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Prediction of glass structure by using multiple regression analysis

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Multiple regression analyses were applied to the prediction of glass structures, such as O 1s binding energy and fraction of four-fold coordinated boron atoms, N_4. In the case of linear combination of the content of glass constituents, an acceptable prediction accuracy was obtained for O 1s binding energy, and as for N_4, however, a poor agreement was observed between the prediction and measurement. After introducing quadratic and cubic interaction terms into the regression formula, a drastic improvement was achieved in the prediction of N_4. Some regression coefficients were dependent on basicity of each glass constituent, suggesting the feasibility of prediction for the glasses containing novel constituents whose regression parameters have never been determined.

Key-words : Glass, Structure, Database, Regression analysis, XPS, NMR

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1. Introduction

Commercial glasses have complicated compositions, and the development of new glasses has been empirically done by repetitions of trial and error. For the design of glass composition, the use of database must be effective. As for glass, two commercial databases, SciGlass[1)] and INTERGLAD[2)] are available. In glass, composition is continuously variable, and hence properties also change continuously along with the composition. Additivity rule[3)] has been used to predict the properties and structures of glass, which is based on the assumption that a property of a glass is expressed by a linear approximation connecting both ends of the glass composition. In some glass systems, such as borate glass, non-linear changes in properties against glass composition are seen, and applicability of the additivity rule is limited. Both Sciglass and INTERGLAD provide predictions of properties, and in INTERGLAD, a multiple regression analysis incorporating a cross-product regression is available, which enables higher accuracy in the prediction of properties with complicated change.

Originally, properties of a material are dependent on its structure, and the property prediction with high accuracy is therefore expected if not only the relationships between composition and property but also trilateral relationships among composition, structure and property are clarified. In the latest version of INTERGLAD released in 2009, a structure database was added,[4)] in which structural information collected by various analytical methods, such as infrared, Raman, NMR, etc. was included. In the current edition, however, prediction of glass structure is not provided. Then, in the present study, the multiple regression analysis provided for the prediction of glass properties has been applied to obtain correlating equations between glass composition and structures.

In this paper, O 1s binding energy measured with X-ray photoelectron spectroscopy (XPS) and fraction of four-fold coordinated boron atoms (N_4) determined from ^{11}B MAS-NMR spectrometry were chosen for the regression analyses. O 1s binding energy changes according to the local structure around oxygen, that is, kind and distance of the neighboring atoms, and information for coordination state and nature of chemical bonds is also obtained. The authors' research group has proposed a prediction expression of O 1s binding energy in alkali binary oxide glasses,[5)] and in the glasses with more than three constituents, however, prediction expression has never been proposed. As for N_4, Dell et al. have proposed a prediction formula,[6)] and the applicability is however limited in borosilicate glasses. When the prediction of structure in multi-component glasses is enabled, it is expected to contribute the development in compositional design of commercial glasses. Statistical analysis such as multiple regression analysis is suitable for the commercial glasses consisting of a number of components. In the present study, INTERGLAD was chosen as a database, and multiple regression analyses were performed for the predictions of O 1s binding energy and N_4.

2. Experimental

The structural data that have been collected in the authors' group were registered in INTERGLAD.[7)] As for XPS, the registered data are as follows: date of measurement, name of operator, measurement conditions, glass composition, preparative conditions, XPS spectrum of whole energy region (wide scan data), XPS core spectra of glass constituents such as O 1s, Si 2p, Na 1s, C 1s, etc., results of peak separation such as peak position, width, relative intensity of peak components, and XPS valence band spectrum. As for ^{11}B MAS-NMR, NMR spectrum and results of peak separation were registered as well as NMR measurement conditions and glass preparing conditions. Details of the XPS and NMR measurements are given in the literatures.[8),9)]

Multiple regression analyses provided in the current edition of INTERGLAD are available only for the data stored in property database. Then, the registered data in structure database were re-registered in property database, and the regression analyses were performed. Regression expression which can be used in INTERGLAD is given in Eq. (1).

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$$\begin{cases} y = \sum_i a_i x_i + \sum_{i<j} a_{ij} x_i x_j + \sum_{i<j<k} a_{ijk} x_i x_j x_k + a_{\text{other}} x_{\text{othre}} \\ x_{\text{othre}} = 1 - \sum_i x_i \end{cases} \quad (1)$$

where y is the dependent variable, that is, the predicted values of O1s binding energy or N_4, x_i is the i-th independent variable, that is, content of i-th component, and a_i is the regression coefficient for x_i. The second and third terms are introduced to reproduce the interactions between the independent variables, and the quadratic $x_i x_j$ and cubic $x_i x_j x_k$ interactions are treated as independent variables. The last term represents the contribution of minor components. In the multiple regression analysis, the regression coefficients of a_i, a_{ij}, a_{ijk} and a_{other} are optimized. In the present study, the structural data of the glasses given in **Table 1** were used in the regression analyses, in which no glass component was regarded as negligible minor component, and hence $x_{\text{other}} = 0$ and a_{other} was not evaluated in the regression analyses.

Explanatory variables which have less contribution to the dependent variable should be removed in regression analyses, and the choice of the explanatory variables is done according to t-values. The t-value is given by dividing the regression coefficient by standard error, which is determined for each explanatory variable. In general, an explanatory variable with $|t| < 2.0$ is removed, and hence the same condition is applied in the present study.

3. Results

Tables 2 and **3** show the optimal regression coefficients, a_i, a_{ij} and a_{ijk} in Eq. (1) obtained from the multiple regression analyses for O 1s binding energy of 487 compositional glasses and the fraction of four-fold coordinated boron atoms, N_4 obtained from NMR measurements of 317 glasses, respectively. Comparing the regression coefficients between the regression models, larger change is seen in a_i for N_4, suggesting the larger interaction between the glass constituents for the formation of four-fold boron atoms.

Figure 1 shows the results of multiple regression analyses of O 1s binding energy, where Fig. 1(a) was obtained by assuming the linear relationship expressed by the first and fourth terms in Eq. (1), and Figs. 1(b) and 1(c) were derived by adding the quadratic and cubic interactions of the explanatory variables, that is, the second and third terms in Eq. (1), respectively. In the horizontal axis, the experimentally measured values of O 1s binding energy are plotted. In silicate and phosphate glasses, O 1s spectrum can be separated into the components of bridging oxygen and non-bridging oxygen. In this study, the center of gravity of O 1s spectrum was chosen as experimental value of O 1s binding energy. In the vertical axis, the predictive values calculated from Eq. (1) are illustrated. The straight lines indicated in Fig. 1 are drawn connecting the points of equal values between experiment and prediction. The points plotted are localized close to the lines. The coefficient of determination, R^2 increases along with the addition of quadratic and cubic interactions, and the difference in the coefficients is, however, not so large.

Figure 2 illustrates the results of multiple regression analyses of the fraction of four-fold coordinated boron atoms, N_4 obtained from NMR measurements. In the linear regression model neglecting the interactions between the explanatory variables, the dispersion of the plotted points is quite large, and the predictive N_4 value reaches a plateau at around 50% as seen in Fig. 2(a). In the quadratic regression model, R^2 increases drastically from the case of linear regression model. As shown in Table 3, the drastic change is also confirmed in a_i, where some a_i become negative, suggesting the larger contribution of quadratic and cubic interactions between the glass constituents.

4. Discussion

As is obvious from Eq. (1), the regression coefficient a_i in the linear independent term of a glass constituent should be ideally equal to the measured value of the pure glass of respective component such as SiO_2 and B_2O_3. Definitely, the values of a_i optimized for O 1s binding energy listed in Table 2 are close to the measured values. For example, the experimental O 1s binding energies of pure B_2O_3 and SiO_2 glasses are 533.3[10] and 532.6[8] eV, respectively. In the case of N_4, however, the meaning of a_i is slightly different. Pure SiO_2 glass contains no B atoms, and hence N_4 is not measurable. If we have to define, a_i for N_4 means the N_4 value of a pure glass containing trace amount of boron. As shown in Table 3, however, some of a_i optimized for N_4 are negative, which does not meet this definition. In the case of B_2O_3, $a_{B_2O_3}$ for N_4 must be ideally equal to the experimental N_4, that is, 0. As shown in Table 3, however, the optimal values of $a_{B_2O_3}$ are 24.72, −37.69 and −27.32 for the linear, quadratic and cubic regression models, respectively, which are far from the ideal value of 0. In the current version of INTERGLAD, it is not allowed to use arbitrary values of regression coefficients, such as $a_{B_2O_3} = 0$. In the future version, the use of fixed values of regression coefficients is expected for the cases with trivial regression coefficients.

Thus, the interpretation of the regression coefficients is quite difficult. However, if we can find some trend in the regression coefficients, it can be used as a criterion of validity. The authors' research group has proposed that the O 1s binding energy can be used as a measure of basicity of oxide glasses.[5] It is hence supposed that glass constituents with higher acidity have larger a_i values and those with higher basicity indicate smaller a_i values. Then, the relations between the regression coefficients and basicity were examined, where basicity Λ of an oxide was calculated from the Pauling electronegativity χ by using the following equation.[11]

$$\Lambda = \frac{1}{1.36(\chi - 0.26)} \quad (2)$$

Figure 3 shows the correlations between the regression coefficients for O 1s binding energy and basicity of the glass constituents. As shown in Fig. 3(a), the regression coefficient a_i in the linear regression model decreases with increasing the basicity Λ. As also reported in the previous study,[5] TeO_2 and Bi_2O_3-containing glasses have lower O 1s binding energy than expected from basicity. In Fig. 3(a), the similar result is also confirmed for PbO, TiO_2 and La_2O_3 besides TeO_2 and Bi_2O_3.

As for La_2O_3, a large increase in a_i is observed after adding quadratic and cubic interaction terms, suggesting the large contribution of the additional terms in the prediction of O 1s binding energy. The contribution of quadratic and cubic interaction terms is reflected by the magnitude of regression coefficients, a_{ij} and a_{ijk}. The coefficients a_{ij} and a_{ijk} associated with La_2O_3 in the cubic regression model are ca. −30 and −45, respectively, which are quite larger among the glass constituents.

The correlations between the regression coefficients for N_4 and basicity are given in **Fig. 4**. The coefficient a_i in the linear regression model is almost constant at around 50, and however, only Al_2O_3 indicates negative coefficient of −97. It is noted that the coefficient a_i in the quadratic and cubic regression models decreases with increasing the basicity Λ, and the coefficients

Table 1. Composition of the glasses provided for the regression analyses

Glass	Composition (molar ratio)
(a) O 1s binding energy determined by XPS	
x_1 $M_2O{\cdot}(1-x_1)$ P_2O_5	x_1 = Li: 0.15–0.5, Na: 0.1–0.6, K: 0.15–0.5
x_1 $CaO{\cdot}(1-x_1)$ P_2O_5	x_1 = 0.2–0.5
x_1 $Na_2O{\cdot}0.1$ $Al_2O_3{\cdot}(0.9-x_1)$ P_2O_5 TeO_2	x_1 = 0.25–0.55
x_1 $M_2O{\cdot}(1-x_1)$ TeO_2	x_1 = Li: 0.15–0.3, Na: 0.1–0.35, K: 0.1–0.25, Rb: 0.1–0.2, Cs: 0.05–0.13
x_1 $WO_3{\cdot}(1-x_1)$ TeO_2	x_1 = 0.1–0.3
0.1 $La_2O_3{\cdot}x_2$ $WO_3{\cdot}(0.9-x_2)$ TeO_2	x_2 = 0.1–0.5
x_1 $K_2O{\cdot}x_2$ $WO_3{\cdot}(1-x_1-x_2)$ TeO_2	x_1 = 0.05–0.25, x_2 = 0.10–0.50
x_1 $Li_2O{\cdot}(0.3-x_1)$ $La_2O_3{\cdot}0.7$ TeO_2 B_2O_3	x_1 = 0.18–0.28
x_1 $M_2O{\cdot}(1-x_1)$ B_2O_3	x_1 = Li: 0.05–0.54, Na: 0.05–0.41, K: 0.05–0.35, Rb: 0.06–0.40, Cs: 0.05–0.40
x_1 $MO{\cdot}(1-x_1)$ B_2O_3	x_1 = Ca: 0.28–0.43, Sr: 0.24–0.44, Ba: 0.20–0.43, Pb: 0.67
x_1 $M_2O_3{\cdot}(1-x_1)$ B_2O_3	x_1 = Bi: 0.06–0.80, La: 0.25
x_1 $Na_2O{\cdot}x_2$ $Al_2O_3{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.2–0.5, x_2 = 0.05–0.2
x_1 $CaO{\cdot}x_2$ $Al_2O_3{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.15–0.5, x_2 = 0.05–0.3
x_1 $Na_2O{\cdot}x_2$ $La_2O_3{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.05–0.23, x_2 = 0.05–0.2
x_1 $La_2O_3{\cdot}x_2$ $TeO_2{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.05–0.2, x_2 = 0.5–0.9
x_1 $Bi_2O_3{\cdot}(1-x_1)$ $TeO_2{\cdot}0.4$ B_2O_3	x_1 = 0.15–0.45
x_1 $Na_2O{\cdot}x_2$ $TiO_2{\cdot}(1-x_1-x_2)$ B_2O_3 0.05 $ZnO{\cdot}0.65$ $PbO{\cdot}0.3$ B_2O_3	x_1 = 0.27–0.33, x_2 = 0.04–0.27
x_1 $Li_2O{\cdot}0.2$ $BaO{\cdot}(0.8-x_1)$ Bi_2O_3 SiO_2	x_1 = 0.2–0.5
x_1 $M_2O{\cdot}(1-x_1)$ SiO_2	x_1 = Li: 0.33, Na: 0.15–0.50, K: 0.20–0.33, Rb: 0.33–0.50, Cs: 0.33, 0.333
x_1 $PbO{\cdot}(1-x_1)$ SiO_2	x_1 = 0.25–0.70
x_1 $Bi_2O_3{\cdot}(1-x_1)$ SiO_2	x_1 = 0.07–0.85
x_1 $Na_2O{\cdot}x_2$ $Al_2O_3{\cdot}(1-x_1-x_2)$ SiO_2	x_1 = 0.06–0.33, x_2 = 0.05–0.40
x_1 $Li_2O{\cdot}(0.33-x_1)Cs_2O{\cdot}0.67$ SiO_2	x_1 = 0.07–0.27
x_1 $M_2O{\cdot}x_2$ $TiO_2{\cdot}(1-x_1-x_2)$ SiO_2	x_1 = K: 0.22–0.33, x_2 = 0.33
	x_1 = Na: 0.20–0.44, x_2 = 0.04–0.40
x_1 $Na_2O{\cdot}(0.5-x_1)$ $ZnO{\cdot}0.5$ SiO_2 GeO_2	x_1 = 0.10–0.40
x_1 $M_2O{\cdot}(1-x_1)$ GeO_2	x_1 = Na: 0.05–0.4, K: 0.1–0.4
x_1 $PbO{\cdot}(1-x_1)$ GeO_2	x_1 = 0.05–0.65
x_1 $MO{\cdot}(1-x_1)$ Al_2O_3	x_1 = Ca: 0.6–0.7, Sr: 0.67–0.7
x_1 $B_2O_3{\cdot}(1-x_1)$ SiO_2	x_1 = 0.33–0.5
x_1 $M_2O{\cdot}x_2$ $B_2O_3{\cdot}(1-x_1-x_2)$ SiO_2	x_1 = Li: 0.23–0.5, x_2 = 0.08–0.51
	x_1 = Na: 0.07–0.6, x_2 = 0.03–0.64
	x_1 = K: 0.12–0.43, x_2 = 0.1–0.59
	x_1 = Cs: 0.2–0.45, x_2 = 0.17–0.53
(b) Fraction of four-fold coordinated boron atoms estimated from ^{11}B MAS-NMR	
x_1 $M_2O{\cdot}(1-x_1)$ B_2O_3	x_1 = Li: 0.09–0.34, Na: 0.17–0.38, K: 0.09–0.38
x_1 $Na_2O{\cdot}x_2$ $MO{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.04–0.28, x_2 = Ca: 0.04–0.28
	x_1 = 0.05–0.28, x_2 = Ba: 0.02–0.27
x_1 $CaO{\cdot}x_2$ $BaO{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = 0.06–0.31, x_2 = 0.07–0.38
x_1 $M_2O{\cdot}x_2$ $Al_2O_3{\cdot}(1-x_1-x_2)$ B_2O_3	x_1 = Li: 0.09–0.45, x_2 = 0.05–0.19
	x_1 = Na: 0.15–0.41, x_2 = 0.06–0.19
	x_1 = K: 0.09–0.41, x_2 = 0.06–0.19
x_1 $M_2O{\cdot}x_2$ $B_2O_3{\cdot}(1-x_1-x_2)$ SiO_2	x_1 = Li: 0.09–0.5, x_2 = 0.13–0.91
	x_1 = Na: 0.11–0.6, x_2 = 0.13–0.83
	x_1 = K: 0.09–0.43, x_2 = 0.14–0.91
x_1 $M_2O{\cdot}x_2$ $Al_2O_3{\cdot}x_3$ $B_2O_3{\cdot}(1-x_1-x_2-x_3)$ SiO_2	x_1 = Li: 0.09–0.5, x_2 = 0.05–0.19, x_3 = 0.11–0.91
	x_1 = Na: 0.11–0.5, x_2 = 0.06–0.19, x_3 = 0.11–0.83
	x_1 = K: 0.09–0.43, x_2 = 0.06–0.19, x_3 = 0.13–0.91
x_1 $Na_2O{\cdot}x_2$ $MO{\cdot}x_3$ $B_2O_3{\cdot}(1-x_1-x_2-x_3)$ SiO_2	x_1 = 0.04–0.38, x_2 = Ca: 0.03–0.35, x_3 = 0.13–0.91
	x_1 = 0.06–0.38, x_2 = Sr: 0.05–0.38, x_3 = 0.25–0.45
	x_1 = 0.05–0.38, x_2 = Ba: 0.03–0.38, x_3 = 0.13–0.91
x_1 $CaO{\cdot}x_2$ $BaO{\cdot}x_3$ $B_2O_3{\cdot}(1-x_1-x_2-x_3)$ SiO_2	x_1 = 0.06–0.38, x_2 = 0.07–0.38, x_3 = 0.13–0.77

Table 2. Regression coefficients of (a) linear (a_i), (b) quadratic (a_{ij}) and (c) cubic (a_{ijk}) terms obtained from, the linear, quadratic and cubic regression models for O 1s binding energy. Basicity, Λ calculated from Eq. (2) is also shown in the parenthesis

Constituents (Basicity, Λ)		Regression models		
		Linear	Quadratic	Cubic
(a) a_i in linear independent term				
P_2O_5	(0.400)	534.75	533.01	533.51
TeO_2	(0.400)	530.20	530.36	530.32
B_2O_3	(0.423)	532.83	533.19	533.29
Bi_2O_3	(0.448)	528.10	530.06	530.21
SiO_2	(0.477)	532.00	532.17	532.58
GeO_2	(0.477)	531.99	531.83	532.09
PbO	(0.477)	528.43	528.38	528.09
WO_3	(0.511)	531.22	531.81	530.96
ZnO	(0.549)	530.86	530.56	530.02
Al_2O_3	(0.593)	531.17	532.38	533.59
TiO_2	(0.593)	528.65	530.27	527.83
La_2O_3	(0.875)	523.93	545.84	544.13
Li_2O	(0.994)	528.68	528.11	528.10
CaO	(0.994)	528.62	528.07	527.74
SrO	(0.994)	529.05	528.27	527.90
Na_2O	(1.149)	528.28	528.29	530.79
BaO	(1.149)	529.05	528.18	527.99
K_2O	(1.362)	527.00	525.67	527.92
Rb_2O	(1.362)	527.02	526.42	526.06
Cs_2O	(1.671)	525.92	525.53	525.87
(b) a_{ij} in quadratic interaction term				
TeO_2	Bi_2O_3		−8.54	
B_2O_3	Bi_2O_3		−5.61	−6.55
Bi_2O_3	SiO_2		−6.33	−7.56
P_2O_5	Al_2O_3		−13.74	
B_2O_3	Al_2O_3		−3.43	−5.41
SiO_2	Al_2O_3			−11.71
TeO_2	La_2O_3		−29.10	−26.57
B_2O_3	La_2O_3		−28.59	−26.60
WO_3	La_2O_3		−33.22	
P_2O_5	Li_2O		6.88	5.39
P_2O_5	CaO		7.34	6.45
P_2O_5	Na_2O		6.53	
TeO_2	Na_2O			−4.23
B_2O_3	Na_2O		−2.18	−6.41
Bi_2O_3	BaO		−14.06	−14.57
SiO_2	Na_2O			−5.31
GeO_2	Na_2O			−5.24
Al_2O_3	Na_2O			−19.22
TiO_2	Na_2O		−6.35	
P_2O_5	K_2O		8.94	3.80
B_2O_3	K_2O			−2.70
SiO_2	K_2O			−2.60
GeO_2	K_2O		4.92	
Li_2O	BaO		12.46	12.74
SiO_2	Cs_2O			−3.94
Li_2O	Cs_2O		−18.16	
(c) a_{ijk} in cubic interaction term				
TeO_2	WO_3	La_2O_3		−45.23
B_2O_3	SiO_2	Li_2O		−9.24
B_2O_3	Al_2O_3	Na_2O		32.97
SiO_2	Al_2O_3	Na_2O		72.66
B_2O_3	SiO_2	K_2O		−20.50
(d) a_{other} in additional term other		—	—	—

Table 3. Regression coefficients in (a) linear (a_i), (b) quadratic (a_{ij}) and (c) cubic (a_{ijk}) terms obtained from, the linear, quadratic and cubic regression models for the fraction of four-fold coordinated boron atoms, N_4. Basicity, Λ calculated from Eq. (2) is also shown in the parenthesis.

Constituents (Basicity, Λ)		Regression models		
		Linear	Quadratic	Cubic
(a) a_i in linear independent term				
B_2O_3	(0.423)	24.72	−37.69	−27.32
SiO_2	(0.477)	59.61	39.58	87.16
Al_2O_3	(0.593)	−97.42	−108.97	−254.40
Li_2O	(0.994)	40.85	−206.75	−152.55
CaO	(0.994)	27.12	−98.56	−48.19
SrO	(0.994)	43.81	111.06	108.55
Na_2O	(1.149)	42.93	−286.88	−262.77
BaO	(1.149)	48.61	−188.55	−167.74
K_2O	(1.362)	36.74	−333.81	−286.39
(b) a_{ij} in quadratic interaction term				
B_2O_3	SiO_2			−184.03
SiO_2	Al_2O_3		−230.44	
B_2O_3	Li_2O		593.08	459.68
B_2O_3	CaO		418.72	310.93
SiO_2	Li_2O		474.16	248.44
SiO_2	CaO		218.11	
B_2O_3	Na_2O		710.64	632.60
B_2O_3	BaO		551.81	486.45
Al_2O_3	Li_2O		484.50	
SiO_2	Na_2O		617.74	426.57
SiO_2	BaO		436.65	262.42
Al_2O_3	Na_2O		624.37	593.46
B_2O_3	K_2O		776.82	659.55
SiO_2	K_2O		633.10	370.57
Al_2O_3	K_2O		670.14	575.73
Na_2O	BaO		81.91	89.37
(c)a_{ijk} in cubic interaction term				
B_2O_3	SiO_2	Li_2O		588.73
B_2O_3	SiO_2	CaO		521.15
B_2O_3	Al_2O_3	Li_2O		1815.57
B_2O_3	SiO_2	Na_2O		657.49
B_2O_3	SiO_2	BaO		562.03
SiO_2	Al_2O_3	Li_2O		804.79
B_2O_3	Al_2O_3	Na2O		1005.27
B_2O_3	SiO_2	K_2O		856.42
B_2O_3	Al_2O_3	K_2O		1061.53
(d) a_{other} in additional term other		—	—	—

become negative except for SiO_2 and SrO. As for the coefficient a_{ij}, most of the glass constituents are positive and widely distributed between 200 and 800. Negative a_{ij} values are obtained for the interactions between the network formers, B_2O_3, SiO_2 and Al_2O_3. Dependency of a_{ij} and a_{ijk} on the basicity is not confirmed in Figs. 4(b) and 4(c), and it is also the case for O 1s binding energy shown in Figs. 3(b) and 3(c). It is hence suggested that the prediction of a_{ij} and a_{ijk} based on basicity is difficult. In the case of a_i, however, dependency on basicity is recognized, and hence it is able to predict the coefficient a_i based on basicity. The authors' research group has reported that N_4 was predictable by using basicity instead of glass composition as an explanatory variable.[9] It is indicated from the present study that prediction of N_4 with high accuracy is also possible even in the case using glass composition as an explanatory variable, and the multiple regression analysis assuming the quadratic and cubic interactions are effective for the predictions of not only properties but also structure of glasses.

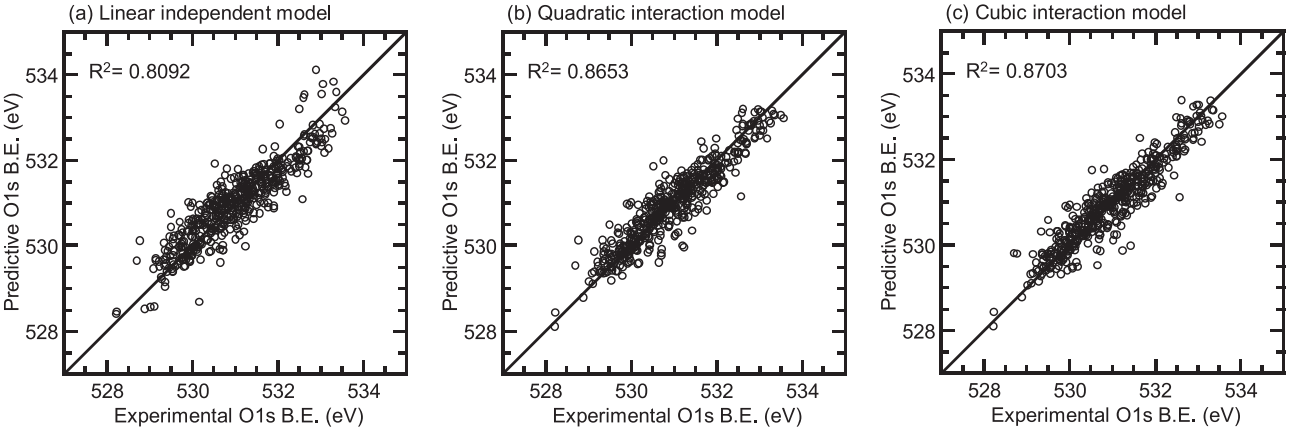

Fig. 1. Results of multiple regression analyses for O 1s binding energy (B.E.) by assuming (a) linear, (b) quadratic and (c) cubic regression models.

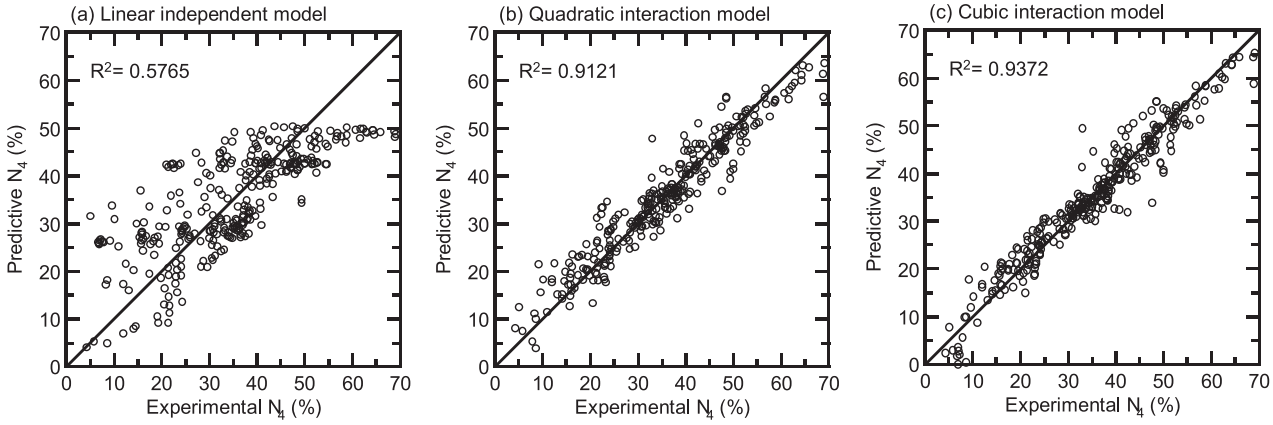

Fig. 2. Results of multiple regression analyses for the fraction of four-fold coordinated boron atoms, N_4 by assuming (a) linear, (b) quadratic and (c) cubic regression models.

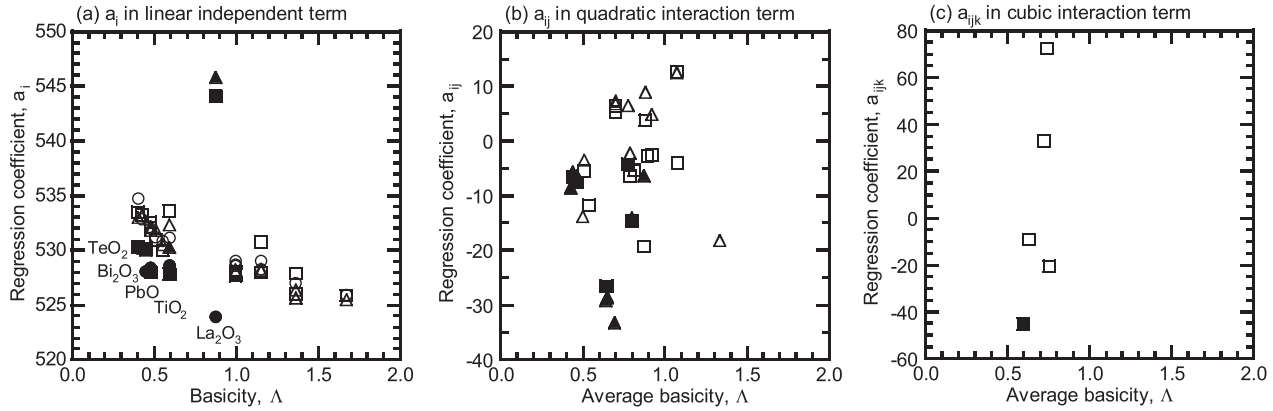

Fig. 3. Regression coefficients in (a) linear (a_i), (b) quadratic (a_{ij}) and (c) cubic (a_{ijk}) terms obtained from the linear (○), quadratic (△) and cubic (□) regression models for O 1s binding energy as a function of basicity. The coefficients concerning TeO_2, Bi_2O_3, PbO, TiO_2 and La_2O_3 are indicated by solid markers.

5. Conclusion

In this study, multiple regression analyses of glass structure were performed by using a commercial glass database system, INTERGLAD, and applicability to the prediction of glass structure was examined. O 1s binding energy measured by XPS and fraction of four-fold coordinated boron atoms, N_4 estimated from ^{11}B MAS-NMR measurement were chosen as glass structures. A regression formula assuming the interactions between the glass constituents was examined. As for N_4, drastic improvement in the prediction accuracy was observed after introducing the quadratic and cubic interaction terms into the regression formula. High effectiveness was also confirmed in the prediction of O 1s binding energy. The regression coefficients can

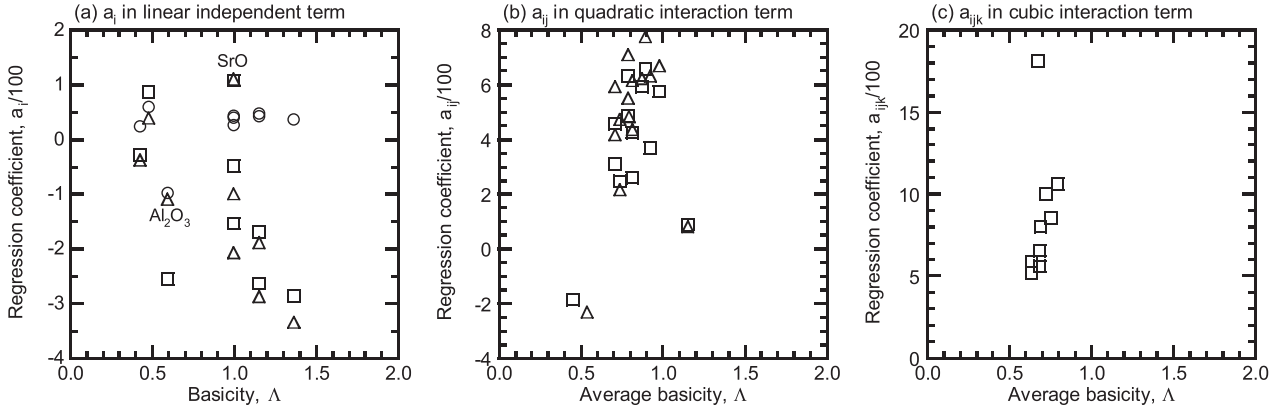

Fig. 4. Regression coefficients in (a) linear (a_i), (b) quadratic (a_{ij}) and (c) cubic (a_{ijk}) terms obtained from the linear (○), quadratic (△) and cubic (□) regression models for the fraction of four-fold coordinated boron atoms, N_4 as a function of basicity.

be empirically determined. If they are correlated to some numerical data, such as basicity, which are non-empirically derived, it is expected that the structure of glasses containing novel constituents will be predicted.

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