Chemometrics and Intelligent Laboratory Systems 104 (2010) 195–204

Contents lists available at ScienceDirect

Chemometrics and Intelligent Laboratory Systems

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

A neural network-based method for estimation of binary gas diffusivity

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ARTICLE INFO

Article history:
Received 20 June 2010
Received in revised form 16 August 2010
Accepted 16 August 2010
Available online 20 August 2010

Keywords:
Neural network
Binary diffusion coefficient
Gas diffusivity

ABSTRACT

In this study, a feedforward three-layer neural network is developed to predict binary diffusion coefficient (D_{AB}) of gases at atmospheric pressure over a wide range of temperatures based on the critical temperature (T_c), critical volume (V_c) and molecular weight (M) of each component in the binary mixture. The accuracy of the method is evaluated through a test data set not used in the training stage of the network. Furthermore, the performance of the neural network model is compared with that of well known correlations suggested in the literature. The results of this comparison show that our developed method outperforms other correlations, with respect to accuracy as well as extrapolation capabilities.

1. Introduction

Estimation of diffusion coefficients in the liquid and gas phases is crucial in the design and simulation of processes involving mass transfer such as liquid–liquid extraction, leaching, gas absorption, reactors, packed and tray towers. For instance, binary diffusion coefficients of gases are needed in the design of gas phase reactors. Some other fields where gas diffusivity plays an important role are global changes, atmospheric chemistry, combustion science, studies of indoor air pollution and atmosphere–biosphere interactions. Diffusion is also a major factor in peak broadening in chromatography. Therefore, development of accurate and reliable correlations for determination of diffusion coefficients has drawn the attention of many researchers. Availability of well known theories for the gas phase such as kinetic theory of gases has motivated scientists to propose several theoretical and semi-empirical equations for estimation of binary diffusivity coefficients of gases. Some of these correlations have been presented in the following:

(i) The Stefan–Maxwell (SM) equation [1]:

$$D_{AB} = \frac{a}{n\sigma_{AB}^2}\left[\frac{8RT}{\pi}\left(\frac{1}{M_A}+\frac{1}{M_B}\right)\right]^{1/2} \quad (1)$$

where a is a constant taking various values ($1/3\pi$, 1/8, $1/2\pi$ and 3/32) based on different researcher studies, n is the number of gas phase molecules per cm^3, σ_{AB} is the collision diameter between gas molecules A and B, R is the gas constant, T is the absolute temperature and M_A and M_B are the molecular masses of solute A and carrier gas B, respectively.

(ii) The Chapman–Enskog equation [1]:

$$D_{AB} = \frac{0.00263T^{3/2}}{P\sigma_{AB}^2}\left(\frac{1/M_A+1/M_B}{2}\right)^{1/2} \quad (2)$$

where P is the gas pressure in atm.

(iii) The Gilliland equation [2]:

$$D_{AB} = \frac{0.0043T^{3/2}(1/M_A+1/M_B)^{1/2}}{P\left(V_A^{1/3}+V_B^{1/3}\right)^2} \quad (3)$$

where V_A and V_B are molar volumes in cm^3 at the boiling points, which can be obtained experimentally, or they can be estimated through group contribution methods.

(iv) The Arnold equation [3]:

$$D_{AB} = \frac{0.0083T^{3/2}(1/M_A+1/M_B)^{1/2}}{P\left(V_A^{1/3}+V_B^{1/3}\right)^2(1+S_{AB}/T)} \quad (4)$$

where S_{AB} is Sutherland's constant, which can be estimated by various ways. The above equation, which introduces a second temperature dependent term in the denominator to account for molecular "softness", shows a dependence varying from $T^{3/2}$ to $T^{5/2}$.

(v) The Hirschfelder–Bird–Spotz (HBS) equation [4]:

$$D_{AB} = \frac{0.00186T^{3/2}(1/M_A+1/M_B)^{1/2}}{P\sigma_{AB}^2\Omega_{AB}} \quad (5)$$

The term Ω_{AB} is the collision integral depending in a complicated way on temperature and the interaction energy of the colliding molecules, ϵ_{AB}. Hirschfelder et al. [5] followed

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doi:10.1016/j.chemolab.2010.08.009

the Chapman–Enskog kinetic approach combined with the Lennard–Jones intermolecular potential function. Ω_{AB} values as a function of the reduced temperature $T^* = kT/\epsilon_{AB}$, where k is the Boltzmann constant, can be obtained from relevant tables [5,6]. The main disadvantage of the HBS equation is the difficulty encountered in evaluating σ_{AB} and Ω_{AB}. There are correlations for estimation of these parameters from viscosity measurements.

(vi) Chen and Othmer [7] provided the most explicit approximation of the HBS equation using the critical values of temperature T_C, and volume, V_C:

$$D_{AB} = \frac{0.43(T/100)^{1.81}(1/M_A + 1/M_B)^{1/2}}{P(T_{CA}T_{CB}/10^4)^{0.1405}\left[(V_{CA}/100)^{0.4} + (V_{CB}/100)^{0.4}\right]^2} \quad (6)$$

Both T_C and V_C values can be estimated in various ways [1,5].

(vii) Fuller–Schettler and Giddings (FSG) [8] developed a successful equation in which atomic and structural volume increments and other parameters were obtained by a least-squares fit to over 300 measured data. In the Fuller et al. [8] method:

$$D_{AB} = \frac{0.00143T^{1.75}(1/M_A + 1/M_B)^{1/2}}{\sqrt{2}P\left[(\sum \upsilon)_A^{1/3} + (\sum \upsilon)_B^{1/3}\right]^2} \quad (7)$$

Σv is determined by summing the atomic contributions.

(viii) Huang et al. [9] investigated the effects of pressure and temperature on the gas diffusivity. Based on their experimental data they modified the Arnold[3] equation as follows:

$$D_{AB} = \frac{5.06T^{1.75}(1/M_A + 1/M_B)^{1/2}}{P^{1.286}\left(V_A^{1/3} + V_B^{1/3}\right)^2} \quad (8)$$

They compared their correlation (Eq. (8)) with most of the other techniques. In this comparison, they used 134 experimental diffusion coefficient values obtained from literature. The results of their work are summarized in Table 1. To achieve higher precision in estimating gaseous diffusion coefficients, more complicated methods derived from HBS equation should be used. These methods, which are based on detailed gas dynamics, are commonly recommended for unusual systems (e.g. large molecules, high temperatures, etc.) which are beyond the valid range of Gilliland and Arnold equations. The Fuller–Schettler–Giddings (FSG) [8] equation provides the best practical combination of simplicity and accuracy.

For binary gas mixtures at low pressure, D_{AB} is inversely proportional to the pressure, increases with increasing temperature, and is almost independent of composition for a given gas-pair. For most binary mixtures, diffusivity data obtained from experiments and correlations are quite limited in range and accuracy. Fuller and Giddings [10] compared the existing theoretical or empirical equations for predicting gaseous diffusion coefficients. Using experimental data for 38 binary gas systems, they examined the prediction accuracy of various proposed correlations, and tabulated the percentage errors for each method. Based on their work, the average absolute percentage errors varied from 4.2 to 20%, depending on the estimation method, and the proposed correlation by Fuller et al. [8] was the most accurate. They also showed the difficulties encountered for theoretical estimation of diffusion coefficients. To overcome these difficulties artificial neural networks can be used as a powerful multivariable function.

Table 1
Overall average relative errors of various methods used for the prediction of binary gaseous diffusion coefficients for 134 experimental test data [9].

Method	Accuracy (%)
Gilliland [2]	6.64
Arnold [3]	11.75
Hirschfelder–Bird–Spotz [4]	18.99
Chen–Othmer [7]	10.85
Fuller–Schettler–Giddings [8]	3.40
Huang–Young–Huang–Kuo [9]	3.51

Neural networks have been used extensively to predict the physical properties in various fields of chemical engineering over the last two decades. Eslamloueyan and Khademi [11] proposed a feedforward three-layer neural network to predict the thermal conductivity of pure gases at atmospheric pressure over a wide range of temperatures based on their critical temperatures, critical pressures and molecular weights. Also, Eslamloueyan and Khademi [12] developed a neural network consisting of two consecutive multilayer perceptrons (MLP) to estimate the conductivities of binary gaseous mixtures at atmospheric pressure. Kauffman and Jurs [13] predicted the surface tension, viscosity, and thermal conductivity for common organic solvents by using a quantitative structure. They used multiple linear regression methods and computational neural networks to train and evaluate their models based on statistical indices and overall root-mean square error. Boozarjomehri et al. [14] characterized the basic properties of pure substances and petroleum fractions by a neural network.

Although different theoretical and semi-empirical correlations have been suggested for estimation of diffusivity coefficients by researchers, there are still considerable differences among their predictions. In the correlations presented by other researchers, some molecular parameters are used that have not been determined experimentally for all substances, and they are usually estimated by auxiliary equations whose accuracy depends on the operating conditions and the type of substances. On the other hand, calculation of these parameters is usually cumbersome and in some cases different equations have been suggested which do not necessarily lead to the same results. Furthermore, the existence of various methods for estimating the gas diffusivity can add more difficulties in the selection of the best prediction scheme. The objective of our work is to develop a neural network model for prediction of binary diffusion coefficient of gases at atmospheric pressure over a wide range of temperatures and substances. The network inputs are the gas temperature, critical temperature, critical volume and molecular weight of each compound. In the next section, the proposed method as well as the data used in its development and validation is presented. Section 3, contains the results and discussion about the proposed method.

2. Methodology

The inherent ability of artificial neural networks to learn and recognize nonlinear and complex relationships can be used to predict binary diffusion coefficient of gases. The development steps of the proposed neuromorphic model for prediction of gas diffusivity have been explained in the following sections. A brief overview of the artificial neural network used in this study has been presented by Eslamloueyan and Khademi [11,12].

2.1. Data acquisition and analysis

One of the most important stages in the development of the suggested neuromorphic model is the availability of reliable experimental data for the model training. These data include experimental published data on critical temperature, critical volume and molecular weight of the pure components as well as binary diffusion coefficients at different temperatures. In this study, 467 experimental data were collected from different literatures. These data were divided into two parts; training data set and test data set. More than two-thirds of the total data were chosen for training to set up the ANN and the other