

MODELING OF WAX DEPOSITION DURING OIL PRODUCTION USING A TWO-PHASE FLASH CALCULATION

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Abstract

Wax deposition is a serious problem in the Petroleum Industry that results in the plugging of flow strings, formation damage, loss of hydrocarbons, increased production cost. The existing models used in the oil industry for predicting wax phase equilibrium parameters over-estimate the amount of wax that is formed in terms of the wax weight fraction and the number of moles of solid. The Coutinho correlation is the latest of these models and is used to calculate the melting temperature and the enthalpy of fusion of the crude oil components. It did not consider the effect of branching of carbon atoms in the hydrocarbon structure/compound. But branching of carbon atoms in the hydrocarbon compounds affects its melting point.

In this work, the correlation for calculating the melting temperature in crude oil developed by Coutinho has been modified to take into account the effect of branching of carbon-carbon chains in isoparaffins and the model has been used to calculate a new set of equilibrium parameter. The modified regular solution theory was used to calculate the activity coefficient ratio that was used as an input to the new model. Wax phase equilibrium flash calculations were carried out with the new set of equilibrium parameters. The wax mixture was then characterized using the weight fraction of component in the solid phase and weight fraction of component in the solid phase in the mixture.

In order to check the reliability of the model, the data presented by Hanquan was used as input into the models developed by Won, Chung, Coutinho and the new one. The new model predicted a more conservative value for the number of moles of solid formed, weight fraction of component in the solid phase and weight fraction of component in the solid phase in the mixture for C₁₈-C₄₀. This is an improvement over the existing models which overestimate the values of the parameters.

Key words: wax; deposition; phase equilibrium; mathematical modeling.

1. Introduction

One of the major problems confronting the petroleum industry is the untimely blockage of oil arteries due to deposition of heavy organics (asphaltene, resin, paraffin wax) present in the oil ^[1]. This deposited organics increase in thickness due to continued deposition and also ages due to an increase in its wax content. The former may lead to the plugging of the complete pipe cross-section, while the latter leads to hardening of the deposited wax. The economic implications of such depositions are tremendous. The phenomena may also take place in the formation near the wellbore.

The precipitated solids adversely affect the economics of exploiting and developing fields prone to solids deposition. Some of the adverse effects are listed as following ^[2]:

- Permeability reduction with formation damage.
- Reservoir fluid composition changes.
- Alterations of reservoir rock wettability (wettability reversal).
- Reduction of the interior diameter and eventual plugging of production strings.
- Deposition of solids in surface facilities reducing separator and tank volume.
- Reduced flow and possible plugging of subsea pipelines.
- Increase service costs and lost production during downtime.

- Decreased pipe diameter- a major concern to oil transportation companies as they represent a major increase in pumping costs.
- Loss of throughput and quality of oil.

Waxes are defined as the relatively high molecular weight C_{18} - C_{60} alkanes, which are deposited as solids where there is a change in thermodynamic equilibrium, such that the temperature falls below the cloud point. The cloud point is a critical point in the rheology of waxy crudes. Above the cloud point, flow is Newtonian and below the cloud point, it is non-Newtonian due to wax/solid precipitation. When the rheology of the fluid changes, they observe a ten-fold or more increase in viscosity. Hence the oil becomes thicker and one needs more compressor horse power to pump such oil in the pipeline. The wax precipitated from oil mixtures primarily consists of C_{18} to C_{36} (paraffin waxes) or C_{30} to C_{60} (microcrystalline waxes), both made up of aligned paraffinic and naphthenic molecules. Paraffin waxes are also called macrocrystalline.

2. Literature review

Deposition of heavy organics from crude oil has been studied ^[1-10]. It has been reported that there are four mechanisms for wax deposition and four mechanisms for asphaltene deposition. They are molecular diffusion, shear dispersion, Brownian diffusion and gravity settling for wax, and polydispersivity effect, colloidal effect, aggregate effect and electrokinetic/wall for asphaltene.

Won ^[1] presented a thermodynamic prediction method of vapor-liquid-solid wax phase equilibrium of paraffinic hydrocarbon mixtures. The result of the study showed that the non-idealities in the condensed phase could be taken care of by introducing new solubility parameters for solid solution constituents.

Svendson ^[2] developed a mathematical model for prediction of wax deposition in both open and close pipeline systems by using a combination of analytical and numerical models. The model predicts that wax deposition can be considerably reduced when the wall temperature is below wax appearance point, provided that the liquid/solid phase transition expressed by the change in moles of liquid with temperature is small at the wall temperature. If, in addition, the coefficient of thermal expansion is sufficiently large, some components may separate and move in opposite radial direction at temperature below wax appearance temperature. Particle transport and sloughing were not considered

Nielsen *et al.* ^[3] performed experiments to measure paraffin deposition rates. Laboratory test procedures and a computational model were developed to predict rate of paraffin deposition and the effects of long term deposition on the pipeline pressure drop and temperature profile. They concluded that deposition by shear dispersion is not significant

Chung ^[2] presented a generalized predictive model which is based on thermodynamic principle for solid-liquid phase equilibrium for organic solid precipitation. The model takes into account the effects of temperatures, composition and pressure. The model assumes that asphaltenes are dissolved in oil in a true liquid state, not in colloidal suspension and that precipitation-dissolution process is reversible by changing thermodynamic conditions. The study concluded that further improvement to the prediction technique can be achieved by developing more accurate correlations for those model parameters of heat of fusion, melting temperature and molal volume.

Brown *et al.* ^[5] developed a thermodynamic model for paraffin formation which accurately accounts for the effect of light ends such as methane, ethane, and propane, and high pressures. The model uses the simplified, perturbed Hard Chain Equation of state for all fluid phases' fugacities and contains no adjustable parameters for the solid-liquid equilibria. Model prediction are compared to solid-liquid, solid-vapor and solid-liquid-vapour equilibria data. They concluded that the simplified, perturbed Hard Chain Theory equation of state was chosen to model all fluid phase properties since it provided a good representation of the vapour-liquid equilibria of high-molecular weight paraffins in light solvents.

Pedersen ^[6] presented a vapour-liquid- solid model for predicting phase equilibria of oil mixtures taking into account the possible formation of a wax phase. The gas and liquid phases were described using the Soave-Redlich-Kwong equation of state while the wax phase was assumed to be an ideal mixture. Only part of the heavy hydrocarbons was considered to be potentially capable of entering into a wax phase. A procedure was developed for estimating the fraction of the heavy hydrocarbon that may potentially form wax.

Huanquan and Firoozabadi [7] modified a multi-solid wax precipitation model to study the effect of pressure and composition on wax precipitation. The result of their study showed, they found that the mixing of crude with a solvent increases the cloud point temperature contrary to the widespread assumption that the addition of solvents such as C₅ increases the cloud point.

Ahmed Hammani *et al.* [8] did a comparison of laboratory result to field data on paraffin deposition from crude oils. They evaluated the characteristics of four reservoir fluid samples from two offshore fields in the Gulf of Mexico as well as their propensities towards paraffin deposition due to temperature and/or pressure variations. State of the art techniques namely a cross-polarized microscopy (CPM) and a laser based solids deflection system (SDS) was used to measure the onsets of paraffin crystallization temperature. Their results were compared to field data and the observed difference and/or similarities between laboratory and field WAT were explained.

Coutinho *et al.* [9] proposed a thermodynamic model for the non-ideality of the solid phase, based on the predictive local composition concept. The extension of the model to high pressures was also discussed. They suggested that using an EoS/G^E model for the fluid phases and introducing the excess volume of the solid solution to account for the effects of the pressure in the solid phase non ideality gives an excellent description of both fluid-fluid and solid-fluid phase boundaries is achieved, even for crude oils.

Kok *et al.* [10] developed a mathematical model for predicting wax deposition in pipeline systems using a combination of analytical and numerical models.

Coutinho *et al.* [11] developed a wax model based on high accuracy thermodynamic data. The model can be used in conjunction with equations of state to perform wax equilibrium calculation for black oil.

2.1 Solid liquid equilibrium thermodynamic modeling

In order to determine the mole fraction of solid composition, the equilibrium solid/liquid K-value should be determined.

At thermodynamic equilibrium between a liquid (oil) and a solid (wax) phase, the fugacity, f_i^L , of component i in the liquid phase equal the fugacity, f_i^S , of component in the solid phase. The fugacity measure of departure of a substance from zero pressure states that:

$$f_i^L = f_i^S \quad (1)$$

Where the fugacities of the i -th component in solid and liquid phases are defined as:

$$f_i^L = \gamma_i^L X_i^L f_i^{0L} \exp \left[\int_0^P \frac{V_i^L}{RT} dp \right] \quad (2)$$

$$f_i^S = \gamma_i^S S_i^S f_i^{0S} \exp \left[\int_0^P \frac{V_i^S}{RT} dp \right] \quad (3)$$

Where γ_i , X_i , V_i and f_i^0 are the activity coefficient, mole fraction, molar volume and standard state fugacity of component- i respectively. The superscript s stands for solid state, and l for liquid state.

The solid-liquid equilibrium coefficient, K_i^{sl} , is then derived by combing Equations 1, 2, and 3

$$K_i^{sl} = \frac{f_i^S}{f_i^L} = \left(\frac{\gamma_i^S}{\gamma_i^L} \right) \left(\frac{f_i^{0L}}{f_i^{0S}} \right) \exp \left[\int_0^P \frac{\Delta V_i}{RT} dp \right] \quad (4)$$

Where $\Delta V_i = V_i^L - V_i^S$

In Equation 4, the equilibrium coefficient is determined by three factors. The first term on the right-hand side is the effect of molecular interaction between components, the second term is the effect of temperature, and the third term is the effect of pressure. Thus Equation 4 is a general representation for solid-liquid equilibrium constant.

In solid-liquid systems, the change in molar volume, ΔV_i is usually small. Thus, the third term usually can be neglected if there is no significant pressure change [3].

The relationship between the standard state fugacity of components i in the solid phase, f_i^{0S} , and that in the liquid phase, f_i^{0L} is given as

$$\ln \left(\frac{f_i^{0L}}{f_i^{0S}} \right) = \frac{\Delta H_i^f}{RT} \left(1 - \frac{T}{T_i^f} \right) \quad (5)$$

Where ΔH^f and T_I^f are the Heat of melting and melting point temperature of solute i respectively, ΔC_p is the difference of heat capacity between two phases, and R is the universal gas constant. The second term is usually small and can be neglected [32].

Hence equation (4) reduces to.

$$K_I^{SL} = \frac{y_i^g}{x_i^l} = \left(\frac{y_i^f}{x_i^f}\right) \exp \left[\frac{\Delta H_i^f}{RT} \left(1 - \frac{T}{T_I^f}\right) \right] \quad (6)$$

The melting point temperature, T_I^f , the enthalpy of fusion, ΔH^f are evaluated for paraffin species according to the following correlations by Won [1]. For normal alkanes

$$T_I^f = 374.5 + 0.02617MW_I - \frac{20172}{MW_I} \quad (7)$$

MW_I is the molecular weight of the component I , by using the melting point temperature, the heat of fusion is calculated as follows.

$$\Delta H_I^f = 1.14 * 0.1426MW_I T_I^f \quad (8)$$

The constant 1.14 is included to obtain a better fit.

Chung [5] improved the Won's [1] correlation as follows.

$$\Delta H_I^f = 0.9T_I^f (MW_I^{0.55}) \quad (9)$$

The correlation proposed by Lira Galeana et al. [7] is used to calculate T_I^f for asphaltenes and iso paraffins; and ΔH_I^f for iso paraffins.

$$T_I^f = 333.46 - 419.01 \exp(-0.008547MW_I) \quad (10)$$

$$\Delta H_I^f = 0.05276MW_I T_I^f \quad (11)$$

Coutinho [11] suggested that when wax precipitates from a real crude oil, the n-paraffins solidify to form an orthorhombic solid solution. For n- paraffin with a carbon number of 42 or more, the normal melting point can be obtained from equation 12

$$T_I^f = 421.63 - 1936112.63 \exp[-7.8945(N - 1^{0.07194})] \quad (12)$$

And the enthalpy of fusion as:

$$\Delta H_I^f = 1000(3.7791N - 12.654) \quad (13)$$

Where N is the carbon number,

For n-paraffins below a carbon number of 42 (but greater than 4), The phase transition is more complicated. At the melting point they form a rotator phase. The melting temperature is given as:

$$T_I^f = 421.63 - 1936112.63 \exp[-7.8945(N - 1^{0.07194})] \quad (14)$$

The enthalpy of fusion can be obtained from the following correlation.

$$\Delta H_I^f = 1000(-0.00355N^3 + 0.2376N^2 - 3.6209N + 18.5391) \quad (15)$$

Note that in this model, the ratio of the activity coefficient of the liquid to that of then solid is assumed to be equal to unity. This was explained by the fact that the inclusion of the activity coefficient has only a small effect on cloud point prediction and that the heat of fusion/melting temperature effect dominates the cloud point more than the activity coefficient does.

2.2 Modifications to the Coutinho model

The original model assumed that the wax phase is formed by only n-alkanes crystallizing from a hydrocarbon fluid as orthorhombic crystals in solid solutions. However, it has been discovered that both normal and iso-paraffins actually deposit as wax. Coutinho assumed that all components with the same carbon number have the same melting temperature. But it has been found out that the presence of a branch in the carbon chain significantly drops the melting temperature of paraffins. Hence, there is the need for a temperature correction for iso-paraffins.

The temperature correction is given thus,

$$T_{I \text{ iso-paraffin}}^f = T_{I \text{ normal}}^f - \left(90 - \frac{80 * CN_I}{100 + CN_I}\right) \quad (16)$$

This is a critical improvement in the Coutinho model as this will perform the necessary corrections for iso paraffins.

In this work also, the activity coefficient was introduced. It has been reported that the ratio of activity coefficient is not unity as was assumed

Using the modified regular solution theory, the effect of the activity coefficient ratio has been estimated. The ratio of the activity coefficient for liquid to solid is:

$$\frac{\gamma_i^L}{\gamma_i^S} = \exp\left(\frac{V_i}{RT}\left(\bar{\sigma} - \sigma_i\right)_L - \left(\bar{\sigma} - \sigma_i\right)_S\right) \tag{17}$$

Where: V_i - molar volume in $(m^3/gmol)$; σ_L - solubility parameter for liquid; σ_s - solubility parameter for solid.

The molar volume V_i^L calculated is shown

$$V_i^L = \frac{MW_i}{0.8155 + 0.6272E - 4MW_i - \frac{13.06}{MW_i}} \tag{18}$$

where MW_i is the molecular weight of component i.

3. Result analysis

3.1 Data analysis and result

A Fortran program has been developed to perform the Equilibrium Flash Calculation. Comparison of mole fractions in solid phase with and without activity coefficient is shown briefly in the table below:

Table 1: Comparison of the mole fractions in solid phase

Model used	Si
Modified	0.5161
Won	0.6323
Coutinho	0.5290
Chung	0.60894

3.2 Analysis- wax weight fraction

One way of characterizing the mixture is to calculate the weight fraction of each component j in the feed, W_f

$$W_f = \frac{Z_j M_j}{\sum_{p=1}^n Z_p M_p} \tag{19}$$

The weight fraction of component j which is in the solid phase is W_{sf} . It is expressed as:

$$W_{sf} = \frac{Z_{sj} M_j}{Z_j M_j} \tag{20}$$

The weight fraction of component j in the solid phase in the mixture is W_{sj} ;

$$W_{sj} = \frac{Z_{sj} M_j}{\sum_{p=1}^n Z_p M_p} = W_f W_{sf} \tag{21}$$

The total weight fraction of wax crystals in the mixture, W_s , is expressed as:

$$W_s = \sum_{j=1}^n W_{sj} \tag{22}$$

The total weight fraction of liquid in the mixture is;

$$W_x = \sum_{f=1}^n W_{x_f} = 1 - W_s \quad (23)$$

Table 2 Comparison of models

Models	Ws	Wx	Wti C ₁₈ - C ₄₀
Modified	0.6592	0.3408	0.6079-0.9239
Won	0.7478	0.2522	0.6909-0.9775
Coutinho	0.5290	0.3221	0.6332-0.9438
Chung	0.7157	0.2843	0.6850-0.9826

It was observed generally that in all the cases considered, the weight fraction of component in the solid phase for C₁₈ to C₄₀ ranges between

0.6079 – 0.9231 Modified Coutinho

0.6232 – 0.9438 Coutinho

0.6909 – 0.9775 Won

0.6850 – 0.9826 Chung

This is observed to be the major reason why the existing model (Won, Chung) indicated high wax tendencies when applied to crude sample.

4. Discussion

Paraffin deposition is a problem that restricts production and causes loss of money. Paraffin deposition mostly results from cooling and precipitation of high molecular weight hydrocarbon during the movement of particles. Precipitation is an example of fluid solid phase equilibrium.

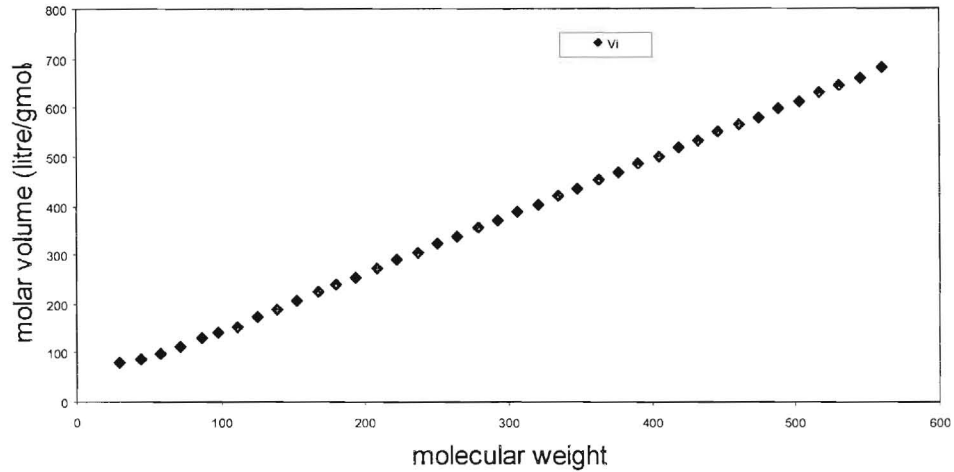
In a typical wax phase, various types of compounds are present; for example, normal paraffins constitute a very large percentage (90-95%) of the wax phase and the iso-normal account for the rest. While the normal paraffins are generally expected to be able to form the wax phase independent of the molecular weight, this is not the case with the iso-paraffins. The reduced melting temperatures lower its tendency or ability to form the wax phase. Alkanes show regular increases in melting point as the molecular weight increases. It has been noticed that non-polar molecules like alkanes are weakly attracted to each other by intermolecular Van der Waal's forces. These force, which operate only over very small distances result from induced polarization of the electron clouds in molecules. Although the electron distribution in a molecule is uniform on average over a period of time, the distribution at any given instant is not uniform. One side of a molecule may by chance have a slight excess of electrons relative to the opposite side. When that occurs, the molecules have a temporary dipole moment. This temporary dipole in one molecule causes a nearby molecule to adopt a temporarily opposite dipole with the result that a tiny attraction is induced between the two molecules. Temporary dipole have a fleeting or brief existence and are constantly changing, but the cumulative effect on a enormous number of these interactions produces attractive forces sufficient to cause the molecules to stay in the liquid state. Only when sufficient heat is applied to overcome these forces does the liquid melt.

As might be expected, Van der Waal's forces increase as molecule size increases. Although other factors are also involved at least part of the increase in melting point on going down the alkane series is due to increased Van der Waal's forces. The effect of branching on melting point can be explained by invoking Van der waal's forces. Branched alkanes are more spherical than the straight-chain alkanes. As a result, they have smaller surface areas, small Van der Waal's forces and consequently lower melting melting. The intermolecular or Van der Waal's are overcome at a lower temperature for iso-paraffins.

Figure 1 shows the relationship between the molar volume V_i and the molecular weight for the sample. A linear relationship is observed between the molar volumes and the molecular weight of paraffin. It has been observed that as the carbon number increases, the molecular weight increases, and if the molar volume is a function of molecular weight, it is reasonable to expect liquid and solid molar volumes of paraffin's to be a linear function of molecular weight.

Figures 2 and 3 show the weight fraction of component in the solid against the molecular weight for each of the models considered. It can be seen from the plot that the modified model of Coutinho gives a fairly lower value than the original one and when compared to the Won and Chung models, one can see that they have a similar shape except the Won and Chung

models give a higher value of number of moles of solid formed and the weight fraction of component in the solid phase.



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Fig. 1 Molar volume vs. molecular weight

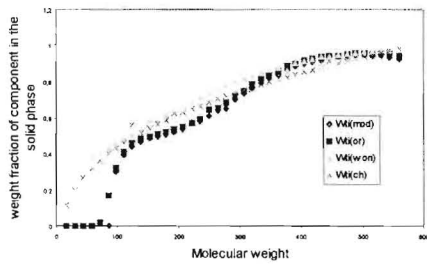


Fig. 2 Comparison of weight fraction of component in the solid vs. molecular weight

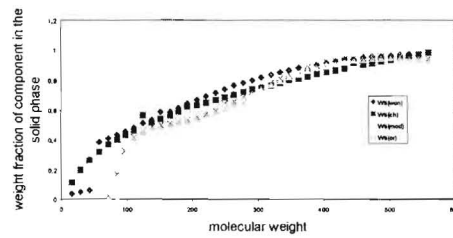


Fig. 3 Comparison of weight fraction of components in the solid vs. molecular weight

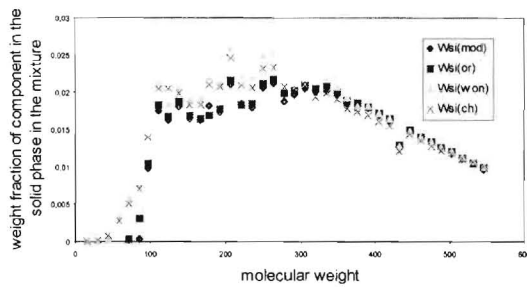


Fig. 4 Comparison of the weight fraction of solid in the mixture

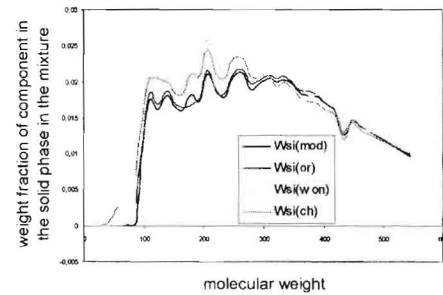


Fig. 5 Comparison of weight fraction of solid in the mixture

Figures 4 and 5, show the weight fraction of each component in the solid phase in the mixture against the molecular weight for each of the models considered. It can be seen from the plots that they have similar shapes, but the fact that one is above the other means that one gives

a higher value than the other does. It can be seen that the Modified Coutinho gives the smallest value of weight fraction in the mixture compared to other.

Figures 6 and 7 show the molar distribution of the oil sample used for this analysis, one shows the solid mole fraction and the other mole % of the component in the feed.

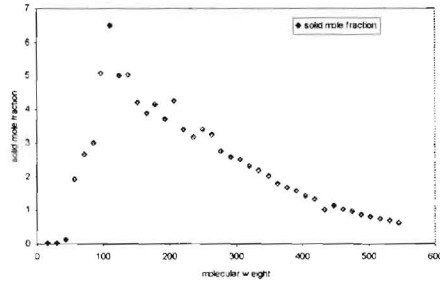


Fig. 6 Molar distribution of oil sample

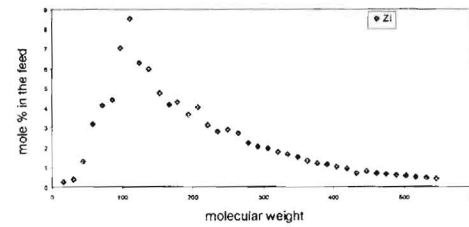


Fig. 7 Mole % of each component in the feed vs. molecular weight

Figure 8 shows the solid mole fraction at 1bar with it relationship to the carbon number.

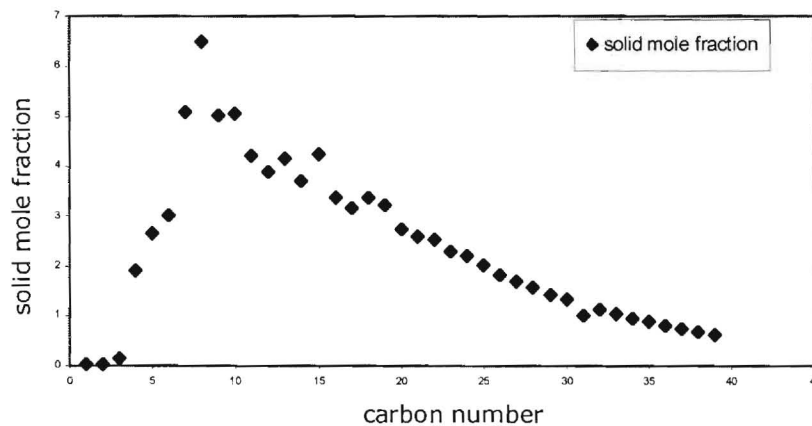


Fig. 8 Solid mole fraction at 1 bar

5. Conclusions

A solid deposition model is proposed for liquid-solid equilibrium based on modified regular solution theory.

A new set of equilibrium parameters was calculated using the most recent correlation for melting temperature and enthalpy of fusion. These two correlations are functions of carbon number.

A temperature correction was made for the presence of iso-paraffin since they also form wax with the normal paraffin.

The result obtained here are more conservative and accurate than some of the existing ones. This confirms the assertion that the existing models actually predict too large amounts of wax precipitation below the cloud point temperature.

Recommendation

The significant difference in the melting temperature of normal paraffins and iso-paraffins of a given carbon number in regard to wax precipitation. It is recommended that there is need to include the iso-paraffin for wax calculations and hence this has been done for the most recent correlation of Coutinho. This was observed to give better results when compared to the old models.

Nomenclature

a	Particle diameter
A	surface area available for deposition
C	Volume fraction concentration of wax in solution
C_w	Volume fraction concentration of wax out of solution at the wall
ΔC_p	Heat capacity difference between solid and liquid
D_m	Molecular diffusion coefficient
D_b	Brownian diffusion coefficient
D_s	Shear dispersion coefficient
f	Fugacity
f^o	Standard state fugacity
Δh^f	Enthalpy of melting
k	Mass transfer coefficient
m	Mixture properties
P	Pressure
r	Radial direction
R	Gas constant
S, L	Solid and Liquid phases
T	Temperature
T_a	Absolute temperature
T^f	Melting temperature
V	Molar volume
w_i	Mole fraction of component i in solid phase
x_i	Mole fraction of component i in liquid phase
γ^o	Oil shear rate at wall
γ	Activity coefficient
δ	Solubility parameter
n	Mass flux of dissolved wax molecules to the pipe wall (Kg/sm^2)
ρ	Density of solid wax (Kg/m^3)
D_m	Molecular diffusion coefficient of dissolved wax molecules (m^2/s)
dC/dr	Concentration gradient (Solubility coefficient) of dissolved wax close to the pipe wall with respect to distance (m^{-1})
dC/dT	Concentration gradient of dissolved wax with respect to temperature, $^{\circ}\text{C}^{-1}$
dT/dr	Radial temperature gradient close to the pipe wall ($^{\circ}\text{C}/\text{m}$)
β	Constant for a particular crude oil (N)

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