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Citation	ISIJ International. 2007, 47(8), p. 1075-1081
Version Type	VoR
URL	https://hdl.handle.net/11094/26404
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ISIJ International, Vol. 47 (2007), No. 8, pp. 1075–1081

Estimation of Surface Tension of Molten Silicates Using Neural Network Computation

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(*Received on December 20, 2006; accepted on May 28, 2007*)

Neural network computation was applied to the estimation of surface tension in ternary silicate melts. In addition, the criterion for designing the units in the middle layer of the layer-type neural network computation was discussed. It was found that the C_p-criterion modified by considering the degrees of freedom in the neural network computation was useful for determining the number of units in the middle layer, which gives an optimal estimation. The surface tension calculated by neural network computation using units determined by the C_p-criterion virtually reproduced the experimental data in molten ternary silicates with high precision.

KEY WORDS: surface tension; silicate; molten slag; neural network computation; estimation.

1. Introduction

Information on the surface tension of mould fluxes is indispensable to continuous-casting processes[1–3)] since the surface tension of mould fluxes, along with the related interfacial tension and wetting, affects slag infiltration into mould/strand channels and slag entrapment in liquid metal. The mould fluxes are usually silicates composed of SiO_2, CaO, MgO, Al_2O_3, Na_2O, K_2O and F with varying additions of B_2O_3, Li_2O, MnO, TiO_2 and ZrO_2. The composition of the mould flux changes according to the reactions with the elements, such as aluminum and titanium in molten steel during the continuous-casting process.[4–6)] Thus, the estimation of the surface tension composition dependence of the mould fluxes is one of the key technologies for improving the experimental process.

A number of models have been employed for estimation of the surface tension of molten slags, by means of the additivity rule,[7,8)] and by using models[9–16)] based on Butler's equation[17)] *etc*. In the former models,[7,8)] there is a poor reproducibility in multi-component systems, since the additivity rule is not reasonable in determining the surface tension of molten slag in these systems. In the latter models,[9–13)] with the exception of the model proposed by Tanaka and co-workers[14–16)] to address the ionic radii, the applicable systems are limited on account of the lack of information such as thermodynamic data. Although the Tanaka and co-workers' model is easily applicable to multi-component systems, and reproduces well the composition dependence of the surface tension in wide composition and temperature ranges, it is not a satisfactory model in the viewpoint of achieving highly accurate estimations.

On the other hand, new estimation methods for the characterization of materials using neural network computation have been proposed recently.[18–20)] In determining the physical properties of molten slags, Hanao and Tanaka *et al.*[20)] succeeded in precisely estimating the viscosity of mould fluxes using neural network computation, although such estimation had previously been considered quite difficult due to the abrupt and complicated viscosity changes with composition and temperature.

In the present work, neural network computation was applied to the estimation of the surface tension of ternary silicate melts consisting of SiO_2, CaO, Al_2O_3, FeO, MgO, MnO, Na_2O, K_2O, Li_2O, CaF_2 and B_2O_3. In addition, assessments of the number of units in the middle layer of layer-type neural network computation were discussed by using methods for determining the number of predictive variables in multiple linear regressions.

2. Calculation of Surface Tension of Silicate Melts with Neural Network Computation

Neural network computation is a processing method based on features known from the physiology of the human brain.[21,22)] The human brain is estimated to contain about 10 billion brain cells (neurons). When the sum of the intensity of input signals received by a neuron from other neurons exceeds a certain critical value, then new signals are transmitted to the next set of neurons as output signals. **Fig-**

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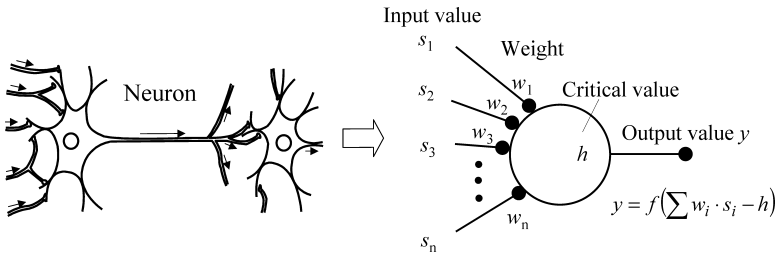

Fig. 1. Concept of a neuron model.

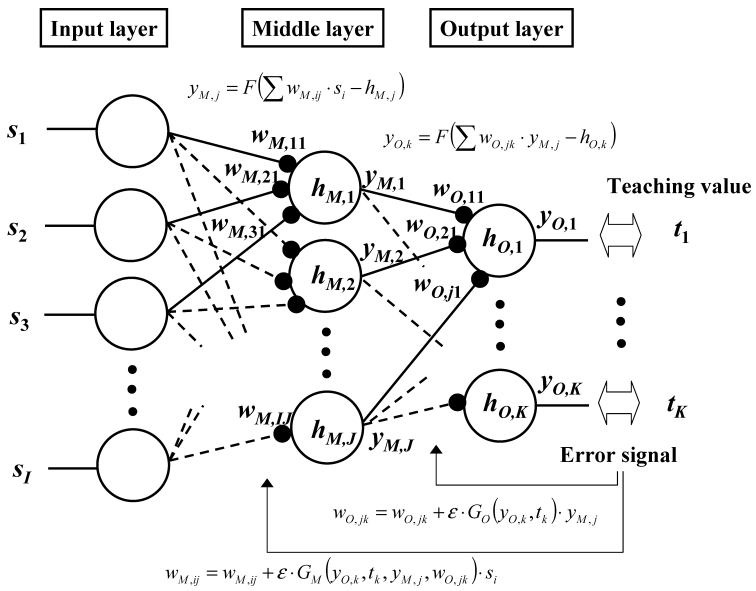

Fig. 2. Schematic diagram of the back-propagation approach in layer-type neural networks.

ure 1 shows this principle of neuron transmittance translated into a computational organization (neuron model). In this model, the situation corresponding to the transmission of signals is represented by a sigmoid function in Eq. (1):

$$f\left(\sum w_i \cdot s_i - h\right) = 1 \Big/ \left\{1 + \exp\left(-\left(\sum w_i \cdot s_i - h\right) \Big/ T\right)\right\} \quad \text{.....(1)}$$

where: s_i is an input value, w_j is a connection weight, h is a critical value and T is a coefficient that specifies the shape of the sigmoid curve.

Figure 2 shows a schematic diagram of the back propagation approach in the layer-type neural network computation. The layer-type neural network consists of three layers, an input layer, a middle layer and an output layer. The units in the middle layer are connected with the units in the input and output layers in the layer-type network structure. After input values have been applied to the units in the input layer, these values are propagated to the output layer through the middle layer. The output values are compared to the teaching values, and the errors are computed for each output unit. Then, these error signals are transmitted back from the output layer to each node between the layers in order to correct the connection weights, as well as correct the critical values in each unit. Here, the critical values are usually treated as one of the weights. These procedures are referred to as "learning". This learning process is repeated until the learning number reaches a targeted value or the error is reduced to an acceptable value. Consequently, a better network with adequate connection weights is constructed for predicting the teaching values from input values. In the present work, the teaching values used are measured surface tensions, and the input values are composed of the measurement temperature and the concentrations of the slag components. The learning process was conducted using the "Neurosim/L" software produced by Fujitsu Ltd.[22] under the following conditions: the number of units in the middle layer ranged from 1 to 12, the learning number was 300 000, and as far as the parameters related to the promotion of learning efficiency concerned, the initial values given in product were used. Then, the surface tensions of molten silicates were calculated using this network. Literature data[7,23–55] used for the silicate systems and temperatures in the present work are listed in **Table 1**. The total number of literature values was 467. These values mainly consisted of almost all of the available values in ternary silicate systems obtained from the Slag Atlas (2nd edition)[23] since the mould fluxes are made up of many kinds of components as described in the introduction. Here, it is assumed that all iron oxides exist as FeO.

3. Results and Discussion

For all systems, the average errors in the experimental values and calculated results obtained using neural network computation were plotted (closed circles, 1st learning process) against the number of units in the middle layer in **Fig. 3**. The average errors were evaluated using Eq. (2):

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Table 1. Literature data used for estimation of the surface tension of silicate melts.

System	Temperature (K)	Ref.
SiO_2-CaO, FeO, MgO, MnO, K_2O, Li_2O, Na_2O	1843	7
SiO_2-Al_2O_3-CaO	1873	23-25
SiO_2-Al_2O_3-FeO	1473, 1573	23, 26-29
SiO_2-Al_2O_3-MnO	1873	23, 30
SiO_2-CaO-FeO	1573, 1623, 1673	23, 31-39
SiO_2-CaO-MgO	1873	23, 40
SiO_2-CaO-MnO	1773	23, 41
SiO_2-FeO-MgO	1623, 1673, 1823	23, 38, 39, 42-44
SiO_2-FeO-MnO	1673	23, 35, 36
SiO_2-CaO-B_2O_3	1673	23, 45
SiO_2-CaO-Na_2O	1473, 1573, 1863	23, 46-49
SiO_2-CaO-CaF_2	1773	23, 50-55

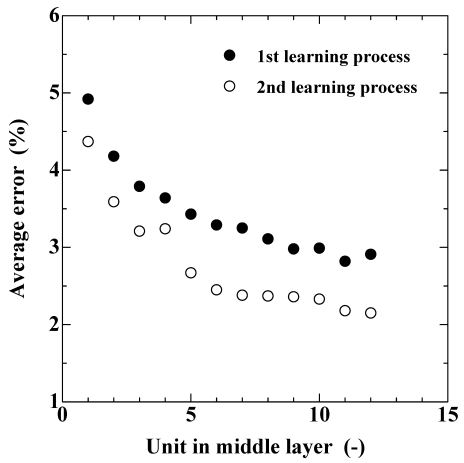

Fig. 3. Change in average error with respect to the number of units in the middle layer.

$$\text{Average error} = \frac{1}{N}\sum_{1}^{N}\left|\frac{\sigma_{\text{calc}}-\sigma_{\text{expe}}}{\sigma_{\text{expe}}}\right|\times 100\,(\%) \quad .....(2)$$

As shown in Eq. (2), the σ_{expe} and σ_{calc} values correspond to the experimental surface tension values found in the literature and the surface tension results calculated by neural network computation, respectively. N corresponds to the number of the literature data. The average error decreases to about 3% by increasing the number of units in the middle layer, as shown in **Fig. 3**. This result indicates that the estimation method for neural network computation can well reproduce the surface tension of molten ternary silicates in the high number of units in the middle layer.

It is advisable to discard the data with gross error from the viewpoint of conducting a preferable estimation as well as in case of common regression analysis, although the neural network computation succeeds in the estimation of the surface tension for silicate melts at the present stage, as shown in Fig. 3. For example, Hanao and Tanaka *et al.*[20] assessed the data by rejecting the upper 5% data points having the largest difference between the experimental and calculated values obtained by neural network computation after the first learning process in the estimation of viscosity for mould fluxes. In this study, the assessment of data was conducted after the 1st learning process by excluding the data with gross error, which is defined as follows:

(i) the data having an error of 10% and above[8] evaluated by Eq. (2)

(ii) the data satisfies (i) in the calculated results with more than 6 different units in the middle layer

16 data points fit the conditions (i) and (ii). The average errors between the experimental values and calculated results obtained by neural network computation without these data are shown as open circles (2nd learning process) in Fig. 3. Rejecting the data with gross error lowers the average errors in all units by about 1%, although the 3–5% errors obtained after the 1st learning process are already acceptable values in the sense of fine estimation. From these results, it was found that the assessment of the data was effective in the estimation of the surface tension in molten ternary silicates by neural network computation.

Here, the number of units in the middle layer has to be determined since the neural network computation with the largest number of units is not guaranteed to be optimal in any sense. For example, a large number of units merely improve the estimation within the experimental uncertainty of the data, although the higher number of units typically yields a better fit, as shown in Fig. 3. Then the following three criteria are applied: r^2-, r^2_{adj}- and C_p-criteria,[56] all commonly employed for determining the optimum number of variables in multi-linear regression, to the optimization of the number of units in the middle layer in neural network computation. These criteria are simply explained below. The r^2-criterion is given by:

$$r^2 = \sum_{1}^{N}(\sigma_{\text{calc}}-\bar{\sigma}_{\text{expe}})^2 \Big/ \sum_{1}^{N}(\sigma_{\text{expe}}-\bar{\sigma}_{\text{expe}})^2 \quad .....(3)$$

where $\bar{\sigma}_{\text{expe}}$ is the mean value of σ_{expe}. r^2 ranges between 0 and 1, with values closer to 1 representing better fits, and r^2 generally increases with adding more predictor variables in the linear regression model. The r^2_{adj}-criterion (adjusted r^2-criterion) is defined as:

$$r^2_{\text{adj}} = 1-\left\{\frac{N-1}{N-(p+1)}\right\}\cdot(1-r^2) \quad .....(4)$$

where p corresponds to the number of predictor valuables in the model form. This value can be regulated by comparing the contribution of the additional variables with the number of degrees of freedom. In practice, initially (for small values of p) r^2_{adj} increases and then begins to decrease as more variables are added to the model. The C_p-criterion based on Mallow's C_p-statistic[57] is expressed by:

$$C_p = \sum_{1}^{N}(\sigma_{\text{expe}}-\sigma_{\text{calc}})^2 \Big/ \sum_{1}^{N}(\sigma_{\text{expe}}-\sigma_{\text{calc}})^2\Bigg|_{p=k} \cdot\{N-(p+1)\}+2\cdot(p+1)-N \quad .....(5)$$

where k is the maximum value of p. This is a measure of

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the predictive ability of a fitted model. C_p first decreases and then increases with an increasing number of predictor variables in the model. The minimum of C_p corresponds to the optimum number of variables in the multi-linear regression.

The degrees of freedom in the neural network computation were taken into consideration in order to apply the r^2_{adj}- and C_p-criteria in the multi-linear regression to the neural network computation. Adding a unit to the middle layer increases the number of variables by {(the number of input values: n_I)+(the number of output values: n_O)+1} since the variables related to the units in the middle layer correspond to the weights between the input layer and the middle layer, the weights between the middle layer and the output layer and its own critical value in the unit. An output value, *i.e.* a unit in the output layer, also possesses its own critical value. These leave $N-\{(n_I+n_O+1)\cdot p'+n_O+1\}$ degrees of freedom for estimating validity in the neural network computation, where p' is the number of units in the middle layer. Then, Eqs. (6) and (7) are derived from Eqs. (4) and (5), respectively:

$$r^2_{adj}=1-\left\{\frac{N-1}{N-[(n_I+n_O+1)\cdot p'+n_O+1]}\right\}\cdot(1-r^2) \quad \text{.....(6)}$$

$$C_p=\sum_1^N(\sigma_{expe}-\sigma_{calc})^2\Big/\sum_1^N(\sigma_{expe}-\sigma_{calc})^2\Big|_{p=k}\cdot\{N-[(n_I+n_O+1)\cdot p'+n_O+1]\}+2\cdot\{(n_I+n_O+1)\cdot p'+n_O+1\}-N \quad \text{.....(7)}$$

In the present work, $n_I=12$ and $n_O=1$, since the input values correspond to the temperature and the concentrations of SiO_2, Al_2O_3, B_2O_3, CaF_2, CaO, FeO, K_2O, Li_2O, MgO, MnO, Na_2O, and the output value is the surface tension. **Figure 4** shows the relations between r^2 and r^2_{adj}, and the number of units in the middle layer. r^2 increases with increasing units, which simply means that including more units in the middle layer gives a better fit. On the other hand, r^2_{adj} is a roughly constant value around 0.9 above 5 units. It is considered that the optimum number of units exists in the range of constant r^2_{adj}. However, it is impossible to determine the number of units in the middle layer since there is no characteristic indication in the results of r^2_{adj} for the determination of single unit numbers. In **Fig. 5**, C_p is plotted against the number of units. This clearly shows that the minimum C_p for 6 units, that is to say, 6 is the optimum number of units in the middle layer in this evaluation using the modified C_p. Therefore, 6 was selected as the unit number in the middle layer for the present calculations.

The results calculated by neural network computation with 6 units in the middle layer are compared with the experimental data in **Fig. 6**. There are two kinds of plots in this figure: closed circles represent the calculated values after the 1st learning process, and open circles correspond to the calculated values after the 2nd learning process, after discarding the data with gross error. It was found that the data with large errors are reasonably removed by using the definition for the data with gross error as described above, and the experimental values can then be reproduced precisely by neural network computation. The average error of the calculated results after the 2nd learning process was 2.45%.

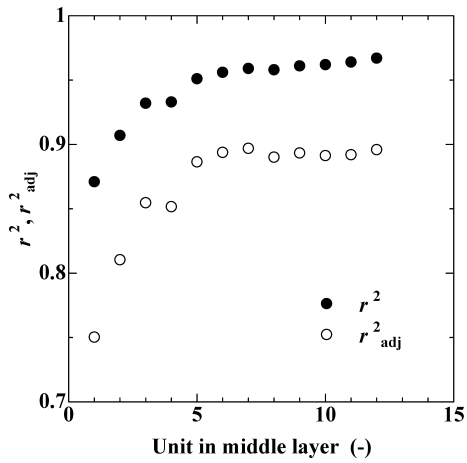

Fig. 4. r^2 and r^2_{adj} as a function of the units in the middle layer.

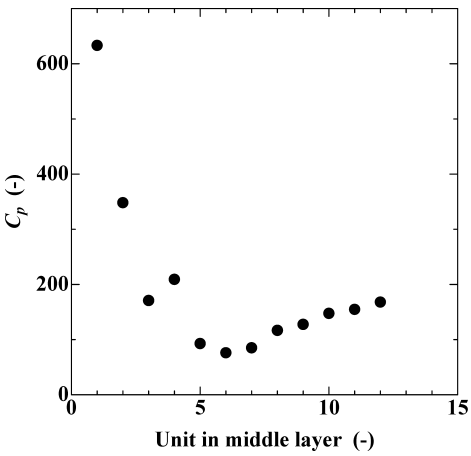

Fig. 5. C_p as a function of unit in middle layer.

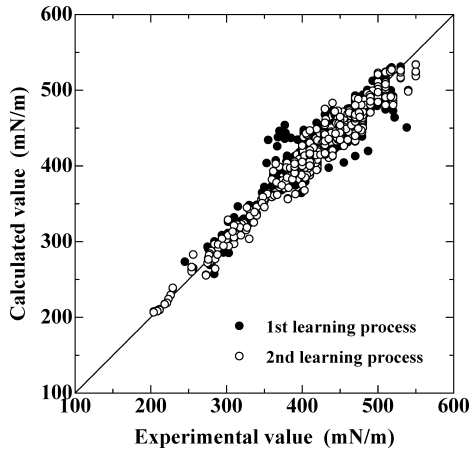

Fig. 6. Comparison between experimental values and calculated values obtained by using neural network computation with 6 units in the middle layer.

Some of the surface tension results for the ternary silicate melts calculated by neural network computation with 6 units in the middle layer after the 2nd learning process are shown in **Figures 7** through **11**. The data rejected before the 2nd learning process are expressed as numbers in parentheses in these figures. It can be seen that these data

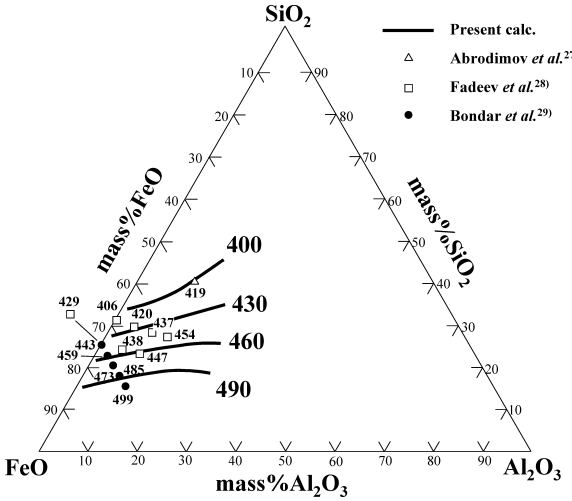

Fig. 7. Surface tension (mN/m) of molten SiO_2–Al_2O_3–FeO system at 1 573 K. Iso-surface tension curves were calculated by using neural network computation with 6 units in the middle layer.

points, with gross errors established on the basis of the definition described above, are also the data with large error in comparison with the other experimental data. The iso-surface tension curves obtained by neural network computation are in good agreement with the experimental data obtained in these systems. **Figures 12** through **14** show the results calculated using the neural network computation with 12 units in the middle layer after the 2nd learning process in SiO_2–FeO–CaO at 1 673 K, SiO_2–FeO–MgO at 1 623 K and SiO_2–CaO–CaF_2 at 1 773 K. These composition dependencies are similar to those obtained using the calculation results in the case where 6 units are present in the middle layer, as shown in Figs. 8(b), 9 and 11, while only a few differences are evident in some of the other areas. These results mean that the neural network computation with 6 units in the middle layer virtually reproduces the experimental data, while the neural network computation with 12 units in the middle layer gives a better fit in local regions. Therefore, the modified C_p derived by considering the number of degrees of freedom in the neural network computation is

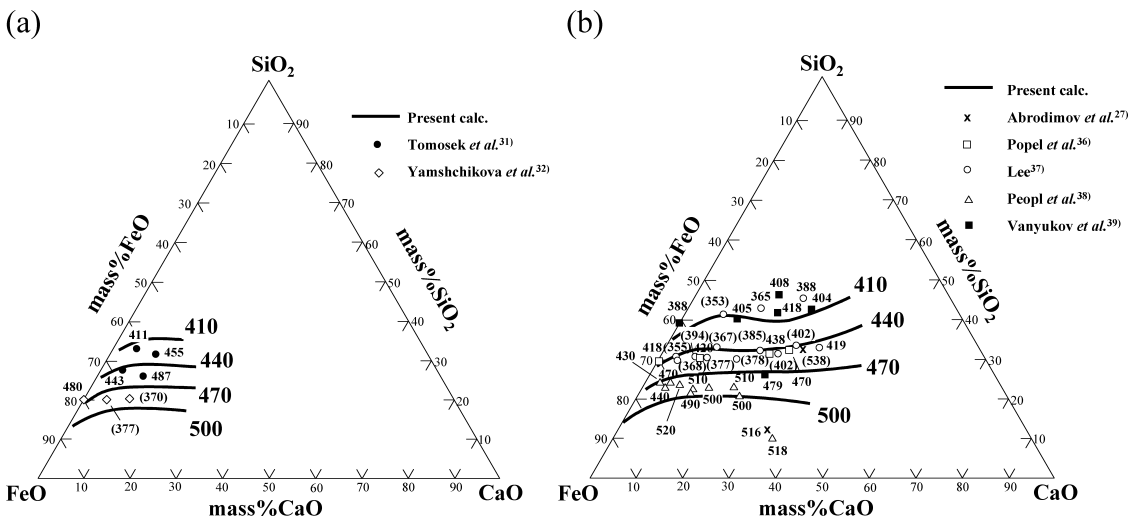

Fig. 8. Surface tension (mN/m) of molten SiO_2–CaO–FeO system at (a) 1 573 K and (b) 1 673 K. Iso-surface tension curves were calculated by using neural network computation with 6 units in the middle layer.

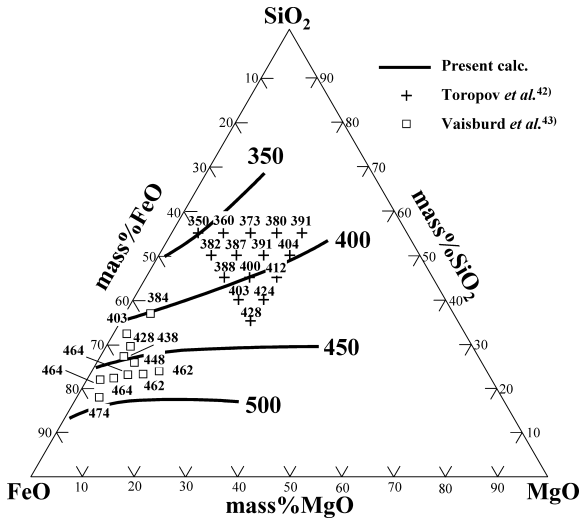

Fig. 9. Surface tension (mN/m) of molten SiO_2–FeO–MgO system at 1 623 K. Iso-surface tension curves were calculated by using neural network computation with 6 units in the middle layer.

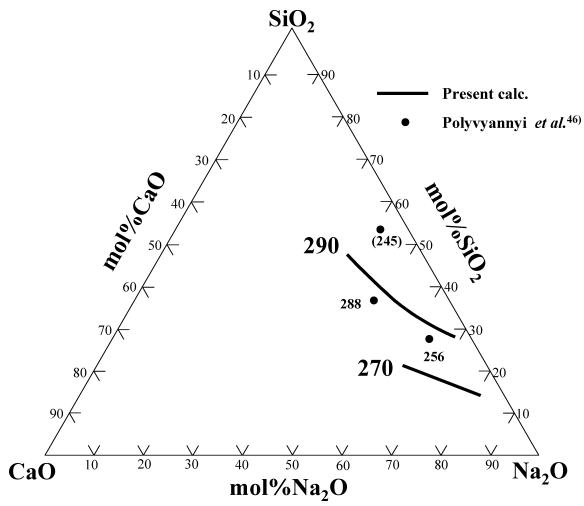

Fig. 10. Surface tension (mN/m) of molten SiO_2–CaO–Na_2O system at 1 473 K. Iso-surface tension curves were calculated by using neural network computation with 6 units in the middle layer.

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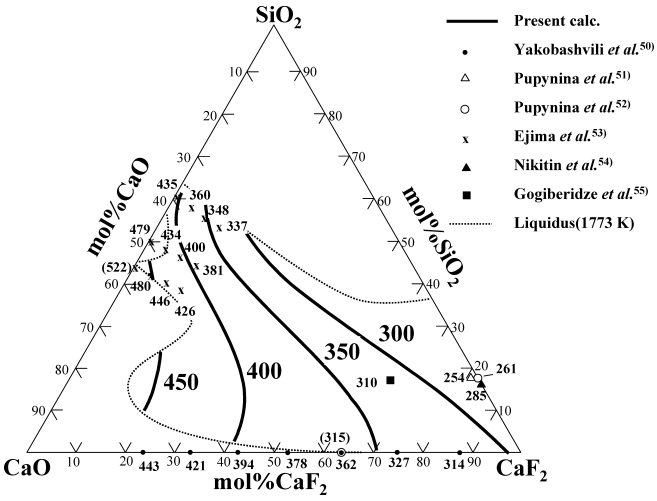

Fig. 11. Surface tension (mN/m) of molten SiO_2–CaO–CaF_2 system at 1 773 K. Iso-surface tension curves were calculated by using neural network computation with 6 units in the middle layer.

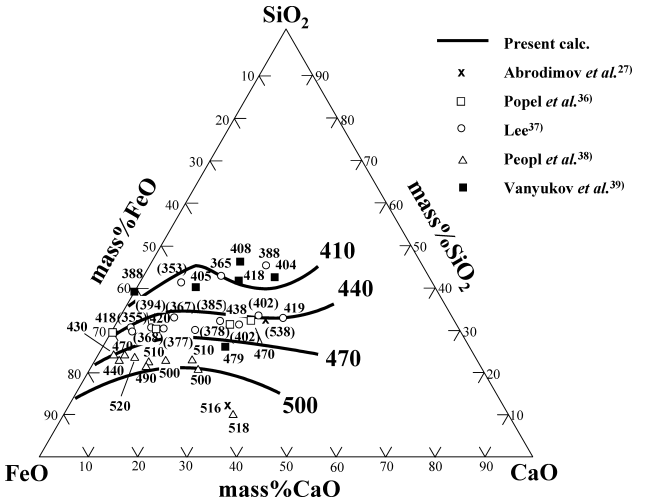

Fig. 12. Surface tension (mN/m) of molten SiO_2–CaO–FeO system at 1 673 K. Iso-surface tension curves were calculated by using neural network computation with 12 units in the middle layer.

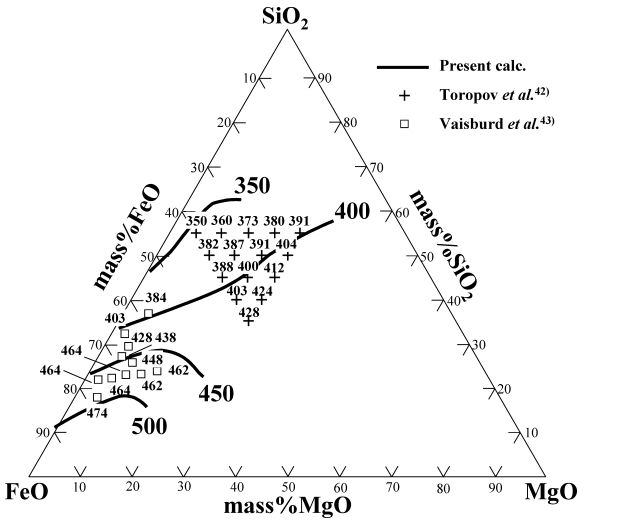

Fig. 13. Surface tension (mN/m) of molten SiO_2–FeO–MgO system at 1 623 K. Iso-surface tension curves were calculated by using neural network computation with 12 units in the middle layer.

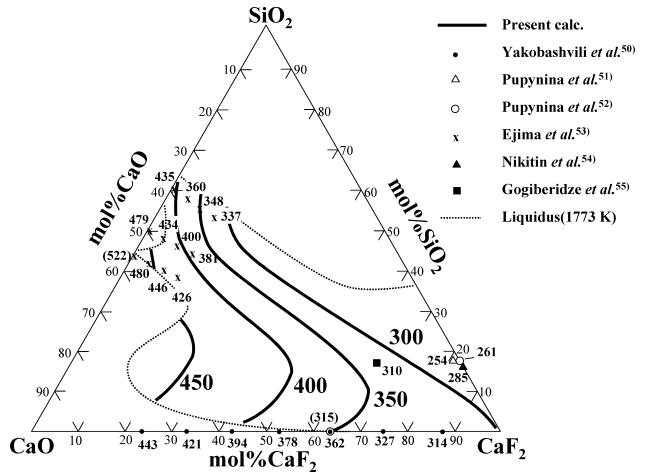

Fig. 14. Surface tension (mN/m) of molten SiO_2–CaO–CaF_2 system at 1 773 K. Iso-surface tension curves were calculated by using neural network computation with 12 units in the middle layer.

found to be one of the practical criteria for designing the number of units in the middle layer.

4. Conclusions

A neural network computation was applied for estimating the surface tension in ternary silicate melts. The discussion on the criteria for designing the number of units in the middle layer of the neural network computation was also conducted. It was found that the modified C_p-criterion, based on the degrees of freedom in the neural network computation, was useful for determining the number of units. The calculated surface tension results obtained using neural network computation with the appropriate unit number, which is assessed satisfactorily by considering the C_p-criterion, showed a virtual reproduction of the experimental data in ternary silicate melts with high precision.

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