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Prediction of wax disappearance temperature using artificial neural networks

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ABSTRACT

In this study, the artificial neural network (ANN) was used for the prediction of WDT. The inputs to network are molar mass and pressure, and the output is WDT at each input. A two-layer network with different hidden neurons and different learning algorithms such as LM, SCG, GDA and BR were examined. The network with 16 hidden neurons and Levenberg–Marquardt (LM) train function showed the best results in comparison with the other networks. Also, the predicted results of this network were compared with the thermodynamic models and better accordance with experimental data for ANN was concluded.

1. Introduction

Crude oil and gas condensate consist of long *n*-alkane chains. In case the temperature reaches a lower temperature than wax appearance temperature, wax would precipitate inside the transferring equipments and pipes. Deposition of wax in the pipes and equipments will cause reduction of the diameter and termination of processes as a result. To prevent wax precipitation in the pipes, the passing fluid flow from the pipeline shall be kept on advanced conditions of appearance of the wax through heating or mixing the oil. Also, the pressure affects the appearance of wax, which is related to the changes in the solution ability of light components with changes in pressure and increasing the melting temperature of paraffins with pressure (Daridon et al., 2002).

Various empirical studies (Pauly et al., 1998; Metivaud et al., 1999; Pauly et al., 2001; Daridon et al., 2002; Pauly et al., 2004; Milhet et al., 2005; Rizzo et al., 2007; Vafaie-Sefti et al., 2007) were accomplished on wax deposition phenomenon. Also several thermodynamic models (Won, 1986; Hansen et al., 1998; Won, 1989; W.B. Pedersen et al., 1991; Lira-Galeana et al., 1996; Coutinho and Ruffier-Me´ray, 1997; Coutinho, 1998; Coutinho, 2000; Zuo et al., 2001; Ji et al., 2004; Coutinho et al., 2006; Esmaeilzadeh et al., 2006; Ghanaei et al., 2007; Dalirsefat and Feyzi, 2007) were presented for prediction of WAT, WDT or amount of precipitated wax.

Won (1986) and Won (1989) proposed two modified regular solutions for wax precipitation. Also, Hansen et al. (1998) proposed a modified regular solution, which used Flory's theory (1953) of multicomponent polymer solutions for activity coefficient of liquid phase. Furthermore W.B. Pedersen et al. (1991) developed the Won (1986) modified model for WAT calculations. Lira-Galeana et al. (1996) presented a multi-solid phase for wax precipitation. They assumed each solid phase was a pure component. Coutinho (1998) modified a predictive UNIQUAC model (basic model developed by Abrams and Prausnitz (1975)) for non-ideal solid and liquid equilibriums. Also a predictive model presented (Coutinho, 2000) for wax formation in jet and diesel fuel. The modified UNIFAC equation for liquid phase and UNIQUAC equation for solid phase (Coutinho, 2000) were used. Vafaie-Sefti et al. (2007) applied multi-solid phase model (basic model developed by Lira-Galeana et al. (1996)) to predict the equilibrium phase in oil mixtures. Liquid and gas phases were described using Peng–Robinson EOS (Robinson and Peng, 1985). Zuo et al. (2001) developed the solid-solution model to predict wax in crude oils and gas condensates. Bhat and Mehrotra (2004) measured and predicted wax–solvent mixtures phase behavior using the Flory free-volume model (Coutinho, 1998) for liquid phase and the predictive UNIQUAC model (Coutinho et al., 1995) for the solid phase. Ji et al. (2004) succeeded to achieve wax disappearance temperature for binary and multi-systems by applying the UNIQUAC thermodynamic model. Firstly they estimated thermodynamic properties of the normal paraffins to calculate terms on fugacity coefficient equation. Esmaeilzadeh et al. (2006) investigated various activity coefficient models and showed that predictive Wilson model (Coutinho and Stenby, 1996) is appropriated to explain the ideal behavior of the solid phase (prediction of wax precipitate in the

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atmospheric pressure). Coutinho and Ruffier-Me´ray (1997) and also Coutinho et al. (2006) studied wax deposition using experimental and thermodynamics methods. They used the predictive UNIQUAC model for modeling the wax formation in hydrocarbon liquids (Coutinho et al., 2006). Also Dalirsefat and Feyzi (2007) used the modified multi-solid phase thermodynamic model (based on Lira-Galeana et al. (1996) model) for predicting WAT and wax precipitate amount. They have utilized Modified Peng–Robinson (MPR) equation (Feyzi et al., 1998) for calculating the fugacity of liquid and gas phases. Ghanaei et al. (2007) have investigated a new predictive thermodynamic model for wax formation under high-pressure conditions. By using various thermodynamic models, they achieved WDT for C_{14}–C_{15} and C_{14}–C_{16} mixtures at various pressures (0.1–100 MPa) and they compared their results with the literature data (Milhet et al., 2005). In other work Nasrifar and Fani Kheshty (2011) used the UNIQUAC model with a pressure dependence term resulting from the Clapeyron equation to determine WDT and the amount of wax. Moreover, recently Ghanaei et al. (2012) used a new modified predictive UNIQUAC model to predict WDT, WAT and amount of wax for different paraffinic mixtures. They used a new generalized heat capacity correlation for liquid, disorder and order solid phases which have been presented to calculate the solid–liquid fugacity ratio. In Table 1 these studies have been listed briefly.

In this study wax disappearance temperature is predicted by using the artificial neural network. A model with two layers (hidden and output layers) was applied on WDT data. The experimental data of Refs. Robles et al. (1996), Metivaud et al. (1999), Daridon et al. (2002), Ji et al. (2004), Pauly et al. (2004), Milhet et al. (2005), Vafaie-Sefti et al. (2007) were used for this model. The proposed model predicts WDT of mixture at several pressures with high accuracy.

2. Thermodynamic wax model

Liquid–solid equilibrium of mixture can be expressed using

$$f_i^S(P,T,x_i^S) = f_i^L(P,T,x_i^L) \tag{1}$$

where

$$f_i^S(P,T,x_i^S) = x_i^S \gamma_i^S f_{i,Pure}^S \tag{2}$$

and

$$f_i^L(P,T,x_i^L) = x_i^L \gamma_i^L f_{i,Pure}^L \tag{3}$$

and equilibrium constant defined as follows:

$$K_i^{SL} = \frac{x_i^S}{x_i^L} = \frac{\gamma_i^L f_{i,Pure}^L}{\gamma_i^S f_{i,Pure}^S} \tag{4}$$

γ_i^L and γ_i^S are the activity coefficients of liquid and solid phases respectively. Several models such as thermodynamic models of Wilson, UNIFAC, UNIQUAC, regular solid solution and cubic equations of state are used to determine them. Fugacity ratio in different phases usually is explained as below:

$$\ln\frac{f_{i,pure}^S(P,T)}{f_{i,pure}^L(P,T)} = \frac{\Delta H_i^f}{RT_i^f}\left(1-\frac{T_i^f}{T}\right) + \frac{\Delta H_i^{tr}}{RT_i^{tr}}\left(1-\frac{T_i^{tr}}{T}\right) + \frac{1}{RT}\int_T^{T_i^f} \Delta C_{pi}^{LS}\,dT - \frac{1}{R}\int_T^{T_i^f} \frac{\Delta C_{pi}^{LS}}{T}\,dT + \frac{1}{RT}\int_P^{P_i^f} \Delta V_i\,dP \tag{5}$$

where ΔH_i^f, T_i^f, ΔH_i^{tr}, T_i^{tr}, ΔC_{pi}^{LS} and ΔV_i are the functions of temperature, pressure, molar mass, carbon number etc., that are defined in several models differently. In this study, presented models in Ghanaei et al. (2007) ('model 1', 'model 2', 'model 3 without k_{ij}', 'model 3 with k_{ij}' and 'new model') and Ji et al. (2004) ('HWWAX', 'ideal solid solution', 'multi-pure-solid' and 'Coutinho's UNIQUAC') works are compared with artificial neural network models. These models are presented below briefly.

2.1. Model 1

Predictive Wilson and ideal solution are used for the solid and liquid phases, respectively. Solid–solid phase transition has been ignored in this model. Poynting correction term has been calculated by category A in Ghanaei et al.'s (2007) work. Liquid molar volume has been calculated from the modified Rackett equation (Rackett,

Table 1
Literature background of wax.

Type of study	Composition of hydrocarbons	Range of pressure	Reference
Modified regular solution model (amount of wax and WAT)			Won (1986, 1989), Hansen et al. (1998), K.J. Pedersen et al. (1991)
Multisolid-phase model (amount of wax)	Oil mixtures	Atmospheric	Lira-Galeana et al. (1996)
Experimental measurements and thermodynamic modeling (amount of wax)	Mixtures of C_{19}–C_{28}	Atmospheric	Coutinho and Ruffier-Me´ray (1997)
Predictive UNIQUAC (amount of wax)	Mixtures of C_{10}–C_{30}	Atmospheric	Coutinho (1998)
Experimental (amount of wax)	Mixtures of C_{10} and C_{18}–C_{30}	Atmospheric	Pauly et al. (1998)
Experimental (WDT)	Ternary mixtures of C_{14}–C_{22}	Atmospheric	Metivaud et al. (1999)
Modified UNIFAC for liquid and UNIQUAC for solid phase (amount of wax)	Jet and diesel fuels	Atmospheric	Coutinho (2000)
Experimental and multisolid phase model (amount of wax)	Oil mixtures (C_1–C_{20}^+)	Atmospheric	Vafaie-Sefti et al. (2007)
Experimental (amount of wax)	Mixtures of C_6–C_{36}	0.1–50 MPa	Pauly et al. (2001)
Multisolid-phase model	Oil mixtures	0.1–60 MPa	Zuo et al. (2001)
Experimental (WDT)	Mixtures of C_{13}–C_{24}	0.1–100 MPa	Daridon et al. (2002)
Experimental (amount of wax)	Mixtures of C_{10}, C_{24}, C_{25} and C_{26}	Atmospheric	Pauly et al. (2004)
UNIQUAC model (WDT and amount of wax)	Binary and ternary mixtures	Atmospheric	Ji et al. (2004)
Experimental (WDT)	Mixtures of C_{14}–C_{15} and C_{14}–C_{16}	0.1–100 MPa	Milhet et al. (2005)
Predictive UNIQUAC	Oil mixtures	0.1–50 MPa	Coutinho et al. (2006)
Several thermodynamic model (amount of wax)	Mixtures of C_{10} and C_{18}–C_{30}, mixtures of C_{10}, C_{24}, C_{25} and C_{26}, mixtures of C_6–C_{36}	Atmospheric	Esmaeilzadeh et al. (2006)
Experimental (WDT)	Mixture of C_{20}–C_{42}	Up to 100 MPa	Rizzo et al. (2007)
Multisolid-phase model (amount of wax and WAT)	Oil mixtures	Atmospheric	Dalirsefat and Feyzi (2007)
Several thermodynamic model (WDT)	Mixtures of C_{14}–C_{15} and C_{14}–C_{16}	0.1–100 MPa	Ghanaei et al. (2007)
Cubic EOS for liquid phase and UNIQUAC with pressure dependence term from Clapeyron equation for solid phase (WDT and amount of wax)	Binary and multicomponent paraffin mixtures	0.1–100 MPa	Nasrifar and Fani Kheshty (2011)
New modified predictive UNIQUAC (WDT, WAT and amount of wax)	Paraffinic mixtures		Ghanaei et al. (2012)