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# Minimal dissipation rate approach to correlate phase inversion data

P. Poesio\*, G.P. Beretta

Università di Brescia, Via Branze 38, 25123 Brescia, Italy

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#### ABSTRACT

Phase inversion refers to the phenomenon whereby a small change in operational flow conditions causes an oil-in-water dispersed flow pattern to suddenly switch to a water-in-oil flow pattern, and viceversa. This paper proposes an interpretation of phase inversion in terms of minimal dissipation rate. To this end, the dissipation rate is computed by a simple homogeneous model together with available correlations for effective viscosity in dispersed flows. It is shown that the data available in the literature can be reasonably interpreted as a manifestation of minimal dissipation rate. Furthermore, if the assumed effective viscosity correlations take into account pipe wettability, the minimal dissipation rate approach is capable to interpret also the so-called ambivalent range (hysteresis effect) and correlate the available data

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# 1. Introduction

The flow of two immiscible liquids is often encountered in industrial applications such as food, pharmaceutical, and oil industries. The possibility to predict the flow characteristics - such as flow pattern or pressure drop - is essential both to design and to maintain industrial facilities. Dispersed flow is a common flow pattern which consists of a continuous phase in which the other phase is dispersed in the form of droplets, Brauner (1998). A water-in-oil dispersion  $(D_{\text{w/o}})$  is a dispersion of aqueous phase in the organic phase. Conversely, oil-in-water dispersion (Do/w) is a dispersion of organic phase in the aqueous phase. At a particular value of the hold-up  $\epsilon$  – in situ oil or water fraction – phase inversion occurs; phase inversion refers to the phenomenon where, with a small change in the operational conditions, the continuous phase and the discrete phase invert. So, for instance, a water-in-oil dispersion (D<sub>w/o</sub>) suddenly switches into an oil-in-water dispersion  $(D_{o/w})$ .

Phase inversion plays a major role in the design of industrial equipment since it affects mass, momentum, and heat transfer efficiency and hence it needs to be accounted for. For instance in petroleum industry where water and oil are frequently transported together, the flow pattern strongly affects the pressure drop; furthermore, corrosion, and hence inhibition agents injection, strongly depends on the wetting phase. In settling-tanks, where the settling time depends on the viscosity of the continuous phase, the correct prediction of the flow pattern  $(D_{\text{W/o}} \text{ or } D_{\text{o/w}})$  proves essential for a proper design of the process. In petrochemical

Studies on phase inversion have been carried out in stirred tanks, batch mixers, continuous mixers, and pipe flows (see Yeo et al. (2000) and references therein) in the attempt to establish a relation that allows to predict the hold-up at phase inversion (i.e., the critical hold-up,  $\epsilon^I$ ) as a function of the fluid properties (viscosity ratio, material wettability, and surface tension). In this paper we focus our attention on phase inversion in pipe flow. However the approach we propose has a broader validity and can be easily extended and applied to different geometries such as stirred vessels.

Although phase inversion has been a problem for more than 40 years, little is known about it and, for the design of industrial equipment, engineers have relied mainly on empirical correlations. For instance, to compute the critical water hold-up  $\epsilon_{\rm w}^l$  at which phase inversion takes place in oil-water flow, Arirachakaran et al. (1989) proposed, as a best fit of their experimental data, the following relation:

$$\epsilon_{\rm w}^{\rm I} = 0.5 - 0.1088 \log_{10} \left( \frac{\mu_{\rm o}}{\mu_{\rm r}} \right),$$
 (1)

where  $\mu_{\rm r}$  = 1 mPa s. Eq. (1) allows to estimate the critical water fraction  $\epsilon_{\rm w}^l$  at which phase inversion occurs as a function of the oil viscosity  $\mu_{\rm o}$ . Another empirical correlation to compute the critical water hold-up  $\epsilon_{\rm w}^l$  was suggested by Nadler and Mewes (1997)

$$\epsilon_{\mathbf{w}}^{I} = \frac{1}{1 + k_{1} \left[ \frac{C_{0}}{C_{\mathbf{w}}} \frac{\rho_{0}^{(1-n_{0})}}{\rho_{0}^{(1-n_{w})}} \frac{\mu_{0}^{(n_{0})}}{\rho_{\mathbf{w}}^{(n_{w})}} (DU_{\mathbf{m}})^{n_{\mathbf{w}} - n_{0}} \right]^{1/k_{2}}}, \tag{2}$$

plants, where for the refinery process oil—water mixtures pass through heat exchangers, the knowledge of which phase is the continuous one is essential for a correct prediction of the heat transfer.

<sup>\*</sup> Corresponding author.

E-mail address: pietro.poesio@ing.unibs.it (P. Poesio).

where D is the pipe diameter,  $\rho_{o,w}$  is the oil or water density,  $\mu_{o,w}$  the oil or water viscosity,  $U_{\rm m}$  the mixture velocity;  $C_{\rm o}$ ,  $C_{\rm w}$ ,  $n_{\rm o}$ , and  $n_{\rm w}$  are the parameters of the friction factor correlation  $CRe^{-n}$  for pure oil and pure water pipe flow;  $k_1$  and  $k_2$  are empirical parameters. All the details on the application of Blasius-like relations to two-phase flows can be found in Brauner and Ullman (2002).

Relations (1) and (2) provide a single value of the hold-up at which inversion takes place once the fluids' properties are known. However, it is found experimentally that a so-called *ambivalent* range of hold-up values exists, in which both phases may be found to be continuous. Only outside this range one phase is always continuous and the other is always discrete. Within the ambivalent range, either one of the two phases can be continuous and the exact value at which phases invert depends on the operating conditions, on surface tension, and on pipe wettability. If the prediction of ambivalent range is neglected, the critical hold-up depends mainly on the viscosity ratio of the two fluids.

Even if empirical correlations are widely used, some attempts to predict theoretically the critical hold-up for phase inversion have been carried out. Several mechanisms have been postulated to predict both phase inversion and the existence of the ambivalent range. The most important are based on

- instability between droplets coalescence and break-up;
- minimization of the total free energy content (including both gravitational potential energy and interfacial energy);
- effects of dynamic forces.

Phase inversion was explained as an instability between breakup and coalescence of dispersed drops. Phase inversion takes place when there is a large coalescence frequency and the break-up mechanism is insufficiently fast. Up to a certain volume fraction, break-up and coalescence processes can reach a dynamical balance; near the inversion point, instead, this balance cannot be sustained anymore and the coalescence rate overcomes the break-up rate. Droplets grow and their shapes change from spherical to cylindrical to lamellae and, eventually, a complex structure is reached, leading to the formation of the new continuous phase. Recently, detailed laser induced fluorescence (LIF) experiments have shown the appearance of a complex flow structure at the inversion point. The presence of secondary dispersions is observed both in stirred vessels - see Liu et al. (2005) - and in pipe flow - Piela et al. (2006). Experiments - Yeo et al. (2000) - have shown that all the factors that affect droplet break-up/coalescence rate (liquid properties, electrostatic interactions, wettability, and shear rate) influence the phase inversion as well as the ambivalent range. So, phase inversion can be regarded as an instability in the dynamic balance between break-up and coalescence. The ambivalent range corresponds to the difference in the onset of this instability in the aqueous and organic continuous dispersions.

Since phase inversion is a phenomenon that occurs spontaneously, other authors proposed that its prediction can be based on a criterion of minimization of the total energy content, including interfacial energy, of the flowing system. Such an approach was originally proposed by Luhning and Sawistowski (1971) and Tidhar et al. (1986). Recently, following this line, Brauner and Ullman (2002) proposed a new model to predict phase inversion. According to this model, the critical oil hold-up at which phase inversion occurs can be computed as

$$\epsilon_{\rm o}^{\rm I} = \frac{[\sigma/d_{32}]_{\rm w/o} + \frac{s}{6}\cos\theta}{[\sigma/d_{32}]_{\rm w/o} + [\sigma/d_{32}]_{\rm o/w}},\tag{3}$$

where  $\theta$  is the water wettability angle,  $\sigma$  the oil–water interfacial tension,  $d_{32}$  the Sauter mean diameter, and s the wetted perimeter. The prediction of the Sauter diameter  $d_{32}$  involves extensive modelling for the break-up/coalescence of droplets. Because in general the

application of the minimum free energy approach relies on a model able to characterize the drop size, several attempts have been made in the literature to predict the typical drop size, for example by means of a population balance equation – Hu et al. (2006) – or a Monte Carlo simulation, Yeo et al. (2002).

The minimum energy criterion *per se* cannot predict the presence of the ambivalent range, nevertheless Yeo et al. (2002) were able to predict it by calculating the drop Sauter mean diameter  $(d_{32})$  from two different correlations in the o/w and w/o dispersions. On the contrary, Brauner and Ullman (2002) claimed that a possible reason for the hysteresis (ambivalent) range is the time needed after phase inversion for the new continuous phase to completely wet the pipe wall. The understanding and the prediction of the ambivalent range require further investigations, both experimental and theoretical.

Yeh et al. (1964) suggested another mechanism by which dispersed flow can be regarded as the flow of three thin layers: the dispersed phase, the interfacial phase, and the continuous one. Dynamic forces are considered to play a major role in the determination of the dispersion type. Assuming no shear at the interface, they derived the concentration ratio of dispersed to continuous phase at inversion as a function of pure liquid viscosities. Assuming all the layers in the model are laminar, the relation obtained by Yeh et al. (1964) can be written as

$$\epsilon_{\rm w}^{\rm I} = \frac{1}{1 + \left(\frac{\mu_0}{\mu_{\rm w}}\right)^{0.5}}.\tag{4}$$

The aim of this paper is to provide an alternative tool to interpret inversion data. In Section 2, we propose our interpretation for phase inversion based on minimal energy dissipation rate reasoning. In Section 3 we interpret literature results (analytical model, fitting relations, and experimental data) in terms of minimal dissipation rate. In Section 4, we present a first attempt to predict and model the ambivalent range. Conclusions are given in Section 5.

# 2. Phase inversion model and minimal dissipation rate

In this paper, we want to show that phase inversion can be interpreted in terms of minimal dissipation rate and, accordingly, the flow pattern that develops (i.e.,  $D_{\text{O/w}}$  or  $D_{\text{W/O}}$ ) is the one that, for a given hold-up, minimizes the dissipation rate. As a first step we compute the dissipation rate as a function of hold-up, first assuming the oil to be continuous and the water discrete, and then the converse. Assuming incompressible Newtonian flow, the dissipation rate is given in general by the relation

$$\Phi = 2 \int_{V_1 \cup V_2} \mu s_{ij} s_{ij} \, dV = 2 \int_{V_1} \mu_1 s_{ij} s_{ij} \, dV_1 + 2 \int_{V_2} \mu_2 s_{ij} s_{ij} \, dV_2, \tag{5}$$

where  $\mu$  is the fluid viscosity and  $s_{ij}$  the shear rate; integration is carried out over the entire two-phase domain  $V_1 \cup V_2$ . In case of pipe flow, the dissipation amounts to

$$\Phi = \left(\frac{\dot{m}_{o}}{\rho_{o}} + \frac{\dot{m}_{w}}{\rho_{w}}\right) \Delta p, \tag{6}$$

where  $\Delta p$  is the pressure drop along the pipe and  $\dot{m}$  are the oil and water mass flow rates, respectively. To use Eq. (6) it is necessary to estimate the pressure drop  $\Delta p$  where  $\Delta p = L \cdot dp/dx$  and L is the pipe length. To this end, we use the homogeneous model according to which the pressure gradient dp/dx can be found as

$$\frac{\mathrm{d}p}{\mathrm{d}x} = 2f_{\mathrm{m}} \frac{\rho_{\mathrm{m}} U_{\mathrm{m}}^2}{D},\tag{7}$$

where  $U_{\rm m}=4(\dot{m}_{\rm o}/\rho_{\rm o}+\dot{m}_{\rm w}/\rho_{\rm w})/\pi D^2$  is the mixture velocity,  $\rho_{\rm m}=\rho_{\rm w}\epsilon_{\rm w}+\rho_{\rm o}\epsilon_{\rm o}$  is the mean mixture density, D is the pipe diameter,

and  $\epsilon_{\rm w}$  = 1  $-\epsilon_{\rm o}$ . For the mixture friction factor  $f_{\rm m}$  we assume a Blasius relation

$$f_{\rm m} = \frac{C}{Re_{\rm m}^n},\tag{8}$$

where the values of C and n depend on the flow regime and the mixture Reynolds number  $Re_{\rm m}$  is defined by means of an effective viscosity  $\mu_{\rm m}$ ,

$$Re_{\rm m} = \frac{\rho_{\rm m} U_{\rm m} D}{\mu_{\rm m}}. \tag{9}$$

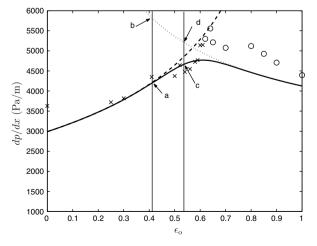
If the flow is laminar, i.e.,  $Re_{\rm m} \leqslant 2300$ , C=64 and n=1; if the flow is turbulent, i.e.,  $Re_{\rm m} \geqslant 2300$ , C=0.079 and n=0.25. Of course, there exist more complex relations to compute  $f_{\rm m}$  that account for pipe roughness and for the non-Newtonian behaviour of highly concentrated mixtures; here, we restrict our analysis to this simple relation.

For the effective viscosity,  $\mu_{\rm m}$ , of a dispersed system, different relations have been proposed and analysed. For dilute dispersions, the well-known Einstein's relation can be used; for highly concentrated mixtures, more sophisticated relations need to be used in order to account for droplet–droplet interactions; a collection of these relations is reported by Brauner (1998) and Hu et al. (2005); the simplest relation suitable for highly concentrated mixtures of solid mono-dispersed spheres is proposed by Ball and Richmond (1980)

$$\mu_{\rm m} = \mu_{\rm c} (1 - K \epsilon_{\rm d})^{-5/(2K)},$$
 (10)

where  $\mu_c$  is the continuous phase viscosity,  $\epsilon_d$  the discrete phase hold-up, and 1/K can be interpreted as the maximum packing fraction. For a mono-dispersed suspension of rigid spheres the maximum packing fraction can be calculated to be 0.58, however it is shown by Pal (1993) that in case of emulsions the maximum packing fraction can be well above 0.58 and up to 0.95. For that reason, in the case of deformable drops, 1/K may be assumed equal to 1.

Using Eqs. (7)–(10) the pressure drop is computed as a function of the hold-up and shown, for example, in Fig. 1. If water is assumed to be the continuous phase for all values of the hold-up, the resulting pressure drop is given by the dashed curve. Similarly, if oil is assumed to be the continuous phase for all values of the hold-up value, the resulting pressure drop is given by the dotted curve. Fig. 1 shows the two predicted pressure drops compared to the measured values. As long as water is actually the continuous



**Fig. 1.** Example of the application of Eqs. (7)–(10) for turbulent flow of oil droplets in water (dashed curve,  $D_{o/w}$ ) and water droplets in oil (dotted curve,  $D_{w/o}$ ). Points represent experimental data: pipe diameter D = 20 mm, water superficial velocity 2 m/s, oil superficial velocity 1.5 m/s, viscosity ratio  $\tilde{\mu} = 24$ , density ratio  $\tilde{\rho} = 0.79$ .

phase, the pressure drop is predicted correctly and the same holds to some approximation when oil is the continuous phase.

Our reading of this result is that the system chooses the flow pattern that, for a given oil hold-up  $\epsilon_0$ , provides the lowest pressure drop, i.e., the minimum energy dissipation. If this is true, as it appears from Fig. 1, the value at which the two curves cross each other is the inversion point and the corresponding hold-up  $\epsilon_0$  is the critical value,  $\epsilon_0'$ .

So, the construction of Fig. 1 involves the following steps:

- the asymptotic behaviours are considered separately; first the pressure drop is computed, using Eqs. (7)–(10) under the assumption that an oil-in-water dispersion occurs for all values of the oil hold-up (dashed curve). Similarly, the pressure drop is computed assuming that a water-in-oil dispersion occurs for all values of the oil hold-up (dotted line).
- An estimate of the actual pressure drop the continuous line in Fig. 1 – is computed by an asymptotic matching interpolation of the two curves, i.e., the resulting pressure drop is computed by the relation

$$\frac{1}{\left(\frac{\mathrm{d}p}{\mathrm{d}x}\right)^a} = \frac{1}{\left(\frac{\mathrm{d}p}{\mathrm{d}x}\right)_{\mathrm{n/w}}^a} + \frac{1}{\left(\frac{\mathrm{d}p}{\mathrm{d}x}\right)_{\mathrm{w/o}}^a},\tag{11}$$

where the exponent a was chosen equal to 10 to best fit the data. • The value of the hold-up corresponding to the maximum of the interpolated pressure drop curve is an estimate of the critical value,  $\epsilon^I$ . A simpler alternative estimate is the value at which the two curves cross.

The predicted pressure drop curve compares reasonably well with the experimental data. Fig. 1 gives us a possible reading key of phase inversion: let us assume to start with a single phase flow of pure water ( $\epsilon_0$  = 0) and then to gradually add oil; since there is a two-phase flow, at each value of  $\epsilon_0$  the system has to choose between  $D_{o/w}$  and  $D_{w/o}$ . For instance, if we look at the case corresponding to  $\epsilon_0$  = 0.42, the system can be either in state a ( $D_{o/w}$ ) or in state b ( $D_{w/o}$ ); experimentally, it is observed that the system is in state a, i.e., the one of minimal dissipation rate. Similarly, if  $\epsilon_0$  = 0.56, state c (rather than state d) is chosen (the corresponding flow rate being  $D_{0/w}$ ) and, again, this is the flow regime for which the dissipation rate is minimal. If we keep adding oil, the pressure gradient increases but the flow pattern does not change up to  $\varepsilon_o \sim 0.61$  where, a small further increase in oil flow rate causes a dramatic change: the flow pattern suddenly switches from Do/w to D<sub>w/o</sub>. Phase inversion has occurred. Fig. 1 shows that phase inversion can be interpreted as the spontaneous tendency of the flow to switch to the flow pattern which minimizes the dissipation rate (pressure gradient). A more systematic analysis of this behaviour is carried out in Section 3. As a final remark, it is worth noting that the ambivalent range is not caught and this limitation is due to the very simple relation for the mixture viscosity assumed with Eq. (10). The possibility to include the behaviour of the system at the ambivalent range is discussed in Section 4.

Notice also that because of the homogeneous model assumption and of setting 1/K = 1 in Eq. (10), our simple model yields, for both laminar and turbulent flow and regardless of the values of  $\rho_{\rm w}$ ,  $\rho_{\rm o}$ , D, and  $U_{\rm m}$ , the following expressions:

$$\epsilon_{\rm w}^{\rm I} = \frac{1}{1 + \left(\mu_{\rm o}/\mu_{\rm w}\right)^{2/5}}, \quad \epsilon_{\rm o}^{\rm I} = \frac{1}{1 + \left(\mu_{\rm o}/\mu_{\rm w}\right)^{-2/5}} \tag{12}$$

for the hold-up at inversion. Of course, this is because in this model we neglect wettability and surface tension. The choice of K = 1 was suggested by Pal (1993) since it gives a good agreement on the pressure drop prediction; if we assume  $K \neq 1$  then the critical hold-up, in case of both laminar and turbulent flow, is given by

$$\epsilon_{\rm o}^{\rm I} = \frac{1 - \tilde{\mu}^{-2/5K} + K\tilde{\mu}^{-2/5K}}{1 + \tilde{\mu}^{-2/5K}}, \tag{13}$$

where  $\tilde{\mu} = \mu_0/\mu_w$ .

# 3. Comparison with theoretical models, empirical correlations, and experimental results

In this section, we extensively check if phase inversion can be interpreted in term of minimal dissipation rate, i.e., if the flow pattern that develops (either  $D_{o/w}$  or  $D_{w/o}$ ) is indeed the one minimizing the dissipation rate. To this end, we compare, by using the homogeneous model, the dissipation rate associated to each flow pattern and we check if the configuration with the lowest dissipation rate is the one that actually obtains. The most suitable way to check if the data can be interpreted in such a way is to look at the critical hold-up since, usually, the data on phase inversion are presented in this form. In Fig. 2 the critical oil hold-up  $\epsilon_o^l$  is plotted as a function of the dynamic viscosity ratio  $\tilde{\mu} = \mu_0/\mu_w$ .

First we check if the most used fitting relations predict values that can be interpreted as a manifestation of the minimal dissipation rate approach. It is not always clear how such fitting relations have been derived, i.e., we are not always aware of the experimental conditions in which the experimental points were taken, nevertheless we trust that this comparison is very important since these empirical relations are widely used to design industrial equipment. From Fig. 2a, it appears that only the relation proposed by Nadler and Mewes (1997) (Eq. (2) with  $n_{\rm w}=n_{\rm o}$ ,  $C_{\rm w}=C_{\rm o}$ ,  $k_1=1$ , and  $k_2=2$ ) can be reasonably interpreted by the minimal dissipation rate approach, while the other relations do not confirm this tendency. Note that the curve proposed by Brauner and Ullman (2002) and plotted in Fig. 2a is an empirical fitting law,  $\epsilon_0^I=\bar{\mu}^{0.22}/(1+\bar{\mu}^{0.22})$ , based on the data shown in Fig. 2c. Brauner and Ullman (2002) collected a large number of data on the critical hold-up for phase inversion and, out of them, they proposed the previous fitting relation. Such a relation, plotted in Fig. 2c, may appear not very good since only part of the experimental data points are plotted here.

Next we compare theoretical models. Fig. 2b shows that the minimal dissipation rate approach correlates fairly well with the predictions of the theoretical model proposed by Yeh et al. (1964), especially at low viscosity ratios. On the same graph, we carry out also the comparison with the model proposed by Brauner and Ullman (2002). To do so, we assume that oil–water surface tension in the pre-inversion and post-inversion dispersions is the

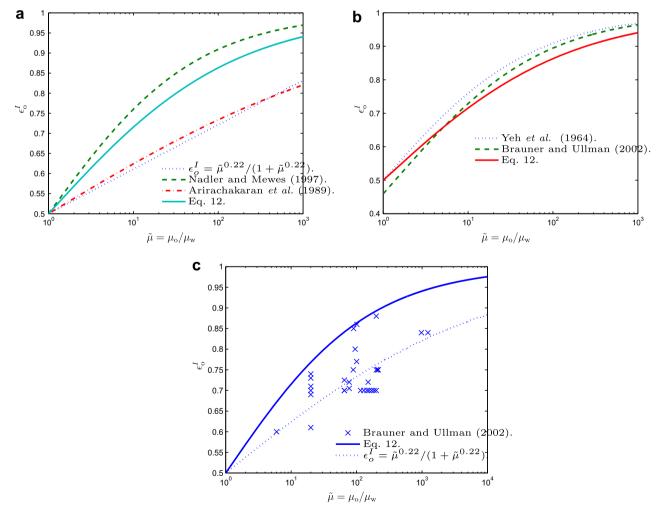


Fig. 2. Comparison of our interpretation of phase inversion data based on minimal dissipation rate with empirical correlations, Fig. 2a, with theoretical models, Fig. 2b, and with experimental data, Fig. 2c. The relation  $\epsilon_0^i = \bar{\mu}^{0.22}/(1 + \bar{\mu}^{0.22})$  is a fitting curve proposed by Brauner and Ullman (2002) and based upon several experimental data points found in the literature. A part of those experimental data are reported in Fig. 2c. This fitting curve may appear not very good since only a part of the experimental data is plotted here. (a) Comparison between our model and some fitting relations. (b) Comparison between Eq. (12) and the theoretical model proposed by Yeh et al. (1964) and the one proposed by Brauner and Ullman (2002). (c) Comparison between Eq. (12) and the experimental values in Brauner and Ullman (2002) which refer to pipe flow. The experimental points correspond to data obtained with diameters between 0.01 to 0.04 cm, with  $\bar{\rho} = 0.825-0.87$ , and  $\bar{\mu} = 4.9-1450$ .

same, that the constants  $(k_d)_{o/w}$  and  $(k_d)_{w/o}$  of Brauner and Ullman (2002) are approximately equal, and that the solid-liquid wettability effects can be neglected. Under these assumptions, it can be proved that Eq. (3) reduces to

$$\epsilon_0^I = \frac{\tilde{\rho}\tilde{v}^{0.4}}{1 + \tilde{\rho}\tilde{v}^{0.4}},\tag{14}$$

where  $\tilde{v}=v_{\rm o}/v_{\rm w}$  is the kinematic viscosity ratio, and  $\tilde{\rho}=\rho_{\rm o}/\rho_{\rm w}$  the density ratio. In order to plot on the same graph Eqs. (12) and (14), in Fig. 2b we further assume a density ratio  $\tilde{\rho}=0.85$  which is a reasonable value for oil–water mixtures (all the experimental points in Fig. 2c are obtained for  $\tilde{\rho}=0.825-0.87$  and  $\tilde{\mu}=4.9-1450$ ). As a result, Fig. 2b shows that our interpretation compares reasonably well with the model proposed by Brauner and Ullman (2002). Of course, for  $\tilde{\rho}=1$ , Eqs. (12) and (14) become identical and the agreement is exact.

Finally, our approach is compared against the experimental data for pipe flow presented by different authors and collected by Brauner and Ullman (2002). The agreement is reasonable, but the experimental data show a large scatter due to the presence of the ambivalent range which is not accounted for in our model. For instance, if we focus our attention on the experiments carried out at  $\tilde{\mu}=21$ , we notice that the critical hold-up ranges from 0.6 to 0.73, clearly demonstrating the existence of the ambivalent range.

# 4. The ambivalent range

As mentioned in the Introduction, it is found experimentally that there exists a range of hold-up values in which, for some specific conditions, both phases can be either dispersed or continuous - the so-called *ambivalent range*. The interpretation of phase inversion proposed in the previous section is not able to describe the existence of such a region due to the overly simple approach of the homogenous model that assumes, by definition, the phases to be homogenously dispersed one in the other. A possible remedy is the use of a modified effective mixture viscosity. Indeed, the ambivalent region can be accounted for in our description provided that Eq. (10) for the effective viscosity is substituted with a more sophisticated relation taking into account the drop size, the non-Newtonian behaviour of the emulsion near the inversion point, and, even more important, the wetting behaviour of the pipe wall. According to some experimental results, the wettability behaviour of the pipe wall plays a key role in the determination of the ambiv-

**Table 1**Fitting constants, Eq. (15), of the experimental data by loannou et al. (2005)

Pipe	Direction	Pattern	$K_1$	K <sub>2</sub>
S	$0 \rightarrow W$	D <sub>o/w</sub>	1.2	1.1
S	$0 \rightarrow W$	$D_{w/o}$	0.93	0.83
S	$W \rightarrow 0$	$D_{o/w}$	1.19	0.98
S	$W \rightarrow 0$	$D_{w/o}$	0.91	1.00
A	$0 \rightarrow W$	$D_{o/w}$	1.1	0.7
Α	$0 \rightarrow W$	$D_{w/o}$	1.03	0.82
Α	$W \rightarrow 0$	$D_{o/w}$	1.21	0.85
Α	$W \rightarrow 0$	$D_{w/o}$	0.98	1.12

S: steel pipe; A: acrylic pipe;  $w \to o$ : pipe flow experiments started with pure water followed by gradual increase of the oil flow rate;  $o \to w$ : experiments started with pure oil followed by gradual increase of the water flow rate.

alent range, so we have tried to include such an effect into a modified effective viscosity. In general the effective viscosity depends on fluids properties and on the drop size, whereas wettability influences the exact flow pattern in that the wetting phase may form a thin layer on the pipe wall thus affecting the pressure drop. In the homogenous model, the effect of pipe wettability on the pressure drop can be included in a straightforward way in the expression for the effective viscosity, as mentioned by Guet et al. (2006), but unfortunately, to the best of our knowledge, there exist no effective viscosity relations that include such effect. Because of this lack of correlations, here we can only prove qualitatively that potentially our approach is able to interpret the ambivalent range providing, in such a way, a more complete and convincing description.

From experimental data presented by Ioannou et al. (2005) where the effect of pipe wettability on phase inversion was investigated, we derive some fitting relations for the mixture viscosity. The effective viscosity is derived for both a steel and an acrylic pipe both when the experiments are started with oil – phase inversion from  $D_{\text{w/o}}$  to  $D_{\text{o/w}}$  – and when the starting phase is water – phase inversion from  $D_{\text{o/w}}$  to  $D_{\text{w/o}}$ . In all cases, the fitting is performed with a function close to the one presented in Eq. (10),

$$\mu_{m} = \mu_{c} (1 - K_{1} \epsilon_{d})^{-5/(2K_{2})}, \tag{15} \label{eq:15}$$

where the two constants  $K_1$  and  $K_2$  account both for the maximum packing of the dispersed droplets and for the wettability of the pipe wall.

More specifically, we have first taken the pressure drop data for the experiments which start with pure oil flow and then gradually

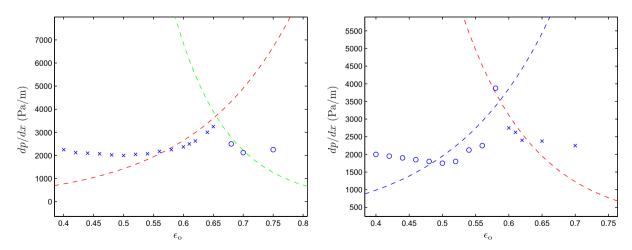


Fig. 3. Pressure gradient as a function of oil hold-up in an acrylic pipe. Experimental data are taken from loannou et al. (2005) while the theoretical predictions are computed by the homogeneous model as described in Section 2, with effective viscosity correlation given by Eq. (15) and Table 1. Points represent experimental data. Left figure:  $w \to o$ . Right figure:  $o \to w$ . Dashed curves:  $D_{o/w}$ . Dotted curves:  $D_{w/o}$ .

**Table 2**Comparison between our prediction of the ambivalent range and the corresponding experimental values, Ioannou et al. (2005)

Pipe	$\epsilon_{\rm o}^{I, \rm l}$ model	$\epsilon_{\mathrm{o}}^{I,\mathrm{u}}$ model	$\epsilon_{\rm o}^{I,{\rm l}}$ experiments	$\epsilon_{\rm o}^{l, \rm u}$ experiments
S	0.56	0.67	0.56	0.66
Α	0.59	0.66	0.58	0.65

S: steel pipe; A: acrylic pipe.

add water, and we split the data set into the  $D_{w/o}$  group and the  $D_{o/w}$  group. For each group, we fit the data by assuming a homogeneous model where the viscosity is given by Eq. (15), and obtain the best fit values of parameters  $K_1$  and  $K_2$ . The same procedure is applied to the data that refers to experiments which start with pure water flow and gradually add oil. The values of the best fitting parameters are listed in Table 1. Fig. 3 shows the experimental data for the acrylic pipe, Ioannou et al. (2005), and the fitting curves.

The model based on Eq. (10) (Section 2) is independent of pipe materials and of direction of phase inversion and, as such, it yields a single value for the critical hold-up. Using Eq. (15) with the values in Table 1, the presence of the ambivalent range is predicted as well as the lower and upper bounding values,  $\epsilon_0^{I,1}$  and  $\epsilon_0^{I,u}$ . In Table 2, the comparison between the predicted range and the measured one is shown, proving an excellent agreement and showing a clear dependence both on pipe wettability and direction of phase inversion. The excellent agreement is of course not surprising since the effective viscosity correlations are ad-hoc relations for this case. Note, however, that if we try to predict the critical hold-up by means of Eq. (12) (being K = 1 in Eq. (10)), we obtain  $\epsilon_0^I = 0.57$ ; this value is in reasonable agreement with those in Table 2.

### 5. Conclusions

We propose an interpretation of phase inversion in liquid–liquid (oil–water) pipe flow based on minimal dissipation rate. The approach is based on estimating the relation between pressure drop dp/dx and hold-up  $\epsilon$  by means of a homogeneous model based on a simple relation for the effective mixture viscosity. This must be done for both water-in-oil dispersed flow  $(D_{w/o})$  and oil-inwater dispersed flow  $(D_{o/w})$  obtaining two dp/dx vs  $\epsilon$  curves, each defined for all values of  $\epsilon$ . Our model, which compares reasonably well with experimental data, as well as with other empirical and theoretical models available in the literature, assumes that at every value of  $\epsilon$ , the flow pattern which actually obtains is the one which minimizes the overall dissipation rate (in pipe flow, this is equivalent to minimizing the pressure gradient dp/dx). As a result, the model implies that phase inversion occurs at the value of the hold-up  $\epsilon$  where the two dp/dx vs  $\epsilon$  curves intersect.

This minimal dissipation rate approach seems to provide a good interpretation of phase inversion, but it is not able to catch the

existence of the ambivalent range (hysteresis effect) if the assumed effective mixture viscosity correlation does not include pipe wettability effects such as that of pipe material and of the direction in which the experiment is performed (e.g., start from pure water and add oil, or start from pure oil and add water). However, our preliminary analysis, limited by scarcity of experimental data in the literature, shows that when such effects are included the minimal dissipation rate assumption implies the ambivalent range and yields bounding values which appear in good agreement with available measurements (the validation is however very limited due to the lack of effective viscosity relations that include wettability effects).

The results and approach presented here can be easily extended to account for the influence of other parameters such as pipe roughness, using more complex relations for the friction factor and for the effective viscosity.

#### References

Arirachakaran, S., Oglesby, K.D., Malinowsky, M.S., Shoham, O., Brill, J.P., 1989. An analysis of oil/water flow phenomena in horizontal pipes. In: SPE Prod Operating Symp. March 13th–14th, 1989.

Ball, R.C., Richmond, P., 1980. Dynamics of colloidal dispersions. J. Phys. Chem. Liquids 9, 99–116.

Brauner, N., 1998. Liquid-liquid two-phase flow, Chapter 2.3.5 in HEDU – Heat Exchanger Design Handbook, 1998.

Brauner, N., Ullman, A., 2002. Modeling phase inversion phenomenon in two-phase pipe flows. Int. J. Multiphase Flow 28, 1177–1204.

Guet, S., Rodriguez, O.M.H., Oliemans, R.V.A., Brauner, N., 2006. An inverse dispersed multiphase flow model for liquid production rate determination. Int. J. Multiphase Flow 32, 553–567.

Hu, B., Angeli, P., Matar, O.K., Hewitt, G.F., 2005. Prediction of phase inversion in agitated vessels using a two-region model. Chem. Eng. Sci. 60, 3487–3495.

Hu, B., Matar, O.K., Hewitt, G.F., Angeli, P., 2006. Population balance modelling of phase inversion in liquid-liquid pipeline flows. Chem. Eng. Sci. 61, 4994-4997. Ioannou, K., Nydal, O.J., Angeli, P., 2005. Phase inversion in dispersed liquid-liquid flows. Exp. Therm. Fluid Sci. 29, 331-339.

Liu, L., Matar, O.K., Perez de Ortiz, E.S., Hewitt, G.F., 2005. Experimental investigation of phase inversion in a stirred vessel using LIF. Chem. Eng. Sci. 60. 85–94.

Luhning, R.W., Sawistowski, H., 1971. Phase inversion in a stirred liquid-liquid systems. In: Proc. Int. Solvent Extr. Conf. Society of Chemical Industry, The Hague, pp. 873–887.

Nadler, M., Mewes, D., 1997. Flow induced emulsification in the flow of two immiscible liquids in horizontal pipes. Int. J. Multiphase Flow 23 (1), 55–68.

Pal, R., 1993. Pipeline flow of unstable and surfactant-stabilized emulsions. AIChE J. 39 (11), 1754–1763.

Piela, K., Delfos, R., Ooms, G., Westerweel, J., Oliemans, R.V.A., Mudde, R.F., 2006. Experimental investigation of phase inversion in an oil-water flow through a horizontal pipe loop. Int. J. Multiphase Flow 32 (9), 1087-1099.

Tidhar, M., Merchuk, J.C., Sembira, A.N., Wolf, D., 1986. Characteristics of a motionless mixer for dispersion of immiscible fluids – II. Phase inversion of liquid-liquid systems. Chem. Eng. Sci. 41 (3), 457–462.

Yeh, G., Haynie Jr., F.H., Moses, R.E., 1964. Phase-volume relationship at the point of phase inversion in liquid dispersion. AIChE J. 10 (2), 260–265.

Yeo, L.Y., Matar, O.K., Perez de Ortiz, Hewitt, G.F., 2000. Phase inversion and associated phenomena. Multiphase Sci. Technol. 12, 51–116.

Yeo, L.Y., Matar, O.K., Perez de Ortiz, Hewitt, G.F., 2002. Simulation studies of phase inversion in agitated vessels using a Monte Carlo technique. J. Colloid Interf. Sci. 248, 443–454.