Multiple Hydrodynamic States in Trickle Flow:
Correlating Pressure Drop and Liquid Holdup

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Multiple hydrodynamic states are possible in trickle flow reactors (Kan & Greenfield, 1978, 1979, Levec et al., 1986, 1988 and Christensen et al., 1986). The hydrodynamic condition has mostly been quantified through pressure drop and liquid holdup variation. It is well established that pressure drop and liquid holdup are influenced by the flow history. In this study, we report the extent of possible variation of these hydrodynamic parameters for the 3 mm glass sphere – air – water system. The ranges of liquid and gas flow rates investigated were 1-9 kg/m^{2}s and 0.013-0.117 kg/m^{2}s respectively. The upper and lower limiting cases are investigated according to specifically defined prewetting procedures (following the approach of Wang et al., 1995 and Van der Merwe & Nicol, 2005):

- Non-prewetted (dry packing)
- Levec prewetted: the bed is flooded and drained and after residual holdup stabilisation, the gas and liquid flows are introduced
- Kan_L prewetted: the bed is operated in the pulse flow regime (by increasing liquid velocity) and liquid flow rate is reduced to the desired set point
- Kan_G prewetted: the bed is operated in the pulse flow regime (by increasing gas velocity) and gas flow rate is reduced to the desired set point
- Super prewetted: the bed is flooded and gas and liquid flow are introduced once draining commences

Some selected results are shown in figures 1 and 2. The different prewetting procedures resulted in three distinct pressure drop regions. The upper region, and hence the upper limiting case for pressure drop, is the Kan_L and Super modes of operation. The lowest region was the non-prewetted and Levec prewetted modes. Interestingly, there was no significant difference between these two modes. The middle region, and as such not considered to be a limiting case, was the Kan_G mode of operation. The difference between the upper and lower regions can be as much as 700%. Liquid holdup is different in all five prewetting modes. The upper limiting case is the Kan_G mode of operation, although this mode is only possible at high liquid velocities. At low liquid velocities the Kan_L or Super modes can be considered to be the upper limiting case. Importantly, the lower limiting case for prewetted beds is the Levec mode. The absolute lower limiting case is the non-prewetted mode.

These results are in qualitative agreement with those reported in literature. Using the present classification, Kan & Greenfield (1979) studied only the Super and Kan_G modes. Levec et al. (1986, 1988) studied the Super and Levec modes for no gas flow and the Kan_L and Levec modes with gas flow. Christensen et al. (1986) studied the Kan_L and Levec modes. Lazzaroni et al. (1988, 1989) compared the Dry mode with the Super mode. Wang et al. (1995) investigated the Kan_L, Kan_G and Levec modes through pressure drop hysteresis only, and therefore dismissed the Kan_G mode as a non-limiting case in their subsequent discussions. Lutran et al. (1991), Ravindra et al. (1997) and Sederman & Gladden (2001) all report visualizations of the Levec and Kan_L modes. Van der Merwe & Nicol (2005) studied the Super, Levec and Dry modes for no gas flow. A host of correlations have been developed in the past 20 years based on data from the Kan_L mode of operation, as this mode is believed to be desirable.

Broadly speaking, there are three approaches that have been used to model the hydrodynamic multiplicity:

- Gas tortuosity effect (Kan & Greenfield, 1979)
- Relative permeability (Levec et al., 1986)
- The rivulet/film concept or multi-zone models (Melli & Scriven, 1991 and Wang et al., 1995)

In modelling two-phase flow, increased pressure drops are seen to be the result of the holdup reducing the cross-sectional flow area for gas flow, while duly considering the gas-liquid interaction forces. More specifically, in an Ergun-type equation, the liquid holdup is subtracted from the porosity in order to yield a lower effective porosity and a higher pressure drop. The difficulty in modelling hysteresis lies therein that non-uniform flows exist and interfacial areas and interstitial velocities in the momentum balance need to be corrected. Any corrective measures are mode specific: For liquid flow rate variation induced hysteresis increased pressure drops are accompanied by increased hold-ups (compare Levec and Kan_L modes), while for gas flow rate induced hystersis lower pressure drops are associated with higher hold-ups (compare Super and Kan_G modes). Present models only consider one of these hysteresis loops. Kan & Greenfield (1979) suggested that there is a gas tortuosity decrease associated with the maximum gas flow rate to which the bed had been subjected. They adopted the Turpin & Huntington (1967) friction factor correlation approach and introduced a maximum gas Reynolds number dependency into the Z factor (see Table 1 for parameter values):

$$\ln(f) = a_0 + a_1 \ln Z + a_2 (\ln Z)^2 \tag{1a}$$

$$f = \frac{d\varepsilon\left(-\Delta P/\Delta z\right)}{\rho_G u_G^2 (1-\varepsilon)} \tag{1b}$$

$$Z = \frac{1}{\mathrm{Re}_L^p \, \mathrm{Re}_G^q \, \mathrm{Re}_{G\max}^s} \tag{1c}$$

$$\frac{\varepsilon_L}{\varepsilon} = \frac{b\,\mathrm{Re}_L^x}{\mathrm{Ga}^{*y}_L} \tag{1d}$$

$$\mathrm{Ga}^*_L = d^3 \rho_L \left[\rho_L g + \left(-\Delta P/\Delta z\right)\right] / \mu_L^2 \tag{1e}$$

Table 1. Parameters for the Kan & Greenfield model

	Kan & Greenfield (1979)		This study	
	Super	Kan_G	Super	Kan_G
a_0	8.46	9.15	7.518	
a_1	-1.19	1.67	-0.663	
a_2	0.043	0.094	-0.063	
p	0.512	-0.39	0.512	
q	-1.265	0.72	-1.265	
s	0	0.4	0	-0.252
b	16.8	14.9	22.93	22.39
x	0.294	0.30	0.30	
y	0.358	0.35	0.35	

The Kan & Greenfield (1979) model underpredicts our pressure drop data when the recommended parameters are used. However, if a_0 to a_2, p and q are fitted to the Super mode data, a satisfactory fit to the Kan_G mode data is obtained by changing s only. That is, introducing a Re_{Gmax} dependency sufficiently compensates for the increased gas velocity history. The holdup values can be predicted by using a version of the Specchia & Baldi (1977) correlation (and fitting parameter b to the data). The difference between the two modes is obtained by changing only s and b. Results are shown in figure 1. The model is highly unsatisfactory due to the large number of parameters and the lack of fundamentality in their values. It is, however, the only correlation available for the Kan_G mode.

The relative permeability approach advocated by Levec et al. (1986) for the prediction of the liquid flow rate variation induced hysteresis, has since been validated for various choices of packing and pressures (Lakota & Levec, 2002 and Nemec et al. 2005) for the Kan_L mode. For liquid holdup in the Levec mode, Levec et al. (1986) suggested that the liquid phase relative permeability – reduced saturation relationship be altered:

$$\frac{1}{k_L}\left(180\frac{\mathrm{Re}^*_L}{\mathrm{Ga}^*_L}+1.8\frac{\mathrm{Re}^{*2}_L}{\mathrm{Ga}^*_L}\right)-\frac{1}{k_G}\left(180\frac{\mathrm{Re}^*_G}{\mathrm{Ga}^*_G}+1.8\frac{\mathrm{Re}^{*2}_G}{\mathrm{Ga}^*_G}\right)\frac{\rho_G}{\rho_L}=1 \tag{2a}$$

$$k_G=\begin{cases}0.4S_G^{3.6} & S_G\le 0.64\\ S_G^{5.5} & S_G>0.64\end{cases} \quad \textit{from Nemec et al. (2005)} \tag{2b}$$

$$k_L=\begin{cases}\delta_L^{2.9} & \delta_L\ge 0.2\\ 0.25\delta_L^{2.0} & \delta_L<0.2\end{cases} \quad \textit{Kan}_L\ \textit{mode} \tag{2c}$$

$$k_L=\delta_L^{2.0} \quad \textit{Levec mode} \tag{2d}$$

$$\frac{1}{k_G}\left(180\frac{\mathrm{Re}^*_G}{\mathrm{Ga}^*_G}+1.8\frac{\mathrm{Re}^{*2}_G}{\mathrm{Ga}^*_G}\right)=1-\frac{\Delta P}{\Delta z}\frac{1}{\rho_G g} \tag{2e}$$

The model is shown for selected data from Levec et al. (1986) and this study in figure 2. One adjusted parameter (the coefficient of the reduced saturation in eq. 2c and 2d) seems capable of compensating for the alternate mode of operation. We note that the pressure drops measured in this study for the two modes are accurately predicted at low liquid Reynolds numbers. In fact, the model performs well at low Reynolds numbers but not at high Reynolds numbers. The reason is apparent from a plot of liquid phase relative permeability against reduced saturation (figure 3). At high saturations (δ_L > 0.33 approx. for these conditions only) the holdups of the two modes are equal, but the permeability is still taken to be much higher for the Levec mode. It is possibly more appropriate to specify the Levec mode liquid relative permeability as a function of the saturation at the trickle-to-pulse flow boundary. Figure 2 also suggests that equation (2b) will also need modification for the Levec mode since the model under-predicts the holdup but over-predicts the pressure drop for this mode at high Reynolds numbers. In fact, following the procedures described in Levec et. al. (1986), plots of relative permeability against saturation can be drawn up for each of the pre-wetting modes investigated (figures 4 and 5). On these figures the exponents (slopes on the log-log plots) of the saturation (compare equations 2b to 2d) have been solved for by a minimum absolute error procedure. For the liquid phase (figure 4) the Levec mode exponent of 1.9 compares well with the value of 2 originally suggested (Levec et. al., 1986). Moreover, the Super, Kan Liquid and Kan Gas modes all have exponents close to the 2.9 suggested by

Nemec et al. (2005). Interestingly, the Non-pre-wetted mode permeability has a nearly linear dependence on the saturation. These observations are consistent with an increased propensity toward rivulet-dominated flow in the order: Kan modes, Super, Levec and Non-pre-wetted modes. For the gas phase (figure 5), the exponent of the gas saturation in the Kan Liquid and Super modes (3.3 for present data) compare well with the 3.6 suggested by Nemec et al. (2005). The Levec and Non-pre-wetted modes also correlate to this exponent at high gas saturations, but deviates from it significantly at low gas saturations. This is surprising, as low gas saturations are expected at high liquid velocities where the rivulet type flow is expected to give way to film flow (Nemec et al., 2005). Present results suggest that rivulet flow prevails in liquid saturated beds in the Levec and Non-pre-wetted modes. The oddity of the Kan Gas mode is particularly evident from figure 5, where a *negative* exponent of the gas saturations is required. This is another way of illustrating that high liquid saturations lead to lower pressure drops in this mode of pre-wetting. Finally, it has to be noted that although the exponents of the permeability-saturation relationships agree with those of prior investigators for the modes that they employed, the pre-exponential factors do not. Generally, holdups in this study are somewhat larger than those reported by Levec et al. (1986), Lakota et al. (2002) and Nemec et al. (2005). Nevertheless, with appropriate empirical adjustment the model seems capable of capturing the trends of hydrodynamic multiplicity.

Significantly more complex hystersis models have been proposed by other authors and are not reproduced here for brevity. Wang et al. (1995) adopted Christensen et al.'s (1986) interpretation and divided the bed into rivulet and film dominated flow cross-sections. The model is unable to model gas flow rate variation induced hysteresis (probably because they measured only pressure drop and not holdup as well). It is more suited to investigating the flow uniformity rather than being a predictive pressure drop model. Melli & Scriven (1991) introduced a 2-D network of pores model (based on pore level hydrodynamics). It is capable of predicting hysteresis, at least in qualitative terms. Van der Merwe & Nicol (2005) introduced a simple momentum balanced based holdup model for stagnant gas conditions. It corrects both the interstitial velocity and liquid-solid interfacial area with a single parameter (volumetric utilization) that is measured independently. In light of the highly empirical nature of other hysteresis models, it is encouraging that accurate predictions of the holdup in the Dry, Levec and Super modes resulted. Unfortunately, the study was limited in scope and has not been generalized to include gas flow.

In this study, we have evaluated the hysteresis modelling approaches in light of new and more complete pressure drop and holdup data. Although the models perform reasonably well for the modes of operation employed by their authors, no general hysteresis model exists for prediction of all multiple hydrodynamic states. Although it is possible to extend these models empirically, it is more desirable to gain further fundamental insights into the hysteresis phenomenon before doing so. The implications of hydrodynamic multiplicity shown in this study are likely to drastically affect trickle bed modelling, design and operation.

References

Christensen, G., McGovern, S. J. and Sundaresan, S. (1986) “Cocurrent Downflow of Air and Water in a Two-Dimensional Packed Column” *A.I.Ch.E. J., 32, (10),* 1677 - 1689.

Kan, K. M. and Greenfield, P. F. (1978) “Multiple Hydrodynamic States in Cocurrent Two-Phase Down-Flow through Packed Beds” *Ind. Eng. Chem. Process Des. Dev., 17,* 482 - 485.

Kan, K. M. and Greenfield, P. F. (1979) “Pressure Drop and Holdup in Two-Phase Cocurrent Trickle Flows through Packed Beds” *Ind. Eng. Chem. Process Des. Dev., 18,* 740 - 745.

Lakota, A., Levec, J. and Carbonell, R.G. (2002) “Hydrodynamics of Trickling Flow in Packed Beds: Relative Permeability Concept” *A.I.Ch.E. J., 48,* 731 - 738.

Lazzaroni, C. L., Keselman, H. R. and Figoli, N. S. (1988) “Colorimetric Evaluation of the Efficiency of Liquid-Solid Contacting in Trickle Flow” *Ind. Eng. Chem. Res., 27,* 1132 - 1135.

Lazzaroni, C. L., Keselman, H. R. and Figoli, N. S. (1989) “Trickle Bed Reactors. Multiplicity of Hydrodynamic States. Relation between the Pressure Drop and the Liquid Holdup” *Ind. Eng. Chem. Res., 28,* 119 - 121.

Levec, J., Grosser, K. and Carbonell, R. G. (1988) “The Hysteretic Behaviour of Pressure Drop and Liquid Holdup in Trickle Beds” *A.I.Ch.E. J., 34,* 1027 - 1030.

Levec, J., Saez, A. E. and Carbonell, R. G. (1986) “The Hydrodynamics of Trickling Flow in Packed Beds, Part II: Experimental Observations” *A.I.Ch.E. J., 32,* 369 - 380.

Lutran, P. G., Ng, K. M. and Delikat, E. P. (1991) “Liquid Distribution in Trickle Beds. An Experimental Study using Computer-Assisted Tomography” *Ind. Eng. Chem. Res., 30,* 1270 - 1280.

Melli, T. R. and Scriven, L. E. (1991) “Theory of Two-Phase Cocurrent Downflow in Networks of Passages” *Ind. Eng. Chem. Res., 30,* 951 - 969.

Ravindra, P. V., Rao, D. P. and Rao, M. S. (1997) “Liquid Flow Texture in Trickle-Bed Reactors: An Experimental Study” *Ind. Eng. Chem. Res., 36,* 5133 - 5145.

Sederman, A. J. and Gladden, L. F. (2001) “Magnetic Resonance Imaging as a Quantitative Probe of Gas-Liquid Distribution and Wetting Efficiency in Trickle-Bed Reactors” *Chem. Eng. Sci., 56,* 2615 - 2628.

Specchia, V. and Baldi, G. (1977) “Pressure drop and liquid holdup for two phase concurrent flow in packed beds” *Chem. Eng. Sci., 32,* 515 – 523.

Turpin, J. L., Huntington, R. L. (1967) *AIChE J.,* 13, 1191.

Van der Merwe, W. and Nicol, W. (2005) “Characterization of Multiple Flow Morphologies within the Trickle Flow Regime” *Ind. Eng. Chem. Res., 44,* 9446 – 9450.

Wang, R., Mao, Z. and Chen, J. (1995) “Experimental and Theoretical Studies of Pressure Drop Hysteresis in Trickle Bed Reactors” *Chem. Eng. Sci., 50, (14),* 2321 - 2328.

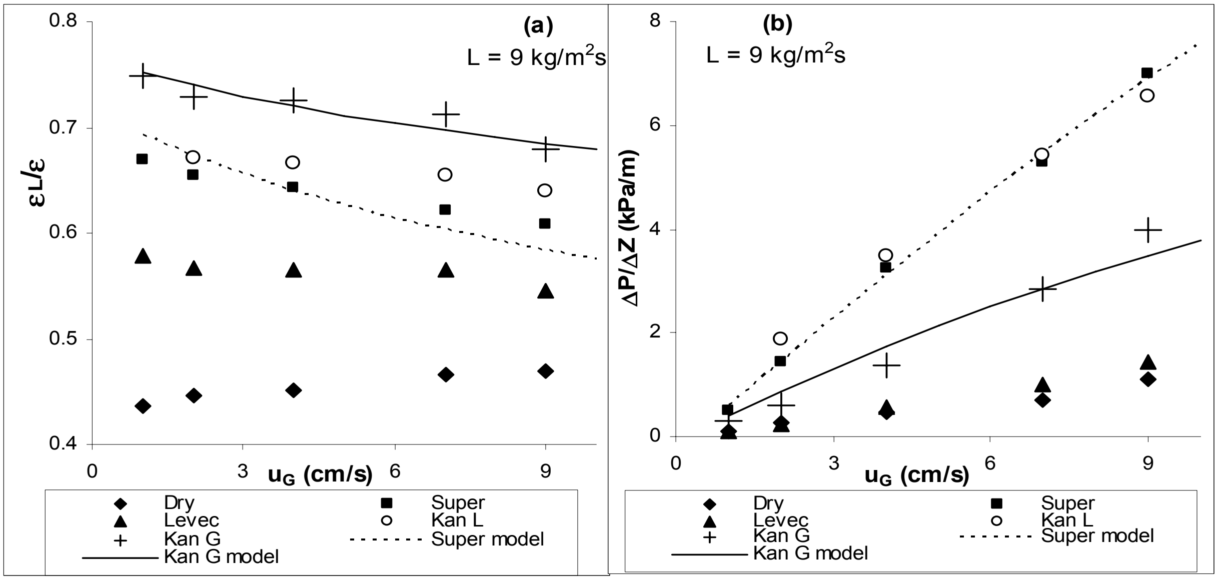

Figure 1. Selected pressure drop and holdup data and Kan & Greenfield model (1979) predictions

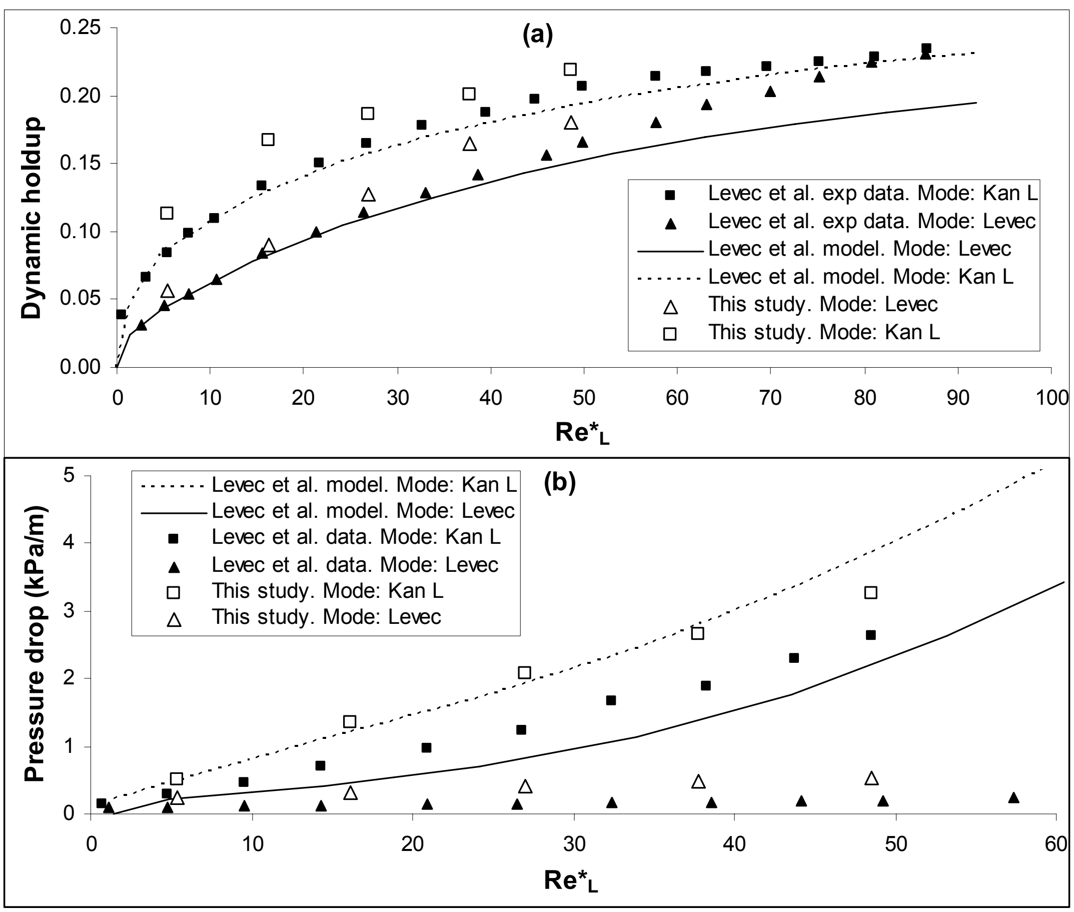

Figure 2. Selected data from Levec et al. (1986) and this study and Levec et al. model predictions

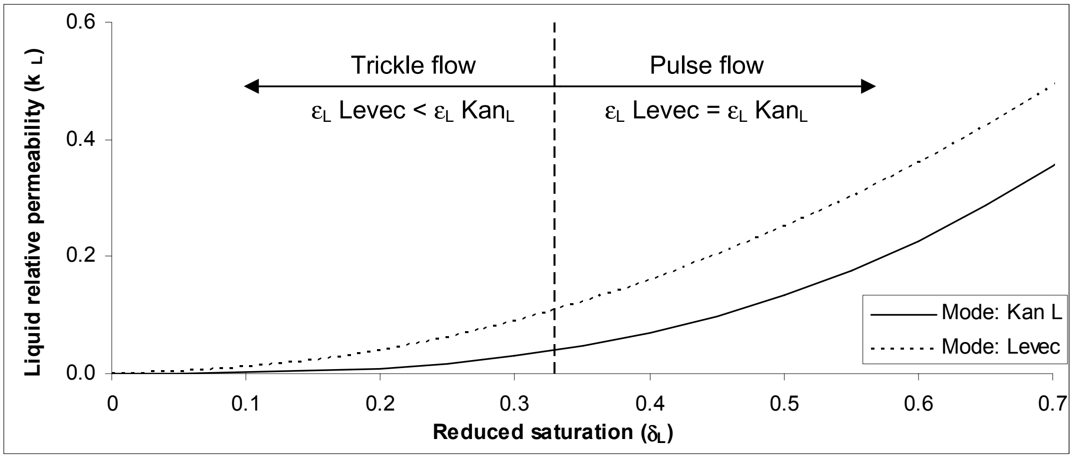

Figure 3. Liquid relative permeability vs. saturation curve (Levec et al. correlation)

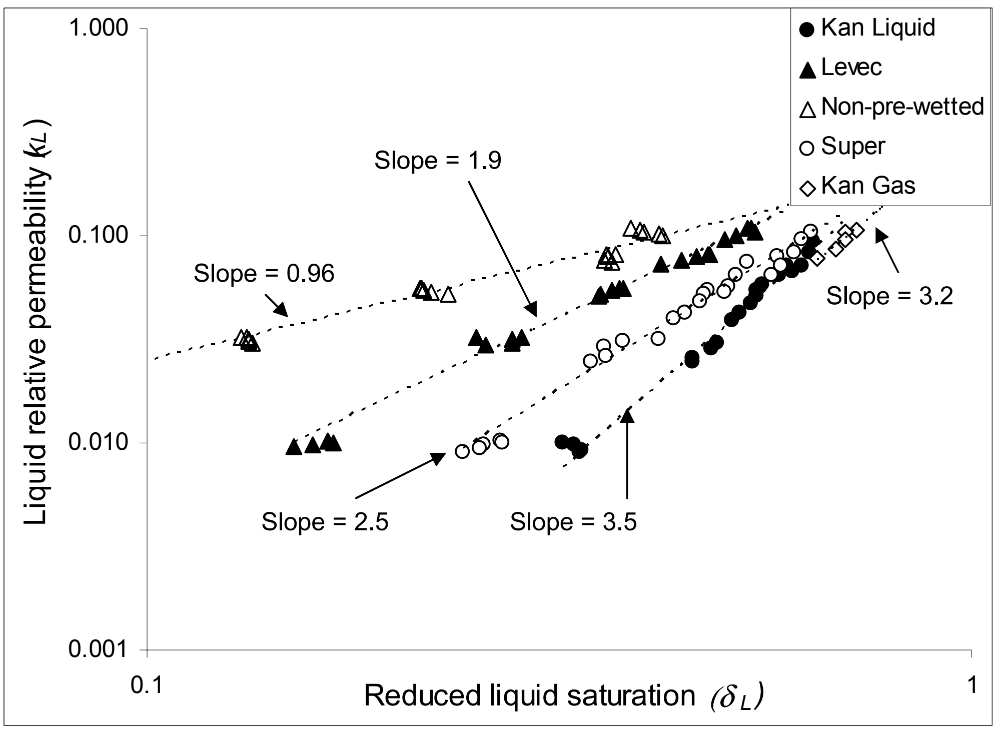

Figure 4. Liquid relative permeability vs. saturation curve(all data from this study)

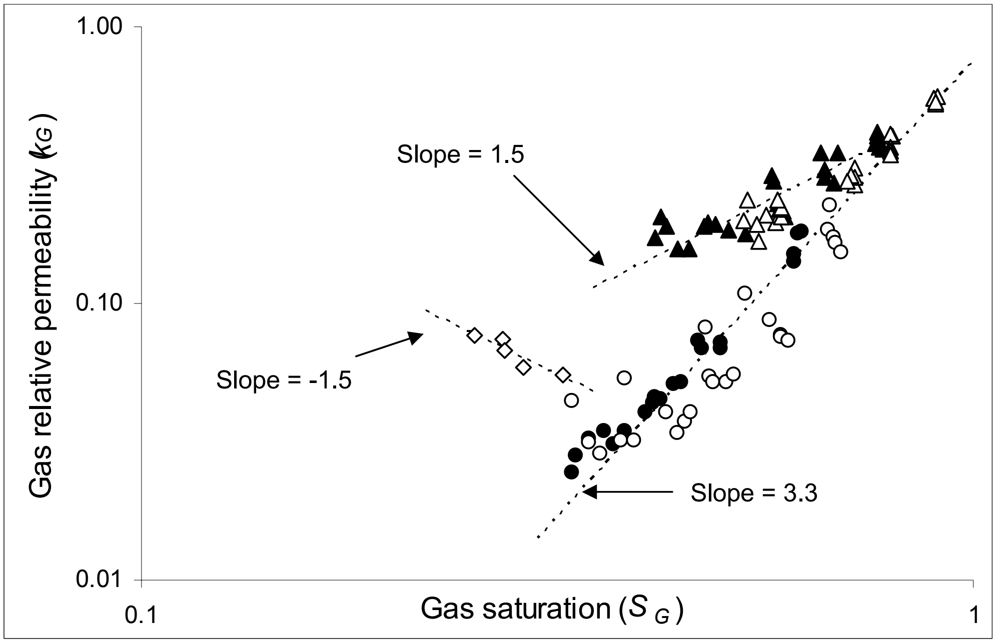

Figure 5. Gas relative permeability vs. saturation curve (all data from this study)