

# Characteristics of Liquid-Liquid Countercurrent Flow in Inclined Tubes: Application to the PTE Process

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A novel and promising extraction process termed Phase Transition Extraction (PTE) (Ullmann [1]; Ullmann et al. [2]) introduced a different approach to conventional extraction processes. The PTE process is based on the use of a class of solvents that have a critical solution temperature (CST) with the feed liquid to be extracted. In the PTE column, heating and cooling sections replace the mixing and settling sections of conventional extraction columns. The countercurrent feed and solvent streams passing those sections are heated and cooled across their coexistence curve and thereby undergo phase transitions, which alternate between states of two distinct liquid phases and a single homogeneous phase. The formation of a single phase in the mixing section results in a substantially superior contact between the solvents, eliminating the need for intense agitation. The continuous change in the composition of the phases during the phase separation process prevents the formation of a stable interface on which solids or emulsion-forming impurities can adhere. Consequently, the coalescence process is very fast, and the process is not sensitive to the presence of impurities or emulsifiers on of the crucial drawback of conventional extractors.

For all practical purposes, the time scale and therefore the throughput of extraction columns, as well as the PTE column, is dominated by the settling section. Although, the coalescence in the PTE process is very rapid, the maximal flow rates in the settling section are determined by flooding limitations. In the present study it is demonstrated that postponing the flooding, and thereby improving the column performance, can be accomplished in off-vertical inclined columns. It is worth noting, that conventional extraction columns must be operated at vertical position (Lo et al., [3]) for preventing segregation of the phases in the mixing sections. In the PTE column, on the other hand, a single phase is obtained in the mixing section and thus, its performance is not expected to deteriorate in an inclined column. The effect of inclination on the characteristics of countercurrent flow is studied both experimentally and theoretically.

The solvent system used in the experiments is a mixture of ethyl-acetate, water and ethanol. This solvent system has an upper "critical solution temperature" (CST) of 42° C. At room temperature, it forms two co-existing phases, with a density ratio of  $\rho_o/\rho_w=0.95$ , viscosity ratio of  $\mu_o/\mu_w=1.5$  and surface tension  $\sigma \approx 0.004$  N/m, where subscript w denotes the heavy (water-rich) phase and o denotes the light (organic-rich) phase.

The test column is mounted on a support system, which permits any inclination between 0 and 90 degree to the horizontal. The test column consists of a one-meter length, 14.4-mm I.D Pyrex pipe. The liquids flow under gravity from two feed reservoirs through a set of rotameters into the test section. Needle valves are used to control the liquids flow. The heavy phase is introduced at the column's upper end and removed from the bottom through an additional set of control valve and a

the light phase storage tank.

The flow patterns and the location of the phases interface are investigated through video recording. In order to avoid optical distortion, the inclined column is placed within a rectangular optical box filled with chemically pure Glycerin. In order to obtain easy identification of the interface level, a striped background was placed behind the optical box. To observe the velocity field, the liquids were seeded with small amount of tracer particles (hollow spherical glass beads 0.01 mm in diameter and density 1.1gr/cm<sup>3</sup>) and a light sheet illumination normal to the camera axis was used.

The effect of the tube inclination on the inception of flooding is shown in Figure 1 for equal flow rates of the heavy and light phases. The effect is dramatic. An off-vertical inclination of about 15<sup>o</sup> of the tube increases three folds the maximum throughput. This effect can be mainly attributed to the flow pattern transition from dispersed flow to stratified flow.

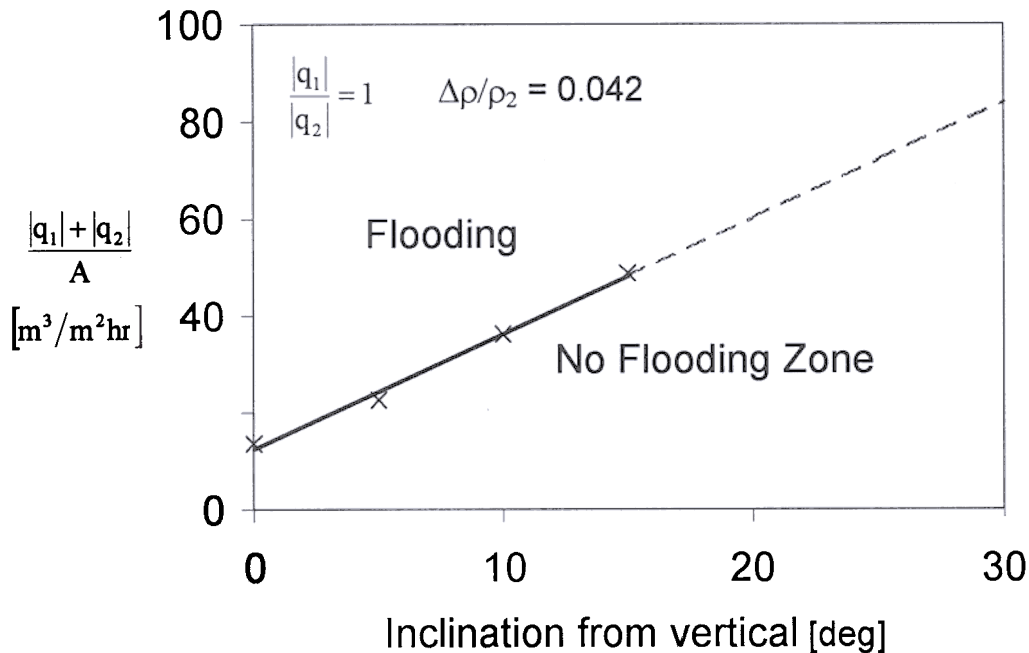


Figure Effect of tube inclination on flooding inception

In a vertical column, the basic flow pattern is dispersed flow, with either the heavy phase dispersed in the light phase (light phase dominated), or the light phase dispersed in the heavy phase (heavy phase dominated). These two configurations of dispersed flow can be simultaneously obtained in the column, separated by an interface. Obviously, each of the two modes is associated with a different in-situ holdup and thus, with a different pressure drop. Therefore, by manipulating the resistance at the heavy phase outlet, the location of the interface between the light-phase-dominated and heavy-phase-dominated zones can be located at any desired position along the column. With a sufficiently low resistance, the flow pattern in the entire column is light phase dominated (the interface is out of the column bottom). On the other hand, with a high resistance, the flow pattern is entirely heavy-phase dominated (the interface is out of the column top). When the interface is set within the column, the heavy phase dominated mode prevails at the lower section of the column, while in the upper section, the light phase dominated mode is obtained. With a slight off-vertical positioning of the column, the phases tend to segregate, even with the liquids of the small density differential used in this study. The two configurations obtained in this case correspond to stratified-dispersed flow of either heavy phase dominated or light phase dominated. In an inclined

flow rates

The experimental results show that, for given liquid flow rates and a fixed inclination, there exist two different stable configurations of stratified flow. One corresponds to a thick layer of the heavy phase flowing counter-currently to a thin layer of the light phase (heavy phase dominated), and the other configuration corresponds to a thin layer of the heavy phase (light phase dominated). Similarly to the operation of a vertical column, adjusting the resistance at the heavy phase outlet can control the location of the interface. Thereby, either of these two flow configurations can occupy the entire column, or both of them can exist simultaneously in the column.

The characteristics of countercurrent inclined flows were studied via investigation of the exact solutions obtained for the model of laminar flow through inclined plates. The flow geometry is schematically described in figure 2. The feed flow rates of the heavy and light phases (per unit width) are  $q_1$  and  $q_2$  respectively.  $H$  is the distance between the two plates and  $h$  is the depth of the heavy phase layer.

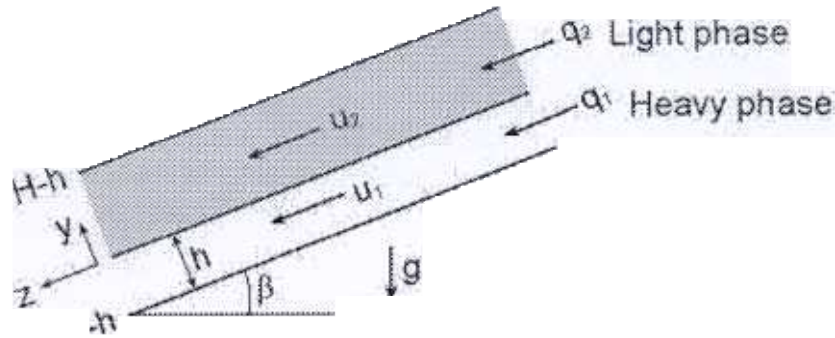


Figure 2. Schematic description of the streams in fully developed laminar flow

For laminar flow the fully developed velocity profiles,  $u_1(y)$ ,  $u_2(y)$  in the two layers are obtained by integration of the following two momentum equations:

$$\mu_1 \frac{\partial^2 u_1}{\partial y^2} = \frac{\partial p}{\partial z} - \rho_1 g \sin(\beta) \quad -h < y < 0 \quad (1)$$

$$\mu_2 \frac{\partial^2 u_2}{\partial y^2} = \frac{\partial p}{\partial z} - \rho_2 g \sin(\beta) \quad 0 < y < H-h \quad (2)$$

subjected to the following boundary conditions:

$$\begin{aligned} u_1 \Big|_{y=-h} &= 0 & u_2 \Big|_{y=H-h} &= 0 \\ u_1 \Big|_{y=0} &= u_2 \Big|_{y=0} & \mu_1 \frac{\partial u_1}{\partial y} \Big|_{y=0} &= \mu_2 \frac{\partial u_2}{\partial y} \Big|_{y=0} \end{aligned} \quad (3)$$

Integrating the resulting velocity profiles over the corresponding layer thickness and equating the results to the feed flow-rates yields explicit analytical expressions for the inclination parameter,  $Y$  and the pressure drop parameter,  $P$ :

$$Y = \frac{1}{4} \frac{\mu q (1 - \tilde{h})^2 [(1 + 2\tilde{h})\mu + (1 - \mu)\tilde{h}(4 - \tilde{h})] - \tilde{h}^2 [(3 - 2\tilde{h})\mu + (1 - \mu)\tilde{h}^2]}{\tilde{h}^3 (1 - \tilde{h})^3 [\tilde{h} + \mu(1 - \tilde{h})]} \quad (4)$$

$$4 \tilde{h}(1 - \tilde{h})^2[(1 + 2\tilde{h})\mu + \tilde{h}(1 - \tilde{h})(4 - \tilde{h}) - 3\tilde{h}] \quad (5)$$

where:

$$Y = \frac{(\rho_1 - \rho_2)g \sin(\beta)}{\left(-\frac{\partial p}{\partial z}\right)_{2s}} \quad P = \frac{\left(\frac{\partial p}{\partial z}\right) - \rho_2 g \sin(\beta)}{\left(-\frac{\partial p}{\partial z}\right)_{2s}} \quad (6)$$

$$q = \frac{q_1}{q_2} \quad \mu = \frac{\mu_1}{\mu_2} \quad \tilde{h} = \frac{h}{H} = \text{Holdup} \quad (7)$$

and  $(-\partial p/\partial z)_{2s} = 12\mu_2 q_2/H^3$  is the superficial frictional pressure drop of the lighter phase.

The model equations are presented in a unified form, that is applicable both to countercurrent and co-current flows note that for countercurrent flow  $q_2$  is negative (the light phase flowing upward), hence  $q$  is negative.

Figure 3 shows the predicted holdup of the heavy phase as a function of the reciprocal of the Martinelli parameter (which for laminar flow is  $X^2 = q\mu$ ) at constant  $Y/X^2$  and  $\mu$ . As one can see, the model predicts the existence of a double-solution for countercurrent inclined flows. The experimental results are also plotted in Figure 3 and more clearly in Figure 4.

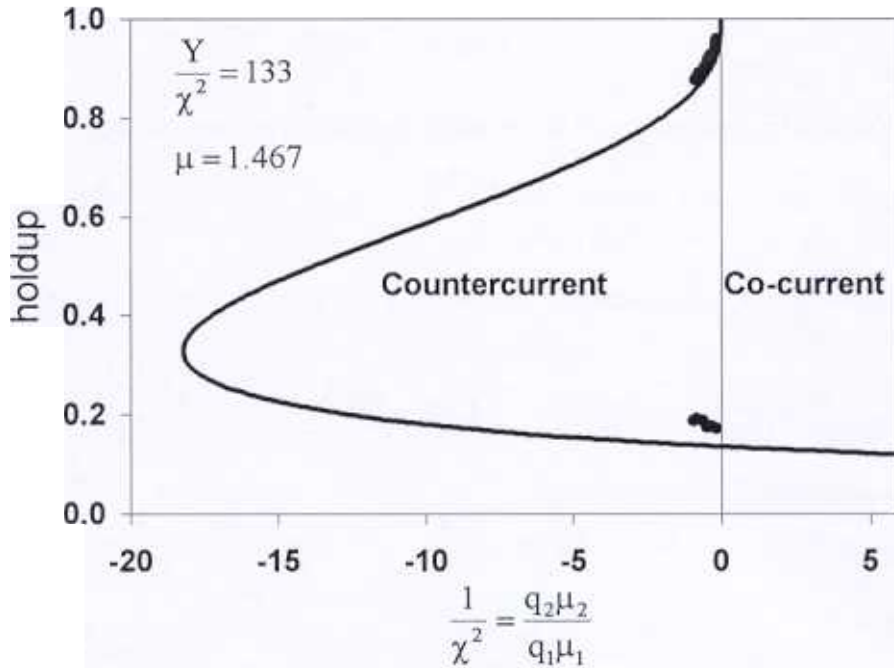


Figure 3. The two-plate model prediction for the heavy phase holdup

The agreement between the model, which was developed for laminar flow between parallel plates, and the experimental results obtained in tube (for the same superficial velocities) is quite remarkable. Moreover, the model confirms the existence of double-solution of the in-situ holdup for specified liquid flow rates.

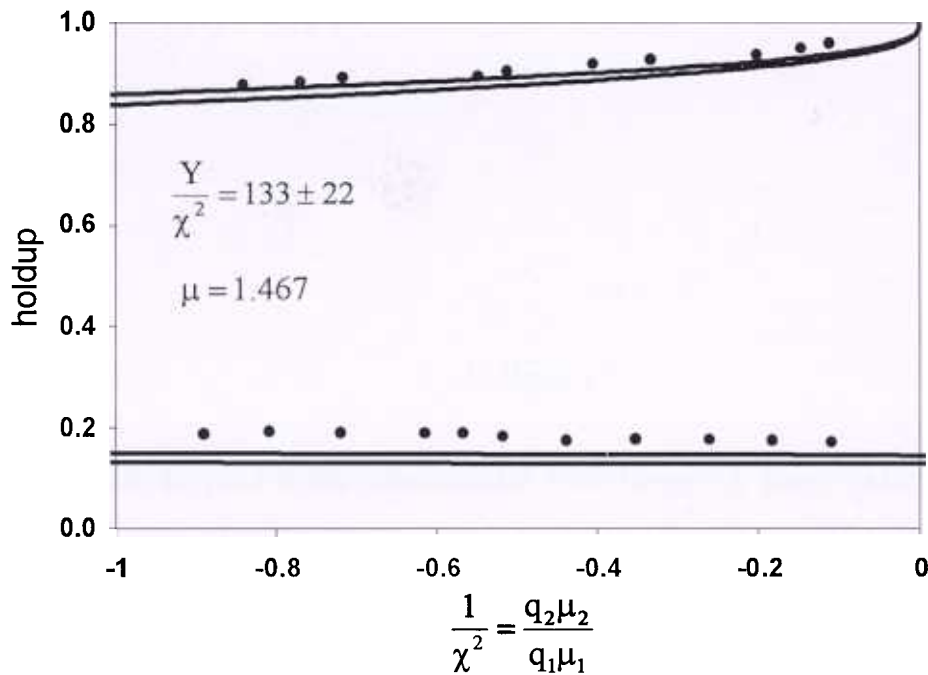


Figure 4. Comparison between the two plates model and experimental results

Experimental study of the flow characteristics was carried out over wide ranges of liquid flow rates and tube inclinations. The results reveal that inclination of pipes postpones the flooding inception via stratification of the countercurrent streams and that countercurrent stratified flow is feasible and stable even for  $\Delta\rho/\rho_2 \ll 1$ . Thus, inclining the PTE column can increase the process throughput. Whereas, owing to the PTE process characteristics, the inclination does not reduce the column efficiency.

#### REFERENCES

1. Ullmann, A., Ludmer, Z., and Shinnar, R., Phase Transition Extraction Using Solvent Mixtures with a Critical Point of Miscibility, *AIChE J.*, 41 (3) (1995) 488-500.
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