the level of significance is below 5%, and the observation that the liquid flow of the lower branch of the hysteresis loop is more severely maldistributed than that of its upper branch is statistically justified.

It was generally assumed that the liquid flow intensity was evenly distributed in the azimuthal direction. With this notion, most authors designed concentric annular collectors for the experimental investigation of liquid flow distribution in the packed bed. However, the measured data on $v_l$ along a circle with $r = 5$ mm indicate that the liquid maldistribution is as severe as that in the radial direction (Fig. 3). Obviously the maldistribution of liquid flow develops both in the radial and azimuthal directions.

When the pulsing flow regime is reached, the difference of liquid distribution between the $L$-increasing and $L$-decreasing paths is hardly discernible. Fig.4 presents the data on $v_l$ for the operation conditions corresponding to the incipient onset of flow pulsation and the point of hysteresis disappearance on the $L$-decreasing path. Nonuniformity of the local liquid flow rates is noticed in contrary to physical intuition. It was usually admitted that for $d_t/d_p$ larger than about 20, the liquid distribution is uniform in the low-interaction flow regime. While in the high-interaction flow regime, the liquid distribution is nearly uniform[9]. In view of this maldistribution which persists even in the pulsing flow regime, the plug flow model, though popular and conventional, should be reexamined for its validity and suitability in describing trickle beds with cocurrent downward flow.

Corresponding to the liquid distribution of Fig.2(b), Figs.5 and 6 show clearly the hysteresis of $c_l$ and $k_{gL}a$ due to the difference in liquid flow distribution and the

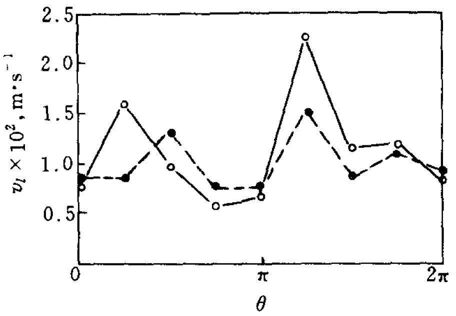


**Figure 3** Local liquid flow rates in 8 equally-spaced points on a concentric circle

($r = 5$ mm, $G = 0.208$ kg·m$^{-2}$·s$^{-1}$, $L = 4.42$ kg·m$^{-2}$·s$^{-1}$)

—— $L$-increasing; – – $L$-decreasing

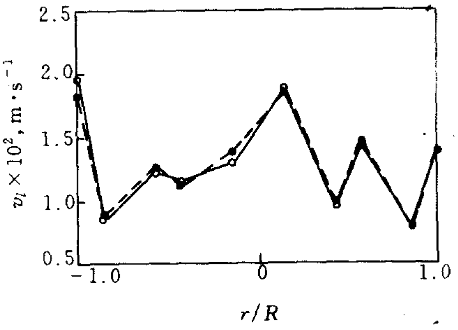


**Figure 4** Liquid distributions under operating condition of pulsation onset

($G = 0.208$ kg·m$^{-2}$·s$^{-1}$, $L = 10.32$ kg·m$^{-2}$·s$^{-1}$)

—— $L$-increasing; – – $L$-decreasing

fluctuation of $c_l$ and $k_{\mathrm{gL}}a$ in the radial direction, respectively, even though the model of plug flow without backmixing was adopted for evaluating $k_{\mathrm{gL}}a$. It is found that the rate of mass transfer with $L$-increasing is lower than that on the $L$-decreasing path at the same liquid flow rate, for in the latter case there exists more uniform radial liquid distribution. The deviation of $k_{\mathrm{gL}}a$ between two curves is up to 20%.

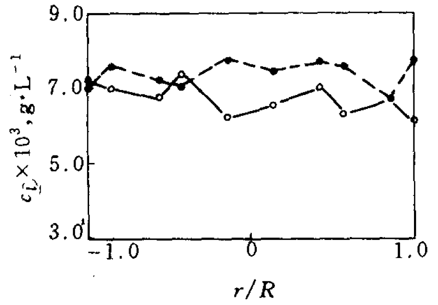


**Figure 5** Radial distributions of $c_l$ on $L$-increasing and $L$-decreasing flow paths

($G = 0.208$ kg·m$^{-2}$·s$^{-1}$, $L = 7.37$ kg·m$^{-2}$·s$^{-1}$)

—— $L$-increasing; – – $L$-decreasing

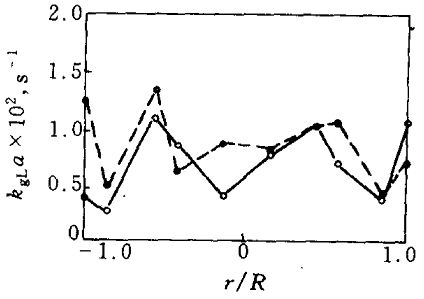


**Figure 6** Radial distributions of $k_{\mathrm{gl}}a$ on $L$-increasing and $L$-decreasing flow paths

($G = 0.208$ kg·m$^{-2}$·s$^{-1}$, $L = 7.37$kg·m$^{-2}$·s$^{-1}$)

—— $L$-increasing; – – $L$-decreasing

In this work, care was taken to minimize the interference of the supporting plate and the liquid collector to liquid flow distribution. The opening fraction of the packing supporting plate was chosen equal to the porosity of the packed bed. If the collector is installed at a distance below the supporting plate, the liquid may migrate radially underneath the plate, giving unreliable experimental data. In this study, the liquid collector was attached to the supporting plate to get the unbiased measurement of liquid distribution. Duplicate runs of measurements performed on different days gave the same experimental data within experimental error.

## 4 CONCLUSIONS

(1) Experimental results indicate that for Levec's mode of operation, the hysteresis of gas-liquid mass transfer originates from the nonuniformity of hydrodynamic states of the gas-liquid flow, and there exists significant radial maldistribution of local gas-liquid mass transfer coefficient in the trickle bed.

(2) The radial liquid distribution with $L$ increasing is less uniform than that on the $L$-decreasing path at the same liquid flow rate, and the difference of two operating paths disappears when flow pulsation takes place.

(3) Local liquid flow rate is not uniform in both the radial and azimuthal directions. The plug flow model based on the assumption of cross-sectional uniformity is not suitable for modeling the trickle bed reactor, even for the pulsing flow pattern and/or a small diameter bed.

## NOMENCLATURE

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|---|---|
| $c_l$ | outlet liquid concentration of dissolved oxygen, $g \cdot L^{-1}$ |
| $d_t$ | column diameter, m |
| $d_p$ | particle diameter, m |
| $F$ | variance ratio |
| $G$ | gas superficial mass velocity, $kg \cdot m^{-2} \cdot s^{-1}$ |
| $k_{gL}a$ | volumetric gas-liquid mass transfer coefficient, $s^{-1}$ |
| $L$ | Liquid superficial mass velocity, $kg \cdot m^{-2} \cdot s^{-1}$ |
| $R$ | column radius, m |
| $r$ | radial coordinate, m |
| $v_l$ | local liquid velocity, $m \cdot s^{-1}$ |
| $\theta$ | azimuthal coordinate, rad |

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