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Modeling heat capacity of ionic liquids using group method of data handling: A hybrid and structure-based approach

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ABSTRACT

Ionic liquids (ILs) are a significant class of chemicals with applications in solar cells, sensors, capacitors, batteries, plasticizers and thermal fluids. These compounds have attracted wide attention due to their low vapor pressure, tunable viscosity, non-flammability, wide liquid region phase diagrams and substantial chemical and thermal stability. Moreover, ILs structures can be easily modified leading to highly tunable physicochemical properties, which widen the application of these compounds. Heat capacity of ILs is an essential property for heat transfer evaluation as well as the estimation of widely used thermodynamic properties. Establishing a generalized and accurate model for predicting the heat capacity of ILs is valuable for their further development. In this manuscript, a hybrid group method of data handling (GMDH) was employed to establish a model estimating the ILs heat capacities. The database employed is an all-inclusive source of data taken from NIST standard, which includes the heat capacities of 56 ILs as a function of temperature and four structural parameters. About 80% of the database was assigned for building the model, and the remainder was used for evaluating the model performance. Statistical parameters and graphical techniques revealed that the model developed in this study is very accurate, with an R^2 value of 0.982 and an average absolute percent relative error (AAPRE) of 1.84%. Moreover, the sensitivity analysis showed that the chemical structure of the cation has the highest impact on the heat capacity of ILs.

1. Introduction

Organic salts containing anionic and cationic species are termed ionic liquids (ILs). Unlike inorganic salts, these compounds remain in the liquid state, even at temperatures below 100 °C [1–6]. The low vapor pressure, tunable viscosity, non-flammability, wide region of the liquid phase, and the substantial chemical and thermal stability are attractive features of the ILs [7–9]. Accordingly, ILs have been used in solar cells, sensors, capacitors, batteries, plasticizers and thermal fluids. The flexibility of altering the length, branching and type of ionic species provides an opportunity to tune the ILs for a particular application [7,10]. Consequently, a large number of different ILs is currently in existence, which creates a need to predict ILs properties and suitability for a given application, even prior to their synthesis [11].

The heat capacity is an important for heat transfer applications and predicting the different thermodynamic properties [11,12]. Different definitions for heat capacity exist in the literature; including:

1. The energy required to increase the temperature rise at a saturated liquid condition (C_{satL});
2. At fixed pressure, changes in enthalpy with respect to the temperature ($C_{\sigma L}$);
3. The Variation of saturated liquid enthalpy against temperature at isobaric condition (C_{PL}).

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In this study, the last definition of heat capacity (i.e., C_{PL}) is used though all other definitions provide similar values of the heat capacity, except at high temperatures [13]. Experimental measurements of the IL heat capacities such as adiabatic calorimetry and differential scanning calorimetry [14] are not feasible for all kinds of ILs at all ranges of operational conditions. Moreover, these experiments are time consuming and expensive. Therefore, the ability to predict the heat capacity for such compounds is very valuable. A thorough survey of the open literature revealed few studies focused on predicting the heat capacity of ILs.

Irrespective of the temperature dependency, a model for predicting IL heat capacity as a function of molar volume was presented by Preiss et al. [15]. Using second order group additivity technique, Gardas and Coutinho [13] estimated IL heat capacity based on database of 19 ILs (about 2400 datapoints). This method could not, however, be generalized to ILs beyond the database. Based on the mass conductivity index, a four-parameter model with five tuning coefficients was extended in the work of Valderrama et al. [16,17] for reliable prediction of heat capacity of ILs. The main drawback of this method is its use of un-synthesized IL heat capacity at room temperature. The value of the heat capacity is calculated from another correlation or experimentally measured works. Moreover, this model was mainly developed for imidazolium-based ILs. Its application to other compounds may lead to large inaccuracies. Implementing Lego and Meccano modeling methods, two predictive models by Nancarrow et al. [18] were constructed using a set of 2396 heat capacity datapoints. The authors concluded that the Lego method is more generally applicable, even though the Meccano method gives higher precision. A structure-based approach was introduced in the work of Farahani et al. [11] using a dataset for 56 ILs heat capacity including 2940 datapoints. Farahani et al.'s model uses five parameters with a wide range of applicability with respect to temperature. Nevertheless, the effect of some molecular parameters, which are commonly available for ILs such as the number oxygen and nitrogen atoms, were not taken into account. Ahmadi et al. [19] proposed a correlation with 12 input variables including 10 molecular parameters, molecular weight of the IL and temperature so as to estimate the heat capacity using 128 different ILs. They concluded that their model could predict the heat capacity of ILs with an average relative error of less than 6%. Barati-Harooni et al. [20] utilized two different types of neural network-based systems; including radial basis function and multi-layer perceptron to estimate IL heat capacity in relation to the same input parameters used by Ahmadi et al. [19]. Even though their smart strategies were highly accurate, the model represented a black box, with no clear mathematical formula. In another study, using the concept of gene expression programming (GEP), support vector machine (SVM) and fuzzy-based systems, three different models were constructed by Barati-Harooni et al. [21]. Their results demonstrated that the SVM-based method gives the most accurate predictions in relation to their own work and other published correlations. The current work was established with the objective of developing a more precise model for predicting heat capacity of ILs with less number of input parameters, so that it could be applicable to different types of ILs at a wider range of physical conditions.

In the current study, a new version of neural network-based system, namely hybrid group method of data handling (GMDH), was employed for estimating the heat capacities of ILs. To the best of the authors' knowledge, there is no literature that applies GMDH modeling to the subject at hand. The developed model employs a comprehensive data source of 56 ILs capturing the heat capacity with respect to the number of methyl groups (CH_3R_c) in the cation counter parts of the ILs, temperature (T), the number of hydrogen atoms in anion (nH) and the atom count (N) in both the cation and the anion. The current model is compared with empirical correlations in the literature using several statistical parameters and graphical techniques. Lastly, a sensitivity analysis was performed to assess the impact of the different model parameters on the model accuracy.

2. Data collection

In any modeling exercise, it is recommended to employ a widespread and reliable source of data in order to develop a dependable and general model [22–39]. With this in mind, a universal database of 2940 heat capacity values was collected from the NIST Standard Reference Database #147. The input variables are the number of methyl groups (CH_3R_c) in the cation counter part of the IL, the number of hydrogen atoms (nH) in the anions, temperature (T) and atom count (N) in both the cation and the anion. Heat capacity of the database varied between 226 and 1413 $J{\cdot}mol^{-1}{\cdot}K^{-1}$, while temperature varied between 188.06 and 663.10 K. A statistical analysis of the dataset used in this study is given in Table 1.

The key reason for using the NIST database is the fact that they report the uncertainty in each measured value which provides the ability to decide whether to use the data for modeling or not. This in turn increases the reliability of the established model. When multi-values of the heat capacities are reported at a particular temperature, the heat capacity with the lowest uncertainty was selected. In the case of identical uncertainty values, the most recently published heat capacity was considered more reliable, by virtue of the higher precision of the more modern instruments [21].

3. Model development

3.1. Group method of data handling

Soft computation strategies including artificial neural network (ANN), fuzzy inference systems (FIS), group method of data handling (GMDH), support vector machine (SVM) and genetic programming (GP), have been utilized in diverse fields of chemical and petroleum engineering [22–27,40,41]. These smart techniques simplify sophisticated and highly nonlinear problems in order to estimate the modeling targets accurately. Initially, GMDH was recognized as a self-organizing approach by Shankar [42]. Subsequently, a comprehensive investigation was implemented by researchers [43] to solve intricate phenomena/problems leading to the GMDH which is the most dominant algorithm for treating long/short-term estimation, function finding and pattern recognition analysis. Originally, it has been represented that the current deterministic techniques in engineering cybernetics have high restrictions in comparison with GMDH approach [44]. In principle, GMDH can be applied in wide areas of science and engineering such as medical diagnostics, environmental systems, marketing and econometric modeling, weather modeling, microprocessor-based hardware, multi-sensor signal processing, acoustic and ultrasonic emissions, seismic and acoustic simulations [45–50]. To the best of our knowledge, there is very few publications on GMDH modeling in chemistry, petroleum and chemical engineering fields of study [40,41].

Polynomial neural network (PNN) or the so-called GMDH method has several layers wherein each layer contains some independent nodes. The nodes in the subsequent layers will be created by the quadratic polynomial functions combining the nodes existing in the previous layers. The optimum representation of the quadratic polynomial equations in GMDH algorithm was illustrated through the work of Ivakhnenko [44]. The following relationships known as Volterra-Kolmogorov-Gabor, is applied in