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Estimating antiwear properties of lubricant additives using a quantitative structure tribo-ability relationship model with back propagation neural network

Kang Dai [a], Xinlei Gao [b,*]

[a] College of Pharmacy, South-Central University for Nationalities, Wuhan, Hubei Province 430074, PR China
[b] School of Chemical and Environmental Engineering, Wuhan Polytechnic University, Wuhan, Hubei Province 430023, PR China

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ABSTRACT

To be able to predict tribological properties of new lubricant additives as well as clarify lubricating mechanisms, one needs to study the relationship between structures of lubricant additives and their lubricating properties. With a focus on estimating antiwear properties of some heterocyclic additives, we use the quantitative structure tribo-ability relationship (QSTR) model to predict tribological data, which introduces the idea of computer-aided design into tribology. This is combined with back propagation neural network (BPNN), a machine-learning method that offers simplicity and robustness. This study determined the feasibility and predictability of developing the BPNN QSTR model to estimate lubricant additive antiwear properties. For 36 additives, 90 structural descriptors, such as octanol-water partition coefficient, quantum indices, 2D topological indices, and 3D Jurs descriptors, were included as BPNN inputs. Antiwear parameters include wear-scar diameters under three loads. Leave-one-out cross-validation was performed to evaluate accuracy and robustness of this BPNN QSTR model. We also evaluate the descriptor sensitivities, from which we can determine the effects of each descriptor and clarify wearing mechanisms. Given a positive assessment, this method warrants further development and validated integration with other tribological properties.

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1. Introduction

Exploring the relationship between chemical structures and properties has been a puzzling but interesting problem for chemists over the years. Using the linear regression method, Hansch et al. [1] revealed a relationship between quantitative structural descriptors and molecular properties, and thereby established predictive models of these properties. Quantitative structure–activity relationship (QSAR) models emerged and have been in wide use ever since. During development, the QSAR method advanced in two directions: one extending the QSAR descriptors from initially electronic, spatial geometry, and lipid–water partition characteristics, to include quantum [2], topology [3–5], and 3D-QSAR [6] characteristics that revealed more subtle structure and conformation information; the other introducing new methods to build QSAR models. From simple linear regression, QSAR methods incorporated machine-learning methods such as artificial neural network [7], partial least square (PLS) [8], and support vector machine (SVM) [9].

In recent years, QSAR has gradually extended to other areas of applications including tribology. However, studies on the dependence of tribological properties on molecular structure are sparse, with only a few available reports. The tribological performance and structure of ionic liquids has been studied by Weimin Liu's team [10,11] and by Jiménez's team [12], and tribological properties of hydrocarbons were also investigated by Himmat Singha et al. [13].

This paper presents the new notion of a quantitative structure tribo-ability relationship (QSTR) with the aim of building a predictive BPNN model for anti-wear-scar properties, and to explore lubrication mechanisms. QSTR uses methods and descriptors from QSAR to build a predictive model for tribological properties. For the present study, we chose the back propagation neural network model (BPNN), which is a classic neural network method known for its simplicity and robustness. The BPNN model (Fig. 1) comprises three layers of neurons: input, output, and hidden. The most important is the hidden layer which provides the key transformation and generates an output value [14]. We chose quantum, 2D topological, and 3D Jurs descriptors as variable inputs to the model. These descriptors are well known for their good performance in the prediction of physical properties of compounds [2–5,15].

* Corresponding author. Tel.: +86 27 83956237; fax: +86 27 83641630.
E-mail address: gaoxl0131@163.com (X. Gao).

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2. Material and methods

A set of 36 compounds and their tribological data were examined [16]. Data included wear-scar area of compounds under three loads: 196, 294 and 392 N. Wear-scar area is used to determine molecule lubricity at the optimal concentration by the simple expression:

$$WS_{(196\ \mathrm{N})} = \log_{10}\frac{S_0 \times MW}{S \times Conc}, \quad (1)$$

$$WS_{(294\ \mathrm{N})} = \log_{10}\frac{\left(S_0^{3/2} - S^{3/2}\right) \times MW}{Conc}, \quad (2)$$

$$WS_{(392\ \mathrm{N})} = \log_{10}\frac{S_0^{3/2} \times MW}{S^{3/2} \times Conc}, \quad (3)$$

where $WS_{(L)}$ expresses the compound's ability to decrease wear-scar area for load L; S_0 is the negative control, which is the wear-scar area formed under pure base paraffin oil; S is the wear-scar area formed using the lubricant compound; MW is the molecular weight of compounds; and $Conc$ is the concentration of the compound to obtain the best lubrication.

The structures of 36 compounds were generated using the Discovery Studio software package; molecular energies were minimized and charges calculated by standard methods. QSAR descriptors included ALogP, quantum descriptors, two-dimensional (2D) topological descriptors and three-dimensional (3D) Jurs descriptors. AlogP is a descriptor determined by liposolubility of compounds, whereas quantum descriptors include energies for HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital), heat of formation, dipole and quadrupole moments, and so on. Dipole and quadrupole descriptors indicate the strength and orientation of molecules in electrostatic fields, and indicate the polarity in the three-dimensional compound. In the presented study, quantum descriptors were calculated by density function methods.

Topological descriptors are a special class of descriptors that do not rely on three-dimensional models. Values of 43 descriptors derive from the two-dimensional topology of the molecule. Topological descriptors indicate graph properties and side-chain characteristics of molecules [4]. Jurs descriptors combine shape and electronic information to characterize molecules [15]. The 30 descriptors are calculated by mapping atomic partial charges on solvent-accessible surface areas around individual atoms. Tables 1 and 2 list all topological and Jurs descriptors.

The predictive models, also built in Discovery Studio, implement a particular type of neural network known as a back-propagation neural network (BPNN), a training method that back-propagates errors of the units of the output layer in determining the errors for the units of the hidden layer. The neuron number in the hidden layer (middle layer) was optimized by the software itself. For all predictive models, the optimal neuron number in the hidden layer is 3, which effectively prevents overfitting. Cross-validation was performed using the leave-one-out (LOO) method. For all models, six compounds were selected randomly to compose the test group. Because inputting a plethora of descriptors would lead to overfitting, we established two BPNN models for WS under each load, one based on input

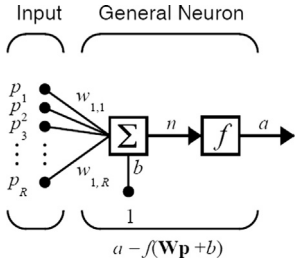

Fig. 1. Scheme of a typical BPNN. (a: Output vector; p: Input vector (R=Number of element in input vector); W: weight; b: intercept vector; n: number of neurons in hidden layer; f: transfer function).

Table 2
Jurs descriptors.

Descriptors	Explanation
Jurs_PPSA_1	Partial positive surface area
Jurs_PNSA_1	Partial negative surface area
Jurs_PPSA_2	Total charge weighted positive surface area
Jurs_PNSA_2	Total charge weighted negative surface area
Jurs_PPSA_3	Atomic charge weighted positive surface area
Jurs_PNSA_3	Atomic charge weighted negative surface area
Jurs_DPSA_1	Difference in charged partial surface areas
Jurs_DPSA_2	Difference in total charge weighted surface areas
Jurs_DPSA_3	Difference in atomic charge weighted surface areas
Jurs_FPSA_1	Fractional charged partial surface areas
Jurs_FPSA_2	
Jurs_FPSA_3	
Jurs_FNSA_1	
Jurs_FNSA_2	
Jurs_FNSA_3	
Jurs_WPSA_1	Surface-weighted charged partial surface areas
Jurs_WPSA_2	
Jurs_WPSA_3	
Jurs_WNSA_1	
Jurs_WNSA_2	
Jurs_WNSA_3	
Jurs_RPCG	Relative positive charge
Jurs_RNCG	Relative negative charge
Jurs_RPCS	Relative positive charge surface area
Jurs_RNCS	Relative negative charge surface area
Jurs_TASA	Total hydrophobic surface area
Jurs_TPSA	Total polar surface area
Jurs_RASA	Relative hydrophobic surface area
Jurs_RPSA	Relative polar surface area
Jurs_SASA	Total molecular solvent-accessible surface area

Table 1
Topological descriptors.

Descriptors	Explanation
JX, JY	Balaban indices
Wiener	Wiener index
Zagreb	Zagreb index
CHI_0, CHI_1, CHI_2, CHI_3_P, CHI_3_C, CHI_3_CH, CHI_V_0, CHI_V_1, CHI_V_2, CHI_V_3_P, CHI_V_3_C, CHI_V_3_CH	Connectivity indices
IC, BIC, CIC, SIC, IAC_Total, IAC_Mean, V_ADJ_mag, V_DIST_mag, V_ADJ_equ, V_DIST_equ, E_ADJ_mag, E_DIST_mag, E_ADJ_equ, E_DIST_equ	Graph-theoretical info Content descriptors
Kappa_1, Kappa_2, Kappa_3, Kappa_1_AM, Kappa_2_AM, Kappa_3_AM, PHI	Kappa shape Indices
SC_0, SC_1, SC_2, SC_3_P, SC_3_C, SC_3_PC	Subgraph counts

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