Saturation-dependencies of three-phase relative permeabilities in mixed-wet and fractionally wet systems

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Abstract

The saturation-dependency behaviour of three-phase relative permeabilities is of central importance for modelling three-phase displacement processes in porous media. In this paper, and in related work, a method has been developed to determine the saturation-dependencies of three-phase relative permeabilities. This method is suitable for all types of mixed-wet and fractionally wet porous systems that contain clusters of oil-wet and water-wet pores with constant but different oil–water contact angles, reflecting weakly wetted conditions. Based on the classification of all allowed pore occupancies in a completely accessible porous medium, saturation-dependencies of the corresponding relative permeabilities are derived. Furthermore, three-phase relative permeabilities that appear to depend only on their own saturations are either linked to the corresponding two-phase relative permeabilities or it is shown that such a link cannot be established. A comparison has been made with existing relative permeability models with respect to their saturation-dependencies.

Keywords: Three-phase flow; Relative permeability; Saturation-dependency; Weakly wetted pores; Contact angles; Interfacial tensions; Mixed-wet; Fractionally wet

1. Introduction

Three-phase flow in porous media is important in many subsurface processes such as those, which occur in oil recovery and in aquifer remediation involving non-aqueous phase liquids (NAPLs). The principal multi-phase flow parameters that appear in the governing flow equations are the three-phase capillary pressures and relative permeabilities. These quantities, especially the three-phase relative permeabilities, are extremely difficult to measure. Therefore, over the past 50 years many attempts have been made to create analytical models of three-phase relative permeability through combination of two-phase relative permeability data. In linking three-phase and two-phase relations it is important to determine for which phases three-phase relative permeabilities depend only on their own saturations, as this may indicate that the three-phase relative permeability relation can directly be identified with one of the two-phase relations. In existing models, saturation-dependencies have been based either on limited knowledge of the underlying pore-scale physics or have been proposed purely as empirical functions that fit available data to some degree.

The papers of Stone [36,37] provide examples of models based on a pore-scale view of the physics, whereas saturation-weighted methods (e.g. [3,4]) are typically empirical models based on data fitting and, in some cases, on their limiting behaviour to corresponding two-phase relative permeabilities [18]. It should be noted that all existing models contain fitting parameters that incorporate the effects of trapping or hysteresis. However, in this work we consider only simple models, which neglect these effects and focus principally on the saturation-dependencies of the three-phase relative permeabilities. Although previously published pore-scale-based models take a certain view on the microscopic flow processes, they are developed assuming a priori that relative permeabilities satisfy certain saturation-dependencies. For purely water-wet systems, it is assumed that oil is the intermediate-wetting phase; thus, water and gas relative permeabilities depend only on their own saturations, while the oil relative permeability
is a function of two saturations. This particular assumption is indeed supported by the pore-scale view that water occupies the small pores, gas occupies the large pores and oil resides in between [9,36]. Many experiments indicate, however, that reservoirs are not purely water-wet but can attain a wide variety of wetting states in which certain clusters of pores are water-wet and others are oil-wet [10,19,28]. For these states, the saturation-dependencies of the relative permeabilities are not systematically known.

Related to assumptions concerning saturation-dependencies are the assumed links between two-phase and three-phase relative permeabilities. In models for water-wet systems, it is assumed that water and gas relative permeabilities are identical to the corresponding relative permeabilities in two-phase water–oil and gas–oil systems, respectively, while the oil relative permeability is a combination of the oil relative permeabilities in both water–oil and gas–oil systems. This assumption is also a consequence of the pore-scale view that the water and gas phases are spatially separated by the oil phase, hence both water and gas have only interfaces to oil, while oil has interfaces to both other phases [36].

More recently, van Dijke et al. [39] have determined all allowed saturation-dependencies that may arise for capillary controlled flow in a mixed-wet bundle of capillary tubes, where the pores are either strongly water-wet or strongly oil-wet, by working through all possible pore filling sequences similar to Stone [37]. It was shown that the values of the various interfacial tensions (thus including spreading effects) also play an important role in determining the saturation-dependencies. From this simple model, it became clear that the saturation space may be divided into two or three regions in which either water, oil or gas is the intermediate-wetting phase (analogous to oil in a water-wet system).

In this study, we generalise the approach of [39] to account also for systems with weakly water-wet or oil-wet pores, reflected by the oil–water contact angle that takes constant but different values in water-wet and oil-wet pores. Furthermore, we consider all types of mixed-wet and fractionally wet systems, in particular also porous media in which pores of the same size may be both water-wet and oil-wet. Not only the saturation-dependencies are analysed, but also the conditions are derived determining whether three-phase relative permeabilities can be linked to the corresponding two-phase relative permeabilities.

In Section 2, the capillary tube model is described together with the appropriate entry conditions. In Section 3, we introduce consistent relations between the three contact angles and the three interfacial tensions that allow us to analyse systems with weakly wetted pores. Section 4 describes the allowed three-phase pore occupancies from which the saturation-dependencies of relative permeabilities are formally derived. Also the possible links between two-phase and three-phase relative permeabilities are addressed. In Section 5, a simulation is first presented with realistic values of the physical parameters to illustrate the existence and extent of regions with different saturation-dependencies in the three-phase saturation space. Then the general appearance of these regions is discussed for all possible combinations of porous media and fluid parameters. Finally, it is indicated in which sections of the saturation-dependency regions two-phase and three-phase relative permeability relations can be related. In Section 6, a comparison is made between the present approach and existing relative permeability models with respect to saturation-dependency and the links between two-phase and three-phase relations. This section also provides a brief overview of the relevant literature.
whose pressure is kept constant during the entire process. The inlets of the pores are connected to a vessel containing a different phase, whose pressure is gradually increased to allow subsequent invasion of the pores. Once a pore is invaded, we assume that its outlet also becomes connected to the vessel with the invading phase. When a prescribed number of pores are invaded, the inlets of the pores are connected to a vessel containing the remaining third phase, whose pressure is gradually increased to allow invasion. During this stage, which is the actual three-phase flow process, the pressures of the first and the second phases are kept constant at the outlets. The pressure changes so slowly that we may assume capillary equilibrium between the phases. Furthermore, all displacements are assumed to occur in a piston-like manner.

It may be clear that each invasion of a pore is a two-phase event, whereas the subsequent filling of pores occupied by different phases constitutes the three-phase flow process [35]. The condition for invasion of phase $i$ into a pore occupied by phase $j$ is given by

$$P_{\text{inv},i} \geq P_{\text{entry},ij},$$

(1)

where $P_{\text{inv},i}$ denotes the pressure of the invading phase, and $P_{\text{entry},ij}$ denotes the entry pressure, which under capillary equilibrium is given by

$$P_{\text{entry},ij} = P_i + P_{c,ij},$$

(2)

for $i \neq j$, where $P_{c,ij}$ denotes the capillary entry pressure of the target pore with respect to the fluids $i$ and $j$ [27,39].

In three-phase flow, the capillary entry pressures $P_{c,ij}$ in a cylindrical pore with radius $r$ for each pair of phases are determined by the interfacial tensions and contact angles through the Young–Laplace equation

$$P_{c,ij} = \frac{2\sigma_{ij} \cos \theta_{ij}}{r},$$

(3)

with $ij = ow$, gw, go. Since we specify only the oil–water contact angle through $\cos \theta_{ow}$, the degree of oil–water wettability of the pore, the values of the remaining contact angles are to be found as functions of $\cos \theta_{ow}$ to establish a complete set of entry conditions (1) and (2).

When gradually increasing the pressure of the invading phase, the entry pressures (2) are met pore by pore and the pores are invaded accordingly. This order is determined by the pressure of the phase that is already present in a pore $P_i$, which may be different from pore to pore, and the capillary entry pressure $P_{c,ij}$. According to Eq. (3), for one fluid pair $ij$ the capillary entry pressures do not only vary with pore size, but also with the oil–water wettability of the pore, which greatly affects the filling order.

From the entry conditions (1) and (2) follows that the conditions for a pore to be occupied by one of the phases can be described in terms of the capillary pressures, the differences between the phase pressures $P_{ij} = P_i - P_j$, in relation to the capillary entry pressures $P_{c,ij}$ as:

\[
\begin{align*}
\text{Gas:} & \quad \begin{cases} P_{gw} > P_{c,gw}, \\ P_{go} > P_{c,go}. \end{cases} \quad (4a) \\
\text{Oil:} & \quad \begin{cases} P_{go} \leq P_{c,go}, \\ P_{ow} > P_{c,ow}. \end{cases} \quad (4b) \\
\text{Water:} & \quad \begin{cases} P_{gw} \leq P_{c,gw}, \\ P_{ow} \leq P_{c,ow}. \end{cases} \quad (4c)
\end{align*}
\]

For consistency of notation, we have used the ‘strictly larger than’ sign (>) and the ‘smaller than or equal to’ sign ($\leq$), which is only correct if water is wetting to oil and oil is wetting to gas. In other words, the more wetting fluid fills the pore if the pressure difference is exactly equal to the capillary entry pressure. Therefore, for different wetting orders the signs should be adapted accordingly. However, because this issue does not affect any of the following analysis, we will use the sign convention of conditions (4a)–(4c).

3. Relations between interfacial tensions and contact angles in three-phase flow

The quantity $\cos \theta_{ow}$ defines the wettability and we now show how the cosines of the remaining contact angles $\cos \theta_{gw}$ and $\cos \theta_{go}$ in the same pore must be related when $\cos \theta_{ow}$ takes arbitrary values between $-1$ and $1$. By combining the three relations for the fluid–solid surface tensions and the related contact angles, Zhou and Blunt [42] obtained a relation between the three contact angles and the fluid–fluid interfacial tensions at thermodynamic equilibrium for a pore of any degree of wettability

$$\sigma_{gw} \cos \theta_{gw} = \sigma_{go} \cos \theta_{go} + \sigma_{ow} \cos \theta_{ow}. \quad (5)$$

In Appendix A, we show that in general condition (5) is equivalent to the relation between the capillary entry pressures

$$P_{c,gw} = P_{c,go} + P_{c,ow}. \quad (6)$$

Moreover, relation (6) is proven to be the necessary and sufficient condition that under capillary equilibrium every point of a porous medium is occupied by exactly one phase.

Eq. (5) implies that we may express both $\cos \theta_{go}$ and $\cos \theta_{gw}$ as functions of $\cos \theta_{ow}$. Since little is known experimentally about such relationships, we adopt linear expressions as follows:

$$\cos \theta_{go} = \frac{1}{2} \left\{ -1 + \frac{\sigma_{gw} - \sigma_{ow}}{\sigma_{go}} \right\} \cos \theta_{ow}$$

$$+ \frac{\sigma_{gw} - \sigma_{ow}}{\sigma_{go}}, \quad (7a)$$

$$\cos \theta_{gw} = \frac{1}{2} \left\{ -1 + \frac{\sigma_{go} - \sigma_{ow}}{\sigma_{gw}} \right\} \cos \theta_{ow}$$

$$+ \frac{\sigma_{go} - \sigma_{ow}}{\sigma_{gw}}, \quad (7b)$$
For a non-spreading oil, we have by definition 
\[ C_{S,o} = \begin{cases} 
\sigma_{gw} - \sigma_{go} - \sigma_{ow} & \text{if } \sigma_{gw} - \sigma_{go} - \sigma_{ow} < 0, \\
0 & \text{if } \sigma_{gw} - \sigma_{go} - \sigma_{ow} \geq 0
\end{cases} \] (10)

For a non-spreading oil, we have by definition \( C_{S,o} < 0 \) and for a spreading oil \( C_{S,o} = 0 \). Although we can choose values of the interfacial tensions such that \( \sigma_{gw} - \sigma_{go} - \sigma_{ow} > 0 \), the corresponding relations between the contact angles for a spreading oil (8a) and (8b) form the limiting case of the relations for a non-spreading oil in (9a) and (9b) by taking \( C_{S,o} = 0 \).

Therefore, for both the spreading and the non-spreading cases we use relations (9a) and (9b).

As suggested in Fig. 1, we have made the following realistic assumptions for the values of the interfacial tensions:

\[ \sigma_{gw} > \sigma_{go} > 0 \text{ and } \sigma_{gw} > \sigma_{ow} > 0 \] (11)

which gives in particular:

\[ 0 < \frac{\sigma_{gw} - \sigma_{ow}}{\sigma_{go}} < 1, \] (12a)

\[ -1 < \frac{\sigma_{go} - \sigma_{ow}}{\sigma_{gw}} < 1, \] (12b)

\[ \frac{\sigma_{gw} - \sigma_{ow}}{\sigma_{go}} < \frac{\sigma_{gw} + \sigma_{ow}}{\sigma_{go}}. \] (12c)

These assumptions affect the wetting order of the fluids within each pore, which is defined through the sign of the contact angles \( \theta_{ij} \); if \( \cos \theta_{ij} > 0 \) phase \( i \) is non-wetting (relative) to phase \( j \) and if \( \cos \theta_{ij} < 0 \) phase \( i \) is wetting (relative) to phase \( j \). The wetting order of water to oil is given by the sign of \( \cos \theta_{ow} \). According to relation (7a), assumption (12a) implies for a non-spreading oil that \( \cos \theta_{go} \) is a decreasing function of \( \cos \theta_{ow} \), but it is positive for all \(-1 \leq \cos \theta_{ow} \leq 1\). Hence, oil is wetting to gas for all possible degrees of oil–water wettability. According to relations (7b) and (8b) assumptions (12b) and (12c) imply that \( \cos \theta_{gw} \) is an increasing function of \( \cos \theta_{ow} \) and it is positive at least for all \( 0 \leq \cos \theta_{ow} \leq 1 \). Furthermore, no assumptions are made on the mutual sizes of \( \sigma_{ow} \) and \( \sigma_{go} \). Therefore, if \( \sigma_{go} < \sigma_{ow} \) a value \(-1 < \cos \theta_{ow} < 0 \) exists such that \( \cos \theta_{gw} \) is negative for \(-1 \leq \cos \theta_{ow} < \cos \theta_{ow}^{*} \) and positive for \( \cos \theta_{ow} \leq \cos \theta_{ow} < 0 \), whereas, if \( \sigma_{go} > \sigma_{ow} \) is positive for all \(-1 \leq \cos \theta_{ow} < 0 \). In other words, only if \( \sigma_{go} < \sigma_{ow} \).
gas is wetting to water for the more strongly oil-wet pores, otherwise water is wetting to gas.

As a result of restrictions (11) on the values of the interfacial tensions, in the water-wet pores the wetting order is always: water wetting, oil intermediate-wetting and gas non-wetting. In the oil-wet pores, the wetting order is either: oil wetting, gas intermediate-wetting and water non-wetting, or: oil wetting, water intermediate-wetting and gas non-wetting, depending on the actual values of \( \cos \theta_{\text{ow}}, \sigma_{\text{ow}} \) and \( \sigma_{\text{go}} \).

4. Three-phase pore occupancies

4.1. Allowed pore occupancies

To derive the possible saturation-dependencies of three-phase relative permeabilities, van Dijke et al. [39] examined the possible three-phase pore occupancies for a bundle of capillary tubes, i.e. which phase occupies which pore. They considered occupancies that may arise in mixed-wet systems (MWL and MWS) with strongly wetted pores, whereas here we consider all kinds of mixed-wet and fractionally wet systems as defined in Section 2, including weakly wet conditions. van Dijke et al. [39] derived two rules for the wetting order of the phases within a bundle of tubes, thus determining which pore occupancies are actually allowed. Based on the wetting order of the phases within each pore, the Young–Laplace equations (3) ensure that within a cluster of either water-wet pores or oil-wet pores the phase pressures are ordered such that the wetting phase has the smallest pressure and the non-wetting phase has the largest pressure. Considering also that the pressure of one phase is the same throughout the porous medium, the invasion conditions (1) and (2), or equivalently (4a)–(4c), lead to the following wetting order rules that govern the pore occupancies:

(a) In one cluster, the phase that is wetting in this cluster occupies smaller pores than the intermediate-wetting phase, which in turn occupies smaller pores than the non-wetting phase.

(b) If the wetting order of two phases is different for water-wet pores and oil-wet pores, then these phases cannot coexist in both the water-wet and the oil-wet pores. This rule also applies if one of the phases is only present as a wetting film.

These rules were derived independent of the sizes of the water-wet pores \( r_1^w \leq r \leq r_2^w \) relative to the size of the oil-wet pores \( r_1^o \leq r \leq r_2^o \). Furthermore, it follows easily that the assumption that the pores were strongly wetted, i.e. \( \cos \theta_{\text{ow}} = 1 \) and \( \cos \theta_{\text{ow}} = -1 \), can be relaxed to allow \( \cos \theta_{\text{ow}} \) to take any constant value between 0 and 1 and \( \cos \theta_{\text{ow}} \) any constant value between -1 and 0, without changing the derivation. Therefore, the above wetting order rules will also apply to any of the weakly wetted porous media that we currently consider.

In Section 3, we derived which wetting orders may occur in the water-wet and the oil-wet pores, respectively, based on the assumptions for the values of the interfacial tensions as in relations (11). Combined with the wetting order rules, this implies that for the currently investigated mixed-wet and fractionally wet systems with constant contact angles only 10 different three-phase pore occupancies may occur. In Fig. 2, two examples of allowed occupancies and two examples of non-allowed pore occupancies are presented (assuming a uniform distribution of pore sizes). The latter occupancies are excluded by wetting order rule (b).

As shown by van Dijke et al. [39] the ordering of the pore sizes may lead to extra restrictions on the allowed pore occupancies. For example, in an MWL system \( (r_2^w \leq r_1^o) \) water cannot be present in the oil-wet pores if the water-wet pores are not completely water-filled as in Fig. 2(b), although this is possible in an MWS system.

As a result of wetting order rule (a), each cluster does contain at most two boundaries between the various phases in the pore occupancy plots. Additionally, as a result of rule (b), at most three boundaries are present in the occupancy plot of the whole system. It can be shown that if three boundaries occur in a three-phase occupancy, two of them (one in the water-wet and one in the oil-wet pores) are of the same type. If two boundaries occur, these must be of different types to constitute a three-phase occupancy. If only one boundary is present, the pore occupancy is called anomalous, as one of the phases must occupy exactly all the water-wet pores or all the oil-wet pores. We do not consider anomalous occupancies separately as they are limiting cases of occupancies with two and three boundaries. In summary, each three-phase occupancy contains two different boundaries and possibly a third that is the same as one of the other two. In Fig. 2(a), an example of the latter is given with two gas–oil boundaries, \( r_{\text{go}}^w \) and \( r_{\text{go}}^o \), and one oil–water boundary, \( r_{\text{ow}} \), whereas Fig. 2(b) shows an occupancy with two gas–water boundaries \( r_{\text{gw}}^w \) and \( r_{\text{gw}}^o \), and one oil–water boundary.

Consequently, in each three-phase pore occupancy only one phase has boundaries to both remaining phases: the “intermediate-wetting” phase. Observe that this definition of “intermediate-wetting” is a generalisation of the usual definition for purely water-wet or purely oil-wet systems. For example, in the pore occupancy of Fig. 2(a) oil is the wetting phase in the oil-wet pores, yet it is the intermediate-wetting phase for the system as a whole because in the water-wet pores oil has boundaries to both water and gas. We specify a three-phase occupancy as type I if oil is intermediate-wetting, as type II if gas is intermediate-wetting and as type III if water is intermediate-wetting, as in [39]. Clearly, the
pore occupancy of Fig. 2(a) is of type I and that of Fig. 2(b) is of type III.

4.2. Saturation-dependencies of relative permeabilities

We now consider how the various pore occupancies lead to particular saturation-dependencies of the relative permeabilities. For each phase $j$, the formal definitions of saturations, $S_j$, and relative permeabilities, $k_{r,j}$, are as follows:

$$S_j = \int_0^\infty \pi_j(r) \phi(r) V(r) \, dr,$$

(13a)

$$k_{r,j} = \int_0^\infty \pi_j(r) \phi(r) g(r) \, dr,$$

(13b)

where $\phi(r)$ denotes the pore size distribution function, which is normalised such that $\int_0^\infty \phi(r) \, dr = 1$. $V(r)$ denotes the volume function and $g(r)$ is the conductance function for each pore, normalised with respect to the total volume and total conductivity of the system, respectively. The function $\pi_j(r)$ denotes the occupancy of phase $j$. For example, in Fig. 2(a), the oil occupancy is given by

$$\pi_o(r) = \begin{cases} 1 & \text{for } r_{ow} < r < r_{go}^w \text{ or } r_{1}^o < r < r_{go}^o, \\ 0 & \text{otherwise.} \end{cases}$$

(14)

By definition $\pi_w + \pi_o + \pi_g = 1$, hence $S_w + S_o + S_g = 1$ and $k_{r,w} + k_{r,o} + k_{r,g} = 1$, whereas the latter relation is true only in our completely accessible capillary tube model.

To show the saturation-dependencies of the relative permeabilities for a certain type of pore occupancy, we consider first the particular occupancy of Fig. 2(a). The saturations and relative permeabilities for this occupancy are given by:

$$S_w = \int_{r_1^w}^{r_{ow}} \phi(r)V(r) \, dr, \quad k_{r,w} = \int_{r_1^w}^{r_{ow}} \phi(r)g(r) \, dr,$$

(15a)

$$S_o = \int_{r_{ow}}^{r_{go}} \phi(r)V(r) \, dr + \int_{r_1^o}^{r_{go}^o} \phi(r)V(r) \, dr,$$

(15b)

$$k_{r,o} = \int_{r_{ow}}^{r_{go}} \phi(r)g(r) \, dr + \int_{r_1^o}^{r_{go}^o} \phi(r)g(r) \, dr,$$

$$S_g = \int_{r_1^g}^{r_{go}^g} \phi(r)V(r) \, dr + \int_{r_{go}}^{r_{go}^g} \phi(r)V(r) \, dr,$$

(15c)

$$k_{r,g} = \int_{r_1^g}^{r_{go}^g} \phi(r)g(r) \, dr + \int_{r_{go}}^{r_{go}^g} \phi(r)g(r) \, dr.$$

Assuming that in one cluster the pore sizes are very close to each other, it follows from relations (4a)–(4c) that the actual capillary pressures are determined by the boundaries between the various phases, hence $P_{ow} = P_{c,ow}(r_{ow})$ and $P_{go} = P_{c,go}(r_{go}^w) = P_{c,go}(r_{go}^o)$. Combining this result with the Young–Laplace equations (3) yields
and

$k_{r,w} = k_{r,w}(S_w)$, $k_{r,o} = k_{r,o}(S_w, S_g)$ and

$k_{r,g} = k_{r,g}(S_g)$.  

(17)

As, by definition, all three-phase occupancies of type I have an oil–water and a gas–oil boundary, dependencies (17) apply to all of them. Furthermore, following similar arguments for type II occupancies the dependencies

$k_{r,w} = k_{r,w}(S_w)$, $k_{r,o} = k_{r,o}(S_o)$ and

$k_{r,g} = k_{r,g}(S_o, S_g)$

(18)

apply and for type III the dependencies

$k_{r,w} = k_{r,w}(S_w, S_g)$, $k_{r,o} = k_{r,o}(S_o)$ and

$k_{r,g} = k_{r,g}(S_g)$.  

(19)

The dependencies (17)–(19) were also found by van Dijke et al. [39] for strongly wetted pores, based on examination of iso-relative permeability plots. The dependencies further support the earlier definition of an “intermediate-wetting” phase, as for each type of occupancy only the relative permeability of this phase depends on more than one saturation. The relative permeability of the intermediate-wetting phase is obviously derived from the remaining two relative permeabilities through the relation $k_{r,w} + k_{r,o} + k_{r,g} = 1$.

4.3. Links between two-phase and three-phase relative permeabilities

From Eqs. (15a)–(15c) we may also derive links between the two-phase and three-phase relative permeabilities for type I occupancies such as given in Fig. 2(a). To achieve this, consider two-phase pore occupancies that are governed by the same respective pressure differences as those involved in the three-phase occupancy. The corresponding oil–water system is governed by $P_{ow} = P_{c,ow}(r_{ow})$, the gas–oil system by $P_{go} = P_{c,go}(r_{go}) = P_{c,go}(r_{go}^*)$ and the gas–water system by $P_{gw} = P_{go} + P_{ow}$. Hence, the boundaries in the oil–water and gas–oil occupancies are the same as those in the three-phase occupancy. The gas–water pressure difference defines one or two gas–water boundaries that are not present in the three-phase occupancy, but it follows below that these are not needed.

In Fig. 3 the corresponding two-phase oil–water and gas–oil systems are shown. Comparison of Fig. 2(a) with Figs. 3(a) and (b) shows that both water and gas occupy the same pores in the three-phase occupancy and in the corresponding two-phase occupancies.

Consequently, applying definitions (13a) and (13b) shows that the water saturation and relative permeability for the oil–water system are given by relations (15a), whereas the gas saturation and relative permeability for the gas–oil system are given by relations (15c). Hence, the three-phase water saturation and relative permeability equal the two-phase water saturation and relative permeability in the oil–water system, yielding

$k_{r,w}(S_w) = k_{r,w}^{ow}(S_w)$,

(20)

where the double superscript refers to the two-phase system. Similarly, the gas relative permeability is given by

$k_{r,g}(S_g) = k_{r,g}^{go}(S_g)$

(21)

Obviously, the relative permeability of oil, the intermediate-wetting phase for occupancy I, is not identical to one of the two-phase oil-relative permeabilities. However, it is given by $k_{r,o}(S_w, S_g) = 1 - k_{r,w}^{ow}(S_w) - k_{r,g}^{go}(S_g)$.
To find the relation between a type II three-phase pore occupancy and the corresponding two-phase occupancies, we go in the “opposite” direction as above. We may start with the allowed gas–oil and gas–water two-phase occupancies as presented in Figs. 4(a) and (b) to construct the corresponding type II three-phase occupancy of Fig. 4(c) for the same pressure differences $P_{go} = P_{cg} (r_{go}) = P_{cgw} (r_{gw})$ and $P_{gw} = P_{cgw} (r_{gw})$. Now, since gas is the intermediate-wetting phase, we want to find the links between the two-phase and the three-phase relations for the oil and the water relative permeabilities, similar to the water and gas relative permeabilities in the previously discussed type I occupancy.

For the two-phase gas–oil occupancy of Fig. 4(a), the oil relative permeability follows from definition (13b) as

$$k^o_{r,o} = \int_{r_1^o}^{r_2^o} \varphi(r) g(r) \, dr + \int_{r_1^o}^{r_2^o} \varphi(r) g(r) \, dr.$$  \hspace{1cm} (22)

On the other hand, for the three-phase occupancy in Fig. 4(c) the oil relative permeability is given by

$$k_{r,o} = \int_{r_1^o}^{r_2^o} \varphi(r) g(r) \, dr.$$  \hspace{1cm} (23)

As the integral in relation (23) depends on the boundary only, the saturation-$r_{gw}$ dependency still follows as $k_{r,o} = k_{r,o} (S_o)$. However, because oil does not occupy the same pores in the two-phase and three-phase occupancies, the expressions on the right-hand sides of (22) and (23) are not identical. Hence, we cannot identify the three-phase with the corresponding two-phase oil relative permeability:

$$k_{r,o} (S_o) \neq k^o_{r,o} (S_o).$$  \hspace{1cm} (24)

We call this saturation-dependency non-genuine, contrary to genuine dependencies, where explicit links between the three-phase and one of the two-phase relative permeabilities can be established. Comparison of the occupancies of Figs. 4(b) and (c) shows that the water relative permeability is still genuinely linked as

$$k_{r,w} (S_w) = k_{r,w} (S_w).$$  \hspace{1cm} (25)

Considering the three-phase occupancy of Fig. 4(c), it appears that the corresponding gas–oil occupancy could also have been the two-phase occupancy of Fig. 4(a) but without the boundary $r_{gw}$, with all the water-wet pores filled by gas. The latter is also an allowed two-phase occupancy. This situation would have led to the genuine link $k_{r,o} (S_o) = k_{r,o} (S_o)$.

Furthermore, the three-phase occupancy of Fig. 4(c) could also relate to a two-phase gas–water occupancy with two boundaries between the phases. In this case also the dependency of $k_{r,w} (S_w)$ would have been non-genuine. The corresponding two-phase occupancies are shown in Fig. 5.

A non-genuine saturation-dependency with respect to the gas–water boundary occurs only if $\cos \theta_{gw} > 0$, i.e., when water is wetting to gas in the oil-wet pores. As a result type II occupancies may have either two genuine saturation-dependencies for the oil and water relative permeabilities.
permeabilities, two non-genuine dependencies or one genuine and one non-genuine dependency.

On the contrary, saturation-dependencies for type I occupancies are always genuine. To show this, we apply wetting order rule (a) of Section 4.1 to find that in type I occupancies oil must be present in the water-wet pores and that, as a result, water is confined to the smallest water-wet pores. Hence, for all three-type phase I occupancies, Fig. 3(a) presents the only possible corresponding two-phase oil–water occupancy. On the other hand, the corresponding two-phase gas–oil occupancy may contain gas in the water-wet pores (e.g. as in Fig. 3(b)), but only such that \( r_{gw} > r_{ow} \), otherwise a type II occupancy would result. Hence, both gas and water occupy the same pores in the three-phase and in the corresponding two-phase occupancies and the same conclusion can be drawn if gas is only present in the oil-wet pores.

Following similar arguments, it can be shown that type III occupancies always show the genuine links

\[
\begin{align*}
\text{Case 1: } & \quad k_{r,g}(S_w) = k_{r,g}^{\text{max}}(S_w), \\
\text{Case 2: } & \quad k_{r,g}(S_w) = k_{r,g}^{\text{min}}(S_w), \\
\text{Case 3: } & \quad k_{r,g}(S_w) = k_{r,g}^{\text{med}}(S_w).
\end{align*}
\]

5. Three-phase saturation space

5.1. Simulation

To show the impact of the various saturation-dependencies, we compute iso-capillary pressure and iso-relative permeability curves as functions of saturation for a given porous medium and a given set of fluid parameters. We consider a FW porous medium where the sizes of the smallest and largest water-wet and oil-wet pores are the same, i.e. \( r_{gw} = r_{ow} = 10 \times 10^{-6} \) m and \( r_{gw}^2 = r_{ow}^2 = 160 \times 10^{-6} \) m. The values of the interfacial tensions are taken as \( \sigma_{ow} = 29 \times 10^{-3} \) N/m, \( \sigma_{go} = 14 \times 10^{-3} \) N/m and \( \sigma_{gw} = 36 \times 10^{-3} \) N/m. Hence, the value of the spreading coefficient is \( C_{S_o} = -7 \times 10^{-3} \) N/m, so we consider a non-spreading oil. The volume function is taken as \( V(\phi) = \phi \) and the conductance function as \( g(r) = r^4 \). Notice that taking 1 for the volume exponent is, strictly speaking, incorrect for cylindrical pores. However, for more general pore geometries this value is realistic and it has the additional advantage of pronouncing some of the features in saturation space. Furthermore, the pore size distribution \( \phi(r) \) is taken to be uniform, with equal fractions of water-wet and oil-wet pores. The degrees of wettability of the water-wet and oil-wet pores are given by the oil–water contact angles \( \cos \theta_{gw} = 0.6 \) and \( \cos \theta_{ow} = -0.1 \), respectively. Using Eq. (9b) the latter implies \( \cos \theta_{gw} > 0 \), i.e., water is wetting to gas in the oil-wet pores.

In Fig. 6, the iso-capillary pressure curves and iso-relative permeability curves for the FW system are presented in ternary saturation diagrams. As shown by van Dijke et al. [39], the diagrams of iso-capillary pressures (Fig. 6(a)) are easily obtained by simulating flow processes, in which one phase is injected into a medium that is filled by two other fluids at various saturation ratios. During one process, the pressure of the invading phase is increased such that its saturation increases from 0 to 1, while keeping the pressure difference between the remaining phases constant as described in Section 2. Hence, a line of constant capillary pressure between the two remaining phases is created. From these flow processes also iso-relative permeability curves (Fig. 6(b)) can be computed.

Both types of saturation diagrams are clearly divided into three regions, reflecting different saturation-dependencies, and thus different types of pore occupancies [39]. For example, the iso-relative permeability lines for a certain phase are curved only when its relative permeability depends on more than one saturation, otherwise the iso-lines are lines of constant saturation of that phase. From the previous sections, we know that for a given combination of saturations, i.e. for a given pore occupancy, only one phase is intermediate-wetting. Only the relative permeability corresponding to this phase depends on more than one saturation. Therefore, the
regions in the saturation diagrams with iso-relative permeability lines as well as those with iso-capillary pressure lines can be identified as regions I, II and III. In region I occupancies of type I occur (of which an example is given in Fig. 2(a)), where oil is intermediate-wetting. In region II gas is intermediate-wetting (as in Fig. 4(c)) and in region III water is intermediate-wetting (as in Fig. 2(b)).

The boundaries between the various saturation-dependency regions are only partly iso-saturation lines. In Sections 6, we consider how variation of the porous medium and fluid parameters affect the non-linear parts of the boundaries, as well as the impact of the earlier discussed non-genuine saturation-dependencies in saturation space.

5.2. Saturation-dependency regions

van Dijke et al. [39] considered MWL systems with strongly wetted pores, for which they found that the six different configurations of the saturation-dependency regions, schematically shown in Fig. 7, can be distinguished. On the contrary, the configuration of the FW system of Fig. 6, only one non-linear boundary, i.e. between regions I and II, may occur. The various configurations differ with respect to the positions of these non-linear boundaries. Note that, strictly speaking, 12 configurations arise as the lower right triangle of the saturation space belongs to either region II or region III. The saturation corresponding to occupancy of exactly all the water-wet pores is defined as

Fig. 6. Ternary saturation diagrams of: (a) iso-capillary pressure curves; (b) iso-relative permeability curves for the simulated non-spreading oil in a FW medium. The curves in (a) are actually saturation paths of consecutive flooding processes, where the combination of capitals indicates the order of the floods: e.g. O–G–W indicates that the paths are obtained from an oil-filled medium in which partial gas floods are followed by complete water-floods. The different curves are identified by the number fraction of pores that are filled with the displaced phase after the first flood: e.g. in O–G–W the residual indicates the number fraction of pores that are still filled with oil after gas flooding. Furthermore, the three saturation-dependency regions are indicated in the O-G-W and gas isoperm diagram.
\[ S_{\text{wet}} = \int_{r_1}^{r_2} \varphi(r) V(r) \, dr. \]

Observe that (part of) the line \( S_w = S_{\text{wet}} \) as well as (part of) the line \( S_o = 1 - S_{\text{wet}} \) are indeed clearly visible in the configuration of Fig. 6. Furthermore, in the configurations of Figs. 7(a) and (f) no non-linear boundary is present. It follows easily that these are the limiting cases of the configurations of Figs. 7(b) and (e), respectively, in which the non-linear boundaries coincide with \( S_w = S_{\text{wet}} \) and \( S_o = 1 - S_{\text{wet}} \), respectively.

Below, we briefly recapitulate the conditions for the different positions of the non-linear boundary between regions I and II for the present, more general, case with weakly wetted pores and indicate the analogous conditions for the non-linear boundary between regions II and III that may additionally arise in the FW system.

van Dijke et al. [39] showed that a non-linear boundary between regions I and II arises when three-phase occupancies can occur, which correspond to two-phase gas–oil occupancies with two gas–oil boundaries, such as shown in Fig. 3(b). The different positions of this non-linear boundary in saturation space are determined by the six possible orderings of the gas–oil capillary entry pressure of the smallest and largest pores of the water-wet and oil-wet clusters, in the terminology of this paper \( P_{c\,go}(r_{1w}^n) \), \( P_{c\,go}(r_{2w}^o) \), \( P_{c\,go}(r_{1o}^o) \) and \( P_{c\,go}(r_{2w}^o) \). This ordering determines in which order the gas–oil boundaries in the occupancy plots may occur within the water-wet or oil-wet clusters or within both, when the gas–oil pressure difference is increased. This is equivalent to the two-phase process of gas invasion into an oil-filled medium. For example, for Fig. 7(d) the ordering

\[ P_{c\,go}(r_{2w}^o) < P_{c\,go}(r_{1w}^n) < P_{c\,go}(r_{1o}^o) < P_{c\,go}(r_{2w}^o) \]  

applies. Hence, when we increase \( P_{go} \), first the threshold value of the largest water-wet pore \( P_{c\,go}(r_{1w}^n) \) is overcome resulting in a gas–oil boundary in the water-wet pores, then \( P_{c\,go}(r_{2w}^o) \) is overcome resulting in an additional boundary in the oil-wet pores. Subsequently, the boundary in the oil-wet pores vanishes when \( P_{go} \) exceeds the threshold value for the smallest oil-wet pore \( P_{c\,go}(r_{2w}^o) \).
and finally the boundary in the water-wet pores vanishes. Applying the Young–Laplace equations (3) to relations (27) yields the following conditions in terms of the fluid and pore size parameters for the position of the non-linear boundary as in Fig. 7(d):

\[
\frac{r_1^w}{r_1} < \frac{\cos \theta^p_{go}}{\cos \theta^p_{go}} < \frac{r_2^w}{r_2}.
\]  

Condition (28) is a generalisation of the corresponding condition given by van Dijke et al. [39] as it incorporates contact angles related to weakly wetted pores.

Furthermore, the conclusion that the configuration of Fig. 7(d) indeed occurs when conditions (27) are satisfied, follows by considering that in this situation three different three-phase occupancies constitute region I [39]. Fig. 2(a) shows one of these pore occupancies,
whereas the other two are given by the same figure but with the oil-wet pores completely filled with gas or completely filled with oil, respectively. The latter two occupancies apply in the top and the bottom parts of region I in saturation space, respectively, whereas the occupancy of Fig. 2(a) applies in the centre part near the non-linear boundary. Only this occupancy corresponds to a two-phase gas–oil occupancy with two gas–oil boundaries. It has been shown that only when a type I pore occupancy is found with two gas–oil boundaries, a non-linear boundary between regions I and II arises. From the latter the position of the non-linear boundary may also be derived as follows.

Near the non-linear boundary in region I the occupancy of Fig. 2(a) applies and transition to region II follows when in the water-wet pores the oil–water boundary coincides with the gas–oil boundary \( r_{ow} = r_{go} \) yielding a type II pore occupancy as shown in Fig. 4(c). As in the configuration in Fig. 2(a) two gas–oil boundaries occur, we have \( P_{c,go}(r_{go}^\ast) = P_{c,go}(r_{go}^\dagger) \). Application of the Young–Laplace equations (3) then yields at the non-linear boundary

\[
r_{ow} = r_{go} \cos \theta_{go}^p \cos \theta_{go}^w
\]

and the corresponding position in saturation space follows from definition (13a). Observe that the non-linear boundary in saturation space can only be determined when the underlying pore occupancies are known.

As observed earlier, in every pore occupancy at most one oil–water boundary can occur because by definition \( \cos \theta_{ow}^p < 0 < \cos \theta_{wo}^p \) and the oil–water capillary entry pressures at the endpoints of the water-wet and oil-wet clusters can be ordered in only one way, irrespective of pore size. Hence, in saturation space \( P_{c,ow} \) never gives rise to non-linear boundaries.

Similarly, if gas is wetting to water in the oil-wet pores, the gas–water capillary entry pressures are always ordered in one way as \( \cos \theta_{gw}^p < 0 < \cos \theta_{wg}^p \), therefore in this situation \( P_{c,gw} \) never leads to a non-linear boundary in saturation space. Furthermore, because gas is wetting to water in the oil-wet pores, water can never be the intermediate-wetting phase for the medium as a whole and the condition \( \cos \theta_{gw}^p < 0 \) determines that region III does not occur. On the other hand, if water is wetting to gas in the oil-wet pores, i.e. if \( \cos \theta_{gw}^p > 0 \), region III must be present and six different orderings of the \( P_{c,gw} \) may arise, similar to the threshold values of the \( P_{c,go} \) in relations (27).

Considering conditions such as (28) shows that the mutual ordering of the sizes of the smallest and largest water-wet and oil-wet pores a priori excludes occurrence of certain configurations. For example, for MWL systems, where \( r_2^\ast \leq r_1^\ast \), while \( \cos \theta_{gw}^p < \cos \theta_{wg}^p \), the \( P_{c,gw} \) threshold values are strictly ordered, even when \( \cos \theta_{gw}^p > 0 \), such that a non-linear boundary between regions II and III can never occur. Similarly, in a MWS system, where \( r_2^\ast \leq r_1^\ast \), while \( \cos \theta_{gw}^p < \cos \theta_{wg}^p \), a non-linear boundary between regions I and II can never arise. However, it may be clear that for a MWS system with \( \cos \theta_{gw}^p > 0 \) six different configurations of a non-linear boundary between regions II and III may arise, determined by the different orderings of the \( P_{c,gw} \) threshold values.

Consequently, in a general fractionally wet system, where the sizes of the water-wet pores are not strictly ordered with respect to those of the oil-wet pores, both the \( P_{c,go} \) and the \( P_{c,gw} \) threshold values (if \( \cos \theta_{gw}^p > 0 \)) may occur in various orderings. Therefore, in these systems non-linear boundaries between regions I and II as well as between regions II and III may occur. The FW system of Fig. 6 is a particular example of such a system, where the ranges of the sizes of the water-wet and oil-wet pores are the same. Taking all the possible orderings of both the gas–oil and the gas–water capillary entry pressures into account when water is wetting to gas in the oil-wet pores, the number of saturation-dependency configurations in a general fractionally wet system could be as large as 36. However, considering the pore occupancies related to regions I and III shows that these regions can never directly border, except for \( S_w = S_{wet} \) or \( S_o = 1 - S_{wet} \). This avoids, for example, that the two non-linear boundaries intersect and also eliminate half the number of possible configurations.

If \( \cos \theta_{gw}^p < 0 \), i.e. if gas is wetting to water in the oil-wet pores, only one non-linear boundary (between regions I and II) may arise and all the possible configurations for a general fractionally wet system are again given by the six configurations of Fig. 7, with the lower right triangle belonging to region II.

Furthermore, observe that with relations (9a) and (9b) the gas–oil contact angles in relation (28) and the gas–water contact angles can be replaced by expressions in terms of the oil–water contact angles \( \cos \theta_{ow}^p \) and \( \cos \theta_{wg}^p \) and the interfacial tensions. Hence, the saturation-dependency configurations are given in terms of the essential parameters, i.e. the interfacial tensions, sizes of the smallest and largest pores of the water-wet and oil-wet clusters and the degree of wettability of the pores.

Regarding spreading oils, van Dijke et al. [39] showed that for MWL systems the threshold values of the \( P_{c,go} \) are always ordered to yield the configuration of Fig. 7(a) only, whereas the remaining five configurations relate exclusively to non-spreading oils. However, for general fractionally wet systems all discussed possible configurations may occur for both non-spreading and spreading oils.

5.3. Non-genuine behaviour

In Section 4.3, we have shown that occupancies of type II may be non-genuine, such that no explicit links
can be made between the corresponding two-phase and three-phase oil or water relative permeability relations. As both kinds of non-genuineness may or may not happen, in saturation space this means that four subregions of region II may emerge, with either zero, one or two kinds of non-genuineness. The boundaries between these subregions follow again by considering the related pore occupancies, in particular the two-phase occupancies of Fig. 5, as these show both two gas–oil and two gas–water boundaries.

The only possible boundary in saturation space between a genuine and a non-genuine subregion II with respect to the oil relative permeability, occurs when the gas–oil boundary of Fig. 5(a) is on the brink of disappearing from the water-wet pores at \( r = r_{go}^{w} \), while the boundary in the oil-wet pores remains present. Obviously, the latter is only possible for (two) particular orderings of the \( P_{c,go} \) threshold values. With respect to the gas-water boundaries, the boundary in the oil-wet pores shown in Fig. 5(b) must vanish at \( r = r_{go}^{w} \) to change from a non-genuine to a genuine type II occupancy.

At the boundary between genuine and non-genuine saturation-dependencies, the disappearing gas–oil boundary requires \( r_{go}^{w} = r_{1}^{w} \), which fixes the remaining gas-oil boundary as \( r_{go}^{o} \) through the Young–Laplace equations: \( P_{c,go}(r_{go}^{o}) = P_{c,go}(r_{1}^{o}) \) whereas the gas–water boundary may vary (see Fig. 4(c)). Hence, the boundary in saturation space separating genuine and non-genuine saturation-dependencies with respect to the oil relative permeability is given by the constant oil saturation

\[
\bar{S}_o = \int_{r_{1}^{o}}^{r_{go}^{o}} \varphi(r)V(r) \, dr.
\]

In Fig. 8, two possible configurations with different subregions of region II are presented for \( \cos \theta_{gw}^w > 0 \) and \( \cos \theta_{gw}^w < 0 \), respectively.

As indicated by Fig. 8 the endpoints of the lines \( S_o = \bar{S}_o \) and \( S_w = \bar{S}_w \) coincide with the intersection of the non-linear boundaries with the \( S_w = 0 \) axis and the \( S_o = 0 \) axis, respectively. This follows by considering that, for example, the non-linear boundary between regions II and III is given by \( r_{gw} = r_{gw}^{o} \) (compare Fig. 2(b) with Fig. 4(c)), whereas at the point where oil is no longer present, we have \( r_{gw} = r_{1}^{o} \). Hence, at the intersection of the non-linear boundary with the axis we find \( r_{gw}^{w} = r_{1}^{w} \), the condition at the boundary between genuine and non-genuine.

Observe that the configuration of Fig. 6 is qualitatively the same as that of Fig. 8(a). The saturation-dependency of the oil relative permeability is non-genuine in almost the entire region II. Observe further that although the part of region III for \( S_w < S_{uw} \) is small, it stretches so far along the \( S_o = 0 \) axis, i.e. \( \bar{S}_w \) is relatively small, that also the saturation-dependency of the water relative permeability is non-genuine in almost

---

**Fig. 8.** Configurations of the saturation-dependency regions with different links between the two-phase and three-phase relative permeabilities in different parts of region II for: (a) \( \cos \theta_{gw}^w > 0 \); (b) \( \cos \theta_{gw}^w < 0 \), in a general fractionally wet system. In 1 both the water and the oil relative permeability saturation-dependencies are genuine, in 2 the oil saturation-dependency is non-genuine, in 3, the water saturation-dependency is non-genuine and in 4 both the oil and water saturation-dependencies are non-genuine.
the entire region II. These quantitative effects are, however, mainly caused by the particular choices of the pore size distribution function and the volume and conductance functions.

From Fig. 8, subregions with genuine and non-genuine saturation-dependencies are easily found for all possible configurations. With respect to the boundaries between the genuine and non-genuine saturation-dependencies of the oil relative permeability, we consider again the configurations of Fig. 7. In Figs. 7(c) and (e), a true boundary between genuine and non-genuine can be found as in Fig. 8. In Fig. 7(f), the limiting case of Fig. 7(e), the endpoint of the non-linear boundary, hence the boundary between genuine and non-genuine, coincides with the line \( S_w = 1 - S_{\text{net}} \). Therefore, the entire region II has genuine saturation-dependencies, irrespective of the sign of \( \cos \theta_{gw} \) (compare Figs. 8(a) and (b)). On the other hand, in Figs. 7(b) and (d) (and similarly in Fig. 7(a) as the limiting case of Fig. 7(b)) the endpoint of the non-linear boundary is such that \( S_w = 0 \), i.e. in the entire region II the oil relative permeability has a non-genuine saturation-dependency. Obviously, similar conclusions can be drawn for the non-genuineness with respect to the water relative permeability, based on the position of \( S_w = 1 - S_{\text{net}} \).

5.4. Discussion

Under certain simplifying assumptions, we have shown how the saturation space may be divided into regions with different saturation-dependencies of the relative permeabilities, based on a number of underlying pore-scale parameters. The required parameters are the values of the interfacial tensions, the ranges of pore sizes of the water-wet and oil-wet pores and the values of the oil-water contact angles. Knowledge of these essential parameters determines the qualitative picture of the subdivision of the saturation space, such as Figs. 7 and 8, i.e. it indicates how many saturation-dependency regions occur and whether part of these regions has genuine saturation-dependencies. Additional information, such as the form of the pore size distribution function and the volume and conductance functions, adds only quantitative features, such as the exact location of non-linear boundaries and the precise curvature of isoperms for relative permeabilities that depend on more than one saturation.

The above-listed essential parameters may be determined from experiments. For example, the interfacial tensions can be measured directly. The pore size distribution can be estimated from mercury porosimetry and, in particular, the capillary pressure curve yields information on the values of the smallest and largest pore sizes. As shown above, \( S_{\text{net}} \) is a major indicator in the analysis of the saturation space and it provides information on the values of the largest water-wet and the smallest oil-wet pores. The value of \( S_{\text{net}} \), the saturation related to exactly filling all the water-wet pores, can be estimated from the transition of the two-phase \( P_{gw} \) curve from positive to negative values and also from Amott and USBM wettability tests [13]. Obtaining the precise values of the contact angles is more difficult, but for the outlined qualitative saturation-dependency analysis, it is sufficient to determine the wetting orders only. For example, if the \( P_{gw} \) curve changes sign, then this indicates that gas is wetting to water in the oil-wet pores.

Clearly, the interconnectedness of pores in real porous systems leads to effects such as multiple displacements and trapping that cannot be captured by the present capillary tube model. However, as argued in the next section, this does not fundamentally undermine the qualitative analysis of the saturation-dependencies. An additional complication that may arise in real porous systems is that degrees of wettabilities cannot be expected to be constant, as we assumed so far, but may vary within certain ranges (see, for example, the simulation in [40]). This issue can be addressed within a capillary tube model, hence in a forthcoming paper we will more thoroughly analyse cases with oil–water contact angles that are randomly distributed, by considering pore occupancies as probabilities that the corresponding capillary entry pressures in relations (4a)–(4c) are overcome. Using this approach, it can be shown that parts of the saturation space exist in which none of the three relative permeabilities depend only on its own saturation. Furthermore, although still regions remain where only one set of isoperms is curved (indicated as regions I, II and III in this paper), not only in region II, but also in parts of region I, saturation-dependencies may be non-genuine.

Even from the present simple pore-scale model it has become clear that in many situations it is theoretically impossible to directly construct a model for three-phase relative permeability relations from two-phase data in the entire saturation space. Instead, the above-mentioned essential pore-scale parameters, which may be derived from two-phase data, must be used in a process-based pore-scale model, such as a network model, to predict three-phase relative permeability relations.

Having noted that there are certain saturation-dependency regions where we cannot relate three-phase and two-phase relative permeabilities, we now indicate some cases where we can say something definitive based on the present model. In regions of the saturation space where either oil or water appears to be the intermediate-wetting phase (regions I and III), the three-phase relative permeabilities can always be linked to the two-phase relations. In the region where gas is the intermediate-wetting phase (region II) this identification is only possible if the two-phase gas–oil filling sequences indicate that gas invades all water-wet pores before filling any oil-wet pores and, similarly, if the two-phase gas–
water filling sequences indicate that gas invades all oil-wet pores before filling any water-wet pores. For example, in a MWL system these conditions are met only if the spreading coefficient is strongly negative and the pore size distribution is very narrow (configuration of Fig. 7(f)). Alternatively, region II could be absent, which happens for example for a spreading oil in a MWL system when water is wetting to gas in the oil-wet pores (configuration of Fig. 7(a), with region III present).

6. Comparison with other three-phase relative permeability relations

In recent years, several authors (e.g. [3,8,11]) have given overviews of existing three-phase relative permeability relations and corresponding experimental data. Existing relations often assume that the relative permeabilities of two of the phases depend only on their own saturations (e.g. [9,24,34,36,37]) and are further assumed to be equal to one of the two-phase relative permeabilities. More recently, there has been a growing awareness of wettability and of its effect on relative permeabilities [8,12,16,22,25,28,30,32,33,41]. In this work, we seek to compare the current model predictions for three-phase relative permeabilities in weakly wetted mixed-wet and fractionally wet systems on: (i) the possible phase saturation-dependency regions in a fully connected system (where there is no phase trapping or hysteresis); (ii) where these saturation-dependency regions are and are not expected to relate to corresponding two-phase occupancies and relative permeabilities; and (iii) how the various empirical models relate to well-founded pore scale physics as expressed by the various authors. It is important to note that the broad saturation-dependency regions predicted by our model will still exist even in complex inter-connected pore networks at capillary equilibrium when all three phases are well connected (even when there are some trapped and isolated phase ganglia). Hence, the central findings of this work should still be used to compare with the phase saturation regions predicted by empirical models as is done below.

In Table 1, we summarise the models that have given original contributions with respect to saturation-dependencies and links between two-phase and three-phase relations, especially in view of variable wettability states and compare these with the findings of the present model. A more detailed comparison is given in [40]. Note that in Table 1 only the completely accessible versions of the existing models are considered, i.e. without the effects of residual saturations, trapping and hysteresis. In this table, we present the quantity \( \sum_{i=0,w,g} k_{i,r} \), which, in a fully accessible capillary bundle model in the absence of intra-pore effects equals 1. We note that this is a theoretical requirement of such models although this behaviour is not expected experimentally in real rocks. We make this comparison here simply to examine the theoretical consistency between various empirical formulations and the limiting case of a capillary bundle model, as presented in this paper.

The model of Corey et al. [9] was the first reported three-phase relative permeability. They considered a water-wet system and correctly assumed, referring to Leverett and Lewis [26], that oil occupies the intermediate-sized pores, and generalised the two-phase expressions of Burdine [7] for a tortuous capillary bundle model. Models based on the same assumption but involving different tortuosity relations, and thus different functional forms, have been developed by Naar and Wygal [31] and Parker et al. [34].

Stone [36], referred to as Stone I, assumed identically to [9] that in a water-wet system the oil occupies the intermediate-sized pores, but with an expression for the oil relative permeability that explicitly incorporates the two corresponding two-phase oil relative permeabilities. In a second model, referred to as Stone II, Stone [37] developed relative permeability relations based on the consistency relation for a fully accessible capillary bundle model, assuming \( \sum k_{i,r} = 1 \). In an appendix of [37] possible pore filling sequences in a mixed-wet system with a fraction of the (smaller) pores strongly water-wet and the remaining pores strongly oil-wet were discussed.

Starting with an oil-filled medium it was hypothesised how subsequent water and gas floods invade. It was correctly assumed that water starts invading the water-wet pores in increasing order of size, followed by the oil-wet pores in decreasing order of size. Gas was assumed to invade oil-filled pores starting in decreasing order of size, starting at the largest irrespective of the oil–water wettability, which, as we note in Table 1, is generally not correct [39]. If water and gas saturations are increased such that they are no longer separated by oil, Stone assumes that water will separate oil and gas, leading to relative permeabilities corresponding to an oil-wet system with water as the intermediate-wetting phase. Although this assumption is generally incorrect, the underlying argument leads to the conjecture that for a mixed-wet system different phases are intermediate-wet in different areas of the saturation space. This idea has been verified in [39] and in the present study. Several authors have incorporated additional phase trapping and hysteresis models in Stone I and II, but did not change the basic forms of the method to include wettability effects [2,14,15,23].

Heiba et al. [17] considered pore filling sequences in a system in which the larger pores have become oil-wet, a case of mixed-wettability. They observed quite complex saturation-dependency behaviour but based on the wrong pore entry conditions [40]. Since a three-phase relative permeability model was not explicitly derived,
the ideas cannot be compared any further with the present work.

Aleman and Slattery [1] used the physics of flow on the pore-scale to obtain a new functional form for two-phase and three-phase relative permeability relations in water-wet pores. However, the calculated saturation-dependencies were merely a result of the traditional assumption that in water-wet systems the intermediate-wetting phase (oil) occupies the intermediate-sized pores.

The model of Temeng [38], see Table 1, included wettability effects in relative permeability relations. Although the model is equivalent to the present model

for strongly water-wet and oil-wet conditions (assuming that water is intermediate-wetting in the oil-wet pores), for all weakly-wet conditions water and oil relative permeabilities depend on more than one saturation, which is certainly not in agreement with the present model. Both Temeng and Bradford [6], see Table 1, assume that only the gas relative permeability depends on its own saturation, whereas oil and water relative permeabilities are both functions of more than one saturation. This is not in accordance with the findings of the present model, as in different saturation ranges either oil, water or gas relative permeabilities may depend on more than one saturation.
Baker [3] took empirical saturation-weighted relative permeabilities to interpolate between various two-phase data for water-wet systems. Later, Baker [4] used a similar approach to analyse three-phase relative permeability data from water-wet, oil-wet and intermediate-wet cores. Hustad and Hansen [18] also employed the saturation-weighted method for the oil relative permeability, but used a sophisticated scaling of the oil saturation to include residual saturations. In addition, Blunt [8] used saturation weighting for all three relative permeabilities, as he explicitly recognised that gas may also be intermediate-wetting in oil-wet pores. Similarly, Jerauld [20] adopted a saturation-weighted method for the water relative permeability as a basic form. Although the saturation-weighted method is quite general and may match specific experimental data quite well, it lacks a pore-scale basis and does not recognise that in some saturation ranges two of the relative permeabilities may still be functions of their own saturation.

The methods of Jerauld [20], Larsen and Skauge [25] and Blunt [8] also include provision for wettability variations through parameters reflecting trapped water saturation, as in a mixed-wet system both oil and gas may trap water. Finally, new forms of relative permeability relations have been introduced as a result of a more detailed view of the intra-pore-scale physics, such as flow in surface roughness. For example, Moulu et al. [29] proposed a new method based on a fractal representation of a capillary bundle model for a water-wet system, which was extended to mixed-wettable within a pore [30]. Blunt [8] included oil flow through layers to reflect fluid configurations in pore crevices. These concepts are not yet included in the present model but will in due course constitute an important extension, as surface roughness also plays a major role in pore wettability.

In conclusion, we have found that most existing models for three-phase relative permeabilities are based on saturation-dependencies for strongly water-wet systems only, where oil is considered as the intermediate-wetting phase. Furthermore, saturation-dependency regions where two of the relative permeabilities depend on only their own saturations, except region I where oil is intermediate-wetting, have not been generally appreciated. The incorporation of wettability effects in relative permeability relations has not been done for the general case on the basis of the present authors’ understanding of the pore-scale physics. All relative permeability models, apart from the tortuosity-based methods, link three-phase to two-phase relations. Therefore, almost all models approach the corresponding two-phase relations when one of the saturations vanishes. However, non-generic saturation-dependencies have not yet been recognised. To be useful for realistic flow simulations the present model is being extended to include distributed instead of constant contact angles. Furthermore, interconnectivity of pores as well as additional intra-pore-scale effects need to be incorporated.

7. Summary and conclusions

A method has been developed to determine saturation-dependencies of relative permeabilities in three-phase flow which is suitable for all types of mixed-wet and fractionally wet porous systems that contain clusters of oil-wet and water-wet pores with constant but different oil–water contact angles. Based on classification of all allowed pore occupancies in a completely accessible porous medium, saturation-dependencies of the corresponding relative permeabilities are derived. Furthermore, three-phase relative permeabilities that appear to depend only on their own saturations are either linked to the corresponding two-phase relative permeabilities or it is shown that such a link cannot be established. A comparison has been made with existing relative permeability models with respect to their saturation-dependencies.

The specific conclusions from this work are:

(i) To analyse weakly wetted systems, linear relations between the cosines of the contact angles are adopted which are consistent with strongly wetting conditions.

(ii) Depending on the interfacial tensions, the ranges of the pore sizes and the degree of wettability of the pores, up to three regions in saturation space can be identified and related to the saturation-dependencies of three-phase capillary pressures and relative permeabilities. The values of the various capillary entry pressures for the smallest and largest pores of the water-wet and oil-wet pore clusters determine the boundaries of these regions.

(iii) In each region of saturation space, one phase turns out to be “intermediate-wetting” for the porous medium as a whole. In such a region, then only the relative permeability of the intermediate-wetting phase depends on more than one saturation.

(iv) When water is wetting to gas in oil-wet pores (cos \( \theta_{gw}^0 > 0 \)), three regions occur, one where oil is the intermediate-wetting phase (region I), one where gas is the intermediate-wetting phase (region II) and one where water is the intermediate-wetting phase (region III). For gas wetting to water in oil-wet pores (cos \( \theta_{gw}^0 < 0 \)), two regions occur, regions I and II.

(v) Boundaries between regions I and II and between regions II and III may occur that do not coincide with iso-saturation lines and, therefore, cannot be predicted without knowledge of the underlying pore occupancies. The exact location of the boundary between regions I and II is determined by the two
boundaries that occur between the phases in the corresponding two-phase gas–oil pore occupancy. The exact location of the boundary between regions II and III is similarly determined by the corresponding two-phase gas–water occupancy.

(vi) In region II, the water and oil relative permeabilities, which depend only on their own saturations, are not always identical to one of the corresponding two-phase relative permeabilities. This saturation-dependency is called non-genuine. For the presently considered systems, saturation-dependencies for gas and water in region I and for gas and oil in region III are always genuine.

(vii) The saturation-dependency configuration occurring for a spreading oil can be considered as a particular case of that for a non-spreading oil. We note that, in a fully connected network model, other aspects of the physics of spreading and non-spreading systems, such as film drainage, accessibility and conductivity behaviour, may lead to differences between spreading and non-spreading oils.

General conclusions are as follows:

(viii) The present model suggests that, in order to analyse the various saturation-dependency regions for experimental three-phase relative permeabilities, it is essential to measure the three interfacial tensions, the sizes of the smallest and largest pores of water-wet and oil-wet clusters and the degree of wettability of the pores.

(ix) The occurrence of non-linear boundaries in saturation space between saturation-dependency regions, as well as possibility of both genuine and non-genuine saturation-dependencies stresses the importance of knowledge of underlying pore occupancies. Therefore, only a process-based approach to three-phase flow modelling will lead to a correct determination of three-phase relative permeabilities.

(x) Existing three-phase relative permeability models incorporate a correct view of the pore-scale physics with respect to saturation-dependencies for systems with strongly water-wet pores. The present work indicates that further development is required in order to incorporate the more complex saturation-dependencies predicted for mixed-wet and fractionally wet systems.

Appendix A. Consistency relation between the capillary entry pressures

By definition, the capillary pressures satisfy

\[ P_{gw} = P_{go} + P_{ow} \]

but the analogous equation (6) for the capillary entry pressures is not a priori known. This relation follows from the consistency requirement that each point of the pore space must be filled with either water, oil or gas. Therefore, if for a single pore two of the conditions (4a)–(4c) are not satisfied, the third must be true. For example, if (4a) and (4b) are not satisfied, then either

\[ P_{gw} \leq P_{c, gw} \quad \text{and} \quad P_{go} > P_{c, go} \]

or

\[ P_{ow} \leq P_{c, ow} \quad \text{and} \quad P_{go} < P_{c, go} \]

applies. If conditions (A.1a) are satisfied, but \( P_{ow} > P_{c, ow} \) is true (which would contradict conditions (4c), hence occupancy by water) then

\[ P_{c, gw} \geq P_{gw} = P_{go} + P_{ow} > P_{c, go} + P_{c, ow}. \]  

If conditions (A.1b) are satisfied, but \( P_{gw} > P_{c, gw} \) is true (which would also contradict occupancy by water), then

\[ P_{c, gw} < P_{gw} = P_{go} + P_{ow} < P_{c, go} + P_{c, ow}. \] 

For consistency both inequalities (A.2) and (A.3) must be false, yielding the desired relation (6) for the capillary entry pressures.

For each pore the entry pressures are given as

\[ P_{c, ij} = \sigma_{ij} \cos \theta_{ij} / R, \]

where \( R / \cos \theta_{ij} \) denotes the radius of curvature of the capillary interface between the phases \( i \) and \( j \) and \( R \) is the effective entry radius of the pore [5]. Hence, Eqs. (5) and (6) are equivalent, in particular if \( R = r / 2 \) for cylindrical pores. However, the above derivation does not depend on the geometry of the pore, therefore Eqs. (5) and (6) are not only valid for cylinders but for any pore shape for which an effective entry radius can be defined. Zhou and Blunt [42] indicated that in a porous medium with three phases at gravity-capillarity equilibrium Eq. (5) ensures continuity of the saturation distributions at the level where the third, organic, phase is about to vanish. We have generalised their result (i.e. concerning relation (6) between the capillary entry pressures and the continuity of phases) since our argument does not depend on the vertical equilibrium condition.

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