Journal of Petroleum Science and Engineering 136 (2015) 78–87

Contents lists available at ScienceDirect

Journal of Petroleum Science and Engineering

journal homepage: www.elsevier.com/locate/petrol

Comparing the capability of artificial neural network (ANN) and CSMHYD program for predicting of hydrate formation pressure in binary mixtures

Saheb Maghsoodloo Babakhani, Mahmoud Bahmani *, Jafar Shariati, Kiumars Badr, Yahya Balouchi

Young Researchers and Elite Club, Darab Branch, Islamic Azad University, Darab, Iran

ARTICLE INFO

Article history:
Received 21 June 2015
Received in revised form
22 October 2015
Accepted 1 November 2015
Available online 5 November 2015

Keywords:
Hydrate formation pressure
Binary mixtures
Artificial neural network
CSMHYD program

ABSTRACT

In the present study, we investigated forecasting of hydrate formation pressure of binary mixtures including at least one hydrocarbon using a feed-forward multi-layer artificial neural network. For this purpose, 895 experimental data which cover a wide range of temperatures and compositions were collected from different studies cited in the literatures. In order to find the best model, different ANN types are tested through the absolute average relative deviation percent (AARD), mean square error (MSE) and the regression coefficient (R^2) and the optimal configuration is selected. It is found that the selected ANN model is based on the statistical analysis has an excellent agreement (AARD = 1.02, MSE = 1.27×10^{-5} and $R^2 = 0.9938$) with the collected experimental data. The obtained results reveal that the developed MLPNN model is an applicable and feasible tool to predict hydrate formation pressure with high accuracy with respect to Colorado School of Mines Hydrate (CSMHYD) Program.

1. Introduction

Gas hydrates known as clathrate are referred to ice like crystals in which intra-matrix gas molecules derived from water molecules are bounded at specific temperature and pressure. Based on the gas particles sizes, hydrates are classified into three different types: I, II, H. Type I includes small gaseous compounds such as methane and ethane. Type II is composed of larger molecules like propane and type H includes both small and large molecules (Sloan, 1998). Large amounts of gaseous hydrocarbons can be located in the voids of hydrate matrices. For example, 180 m^3 of gas is located in the crystal matrices of water per 1 m^3 of formed hydrates at standard conditions. Since natural gas hydrate formation annually has a great cost for petroleum industry, prediction of hydrate formation conditions has great influence on the elimination of this phenomenon in the pipelines. There are numerous studies on the estimation of hydrate formation conditions.

Wilcox et al. (1941) initiated the *K*-value method based on coefficients distribution (K_i values) for components on a water-free basis. They also, generated the gas-gravity plot that relates the hydrate pressure and temperature to a specific gravity of natural gas (wlicox et al., 1941; Kobayashi et al., 1987). Hoseininasab et al. (2011) developed a new correlation for predicting the hydrate-formation temperatures for pure and mixture of hydrocarbon systems based on the gravity method using Statistical Analysis Software (SAS).

ANNs have received considerable interest in the last two decades because of their great capability in the modeling of linear and non-linear systems without needing the prior empirical model (Zupan and Gasteiger, 1991; Anderson and Kaufmann, 2000). In comparison with the conventional fitting methods, ANNs are more powerful tools for experimental correlations and fitting parameters (Jouyban et al., 2004).

A lot of studies have been done on hydrate formation in gaseous systems (Balakin, 2010; Shahnazar and Hasan, 2013; Dalmazzone et al., 2006); however, little information is available on how hydrate formation or dissociation happens in pipelines and how it can be possible to predict a model to simulate the hydrate formation/dissociation due to difficulties in measuring the thermodynamic and kinetics of hydrate stability zone at high pressure and low temperature. The benefit of our work is that it works based on real data of hydrate formation conditions. So it has not any limitations like other programs that use analytical equations.

In this study, Levenberg Marquardt algorithm is applied to train the network in order to predict hydrate formation pressure. The proposed ANN model results are compared with the results of Colorado School of Mines Hydrate (CSMHYD) program (Sloan,

* Corresponding author. Fax: +98 711 6287294.
E-mail addresses: saheb.maghsoodloo@gmail.com (S.M. Babakhani), m.bahmani67@gmail.com (M. Bahmani), j.shariati84@yahoo.com (J. Shariati), kiumarsbadr@gmail.com (K. Badr), yahyabalouchi@gmail.com (Y. Balouchi).

http://dx.doi.org/10.1016/j.petrol.2015.11.002
0920-4105/

Nomenclature

a	Plank's constant
h	enthalpy
H	hydrate
k	Boltzmann's constant
m	mass of a molecule
P	pressure
q	partition function
r	the distance of the wandering molecule from its lattice site
R	universal gas constant
T	temperature
W	water
y	mole fraction

Greek letters

θ	polar angle
ϕ	azimuthal angle
$\omega(r)$	Total intermolecular force when the wanderer is at distance r from its lattice point
Δ	difference
μ	chemical potential
υ	specific volume (V/N)

1998). A proper experimental hydrate formation pressure of binary mixtures database is used to validate the model results.

2. Gas hydrates

2.1. Hydrate description

There are several kinds of apparatus to investigate the thermodynamic equilibrium conditions of gas hydrates (pressure and temperature) and determine the composition of all existing phases (gas, liquid and hydrate). For example Herri et al. (2011) used an experimental set-up (Fig. 1) consisting of a stainless steel high pressure batch reactor with a double jacket connected to an external cooler. A Pyrex cell is located in the stainless steel autoclave in which the pressure can be raised up to 10 MPa. Any variations into the hydrate phase can be observed by the two sapphire windows located on both sides of the reactor. A High-Performance Liquid Chromatography (HPLC) pump (which is well-known as high pressure pump for their compactness and reliability) injects liquid to pressurize the reactor. A four vertical-blade turbine impeller ensures stirring of the suspension during crystallization. The temperature is monitored by two probes one in the liquid bulk and other in the gas phase. A pressure transducer (range: 0–10 MPa) is used to measure the pressure. A gas chromatograph determines the composition of the gas phase and collects a controlled volume of gas which is directly injected into the loop of the gas chromatograph (Herri et al., 2011).

2.2. Explanation of the most common experimental procedure

At first stage, the hydrate is formed by crystallization of gas mixtures in the presence of a liquid phase (Fig. 1.) Firstly, the reactor cell is closed and evacuated using a vacuum pump. Subsequently, the cell is flushed three different times with methane (depending on the experiment) to delete any trace of the other gases. After this cleaning procedure, the reactor has been evacuated again.

At the start of the real experimental run, the reactor is pressurized with gas. The gas mixture is stirred and cooled down, then kept at the operation temperature. The stirrer is then stopped and the liquid solution is poured into the reactor using the pump. Upon injection liquid at ambient temperature which leads to the gas volume reduction, the enhancement of both temperature and pressure are achieved simultaneously. Then the stirrer starts agitating. A pressure reduction is observed due to the gas partial dissolution in the liquid phase. After a while (ranging from some minutes to several hours, since nucleation is a stochastic phenomenon), crystallization (exothermic process) begins along with a sudden increase of temperature which depends on the intensity of the crystallization. During the solid formation, as gas is consumed to form hydrate, the pressure decreases. In order to measure the gas species values after determination of the crystallization time, the gas phase is sampled with a special instrument and then analyzed by in-line gas chromatography. Moreover, analyzing the liquid phase is done by an off-line ion-exchange chromatography. After reaching equilibrium (end of crystallization), the pressure and temperature approach to constant values.

The gas hydrate dissociation is carried out at constant volume and started by heating the reactor in the increments of 1 °C. After

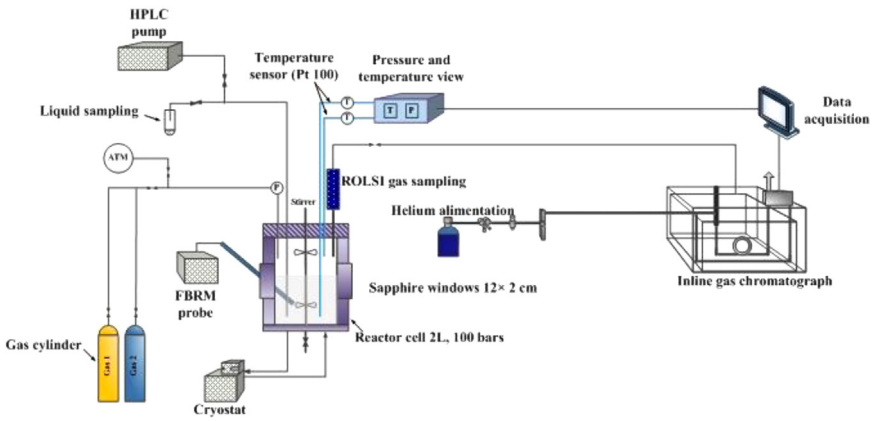

Fig. 1. Experimental set-up for hydrate formation pressure measurement (Herri et al., 2011).