ELSEVIER

Catalysis Today 79–80 (2003) 235–240

CATALYSIS TODAY

www.elsevier.com/locate/cattod

Bed length effect on the liquid phase non-idealities and holdup in pilot scale upflow reactors

A.M. Thanos [a], G.D. Bellos [a], P.A. Galtier [b], N.G. Papayannakos [a,*]

[a] *School of Chemical Engineering, National Technical University of Athens, 9, Heroon Polytechniou Street, Gr-157 80 Zografos, Greece*
[b] *Institut Français du Pétrole, C.E.D.I., B.P. 3-69390 Vernaison, France*

Abstract

Residence time distribution experiments were carried out at ambient conditions in pilot scale upflow reactors. Diluted and non-diluted beds of various lengths and diameters were tested. The axial dispersion model was used to describe the liquid phase non-idealities. Results of Peclet number (*Pe*), liquid holdup and axial dispersion coefficient for various bed lengths and fluid velocities have been collected. The dependency of the non-ideal flow parameters on bed length is discussed.

Keywords: Non-ideal flow; Axial dispersion; Holdup; Upflow reactors

1. Introduction

In evaluating catalyst efficiency and predicting the performance of industrial hydroteaters, bench and pilot scale reactors with gas and liquid concurrent upflow are used as an alternative to the well known downflow ones (trickle bed reactors). Upflow reactors are characterized by certain advantages compared to the corresponding downflow ones [1] and these advantages are mainly attributed to the high liquid holdup values. However, upflow reactor performance is known to be affected by liquid phase mixing [2,3]. Catalyst beds of various diameters (10–30 mm) and lengths (10–150 cm) are widely used to obtain experimental data. Incorporation of dispersion effects into simulation codes demands values of the axial dispersion coefficient. The dependence of these values as well as of the liquid holdup values upon the bed length are important factors for both the reliable simulation and the evaluation of data from bench and pilot scale reactors.

Limited experimental data exists on the variation of non-ideal flow characteristics for different bed lengths [4] for pilot scale reactors and only for higher velocities and larger packing particles than the ones used in pilot scale hydrotreaters. As a consequence, no validated dependence of liquid holdup and axial dispersion on bed length is available. The scope of this paper is to present and discuss results concerning the bed length effect on the liquid flow non-idealities and holdup for pilot scale upflow reactors with diluted and non-diluted beds.

2. Experimental

The RTD experiments were carried out at ambient conditions in pilot scale upflow reactors, using the experimental set-up described in a previous work [5]. The gas–liquid feed system used was toluene–nitrogen, which resembles at ambient

* Corresponding author. Fax: +30-10-7723155.
E-mail address: npap@central.ntua.gr (N.G. Papayannakos).

0920-5861/03/$ – see front matter
doi:10.1016/S0920-5861(03)00010-5

Nomenclature

d_p	extrudate diameter (mm)
D_{ax}	axial dispersion coefficient (cm^2/s)
D_r	reactor diameter (mm)
H_l	liquid holdup
L	reactor length (cm)
Pe	Peclet number
Q_l	liquid volumetric flow (cm^3/s)
u_{gs}	gas feed superficial velocity (mm/s)
u_{ls}	liquid feed superficial velocity (mm/s)
U	liquid phase real velocity
V_l	liquid in the reactor (cm^3)
V_r	reactor volume (cm^3)
V_{void}	bed void volume (cm^3)
x	axial coordinate
z	dimensionless axial coordinate (x/L)

conditions the properties of gas feed (hydrogen) and liquid feeds (gas oil) of hydrotreaters at reaction conditions. The diameter of the beds tested was $D_r = 10$, 16 and 25 mm and the bed length varied within $L_r = 16$–250 cm. Non-porous cylindrical extrudates with dimensions close to those of typical commercial hydrotreatment catalysts ($d_p = 1.2$ mm) were used as packing. In the case of the diluted beds, SiC particles (Carborundum) with a mean diameter of 0.25 mm were also used as the dilution medium. The tracer used in the liquid phase was *p*-nitrophenol. The step increase/decrease tracer concentration technique was used to determine the liquid flow non-ideal characteristics [5].

3. Results

The deviations of the liquid flow from the ideal plug flow model were evaluated by the axial dispersion model (Eq. (1)). The boundary equations used in solving Eq. (1) were the Danckwerts (closed vessel) ones:

$$\frac{L}{U}\frac{\partial C}{\partial t} = \frac{1}{Pe}\frac{\partial^2 C}{\partial z^2} - \frac{\partial C}{\partial z} \tag{1}$$

The liquid holdup is defined (Eq. (2)) as the ratio of the liquid in the reactor to the bed void volume. The characteristic length used in the Peclet number (Pe) (Eq. (3)) is the bed length, and the velocity used in Pe is the real liquid phase velocity.

$$H_l = \frac{V_1}{V_{void}} = \frac{LQ_l}{UV_{void}} \tag{2}$$

$$Pe = \frac{UL}{D_{ax}} \tag{3}$$

Eq. (1) was solved in the time domain applying the control volume method and using the Crank–Nicholson approach. The solution algorithm was coupled with the Marquardt optimization algorithm [5] in order to estimate the liquid flow

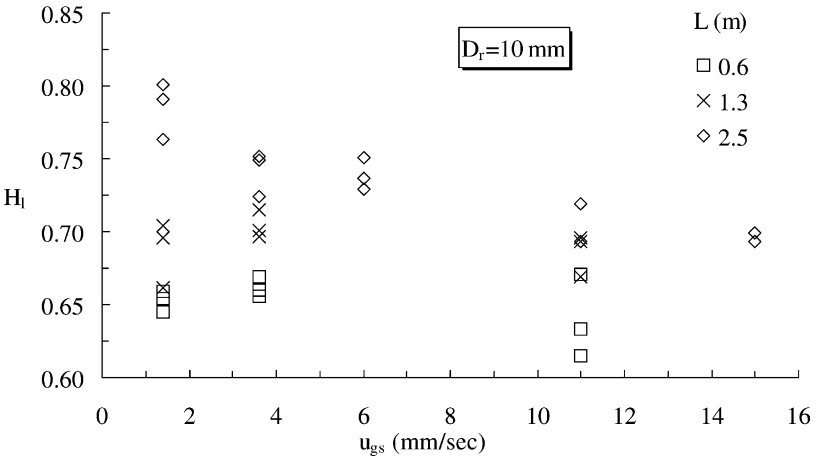

Fig. 1. Liquid holdup dependency on bed length (non-diluted beds).

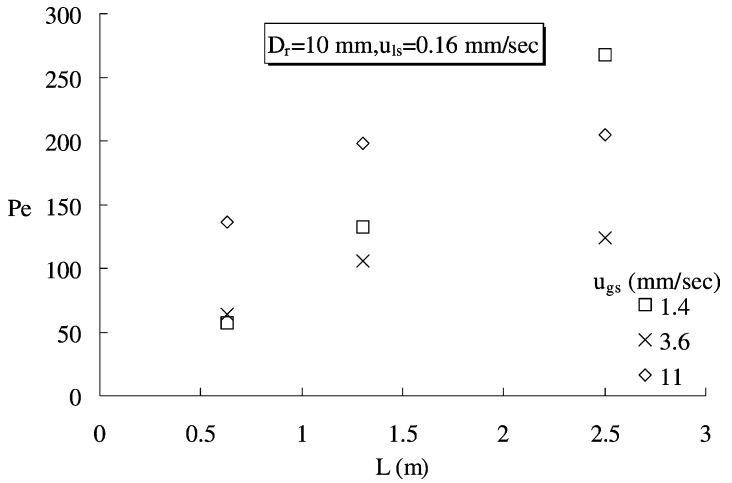

Fig. 2. Peclet number dependency on bed length (non-diluted beds).

parameters (*Pe* and H_l). The objective function to be minimized in the Marquardt algorithm was the sum of squares of the differences between experimental and calculated values of the tracer concentration at the system exit for the various sampling times. The parameters to be fitted in order to minimize the objective function was *Pe* and H_l. The convergence criterion was that the objective function for two successive predictions of the model parameters was relatively reduced by less than 10^{-4}.

The mean bed void fraction did not show any trend with bed length and the small variations of bed void fraction measured for the various beds built (maximum ±5%) ensured that the bed loading procedure used (described in detail in [6]) leaded to reproducible loading of diluted and non-diluted beds.

The liquid holdup was found to increase with bed length for constant superficial gas velocities (Fig. 1), for either non-diluted or diluted beds. In all cases, the liquid holdup values were higher than 0.65 for gas superficial velocities lower than 6 mm/s. The liquid holdup increase was higher for the non-diluted beds (mean liquid holdup increase by 0.05 for doubling the bed length) than for the diluted ones (mean liquid

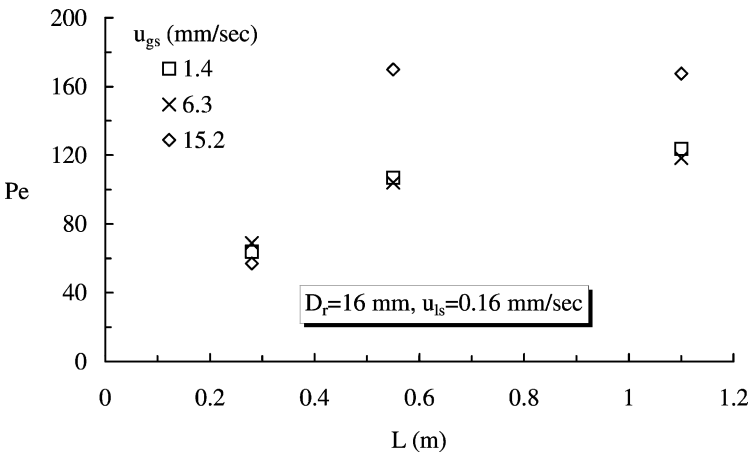

Fig. 3. Peclet number dependency on bed length (non-diluted beds).

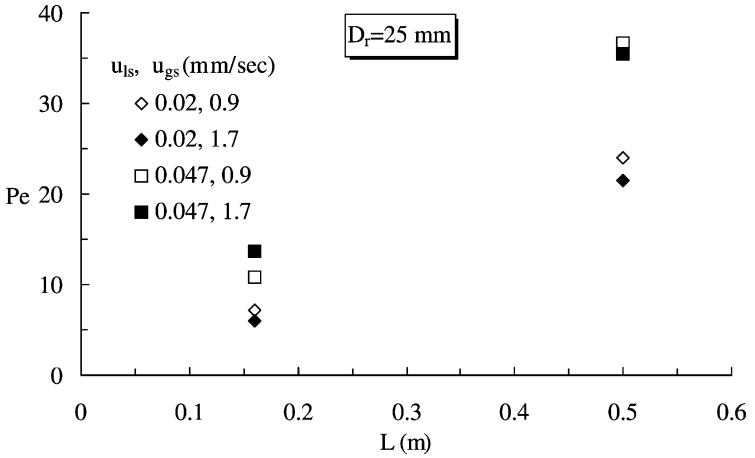

Fig. 4. Peclet number dependency on bed length (non-diluted beds).

holdup increase by 0.05 for a 200% bed length increase). Stiegel and Shah [4] found a smaller but also decreasing trend of liquid holdup when the bed length increased from 91 to 152 cm, although they used water and air as liquid-gas feed system, bigger packing particles ($d_p \approx 3$ mm) and much higher fluid velocities.

In Figs. 2–4, representative results are presented for the dependence of *Pe* on bed length of non-diluted beds. In general, a rather linear increase of *Pe* with bed length is observed for short beds, but as bed length increases *Pe* approaches a plateau. The only exception of this tendency is noticed for the small diameter beds, $D_r = 10$ mm without thermowell (Fig. 2), operating at low gas velocity ($u_{gs} = 1.4$ mm/s). In this case, *Pe* increased linearly with bed length within the whole range of the bed lengths examined in this work. For higher gas velocities an increase of *Pe* with bed length is observed up to 130 cm and thereafter *Pe* is practically constant. In Fig. 3, it is shown that for 16 mm diameter beds, *Pe* increases up to 55 cm bed length while for longer beds it appears constant. In Fig. 4, an increase of *Pe* with bed length is depicted for beds with a diameter of 25 mm. In Fig. 5, the dependence of liquid axial dispersion on bed height for 16 mm

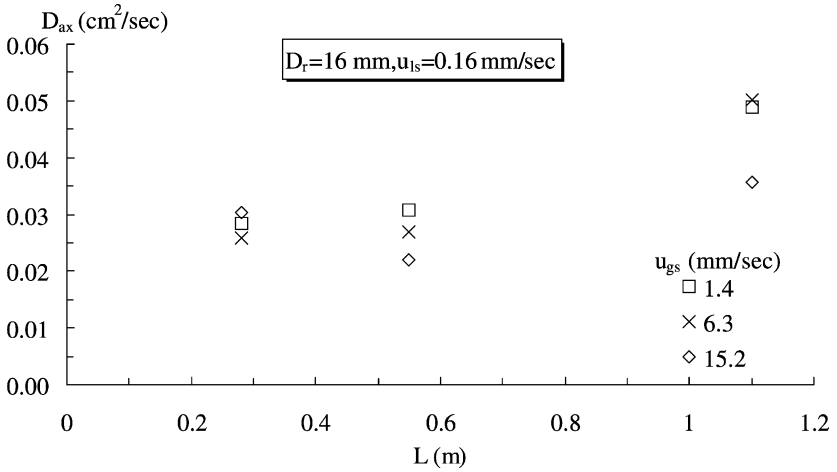

Fig. 5. Axial dispersion coefficient dependency on bed length (non-diluted beds).

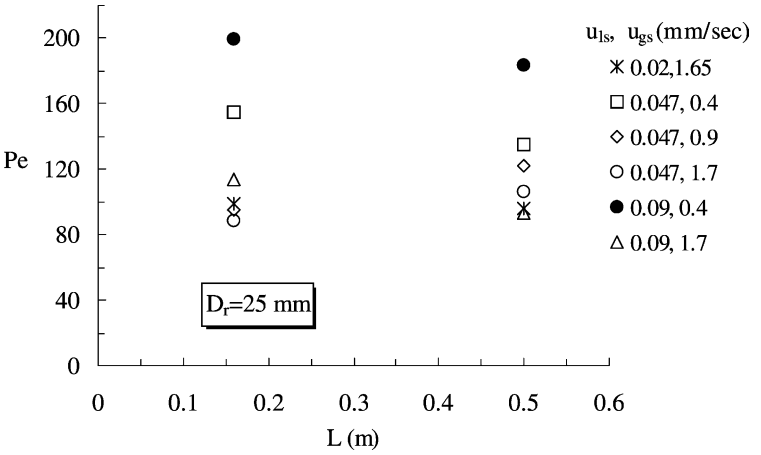

Fig. 6. Peclet number dependency on bed length (diluted beds).

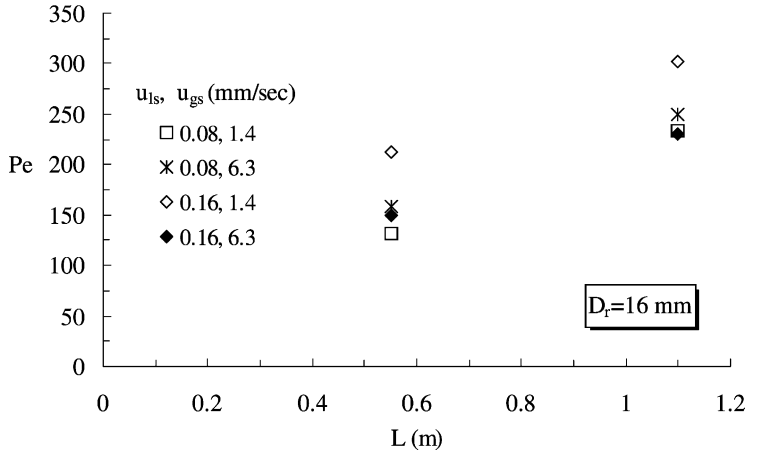

Fig. 7. Peclet number dependency on bed length (diluted beds).

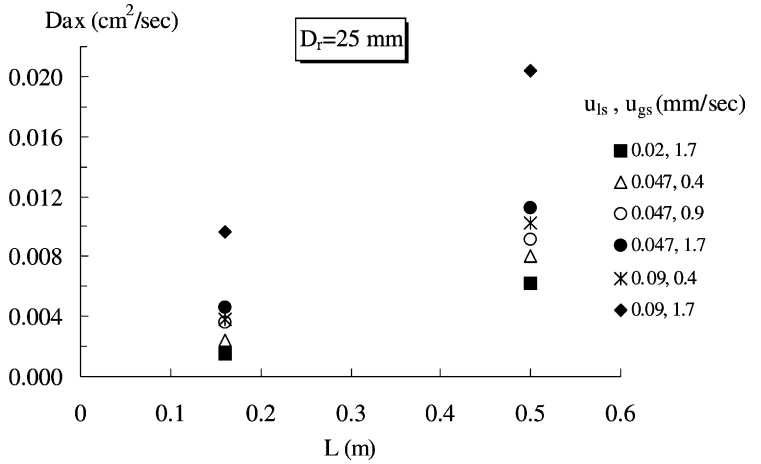

Fig. 8. Axial dispersion coefficient dependency on bed length (diluted beds).

diameter beds is presented, corresponding to *Pe* variations illustrated in Fig. 3. Up to a height of 55 cm, the value of axial dispersion coefficient is rather constant and above this bed length it increases.

A different dependence of *Pe* on the bed height has been obtained for the diluted beds. *Pe* number appears fairly constant up to 50 cm bed length, as shown in Fig. 6, for beds with a diameter of 25 mm. For longer than 60 cm diluted beds, *Pe* number increases with bed length as shown in Fig. 7 for 16 mm diameter beds. In Fig. 8, the linear increase of D_{ax} with bed length is illustrated for 25 mm diameter beds shorter than 50 cm.

The axial dispersion coefficient dependency on bed length presented in this paper implies that the mixing conditions along the reactor axis are not constant, even for constant feed superficial velocities and packing type. As a consequence, the mixing phenomena within the reactor cannot be considered a priori as linear. In scaling-up and kinetic studies the hypothesis of *Pe* linearity with bed length should always be checked.

4. Concluding remarks

Liquid holdup increases with bed length in diluted and non-diluted beds and in diluted beds the increase was smaller than in non-diluted beds. The dependence of liquid axial dispersion on bed length is different in diluted beds and non-diluted beds. For the non-diluted beds, D_{ax} does not change in beds shorter than 60 cm but for larger beds D_{ax} increases. On the contrary, for the diluted beds a linear increase of D_{ax} with bed length was obtained for beds shorter than 60 cm, although for larger beds D_{ax} appeared constant.

References

[1] M.R. Khadilkar, Y.X. Wu, M.H. Al-Dahhan, M.P. Duducovic, Comparison of trickle and upflow reactor performance at high pressure: model predictions and experimental observations, Chem. Eng. Sci. 51 (1996) 2139–2148.
[2] N.G. Papayannakos, P.A. Galtier, P.H. Bigeard, S. Kasztelan, Hydrodynamic effects in bench scale hydrotreaters operating in cocurrent gas–liquid upflow mode, Chem. Eng. Sci. 47 (1992) 2275–2280.
[3] A.M. Thanos, P.A. Galtier, N.G. Papayannakos, Liquid dispersion and holdup in a small-scale upflow hydrotreater at high temperatures and pressure, Chem. Eng. Sci. 56 (2001) 693–698.
[4] G.J. Stiegel, Y.T. Shah, Backmixing an liquid holdup in a gas–liquid cocurrent upflow packed column, Ind. Eng. Chem. Process Design Dev. 16 (1) (1977) 37–43.
[5] A.M. Thanos, P.A. Galtier, N.G. Papayannakos, Liquid flow non-idealities and holdup in a pilot scale packed bed reactor with cocurrent gas–liquid upflow, Chem. Eng. Sci. 51 (1996) 2709–2714.
[6] A.M. Thanos, P.A. Galtier, N.G. Papayannakos, The effect of dilution on the liquid-phase non-idealities in pilot scale upflow reactors, Chem. Eng. Sci. 54 (1999) 2781–2785.