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Prediction of solubility of carbon dioxide in different polymers using support vector machine algorithm

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ABSTRACT

This paper concerns with implementation of support vector machine algorithm for developing improved models capable of predicting the solubility of CO_2 in five different polymers namely polystyrene (PS), poly vinyl acetate (PVAC), polypropylene (PP), poly butylene succinate-*co*-adipate (PBSA) and poly butylene succinate (PBS). Validity of the presented models has been evaluated by utilizing several statistical parameters. The predictions of the developed models for polymers of PS, PVAC, PP, PBSA, PBS are in excellent agreement with corresponding experimental data with the average absolute relative deviation percent (%AARD) equal to %0.151, %0.500, %1.381, %0.158, %0.239 and R^2 values of greater than 0.999. Furthermore, the estimation capability of the proposed models has been compared to a well-known equation of state (EOS) as well as artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS) models. According to the results of comparative studies, it was found that the developed models are more robust, reliable and efficient than other existing techniques for improved analysis and design of polymer processing technology.

1. Introduction

Solubility of gases in polymers have been of continuous experimental and theoretical interest because gas and vapor separation membranes are based on the so-called solution–diffusion mechanism, according to which thermodynamic parameters of gas sorption determine the mass transfer driving force [1]. This subject is one of the most important interests of many chemical engineers due to its wide application in industries such as polymer foaming processes.

During the past decades, several attempts have been made to describe the solubility of gases in polymers. The main experimental methods for determination of gas solubility in polymers are volumetric method [2,3], gravimetric method [4], chromatographic method [5], separation method [5] *etc.* Due to the fact that development of suitable conditions for implementation these experiments is very difficult, time consuming and costly process, several studies have been focused on development of new ways for simple and accurate determination of solubility of gases in polymers [6,7].

Carbon dioxide also has a great use in polymer processing. Micro-molecular foams can be formed by using CO_2 as a blowing agent [8]. Some polymers (polypropylene) can be used in heat insulators and support materials after foaming with gases like CO_2 [8]. For these reasons the prediction of CO_2 solubility in different polymers has been a research topic for many years [9–11]. In recent years new intelligent methods have been developed to predict the answer of a problem without difficulty of experimental works [7,11–14]. For example in artificial neural networks (ANNs) the experimental data are used for learning the network and due to high computational rate of this method it can improve the accuracy of prediction in compared to thermodynamic models [11]. Another intelligent model is adapted neuro-fuzzy inference system (ANFIS) which has a potential for solving nonlinear systems and reduces the computational time [7]. For more examples of intelligent

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Nomenclature

AARD	= average absolute relative deviations (%)
b	= bias term
C	= a positive constant
$K(x,x_k)$	= Kernel function
LSSVM	= least-squares supported vector machine
MSE	= mean square error
R^2	= R-squared
RMSE	= root mean square error
SA	= simulated annealing
T	= transpose
w	= a nonlinear function
y	= outputs
α_i	= Lagrange multipliers
γ	= relative weight of the summation of the regression errors
σ^2	= squared bandwidth
PS	= polystyrene
PP	= polypropylene
PVAC	= poly vinyl acetate
PBS	= poly butylene succinate
PBSA	= poly butylene succinate-*co*-adipate

systems, fuzzy logic, wavelet analysis, colony optimization algorithm, particle swarm optimization algorithm, radial basis neural networks and support vector machine can also be mentioned [12,15–23].

Khajeh et al. [7] applied adaptive neuro-fuzzy inference systems for prediction of CO_2 solubility in seven different polymers and compared the prediction results with artificial neural network and EOSs. They concluded that the prediction of ANFIS is more accurate than other predictive tools. More recently, Li et al. [6] developed an intelligent model for solubility prediction of different gases in polymers using radial basis function neural network based on chaotic self-adaptive particle swarm optimization and a clustering method. The statistical quality measures showed that their model is accurate for gas-polymer solubility prediction.

In another study Li et al. [24] attempted to improve back propagation artificial neural network model by chaotic self-adaptive particle swarm optimization techniques (CSPSO) for the same purpose. Their results showed that developed model has high capability for prediction of solubility of CO_2 and N_2 in polystyrene and polypropylene. Khajeh et al. [25] also used an adaptive neuro-fuzzy inference system and radial basis function neural network for solubility prediction of some gases in polystyrene such as butane, isobutene, carbon dioxide, 1,1,1,2-tetrafluoroethane, 1-chloro-1,1-difluoroethane,1,1-difluoroethane.

In the past decades, the developments in various statistical and intelligent methods make them attractive for modeling of complex systems [12,26,27]. Nowadays, support vector machine (SVM) is gaining more popularity among researchers [12,14,28]. The SVM is a new and supervised machine learning technique which works based on the statistical learning theory [29]. The least square version of the SVM (LSSVM) which widely used in complex system studies for modeling, regression or parameter prediction, was described in Suykens and Vandewalle [29]. However its application to the gas-polymer systems is very limited. Moreover, to the best of authors' knowledge, no work has been published on the subject of modeling of CO_2 gas solubility prediction in polymer with this approach. Hence, In this work the LSSVM algorithm was applied for accurate determination of CO_2 solubility in five different polymers including polystyrene (PS), poly vinyl acetate (PVAC), polypropylene (PP), poly butylene succinate (PBS) and poly butylene succinate-*co*-adipate (PBSA). To follow our objective in this study, statistical error analysis (*i.e.*, APRE, AAPRE, RMSE and R^2) and graphical error analysis (*i.e.*, cross plots) have been performed. In addition accuracy of the developed models is compared with other intelligent models (*i.e.*, ANN and ANFIS) as well as EOS. Moreover, the applied data base is evaluated by the leverage value statistical approach.

2. LSSVM modeling

The SVM is a supervised learning model which was proposed by Vapnik [30]. SVMs use the spirit of the structural risk minimization principle [30,31]. However the major drawback of the SVM is its higher computational burden because of required constrained optimization programing [29]. Suykens and Vandewalle [29] presented a modification to the traditional SVM called least-squares SVM (LSSVM) so as to facilitate the solution of the original SVM framework. LSSVM appears to offer advantages similar to those of SVM, but its great advantage is that LSSVM applies a set of linear equations (linear programming), instead of quadratic programming problems in order to reduce the complexity of optimization process [29]. Considering the problem of approximating a given dataset $\{(x_1,y_1),(x_2,y_2),\ldots,(x_N,y_N)\}$ with a nonlinear function:

$$f(x) = \langle w, \Phi(x)\rangle + b \tag{1}$$

where, $\langle .,. \rangle$ represent dot product; $\Phi(x)$ represents the nonlinear function that performs linear regression; w and b are weight vector and bias terms, respectively. In LSSVM for function prediction, the optimization problem is expressed as [32]:

$$\min_{w,b,e} j(w,e) = \frac{1}{2}\|w\|^2 + \frac{1}{2}\gamma\sum_{k=1}^{N} e_k^2 \tag{2}$$

$$s.t.\, y_k = \langle w, \Phi(x_k)\rangle + b + e_k \quad k = 1,\ldots,N \tag{3}$$

where, $e_k \in R$ are error variables; $\gamma \geq 0$ is a regularization constant. The Lagrangian of the problem is defined by [32]:

$$L_{\mathrm{LSSVM}} = \frac{1}{2}\|w\|^2 + \frac{1}{2}\gamma\sum_{k=1}^{N} e_k^2 - \sum_{k=1}^{N}\alpha_k\{\langle w, \Phi(x_k)\rangle + b + e_k - y_k\} \tag{4}$$

with Lagrange multipliers $\alpha_k \in R$. The condition for optimally is determined by [32]:

$$\begin{cases} \dfrac{\partial L_{\mathrm{LSSVM}}}{\partial w} = 0 \rightarrow w = \displaystyle\sum_{k=1}^{N}\alpha_k\Phi(x_k) \\ \dfrac{\partial L_{\mathrm{LSSVM}}}{\partial b} = 0 \rightarrow \displaystyle\sum_{k=1}^{N}\alpha_k = 0 \\ \dfrac{\partial L_{\mathrm{LSSVM}}}{\partial e_k} = 0 \rightarrow \alpha_k = \gamma e_k \\ \dfrac{\partial L_{\mathrm{LSSVM}}}{\partial \alpha_k} = 0 \rightarrow \langle w, \Phi(x_k)\rangle + b + e_k - y_k = 0 \end{cases} \tag{5}$$

By specifying $Y = [y_1;\ldots;y_N]$, $1_v = [1;\ldots;1]$, $\alpha = [\alpha_1;\ldots;\alpha_N]$ and eliminating e_k and w, following equations are achieved [32]:

$$\begin{bmatrix} 0 & 1_N^T \\ 1_N & \Omega + \gamma^{-1} I_N \end{bmatrix}\begin{bmatrix} b \\ \alpha \end{bmatrix} = \begin{bmatrix} 0 \\ Y \end{bmatrix} \tag{6}$$

where, 1_N is an $N \times N$ identity matrix, and $\Omega \in R^{N\times N}$ is the kernel matrix presented by:

$$\Omega_{kl} = \Phi(x_k)\Phi(x_l) = K(x_k,x_l), \quad k,l = 1,\ldots N \tag{7}$$

As mentioned earlier, has a tuning parameter γ. From the other point, as the LSSVM is a kernel based method, parameters of kernel