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Prediction of the electric conductivity of ionic liquids by two chemometrics methods

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Abstract: In recent years, the study of the properties of ionic liquids (ILs) and their structures has developed largely. Among the common physicochemical properties of pure ILs, electric conductivity (*EC*) is of crucial importance for both practical and fundamental viewpoint. In order to develop effective models for predicting the *EC* value of various ILs, the relationship between the structural descriptors and the *EC* of thirty-five ionic liquids at different temperatures was investigated by multi-linear regression (MLR) and a back propagation artificial neural network (ANN). As a result, a three layer ANN with four variables selected by the MLR model as input nodes was successfully set up. The descriptors selected by MLR were suitable and significant to be the input nodes of the ANN model in this study. Moreover, the ionic conductivities calculated by the ANN model, having a high correlation coefficient and low root mean squared error, were quantitatively in good agreement with the experimental values. The ANN model was proved to be better than the MLR model.

Keywords: electroconductibility; ionic liquids; multi-linear regression; artificial neural network.

INTRODUCTION

In the last decades, ionic liquids (ILs) have attracted increasing interest in countless fields of physical and chemical sciences as “designer solvents”.[1] Their properties, such as high chemical and thermal stability, low vapor pressure and high ionic conductivity, drive their popular application as useful media for synthesis, catalysis, extraction and separation processes.[2–6] Among the common properties of pure ILs, conductivity is of crucial importance for both practical and fundamental viewpoints.[7] The study of the conductivity of ILs was fully reviewed by Galinski *et al.*[8] In recent years, methods to investigate the properties of ILs

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have been developed to a great extent. Especially with the development of chemometrics, the computational method has been widely used in fundamental research of ILs.[9,10] The early theoretical methods were summarized in a paper by Picálek *et al.*,[11] which included quantum calculations, molecular dynamics (MD) simulation and some Monte Carlo simulations. Zhao *et al.*[12] reported a model based on the conventional hole theory to study the conductivity and the calculated values were successfully fitted to the experimental data. However, the model was weak in predicting the conductivity of new ionic liquids. Matsuda and his coworkers[13] developed a method using computer-aided reverse design for ionic liquids. The conductivity of ionic liquids was conveniently predicted by computer-aided reverse design, but the model required complex parameters to fit. As for MD simulation, the presented studies on calculating the conductivity of ionic liquids were usually limited to one type of ionic liquids.[9,11,14] Recently, some researchers attempted to adopt simple models to predict the conductivity of different ILs at different temperatures, *e.g.*, the study of Eiden *et al.*[15] Moreover, the quantitative structure–property relationships (QSPR) method was popularly used to study the properties of ionic liquids and Zheng *et al.*[16] reviewed these meaningful studies. The back-propagation artificial neural network (BP ANN) method has been widely used by QSPR models in the study of other properties of ILs and good results were obtained.[17–19] To the best of our knowledge, the BP ANN method has not been reported in the study of the conductivity of ILs. This study aimed to explore multi-linear regression (MLR) and BP ANN methods to fit and predict the conductivity of ILs. The BP ANN was proved to have a better fitting and prediction capability than the MLR model. Furthermore, the developed ANN model has only four input nodes, which make it simpler than the other models.[12,13] The structures of the ionic liquids investigated in this study included the cations and anions given in Fig. 1, which represent the common anion–cation compositions of ILs.

COMPUTATIONAL DETAILS

Dataset

Thirty-five ionic liquids (Table I) with 364 electric conductivity (*EC*) data at different temperatures from the Ionic Liquid Database (ILD)[20] were collected to investigate the relationship between the structure and conductivity of ILs. The 3D structural coordinates of the cations and anions[21-30] of these ILs were obtained from the Cambridge Structural Database (CSD).[31] All 364 data were divided into three data sets that contained a training set, a validation set and a test set with the percentage of 55, 18 and 27 %, respectively. In order to reflect the prediction capability of model, the data points of the same structural ILs were not divided into different datasets.

Descriptors

All the structures of the cations and anions were separately optimized at the B3LYP[32,33] level of calculation, using the 6-31G(d) basis set. These calculations were performed using the

Gaussian03 program.[34] Quantum mechanics descriptors of cations and anions were collected, which included total molecular energy (E), volume (V), the highest occupied molecular orbital energy (E_{HOMO}), the lowest unoccupied molecular orbital energy (E_{LUMO}), dipole moment (μ) *etc*. (Table II).

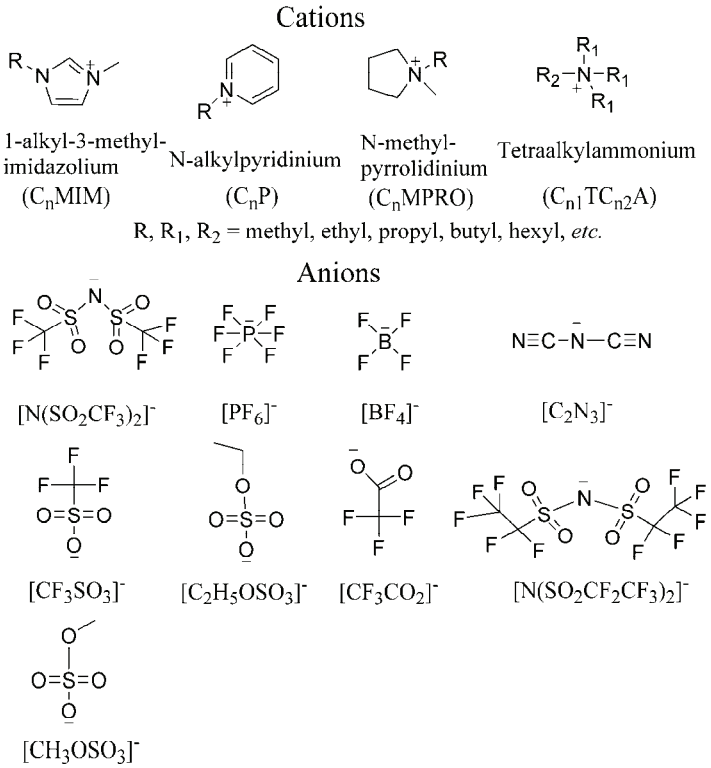

Fig. 1. The optimized cations and anions of the investigated ionic liquids.

TABLE I. The studied ionic liquids and data sets; training set: entries 1–18, validation set: entries 19–26, test set: entries 27–35

Entry	ILs	Temperature range, K	Data points
1	$[BP]^+$ $[N(SO_2CF_3)_2]^-$	263.1–373.1	10
2	$[BMIM]^+[PF_6]^-$	268.15–468.15	21
3	$[HMIM]^+[BF_4]^-$	238.1–468.1	24
4	$[BMIM]^+[N(SO_2CF_2CF_3)_2]^-$	263.1–373.1	10
5	$[BTMA]^+[N(SO_2CF_3)_2]^-$	263.1–373.1	10
6	$[HMIM]^+[PF_6]^-$	295.1	1
7	$[OMIM]^+[PF_6]^-$	273.22–353.17	17
8	$[BMIM]^+[CF_3SO_3]^-$	268.15–468.15	21
9	$[MMIM]^+[N(SO_2CF_3)_2]^-$	263.15–373.15	12
10	$[EMIM]^+[C_2H_5OSO_3]^-$	258.1–433.1	36
11	$[HepTMA]^+[N(SO_2CF_3)_2]^-$	298.1	1
12	$[HTEA]^+[N(SO_2CF_3)_2]^-$	298.1	1

TABLE I. Continued

Entry	ILs	Temperature range, K	Data points
13	$[PMIM]^+[BF_4]^-$	298.1	1
14	$[HTBA]^+[N(SO_2CF_3)_2]^-$	298.1	1
15	$[OTBA]^+[CF_3SO_3]^-$	298.1	1
16	$[DMIM]^+[N(SO_2CF_3)_2]^-$	295.1	1
17	$[BMPRO]^+[N(SO_2CF_3)_2]^-$	263.1–373.1	10
18	$[BMIM]^+[C_2N_3]^-$	248.15–468.15	23
19	$[BMIM]^+[BF_4]^-$	238.1–468.1	24
20	$[HTMA]^+[N(SO_2CF_3)_2]^-$	298.1	1
21	$[HMIM]^+[N(SO_2CF_3)_2]^-$	263.15–373.15	12
22	$[BMIM]^+[N(SO_2CF_3)_2]^-$	263.15–373.15	12
23	$[OTEA]^+[N(SO_2CF_3)_2]^-$	298.1	1
24	$[HepTBA]^+[N(SO_2CF_3)_2]^-$	298.1	1
25	$[PMPRO]^+[N(SO_2CF_3)_2]^-$	298.1	1
26	$[BMIM]^+[CF_3CO_2]^-$	248.15–368.15	13
27	$[OMIM]^+[BF_4]^-$	238.1–468.1	24
28	$[OTMA]^+[N(SO_2CF_3)_2]^-$	298.1	1
29	$[EMIM]^+[N(SO_2CF_3)_2]^-$	248.15–353.15	12
30	$[HepTEA]^+[N(SO_2CF_3)_2]^-$	298.1	1
31	$[OTBA]^+[N(SO_2CF_3)_2]^-$	298.1	1
32	$[EMIM]^+[C_2N_3]^-$	238.15–353.15	13
33	$[BMIM]^+[CH_3OSO_3]^-$	303.2–353.2	11
34	$[OMIM]^+[N(SO_2CF_3)_2]^-$	263.15–373.15	12
35	$[EMIM]^+[BF_4]^-$	248.1–468.1	23

TABLE II. Data on the calculated quantum mechanics descriptors of the cations and anions

Ion	E / Hartree	E_{HOMO} / eV	E_{LUMO} / eV	M / D	V / cm^3 mol^{-1}
$[N(SO_2CF_3)_2]^-$	–1827.204081	–0.13542	0.17532	5.4169	127.003
$[PF_6]^-$	–940.6433857	–0.15668	0.281	0.0039	60.874
$[BF_4]^-$	–424.4990828	–0.11504	0.45846	0.0056	39.883
$[CF_3SO_3]^-$	–961.4979779	–0.07422	0.27653	4.4049	66.57
$[C_2H_5OSO_3]^-$	–778.313977	–0.06373	0.23851	4.9715	102.525
$[CF_3CO_2]^-$	–526.2375255	–0.03684	0.23386	4.7109	52.332
$[C_2N_3]^-$	–240.4860307	–0.0356	0.24231	0.8802	52.9
$[CH_3OSO_3]^-$	–738.9948944	–0.06002	0.25643	3.7771	72.761
$[N(SO_2CF_2CF_3)_2]^-$	–2302.767629	–0.14171	0.14446	6.3196	152.803
$[BP]^+$	–405.9244466	–0.45036	–0.23699	4.654	125.055
$[BMIM]^+$	–423.1786802	–0.42798	–0.17697	4.7649	131.637
$[HMIM]^+$	–501.8072113	–0.407	–0.17572	10.7448	176.128
$[OMIM]^+$	–580.4349008	–0.37963	–0.17545	16.0574	166.481
$[BTMA]^+$	–332.1108499	–0.45713	–0.11563	5.1609	124.465
$[MMIM]^+$	–305.2307599	–0.43626	–0.18467	0.7255	79.19
$[HTMA]^+$	–410.739132	–0.41514	–0.11419	10.015	151.489
$[EMIM]^+$	–344.5497093	–0.43158	–0.1805	1.6718	91.159
$[HepTMA]^+$	–450.0530362	–0.40016	–0.11389	12.6447	124.016
$[OTMA]^+$	–489.3668758	–0.38626	–0.11367	15.3359	175.687

TABLE II. Continued

Ion	E / Hartree	E_{HOMO} / eV	E_{LUMO} / eV	μ / D	V / cm^3 mol^{-1}
$[HTEA]^+$	–528.6795481	–0.41196	–0.08612	7.3707	192.29
$[PMIM]^+$	–383.8648154	–0.42903	–0.17839	3.5943	98.59
$[HepTEA]^+$	–567.9934591	–0.39697	–0.0858	9.6601	197.391
$[OTEA]^+$	–607.3072645	–0.38367	–0.08561	12.0649	211.196
$[HTBA]^+$	–764.5659048	–0.4091	–0.07486	3.6835	286.142
$[HepTBA]^+$	–803.879782	–0.39416	–0.07472	5.3987	318.19
$[OTBA]^+$	–843.1935931	–0.38139	–0.07462	7.3244	263.86
$[DMIM]^+$	–659.0624362	–0.36004	–0.17544	21.6069	181.37
$[PMPRO]^+$	–370.2219501	–0.48125	–0.10628	1.4382	115.118
$[BMPRO]^+$	–409.5364044	–0.45239	–0.10493	3.3716	140.102

Moreover, the molecular connectivity index is a term that reflects the relative accessibility of each bond to encounter other bonds of the same molecule in a milieu. The essence of molecular connectivity is the encoding of structure in a non-empirical way, which is thought to be helpful in the investigation of the physical properties.[35] Molecular connectivity index used in this study contained a chi index of zero order (^0X), first order (^1X) and second order (^2X). Eleven functions, such as linear, multinomial, exponent, logarithm and power function *etc*., were explored to fit the relationship between each descriptor and conductivity. $^0X_{cation}$ was found to have a good S function relationship with the conductivity of ILs.

To predict the conductivity of ionic liquids at different temperatures, the temperature (T) must be considered as a variable in the models. In addition, the temperature and *EC* had a good quadratic polynomial relationship. Thus, the temperature (T), the S function of $^0X_{cation}$ (S) and the square of temperature (T^2) were also explored in this study. To decrease the redundancy existing in the descriptor data matrix, the descriptors correlation with each other and with the conductivity of the ILs were examined and collinear descriptors (*i.e*., $r^2 > 0.9$) were detected.[36] Among the collinear descriptors, the one with the highest correlation with the conductivity was retained and the others were removed from the data matrix.

Prediction models

The prediction models for the conductivity of ILs were obtained by correlating the selected descriptors with the conductivity of corresponding ILs by the MLR and BP ANN methods. For a comparison of the different results from the MLR and BP ANN models, the training set and validation set were combined as one data set (called the construction set in the following text) and regressed to construct the MLR model, and the test set was used to evaluate the prediction capability of the models. The linear model was found with MLR method by the statistical package of the Social Sciences (SPSS) software. The model which had the highest squared correlation coefficient (R^2) was selected as the MLR prediction model to investigate the conductivities of the ILs. The descriptors determined in the linear model were selected as the input node of the ANN, and the conductivity of the ILs was considered as the output node. All parameters were first normalized to a scale of zero to one prior to use as the training, validation or testing data to avoid numerical overflows during the ANN process. The initial weights of the training network and momentum factor were random numbers. Then different training times, learning rates and number of neurons in the hidden layer were inputted to calculate the conductivity of ILs in training sets by means of the BP ANN method. The ANN algorithms were implemented in MATLAB programming language.

RESULTS AND DISCUSSION

The relationship between the descriptors and EC

Twenty-eight ionic liquids having conductivity data at 298.1K were used to investigate the relationship between the descriptors and conductivity. Eleven common functions were explored to fit each descriptor and conductivity. The EC was found to have a good S-function relationship with ${}^0X_{\text{cation}}$ (*S*), and the S function between ${}^0X_{\text{cation}}$ and the *EC* is shown in Fig. 2.

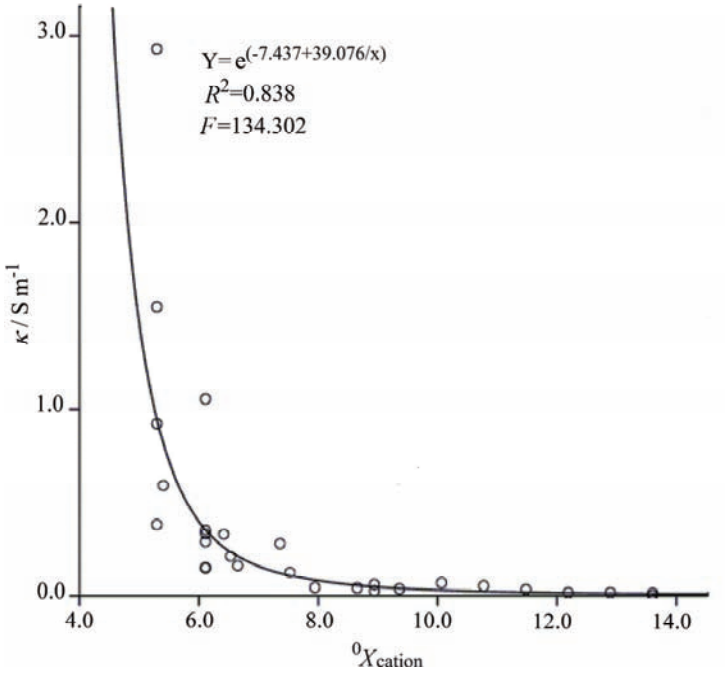

Fig. 2. The S-function relationship between ${}^0X_{\text{cation}}$ and electric conductivities.

The effect of temperature (*T*) was also studied. The relationship between the temperature and *EC* of the ionic liquids having more than one data point was all investigated. The rule was found that the temperature had quadratic polynomial relationship with *EC* for all the studied ionic liquids and had R^2 values higher than 0.98.

MLR Study

The descriptors were potential variables for the MLR model, which were chosen through correlation analysis. The linear relationship between these descriptors and *EC* was set up using the MLR model using SPSS software. The good correlations with experimental conductivity were selected based on R^2. Then, those descriptors without notable influence on R^2 were deleted and the model

with least number of descriptors and a high R^2 was founded. The correlation obtained for 364 data points of ILs conductivity is presented by the four-parameter equation as following:

$$EC=14.577\ E_{\text{HOMO-anion}} - 0.117\ \mu_{\text{anion}} + 0.422\ S + (5.751\text{E}{-5})\ T^2 - 2.941 \quad (1)$$

$$R^2 = 0.836,\ F = 331.818,\ sig = 0.000 \text{ (for all variables and the model)}$$

In the SPSS software, a *sig* value smaller than 0.05 indicates that the MLR model is of obvious statistical significance. In order to evaluate the model better, two aspects were compared concerning the prediction capability for different ionic liquids and the prediction capability for the same kind of ionic liquid but at different temperatures.

To illustrate the capability for different ionic liquids, the *EC* in the middle point of the temperature range for every ionic liquid was collected and analyzed. The calculated results for different ILs in the construction set and test set by MLR model are listed in Table III.

TABLE III. The conductivities of ILs predicted by the MLR and ANN models

Entry	T / K	κ_{exp} / S m^{-1}	κ_{MLR} / S m^{-1}	κ_{ANN} / S m^{-1}	ΔD (MLR)	ΔD (ANN)
1	298.10	0.33	–0.33	0.25	0.66	0.08
2	368.15	1.825	2.718	1.853	0.893	0.028
3	348.10	0.926	2.395	0.933	1.469	0.007
4	298.10	0.15	–0.49	0.15	0.64	0.00
5	298.10	0.21	–0.34	0.24	0.55	0.03
6	295.10	0.11	–0.17	0.04	0.28	0.07
7	313.15	0.06553	0.43389	0.06880	0.36836	0.00327
8	368.15	2.16	3.41	2.21	1.25	0.05
9	313.15	1.374	1.335	1.364	0.039	0.010
10	343.10	1.747	2.717	1.735	0.97	0.012
11	298.10	0.04	–0.42	0.18	0.46	0.14
12	298.10	0.067	–0.426	0.169	0.493	0.102
13	298.10	0.59	0.84	0.59	0.25	0.00
14	298.10	0.016	–0.432	0.164	0.448	0.148
15	298.10	0.0017	0.5767	0.1352	0.5750	0.1335
16	295.10	0.13	–0.53	0.14	0.66	0.01
17	298.10	0.28	–0.39	0.20	0.67	0.08
18	358.15	4.69	3.96	4.70	0.73	0.01
19	348.10	2.00	2.50	1.94	0.50	0.06
20	298.10	0.043	–0.404	0.187	0.447	0.144
21	313.15	0.3964	0.1355	0.3789	0.2609	0.0175
22	313.15	0.6575	0.2396	0.5115	0.4179	0.1460
23	298.10	0.033	–0.431	0.165	0.464	0.132
24	298.10	0.016	–0.433	0.163	0.449	0.147
25	298.10	0.16	–0.35	0.23	0.51	0.07
26	308.15	0.504	1.581	0.469	1.077	0.035
27	348.1	0.512	2.370	0.740	1.858	0.228
28	298.1	0.035	–0.422	0.172	0.457	0.137

TABLE III. Continued

Entry	T / K	κ_{exp} / S m^{-1}	κ_{MLR} / S m^{-1}	κ_{ANN} / S m^{-1}	ΔD (MLR)	ΔD (ANN)
29	298.1	0.921	–0.038	0.381	0.959	0.540
30	298.1	0.051	–0.429	0.167	0.480	0.116
31	298.1	0.013	–0.434	0.162	0.447	0.149
32	298.15	2.9281	1.9481	2.9390	0.9800	0.0109
33	328.2	0.626	2.086	0.688	1.460	0.062
34	313.15	0.2465	0.1105	0.3437	0.1360	0.0972
35	358.1	6.2	3.2	4.7	3.0	1.5

The results of the construction set reflected the fitting capability of the models. The remaining data, which did not construct the model, were collected as the test set to evaluate the prediction capability of the related models. For MLR model, the predicted conductivity of ILs in test set was calculated according to Eq. (1). The results in Table III suggested a minimum absolute error of 0.039, a maximum absolute error of 1.469 for the construction set, and a minimum error of 0.136 and a maximum error of 3.0 for the test set. The differences between the actual and estimated values were measured by the root mean squared error (*RMSE*), which is calculated as:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\left(y_i^{exp} - y_i^{cal}\right)^2}{n}} \quad (2)$$

where i represents the i^{th} sample, y_i^{exp} is the experimental values, and y_i^{cal} is the predicted value by the model, n is the number of samples in the data set.[37] The *RMSE* of MLR model was 0.674 for construction set and 1.380 for test set, respectively. Fig. 3 shows the plots of the calculated electric conductivities by MLR model *versus* the experimental ones, with R^2 = 0.8044 for construction set and 0.5264 for test set. The fitting and prediction capabilities for the same kind of ionic liquid at different temperatures of MLR model were indicated by the *RMSE* of each kind of ILs in construction set and test set, respectively (Table IV).

The values of *RMSE* and R^2 for the MLR model suggested that the fitting and prediction capability of the MLR model were not good, especially of the latter. This means that the relationship between the structure and the conductivity of ionic liquids is not perfectly linear. However, compared with other methods used for the selection of descriptors, the MLR method is always faster and easier to implement.

BP ANN study

For the training, validation and test data sets, their ANN models were established by BP ANN analysis based on their descriptors obtained by the MLR model. The employed neural network in this modeling problem had four input

nodes, corresponding to the four operating variables, *i.e.*, $E_{\text{HOMO-anion}}$, μ_{anion}, S and T^2. The experimental data of the conductivity was the output node of the BP ANN. After optimization, a three-layer BP ANN was set up. The number of neurons in the hidden layers was 16, and the learning rate was 0.01. A log-sigmoid transfer function within the hidden layer and a linear transfer function within the output layer were used for the ANN model. According to the literature,[36,38] a validation set was employed to avoid over-fitting and optimize the weight value in this study. As the initial weights were random numbers produced by the computer, the weights may be converged on a local optimal. In order to avoid local optimal results, the ANN was calculated many times with different initial weights to obtain the global optimal result. Finally, the obtained ANN model was selected as a prediction model of the conductivity of ILs, which achieved the goal of training set and had good performance for validation set.

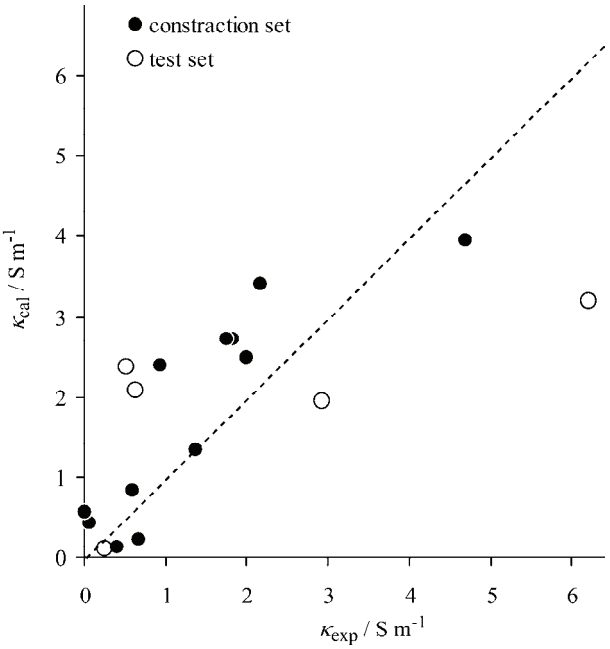

Fig. 3. The plots of the electric conductivities calculated by the MLR model *vs*. the experimental ones, the dotted line represents $y = x$.

Similar to the MLR model, the ANN prediction model was evaluated from two aspects. The experimental values and calculated values of conductivity by ANN model for different ionic liquids are given in Table III, showing that the minimum errors of the training set, validation set and test set achieved by the ANN model were 0.00, 0.0175 and 0.0109, respectively. The maximum error of the training set, validation set and test set were 0.148, 0.147 and 1.5, respectively. The *RMSE* values of the training set, validation set and test set obtained the ANN model were 0.072, 0.107 and 0.544, respectively. Furthermore, plots of the elec-

tric conductivities calculated by the ANN model *vs*. the experimental ones are shown in Fig. 4, with R^2 values of 0.9966, 0.9803 and 0.9703 for the training set, validation set and test set, respectively.

TABLE IV. The *RMSE* of the ionic liquids by MLR and ANN models

Entry	*RMSE* (MLR)	*RMSE* (ANN)	Data points
1	0.795	0.106	10
2	0.613	0.017	21
3	1.324	0.013	24
4	0.914	0.011	10
5	0.800	0.120	10
6	0.282	0.073	1
7	0.859	0.007	17
8	0.946	0.055	21
9	0.297	0.012	12
10	0.632	0.029	36
11	0.455	0.138	1
12	0.493	0.102	1
13	0.246	0.003	1
14	0.448	0.148	1
15	0.575	0.134	1
16	0.660	0.015	1
17	0.832	0.069	10
18	2.574	0.066	23
19	1.111	0.089	24
20	0.447	0.144	1
21	0.778	0.088	12
22	0.704	0.147	12
23	0.464	0.132	1
24	0.449	0.147	1
25	0.510	0.075	1
26	1.042	0.058	13
27	2.234	0.610	24
28	0.457	0.137	1
29	1.154	0.808	12
30	0.480	0.116	1
31	0.447	0.149	1
32	2.053	0.866	13
33	1.387	0.087	11
34	0.896	0.303	12
35	5.338	1.762	23

Similarly, the fitting and prediction capabilities for the same ionic liquids at different temperatures are given in Table IV. It could be concluded from Table III and Fig. 4 that the conductivities of the ionic liquids were mostly well predicted by the ANN model, and the results proved the good fitting and prediction capabilities of the constructed ANN model for these samples. Compared with the MLR

model, the ANN model had larger R^2 and smaller *RMSE* values than those of the MLR model. Moreover, the *RMSE* of the ionic liquids calculated by the ANN in Table IV were all smaller than those calculated by the MLR model. Based on above analysis, the prediction precision of this ANN model was high, and better than that of MLR model. In addition, the descriptors selected by the MLR model as the input node of the ANN model performed well in fitting and predicting the experimental data.

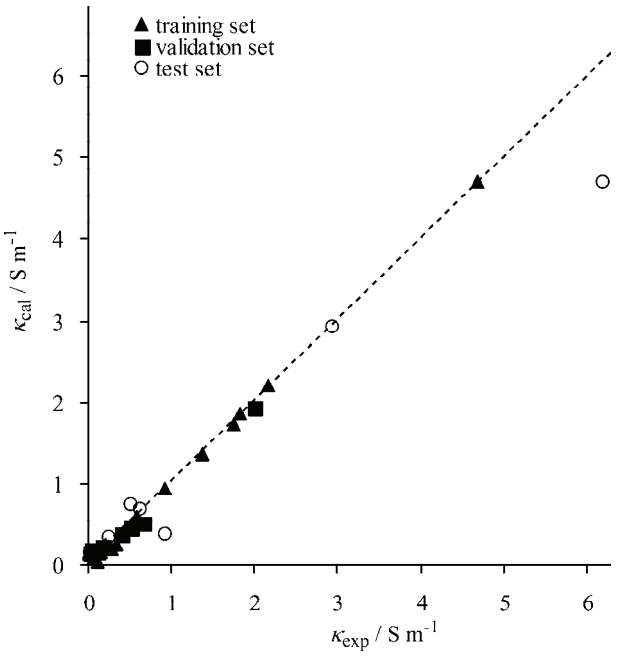

Fig. 4. The plots of the electric conductivities calculated by the ANN model *vs*. the experimental ones, the dotted line represents $y = x$.

On the other hand, both Tables III and IV suggested that the results for the test set were worse than those for the construction set. It was discussed in MLR study section that the ANN model had a much stronger fitting capability and better prediction precision than the MLR model. However, the results showed that some ionic liquids in the test set were not predicted well by ANN. For instance, the ionic liquid **29** was not predicted well, so all the ionic liquids with the same structural type as **29** were analyzed. The number of data points for this type of IL was 49, but only 13 data were in training test. In addition, the conductivity of ionic liquid **4** with ten data points in training set was very different from the others. For example, its conductivity was over 4 S m^{-1} at 373.15 K, while the others were all smaller than 3 S m^{-1}. Above all, the lack of experimental data for the training set may be the reason for the large deviation of IL **29**. Moreover, in the region of ionic conductivities lower than 0.1 S m^{-1}, a decrease in the prediction accuracy was found. It was noteworthy that the same phenomenon also

emerged in another study.[13] However, compared with the results in the previous study, the range of prediction accuracy was extended from 0.6 to 0.1 S m^{-1} in the present model. The ANN model was proved to be a relatively accurate prediction model and much better than the MLR model.

Analysis of the variables

The four descriptors employed in the MLR and ANN models were $E_{\text{HOMO-anion}}$, μ_{anion}, S and T^2. Multi-co-linearity between the four descriptors was detected by calculating their variation inflation factor (*VIF*) as:

$$VIF = \frac{1}{1-r^2} \tag{3}$$

where r is the correlation coefficient of the multiple regression equation between one descriptor and the others. If *VIF* equals one, no inter-correlation exists between each of the descriptors; if *VIF* is within the range 1.0–5.0, the corresponding model is acceptable; if *VIF* is larger than 10.0, the corresponding model is unstable.[37,39] The r^2 values for the four employed descriptors were 0.047, 0.164, 0.102 and 0.090, respectively. Accordingly, their *VIF* values were 1.049, 1.196, 1.136 and 1.099, respectively. Thus, the *VIF* values were less than five for each descriptor, which indicated that the constructed model was of obvious statistical significance.[37] To analyze the influence of descriptors on the EC values, the weights between input layer and hidden layer (shown in Table V) were studied.

The results given in Table V suggested that temperature, which is a thermodynamics variable, was the most important influencing factor for the *EC* of the ILs. Except temperature, the other three descriptors reflected the relationship between the structural characteristics and the *EC* of the ILs. According to previous analysis presented above, ${}^0X_{\text{cation}}$ is a good *S* function of *EC* at 298.1 K. Among the three variables, *S* founction of ${}^0X_{\text{cation}}$, which is related to the structural character of the cation, had the largest total weight value. ${}^0X_{\text{cation}}$ is the molecular connectivity index of a cation and it could represent information of structural characteristics containing size, number of valence electrons and the number of hydrogen atom attached to non-hydrogen atoms in the molecule.[40] The meaning of ${}^0X_{\text{cation}}$ and its total weight value analysis suggested that *EC* was related to the structural type of the cation, which was also proved by Matsuda *et al.*[13] in another model. μ_{anion} and $E_{\text{HOMO-anion}}$ are both related to the activity of anion. Affirmatively, different E_{HOMO}–E_{LUMO} gaps will lead to different conductivities. If the HOMO energy is lower, the electron work function will increase and electron transfer becomes more difficult. Dipole moment can reflect polarizability and is closely related with the capacity to conduct electricity. This indicated that the *EC* of the ionic liquid in this study might be determined by the

kind of the cation to a great degree and simultaneously influenced by the activity characteristics of the anion.

TABLE V. The weights between input layer and hidden layer

Neurons No.	Weight (absolute value)			
	μ_{anion}	$E_{HOMO\text{-}anion}$	S	T^2
1	0.1045	0.1650	8.8672	5.5500
2	2.7061	4.9088	5.5977	5.0415
3	4.6532	4.1600	5.5167	6.2565
4	10.1419	1.8552	3.8925	2.5628
5	3.4325	6.9361	8.4247	3.7612
6	4.9101	0.5836	10.7555	4.5674
7	5.9070	5.4873	7.9757	0.4859
8	6.6151	0.5055	1.3238	4.8877
9	1.1565	0.5655	0.9835	7.2336
10	6.0981	8.4227	1.2909	5.1381
11	1.5735	1.6982	5.0830	10.6385
12	10.2083	2.4696	9.5344	1.4208
13	4.1750	0.6770	4.6322	10.1808
14	6.1998	3.5529	6.1921	6.4718
15	8.4252	7.4568	0.2939	0.8942
16	1.3254	6.6955	1.5020	8.7378
Total	77.6322	56.1397	81.8658	83.8286

CONCLUSIONS

In this study, linear and ANN models were established based on the MLR and BP ANN methods to fit and predict the conductivity of ILs. 364 experimental conductivity data of the ILs were used to establish the models and test their predictive capability. The ANN model with R^2 = 0.9966 and *RMSE* = 0.072 for training set showed good fitting capability. The predictive capability of the ANN model was also reflected by its R^2 = 0.9703 and *RMSE* = 0.544 for the test set. The two models were found stable by *VIF* analysis. The R^2 and *RMSE* values of two models proved the ANN model was better than the MLR model. The results indicated the non-linear modeling might be more rational for the prediction of conductivity. It is worth mentioning that the descriptors selected by MLR were suitable and significant to be the input nodes of the ANN model in this study. The weights between the input layer and hidden layer were also analyzed, which indicated the conductivity of ionic liquid in this study might be determined to a great degree by the kind of cation and influenced by the activity characteristics of the anion. In summary, the developed ANN model proved itself to be promising for the prediction of the conductivity of various kinds of ionic liquids at different temperatures.

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И З В О Д

ПРЕДВИЂАЊЕ ЕЛЕКТРИЧНЕ ПРОВОДЉИВОСТИ ЈОНСКИХ ТЕЧНОСТИ ДВЕМА ХЕМОМЕТРИЈСКИМ МЕТОДАМА

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У последњих неколико година постигнут је знатан напредак у проучавању особина јонских течности (ILs) и њихове структуре. Од обичних физичко–хемијских особина чистих ILs, електрична проводљивост (EC) је од битне важности како са практичног, тако и са фундаменталног аспекта. Да бисмо развили ефикасне моделе за предвиђање EC разних ILs, испитана је зависност између разних структурних дескриптора и EC за 35 IL на различитим температурама. То је учињено помоћу мултилинеарне регресије (MLR) и методом неуронских мрежа (ANN). Нађено је да је MLR модел одређен тростепеном ANN методом са 4 променљиве био успешан. Јонска проводљивост одређена ANN моделом има висок коефицијент корелације и малу средњу квадратну грешку и добро се слаже са експерименталним вредностима. ANN модел се показао бољим од MLR модела.

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