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Density prediction of liquid alkali metals and their mixtures using an artificial neural network method over the whole liquid range

Shiva Sabzevari, Majid Moosavi*

Department of Chemistry, Faculty of Sciences, University of Isfahan, Isfahan 81746-73441, Iran

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ABSTRACT

In this study, the application of artificial neural network (ANN) method in predicting the density of alkali metals and their mixtures is investigated. A total number of 595 different data points of these compounds were used to train, validate and test the model. A typical three-layer feedforward backpropagation neural network has been trained by the Levenberg Marquardt algorithm. The tansig-tansig transfer functions with 15 neurons in the hidden layer makes the least error, so a network with (8-15-1) structure was used to design the ANN model. The average relative deviations for train, validation, and test sets are 0.1029, 0.1396, and 0.1002, respectively. A comparison between our results and those obtained from some previous works shows that this work, as an excellent alternative, can provide a simple procedure to predict the density of these compounds in a better accord with experimental data up to high temperature, high pressure (HTHP) conditions.

1. Introduction

The study of alkali metals is an interesting field for many researchers, due mainly to their specific advantages for high-temperature applications. Alkali metals' desirable properties such as low vapor pressure, high boiling point, high thermal conductivity, and good thermal and reaction stability, permit low-pressure operation of coolant systems at high temperatures and radiation fluxes. Therefore, they are widely used in modern science and technology [1–5]. The construction of high energy electrochemical cells as well as thermionic and magneto-hydrodynamic converters is only possible using alkali metals. They could be more effectively used in extraction metallurgy, especially in that of some precious metals from their ores and wastes. Liquid alkali metals also act as coolant in nuclear power plants [6]. The temperatures envisaged in these applications are frequently above the range of temperatures for which the experimental data on the physical properties of the liquid metals are available.

The experimental difficulty in obtaining data for alkali metals at elevated temperatures arises from the high chemical reactivity of these elements. They can readily react with components of the surrounding atmosphere such as oxygen, nitrogen, carbon dioxide, and water vapor. It is difficult to obtain materials of high purity, and it is easy to contaminate samples during measurements. So it is important to predict alkali metals' properties using an accurate and trustworthy method.

The information on the physical and thermodynamic properties of alkali metals is important in technological application of these groups of elements. During the last decades, many studies have been devoted to prediction of thermodynamic properties of liquid alkali metals [7–24], and their alloys [25–29]. Different authors used different equations and methods to predict and reproduce the thermodynamic properties of these systems. Some of these attempts are restricted to the limited ranges of temperature and pressure and their results to predict the thermodynamic properties of these systems show different degrees of accuracy. It still seems essential to find a new method by which the thermodynamic properties of alkali metals and their alloys can be predicted more accurately.

An artificial neural network (ANN) can be a suitable alternative to model the different thermodynamic properties. The relationship between the physical and thermodynamic properties is highly nonlinear, and an artificial neural network (ANN) is an especially efficient algorithm to approximate a certain function (such as density) by learning the relationships between the input and output vectors [30]. Consequently, ANN method can be an alternative tool to model the different thermodynamic properties [31,32]. In the past decades, ANNs have been intensively used in various fields. The major reason for this rapid growth and diverse applications of neural networks is their ability to virtually approximate any function in a stable and efficient way. This method also is widely used to estimate the different thermodynamic properties such as density, melting point, vapor pressure, etc. for different classes of materials

* Corresponding author. Tel.: +98 311 7932722; fax: +98 311 668 9732.
E-mail addresses: m.mousavi@sci.ui.ac.ir, majid_822002@yahoo.com (M. Moosavi).

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[33–48]. To the best of the authors' knowledge, there isn't any publication on the application of the ANN for the prediction of density of liquid alkali metals, neither for pure alkali metals nor for their mixtures. In this study, the application of the ANN method in predicting the density of alkali metals and their mixtures is investigated.

2. The neural network used

Artificial neural network is a simple method for modeling, which does not need explicit formulation between input and output data. It carries out the modeling based on simple mathematical functions even in the case of complex systems. The first idea of the neural networks came from the structure of the human brain. The purpose of designing the neural network was simulating parallel structure of biological neural system. Accordingly, artificial neural networks generally consist of a number of interconnected processing elements called "Neurons", which are connected in a massively parallel structure. How the inter-neuron connections are arranged determine the structure of a network. How the strengths of the connections are adjusted or trained to achieve a desired overall behavior of the network is governed by its learning algorithm.

All neural networks consist of different layers: three main layers which are called input, hidden, and output layers. The hidden layer itself may consist of multiple layers. The input layer consists of one neuron for each variable. The number of neurons in the output layer is also equal to the number of output variables. Since, there is no specific approach to determine the number of neurons of the hidden layer, the optimum number of neurons was determined by adding neurons in a systematic way during the learning process. The neurons of each layer are connected to the next one by weighted connections. Weighted connections make a relation between the inputs and outputs of the network. It has been showed that any complicated nonlinear function can be modeled by a multilayer feed forward neural network with one hidden layer to an arbitrary degree of accuracy [47]. In this work, a typical three-layer feedforward backpropagation neural network was used. The network was programmed with the software MATLAB. This network is very effective for representing nonlinear relationships among variables. In this network, the flow of information spreads forward through the layers while the propagation of the error is back. Back propagation method usually gives out better results in the case of chemical engineering systems [39].

The output variable (density) is calculated using the input variables according to the following steps: The net inputs (N) for the hidden neurons are calculated from the input neurons via:

$$N_j^h = \sum_i^n w_{ij}^h p_i + b_j^h \tag{1}$$

where p corresponds to the vector of the inputs of the training, j is the hidden neuron, w_{ij} is the weight of the connection among the input neurons with the hidden layer, and the term b_j corresponds to the bias of the neuron j of the hidden layer. A network with bias developed relations between input and output easier than a network without bias. Starting from these inputs, the outputs of the hidden neurons (y) are calculated using a transfer function f^h associated with the neurons of this layer.

$$y_j^h = f_j^h \left(\sum_i^n w_{ij}^h p_i + b_j^h \right) \tag{2}$$

Similar calculations are carried out to obtain the results of each neuron of the following layer until the output layer.

The utmost advantage of an ANN is eliminating the complex equations, and replacing them with popular transfer functions. Different types of transfer functions have been proposed for artificial neural networks such as linear (purelin) function, logarithmic sigmoid (logsig), and hyperbolic tangent sigmoid (tansig) [30]. In the present study, different combinations of these mentioned transfer functions have been tested to choose the best. The purelin, logsig, and tansig transfer functions are defined in Eqs. (3)–(5), respectively:

$$\text{purelin}(n) = n \tag{3}$$

$$\text{logsig}(n) = \frac{1}{1+\exp(-n)} \tag{4}$$

$$\text{tansig}(n) = \frac{2}{(1+\exp(-2n))-1} \tag{5}$$

There are three main stages in the operation of the neural networks: learning, validation and test. The learning or training is the process in which the neural networks modify the weights and biases in answer to the initial information. The validation is used to measure the generalization of a network, and to halt training when generalization stops improving. The error on the validation subset is monitored during the training process. The validation error normally decreases during the initial phase of training, as does the training set error. However, when the network begins to overfit the data, the error on the validation set typically begins to rise. When the validation error increases for a specified number of iterations (the training is stopped, and the weights and biases at the minimum of the validation error are returned. Testing stage has no effect on training and so provides an independent measure of network's performance.

Overfitting occurs when the error on the training set is driven to a very small value, but when new data is presented to the network, the error is large. In the other words, the network has memorized the training examples, but it has not learned to generalize to the new situations. Note that in this study, the number of parameters in the network is much smaller than the total number of points in the training set and in this case, there is little or no chance of overfitting. Also, there are some techniques to prevent overfitting. Early stopping is automatically provided for all of the supervised network functions including the back propagation in which the data are divided into three subsets and prevent the overfitting. Regularization is the other technique that involves modifying the performance function. In this study, the early stopping has been used.

During the training process, input data are fed to the input layer of the network and the difference between the results from the output layer and the desired outputs (i.e., network error) is used as a criterion for adjustment of the network's synaptic weights and biases. At the beginning, all synaptic weights and biases are initialized randomly. Then, the network is trained (i.e., its synaptic weights are adjusted) by an optimization algorithm until it correctly emulates the input/output mapping [48,49]. Scheme 1 shows a block diagram developed to create an ANN model to predict the density of alkali metal systems and their mixtures.

In this study, 595 different data points of alkali metals and their alloys were used to train, validate and test the network. Table 1 shows the names of investigated alkali metal systems and the temperature and pressure ranges of experimental data [50–54] used in this work. The input parameters of the network were temperature (T), pressure (P), molecular weight (in the case of mixtures, average molecular weight) (M_w), and the composition of each system (x_i). Experimental density data at several temperatures and pressures were collected from the literature for both pure alkali metals [50–53] and their alloys [54]. In this work, all data were divided randomly to three subsets: 70% were randomly chosen to train the network, 15% for validation, and 15% to test it. In addition, the feed-forward multi-layer neural network has been trained by