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Phase equilibrium modelling of natural gas hydrate formation conditions using LSSVM approach

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ABSTRACT
The formation of gas hydrates in industries and chemical plants, especially in natural gas production and transmission, is an important factor that can lead to operational and economic risks. Hence, if the hydrate conditions are well addressed, it is possible overcome hydrate-related problems. To that end, evolving an accurate and simple-to-apply approach for estimating gas hydrate formation is vitally important. In this contribution, the least square support vector machine (LSSVM) approach has been developed based on Katz chart data points to estimate natural gas hydrate formation temperature as function of the pressure and gas gravity. In addition, a genetic algorithm has been employed to optimize hyper parameters of the LSSVM. Moreover, the present model has been compared with five popular correlations and was concluded that the LSSVM approach has fewer deviations than these correlations so to estimate hydrate formation temperature. According to statistical analyses, the obtained values of MSE and R^2 were 0.278634 and 0.9973, respectively. This predictive tool is simple to apply and has great potential for estimating natural gas hydrate formation temperature and can be of immense value for engineers who deal with the natural gas utilities.

KEYWORDS
Correlation; GA, hydrate; LSSVM; natural gas; temperature

1. Introduction

By development of natural gas extraction, methods of production should be more economical because the natural gas contains many inclusion and small compounds. The natural gas hydrate phenomena means that some small gas molecules (e.g., light hydrocarbons, water soluble polar compound, acid gases) being stuck among a network of water molecules and trapped in hydrogen bonds of water (Wilcox et al., 1941; Carson and Katz, 1942; McCain, 1990; Carroll et al., 2009). These scientific interests of hydrate formers started in the 19th century (Sloan and Koh, 2007). The first reliable research belonged to Hammerschmidt in 1934 who found gas hydrates as a reason for gas transition line obstructions (considered to have been ice in the past). As a result, scientific and industrial testing process of gas hydrates began (Englezos, 1993). In this case, many researchers tested the impact of restrainers, such as chloride salts, methanol, and monoethylene glycol, on creation of hydrates (Englezos and Bishnoi, 1988; Fan et al.,

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2006; Sloan and Koh, 2007; Cha et al., 2013). Hydrates have three various crystal structures that depend on the hydrate incorporators. By using X-ray diffraction (Englezos, 1993), two usual structures such as sI and sII were disclosed in the 1950s and sH structure was determined in 1987 (Khokhar et al., 1998; Sloan, 2003). Hydrates affect the lines harmfully because they block the transmission line, so oil and gas companies want to find a way to operate their obligation outside the range of the hydrate formation (Sloan and Koh, 2007; Mokhatab and Poe, 2012). Although many theoretical methods and thermodynamics have been discovered, a good development systematic prediction method is rare to use it for natural gases. Furthermore, sour (hydrogen sulfide [H_2S]) and acid gases (carbon dioxide [CO_2]) are more common all around the world scattering in the natural gases. Demand and consume are becoming more and more common around the world, so finding new resources for responding to it is necessary (ZareNezhad and Ziaee, 2013). The main component of sour gas, H_2S, at relatively low pressure and relatively high temperature causes to form hydrate (Sun and Chen, 2005). As a result, it is likely to form in the pipelines of sour gas. Hence, determining the hydrate formation condition is necessary. H_2S is toxic and corrosive; moreover, the real experimental data of its equilibrium are little. Some of the early correlations were presented in the 1930s and 1940s (Hammerschmidt, 1934; Katz, 1945). Katz offered two different methods: gravity approach and K value approach. The gravity method just supported only a special rang of gas gravity, pressure and temperature. The K value strategy was more accurate for pure components and less for mixtures, so it wasn't accepted in the industry. Although several methods have been proposed in past years, the main distinguishing problem between the different types of hydrates remains. The two different types of hydrates (type 1 and type 2) have significant effects on its formation pressure (Koh, 2002).

Nowadays, artificial intelligence schemes have great potential to model in different fields and applications. The commonly applied embranchments of artificial intelligence are the fuzzy logic, artificial neural network (ANN), support vector machine (SVM), and adaptive network-based fuzzy inference system (ANFIS; Shafiei et al., 2014; Ahmadi and Baghban, in press; Baghban et al., 2015a; Baghban et al., 2015b; Amedi et al., 2016; Baghban et al., 2016; Bahadori et al., 2016a). Applications of intelligence modeling of hydrate formation conditions have brought considerable attention (Elgibaly and Elkamel, 1998; Zahedi et al., 2009; Mohammadi et al., 2010; Ghavipour et al., 2013). These techniques are easy to use and can reduce the cost, time, and monotonous parameter adjustment and can be applied in the presence of uncertain experimental data.

In this present study, we used a simple predictive strategy called least square support vector machine (LSSVM) to predict the hydrate formation temperature natural gas. The LSSVM has shown high capability in solving nonlinear and complex problems and it has been applied for estimating of hydrate formation temperature of natural gas. In this study, a collection of 710 data points for the natural gas hydrate temperature as a function of the pressure (5026–39007 KPa) and gas gravity (0.6–1) has been gathered from the Katz chart (Katz, 1959).

2. Theory

The basic concepts of the support vector machines have been presented in our previous papers (Ahmadi and Baghban, in press; Baghban et al., 2015b; Baghban et al., 2016; Bahadori et al., 2016a; Bahadori et al., 2016b). In addition, there are some well-known correlations for predicting natural gas hydrate temperature such as the Bahadori and Vuthaluru (2009) correlation, Berg (1986) correlation, Motiee (1991) correlation, Towler and Mokhatab (2005) correlation, and Hammerschmidt (1934) correlation.

3. Data preparation

Accurate real data points are required to implement the LSSVM strategy. In this regard, 710 data points were applied from the Katz chart (Katz 1959). This set of data points consists of temperatures, which range from 290.69 to 298.90 K and pressures, which are between 5037 and 28464 KPa and gas gravities, which range between 0.6 and 1. We divided the dataset into two subsets of training and testing. Testing data set comprises 135 data points that have SG equal to 0.8. The remaining 575 data points have different SG compared with the testing data points and were considered as training set. It is noteworthy to mention that the test data set has been used to evaluate the prediction capability of the model for unused data.

4. Results and discussion

As mentioned in the LSSVM methodology, there are two parameters, namely the regularization parameters (γ) and the kernel parameter (σ^2), that should be specified before training phase. In the present study, owning to remarkable characteristics of the radial basis function (RBF), we used this function as the kernel function of the LSSVM. The parameters of RBF have been influenced greatly by the number of support vectors. As the number of support vectors increases, the training elapsed time increases. Furthermore, potential of the GA was evaluated as great evolutionary optimization algorithm to obtain the hyper variables of the LSSVM. A schematic illustration of this model is indicated in Figure 1. In addition,

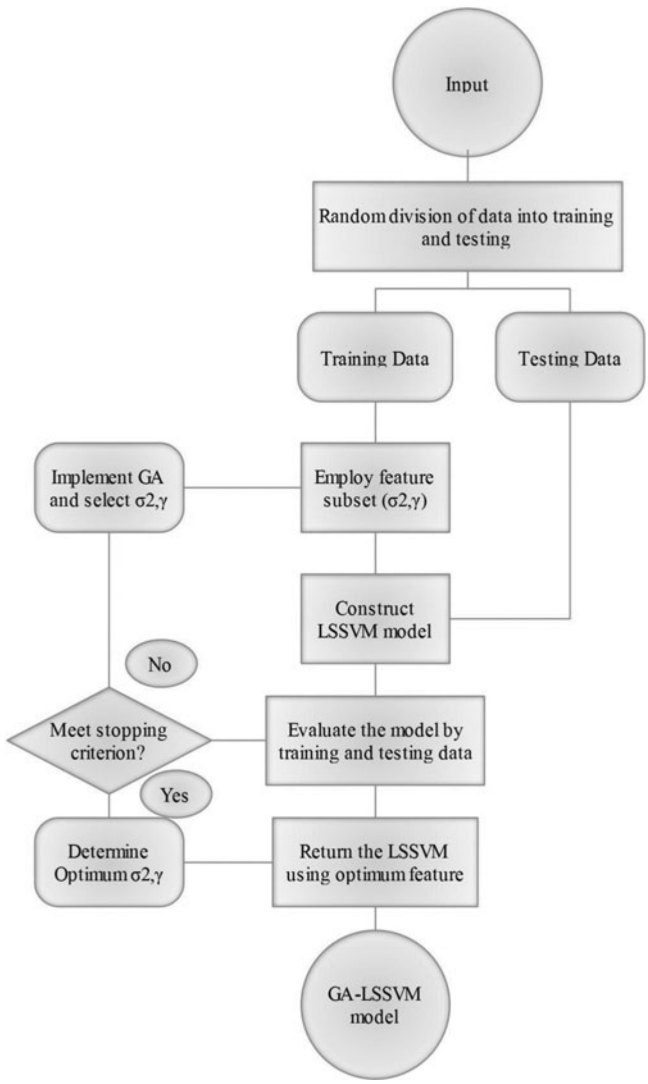

Figure 1. Schematic diagram of combination of the LSSVM with GA.

Table 1. Details of trained LSSVM model.

Type	Value/comment
No. of training data	135
No. of testing data	575
Kernel Function	RBF
γ	11739.4621
σ^2	0.256459582
Optimization method	GA
Pop. size	30
Maximum iterations	200

details of the trained LSSVM model have been summarized in Table 1. Capability of the present LSSVM was investigated by some popular statistical techniques such as the MSE, MRE, MAE, STD, RMSE, and R^2 between the actual and estimated values. These techniques have been presented with details at the following (Landau and Everitt, 2004). Figure 2 indicates the experimental versus predicted hydrate formation temperature of natural gas through the LSSVM strategy for both training and testing samples. Good agreement of predicted and actual values indicates remarkable capability of the LSSVM model to estimating hydrate formation conditions. Furthermore, the regression analysis of aforementioned models has been indicated in Figure 3. Aggregation of the data points around the 45° line for the LSSVM model indicates less deviation of model. The values of R^2 were 0.9999 and 0.9973 for training and testing samples, respectively. In addition, the linear formulations of training and testing samples through the regression analysis obtained as follows:

$$y = x - 0.0037,\ \ R^2 = 0.9999 \tag{1}$$

$$y = 1.2985x - 88.351,\ \ R^2 = 0.9973 \tag{2}$$

The values of MREs obtained 0.003890% and 0.136427% for training and testing samples respectively. In addition, a histogram of the residual data points is indicated in Figure 4 for the training and testing phases. However, because the residual data points relatively followed the normal curve, the model has relatively normal distribution. Furthermore, a comparison was carried out and indicated in Figure 5 between the proposed LSSVM model and five well-known correlations in order to determining hydrate formation temperature such as the Bahadori and Vuthaluru (2009) correlation, Berg (1986) correlation, Motiee (1991) correlation, Towler and Mokhatab (2005) correlation, and Hammerschmidt (1934)

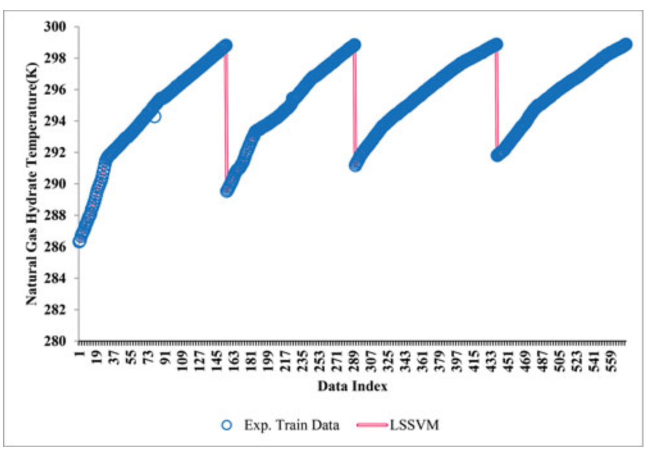

Figure 2. Experimental and predicted hydrate formation temperature of natural gas by the LSSVM versus data number for both training and testing data sets.

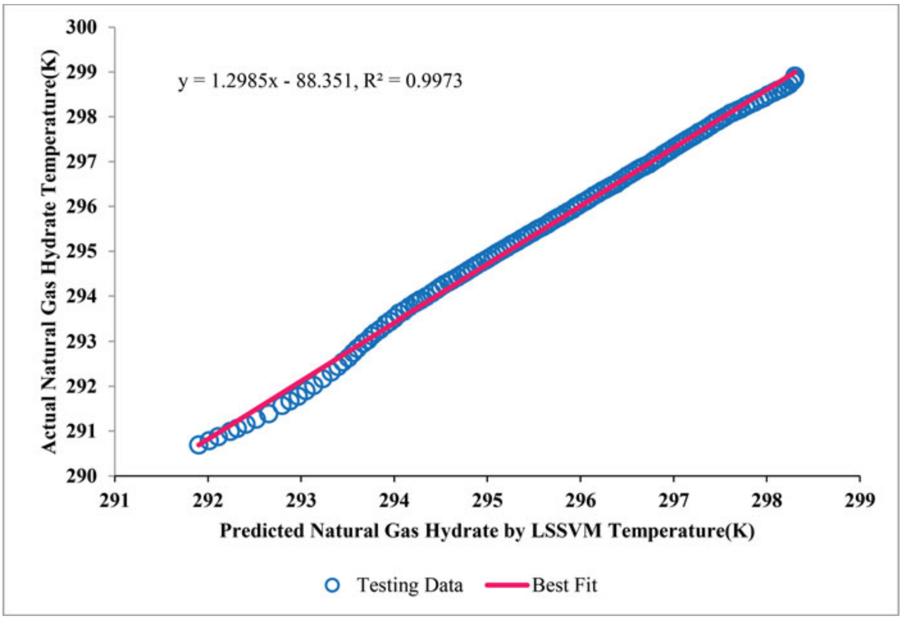

Figure 3. Regression plots between the experimental and predicted hydrate formation temperature by the LSSVM at training and testing stages.

correlation. This comparison showed greater capability of the LSSVM than did the correlations to predict hydrate formation temperature.

The obtained MSEs, MRE, standard deviation (STD), root mean square errors (RMSE), and R^2 are listed in Table 2 for both the testing and training data sets. These statistical analyses also confirmed reasonability of the LSSVM for estimating the hydrate formation temperature.

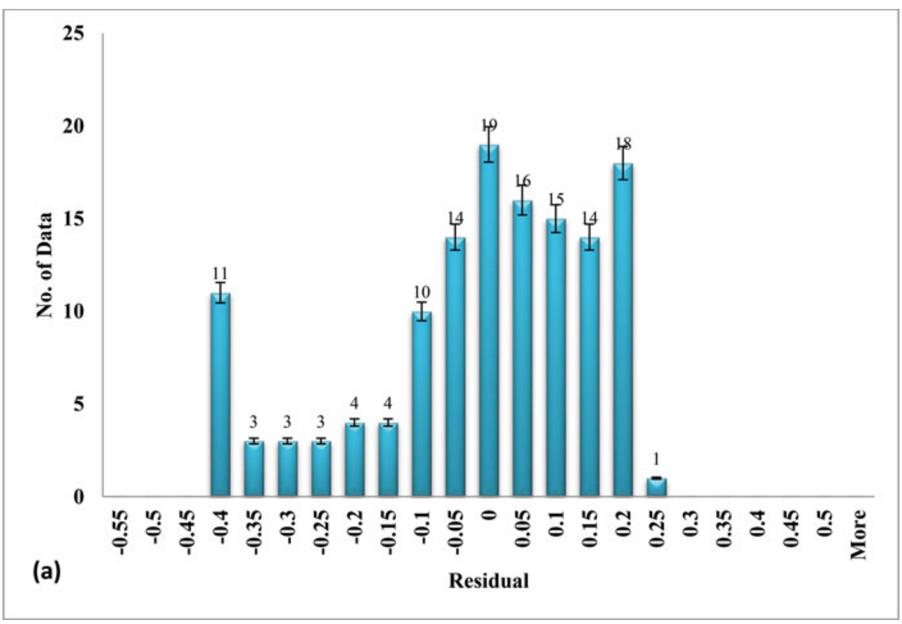

Figure 4. Histograms of residual for testing data.

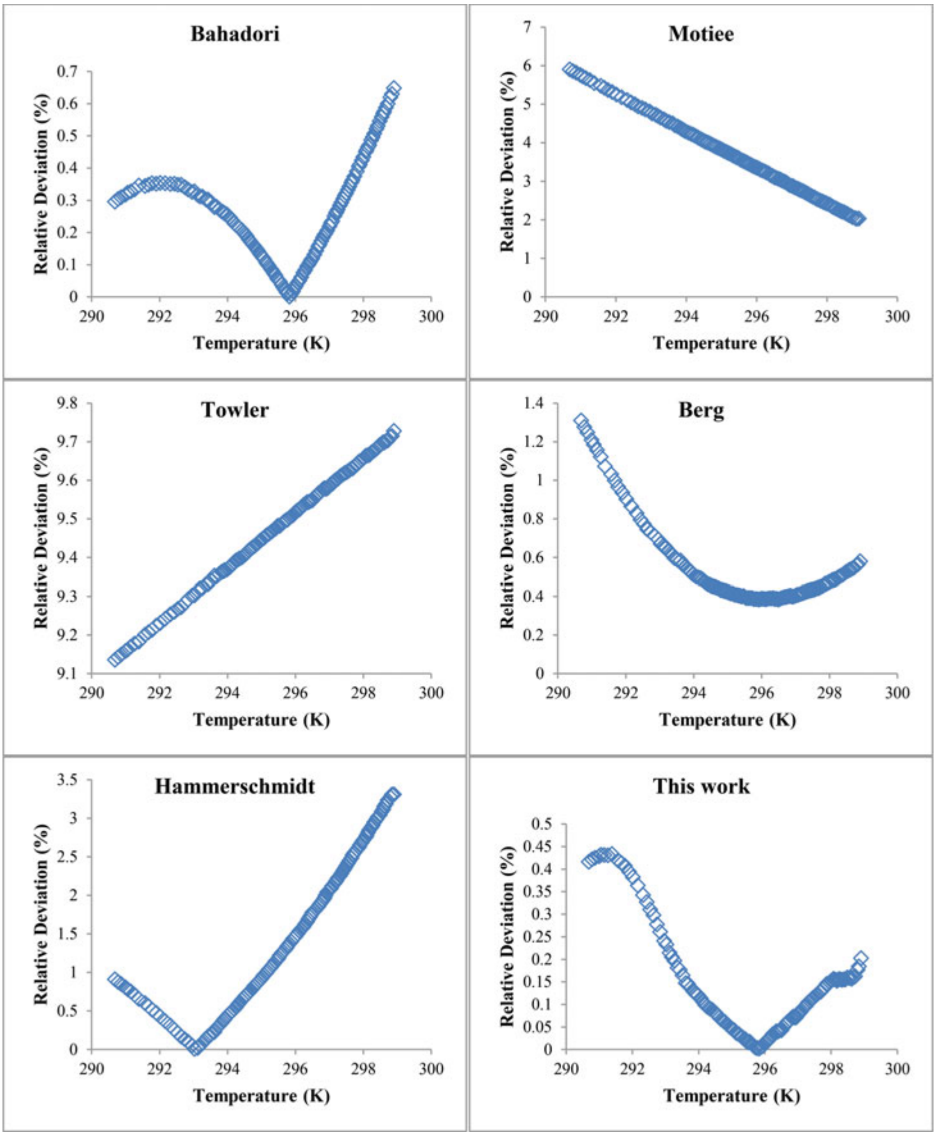

Figure 5. Relative deviations of proposed LSSVM model and other correlations for testing data points.

Table 2. Statistical analyses obtained from the LSSVM model.

Analysis	Training	Testing
MSE	0.000966	0.278634
R^2	0.9999	0.9973
MRE%	0.003890	0.136427
RMSE	0.031088	0.527858
STD	0.028926	0.343187

5. Conclusions

This study was aimed to evolve a simple-to-apply predictive tool for estimating natural gas hydrate formation temperature as function of the pressure and gas gravity by the LSSVM approach. In addition, potential of the GA was evaluated to determine the hyper variables of LSSVM (σ^2 and γ). This evolutionary algorithm has great performance to optimize the LSSVM structure. Moreover, the LSSVM model was compared to five well-known correlations such as the Bahadori & Vuthaluru correlation, Berg correlation, Motiee correlation, Towler and Mokhatab correlation, and Hammerschmidt (1934) correlation. These comparisons showed great capability of LSSVM than the correlations to predict hydrate formation temperature.

Other statistical analyses showed great performance of the LSSVM approach and obtained approximations were to be in good agreement with the experimental reported data points. The efforts in this contribution absolutely covered the manner in order to accurate estimation of natural gas hydrate formation temperature, which can help chemist and engineers to have a simple predictive tool with low dependent parameters for monitoring the operational conditions and phase behavior of the systems.

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