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Neural network as a tool for catalyst development

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Abstract

This article introduces a novel information science technique, an artificial neural network, which will possibly be a powerful tool for catalyst development. As an example of synergistically generated catalytic functions, the neural network has been applied to the estimation of acid strengths of mixed oxides, and it has been shown that not only the interpolation but also the extrapolation of given acid strength data is possible within reasonable experimental error. The neural network also has been successfully applied to the estimation of catalytic performance, such as the catalytic activity of a series of lanthanide oxides in the oxidation of butane and the selectivities to various products in the oxidative dehydrogenation of ethylbenzene on a series of promoted SnO_2 catalysts. On the basis of these results, some remarks are given on its application to catalyst development.

1. Introduction

Computers may be applied to the research and development of catalysts in various ways. Computation based on theoretical chemistry, such as quantum chemistry, molecular dynamics and Monte Carlo simulations, enable us to 'watch' catalytic phenomena on a catalyst surface or in porous crystals, which leads to an in-depth understanding of catalysts and catalysis on an atomic scale. Another application is in the information science approach to catalyst development. After the pioneering works of Yoneda [1], several papers have been devoted to the development of knowledge-based expert systems for catalyst design [2–5].

The present authors have also developed a prototype catalyst design expert system: INCAP (INtegration of Catalyst Activity Pattern) for the selection of single catalyst components [6] and multi-component catalysts [7]. INCAP was applied to the selection of promoter oxide to be added to a predetermined main component, SnO_2, for oxidative dehydrogenation of ethylbenzene; the promoting effect estimated by INCAP was afterward verified through experimental catalyst testing [8]. These results led us to plan an advanced system, ARCADE (ARtificial intelligence system for CAtalyst DEsign) [9]. As a preliminary model of the module of ARCADE, we also developed a prototype knowledge-based system named CREAM (Creation of catalytic REAction Mechanism) [10] on the basis of the concept of A/GRACE which is a sub-system of GRACE (Generalized Reaction Analysis for Creation and Estimation) developed by Yoneda [11]. The A/GRACE can generate all of the possible network of elementary reactions including surface steps without any specific knowledge of the problem reaction.

These works indicate that an expert systems approach is a promising method for catalyst devel-

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opment. It should be noted, however, that the construction of an expert system requires much effort, especially for knowledge acquisition. This article introduces another information science approach, an artificial neural network, which requires less effort and labor.

2. Overview of neural network

The artificial neural network, which arose from attempts to model the functioning of the human brain, is structured to learn the relationship between a set of factors and the results, even if the relation between them is theoretically undefined and is non-linear. Actually, the neural network has been successfully applied predictively in various fields of chemistry, such as structure–activity relationships, structure–spectrum correlations, and so on [12,13]. In catalysis, however, only a few attempts have so far been reported by the present authors: estimation of the acid strengths of mixed oxides [14] and of the catalytic activity and selectivity of oxide catalysts in the oxidative dehydrogenation of ethylbenzene [15].

Fig. 1 shows an overview of the neural network construction used for the estimation of acid strength of mixed oxides. Although a large variety of neural network architectures and methods have been proposed, the training algorithm called back-propagation is almost exclusively used for applications to chemistry because of its strong capability of learning. This is also the case in our studies. The neural network consists of artificial neurons (or units), shown by the circles in the figure, interconnected with each other and arranged into layers: an input layer containing units representing possible factors controlling acid strength, hidden layer(s), and an output layer containing units representing acid strength.

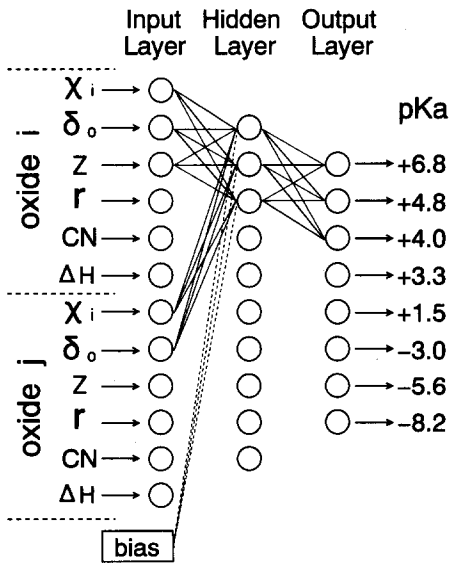

Fig. 1. Construction of the neural network used for the estimation of acid strengths of mixed oxides.

If one gives a training set of known pairs of input data (factors) and output data (results), the neural network calculates the output data from given input data and compares calculated output data with given output data. The error is then propagated backward through the network to readjust the weights of the interconnections shown by straight lines in Fig. 1. This procedure called training is iterated, until the desired level of agreement is achieved between the calculated and the given output data. The network thus trained may be viewed as the 'function' representing the relation between the result (output data; acid strength in this case) and the factors (input data). Thus,

$$\text{Acid strength}_{i,j} = f(Z_i, CN_i, r_i, X_i, \delta o_i, \Delta Hf_i, Z_j, CN_j, r_j, X_j, \delta o_j, \Delta Hf_j) \quad (1)$$

where sub i and j stand for constituent oxides; the factors Z_i through ΔHf_i are described below.

The trained network can be used to estimate or predict unknown output data: if one gives the trained network the input data of the sample not included in the training set, the network calculates corresponding output data. The calculated output data would be correct, if the neural network learns properly. This procedure, viewed as a function approximation, might appear to resemble a least squares fit. It should be noted, however, that the function approximation by a neural network does not require any model equation and it can correlate by using a relatively small number of data.

The accuracy of estimation/prediction can be examined by a 'leave-one-out' test as follows: The input and output data of one catalyst, which is a 'dummy' of a target catalyst, are eliminated from the training set of input and output data, and the neural network is trained by using all of the remaining training set. After training is completed, the input data of the eliminated catalyst are fed into the thus trained neural network through an input layer to calculate the output data. Then, the thus calculated output data are compared with the experimentally observed values to find out the error of estimation. This procedure is carried out for all of the different catalysts.

3. Human task for use of the neural network

Tanabe and co-workers [16] and Seiyama and co-workers [17] proposed correlations of the acid strengths of mixed oxide with the physical/chemical properties of constituent oxides/ions. In Tanabe's correlation, the acid strength is represented as a function of the electronegativity (X_i) of the metal ions or of the partial charge of the oxygen ions (δo) of the constituent oxides, and in Seiyama's correlation, the valence (Z), coordination number (CN), and ionic radius (r). In their cases, not only the controlling factors but also the form of the 'function' need to be explicitly determined, of course, through the elegant activity of the human brain either empirically or theoretically.

In our case, on the other hand, since the form of 'function' is to be determined by the neural network, only the selection of 'possible' factors remains as the human task. At first, we tried to estimate acid strengths by using the above-mentioned five parameters used in Tanabe's and Seiyama's correlations as the input data. Both the training and the estimation of acid strengths were conducted with satisfactory accuracy for the mixed oxides shown in Table 1 except for those underlined, indicating that the acid strength is controlled by some of these five parameters. The trained network, however, could not estimate the acid strength of promoted SnO_2 in the case of promoters which were not included in the training set. In other words, the 'function' composed from the five parameters covers the training set, but does not work predictively outside of the training set. The heat of formation of the oxide (ΔHf) was then tentatively added to input data, which resulted in satisfactory estimations even for the promoted SnO_2 catalysts.

Another task to be accomplished is the representation of output data. In the case of acid strengths of mixed oxides, we have examined the effect of the representation mode on the training efficiency and estimation accuracy. For example, benzeneazodiphenylamine ($pK_a = +1.5$) adsorbed on SnO_2–ZnO changes in colour to the purple of its acid form, while dicinnamalacetone ($pK_a = -3.0$) maintains the yellow of its base form. This colour change pattern indicates that the maximum acid strength of SnO_2–ZnO is given as $-3.0 < pK_a < +1.5$. The most popular way to represent this result is to use pK_a(max) of $+1.5$ for SnO_2–ZnO, i.e., the lowest pK_a value of the indicator which changes its colour on adsorption. In the first phase of our study, this mode of representation was adopted as the output data. The training efficiency, however, was so low that the training set of pK_a(max) could not be reproduced within a reasonable number of training iterations. In the second phase, the acid strength was represented by an average of $+1.5$ and -3.0 for SnO_2–ZnO, i.e., an average of the lowest pK_a of colour-changed indicators and the highest pK_a of colour-unchanged indicators, reflecting that the real acid strength lies between the two values. This mode of representation resulted in a training with satisfactory accuracy, but with insufficient estimation accuracy. Finally, the colour change pattern of Hammett indicators was used. In the case of SnO_2–ZnO, the indicators with pK_a of $+6.8$, $+4.8$, $+4.0$, $+3.3$, and $+1.5$ changed their colours, but not those with pK_a of -3.0, -5.6, and -8.2. This result was represented as '1,1,1,1,1,0,0,0', that is, each unit of the output layer indicates whether or not the indicator of corresponding pK_a changes its colour. This mode of

Table 1
Acid strengths[a] of mixed oxides in comparison with estimated acid strengths

		MgO	Al_2O_3	SiO_2	TiO_2	ZnO	ZrO_2	SnO_2	Bi_2O_3
MgO	Obs.[b]	–	+4.8	−5.6	+3.3	+4.8	*+1.5*	*−3.0*	*+1.5*
	Est.[c]	–	+4.8	−5.6	+1.5	+3.3	*+4.8*	*−3.0*	*+3.3*
Al_2O_3	Obs.[b]	+4.8	–	−8.2	−5.6	+1.5	−5.6	*−8.2*	+1.5
	Est.[c]	+4.8	–	−5.6	−8.2	+3.3	−5.6	*−5.6*	+1.5
SiO_2	Obs.[b]	−5.6	−8.2	–	−8.2	−3.0	−8.2	−5.6	*+1.5*
	Est.[c]	−5.6	−5.6	–	−8.2	−5.6	−8.2	−3.0	*+1.5*
TiO_2	Obs.[b]	+3.3	−5.6	−8.2	–	−3.0	−8.2	−3.0	+1.5
	Est.[c]	+1.5	−8.2	−8.2	–	+1.5	−5.6	−5.6	−3.0
ZnO	Obs.[b]	+4.8	+1.5	−3.0	−3.0	–	+1.5	+1.5	+3.3
	Est.[c]	+3.3	+3.3	−5.6	+1.5	–	+3.3	+1.5	+1.5
ZrO_2	Obs.[b]	*+1.5*	−5.6	−8.2	−8.2	+1.5	–	−5.6	*+1.5*
	Est.[c]	*+4.8*	−5.6	−8.2	−5.6	+3.3	–	−3.0	*+1.5*
SnO_2	Obs.[b]	*−3.0*	*−8.2*	−5.6	−3.0	+1.5	−5.6	–	*+1.5*
	Est.[c]	*−3.0*	*−5.6*	−3.0	−5.6	+1.5	−5.6	–	*+1.5*
Bi_2O_3	Obs.[b]	*+1.5*	+1.5	*+1.5*	+1.5	+3.3	*+1.5*	*+1.5*	–
	Est.[c]	*+3.3*	+1.5	*+1.5*	−3.0	+1.5	*+1.5*	*+1.5*	–

[a] pK_a(max).
[b] Observed acid strength; numbers not in italics, used as output data; numbers in italics, measured after the calculation had been completed.
[c] Estimated acid strength; numbers not in italics, estimated by the leave-one-out test; numbers in italics, estimated by using all the non-italicized data as output data.

representation is found to work well for both training and estimation.

The construction of the hidden layers also has an effect on the training efficiency and estimation accuracy. Generally speaking, more layers and more units result in not only improved training efficiency but also a more complicated 'function'. The latter increases the likelihood of obtaining a local minimum of the error but not a real minimum. However, there is no a priori rule to determine the numbers of layers and units, and they are usually determined empirically through preliminary trial and error experimentation. At first, a two layer model was used for the estimation of acid strength [14a–c], but it was confirmed afterward that a single layer model gives estimates of the acid strengths with essentially identical accuracy [14d]. Thereafter, the single layer model has been preferably used in our studies.

The numbers of training iterations also should be determined, because overlearning with too many iterations possibly results in a rather large

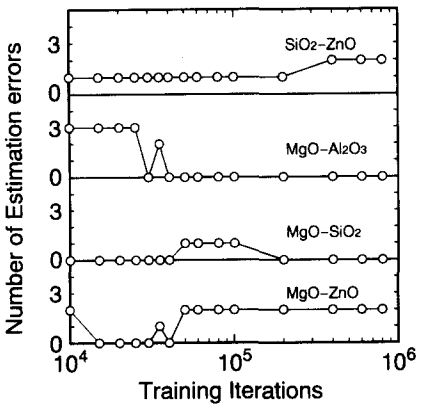

Fig. 2. Change of error with training iteration in the estimation of acid strengths of mixed oxides.

error in the estimation. The strong learning capability of the back propagation neural network allows it to learn not only the tendency of target phenomena but also various 'noise' such as the error of the input and output data, particularly after training with an excess number of iterations. Fig. 2 indicates a few examples of the error in the leave-one-out test as a function of training iterations [14c]. The errors on MgO–ZnO and SiO_2–MgO actually increase with training iterations around 50000 iterations. In this case, 50000 iterations was selected as the standard number of training iterations by taking the changes in estimation errors on all the samples into consideration.

4. Estimation of acid strength of mixed oxides

The neural network has been applied for the estimation of acid strengths of mixed oxides to examine the possibility of estimating the synergistic effect [14]. Although the synergistic effect is a key factor for the development of catalysts and is an important research topic, theoretical prediction is very difficult because of the non-linearity of these systems, and the experimental evaluation is also difficult because of the vast number of possible combinations of constituents. This provides a good situation for examining the applicability of a neural network to catalyst research and development.

4.1. Interpolation ability

The leave-one-out test was conducted to examine the capability of interpolation. Observed acid strengths in Table 1, except for italic ones, were used as the output data, and the numerical values of the properties of the constituent oxides or cations were used as the input data. As shown in Table 1, the neural network gives good estimates of acid strengths for these various mixed oxides. The error is within the interval between adjacent pK_a values of the Hammett indicators. The experimentally observed acid strengths will also be subject to experimental errors which are expected to be, at best, within the adjacent pK_a values. Such experimental errors in the observed acid strengths used as the output data will also contribute to some uncertainty in the calculated values.

When the leave-one-out test was conducted, the acid strengths of some mixed oxides, which are underlined in Table 1, were not known. The neural network was then applied to predict the acid strengths of these mixed oxides for further examination of the interpolation ability [14d]. The acid strengths of these mixed oxides were calculated by introducing the properties of the constituent oxides of these mixed oxides into the neural network which had been trained by using all of the non-underlined data in Table 1. The mixed oxides were prepared by a sol–gel or coprecipitation method, and their acid strengths were measured by using Hammett indicators. As shown in Table 1, the observed acid strengths agree well with the calculated acid strengths except for $MgO–ZrO_2$. The estimation accuracy is essentially identical to that of leave-one-out test and, therefore, to that of experimental accuracy.

4.2. Extrapolation ability

For further examination of the estimation ability, a more severe test was applied to the neural network by taking, as an example, a series of

Table 2
Observed acid strengths of promoted SnO_2 catalysts in comparison with estimated strengths

Promoter		B	P	Cu	Ge	Nb	Mo	Sb	Ce	W
pK_a(max)	Obs.	−3.0	−3.0	+3.3	−3.0	−5.6	−3.0	+1.5	−3.0	−3.0
	Est.[a]	−3.0	−5.6	+1.5	−3.0	−3.0	−3.0	−1.5	−3.0	−3.0

[a]Estimated by using all the non-italicized data in Table 1 as output data.

SnO_2-containing promoter oxides which are not included in Table 1 [14d]. This system is chosen, because we have experienced, in the course of the development of the expert system, INCAP [6,7], that the proposed correlations can not estimate the acid strengths of SnO_2-containing mixed oxides with satisfactory accuracy. This test was conducted essentially in the same way as above. Since four SnO_2-containing mixed oxides are included in the training set, the trained network has 'learned' the relation between the acid strengths of SnO_2-containing mixed oxides and the properties of SnO_2. However, the neural network has not 'learned' anything about the promoter oxides.

Nine mixed oxides containing SnO_2 shown in Table 2 were prepared by a gel mixing or coprecipitation method, and their acid strengths were measured in the same way as above. The observed acid strengths are shown in Table 2 together with the calculated values. As shown, the observed results agree with the predicted results to within reasonable experimental error, i.e., within the unit interval between adjacent pK_a values of the Hammett indicators. Thus, the results shown in Table 2 indicate that the neural network estimates the acid strengths of the mixed oxides with an accuracy similar to that in the experimental measurement, even when one of constituent oxides was not included in the training set.

5. Estimation of catalytic performance

If one were to know the relation between the catalytic performance and the factors controlling catalytic performance, catalyst development would be greatly accelerated. Even if only the major controlling factor is known, it would be a great help for researchers. The difficulty in knowing such factors seems to arise because many factors may affect the catalytic performance and their effects may be non-linearly interconnected. Since the neural network is appropriate for this general kind of problem, we have applied a neural network to the estimation of catalytic performance in the oxidation of butane on a series of lanthanide oxides, and the oxidative dehydrogenation of ethylbenzene on a series of promoted SnO_2 catalysts.

5.1. Activity of lanthanide oxides in the oxidation of butane

One of the authors has conducted in-depth research on the catalytic activity of lanthanide oxides in the oxidation of butane, and correlated the yield of CO_2, taken as a measure of catalytic activity, with the fourth ionization potential (I_4) of the lanthanide cation [18]. This system has been reexamined using the neural network of the construction shown in Fig. 3. The output layer

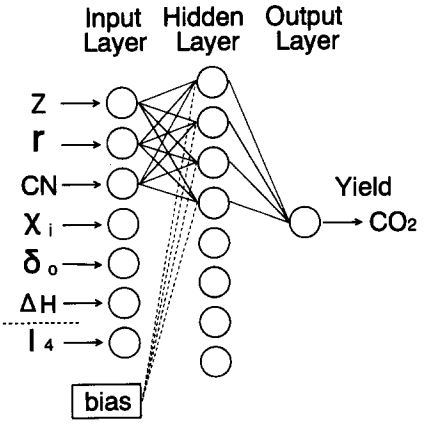

Fig. 3. Construction of the neural network used for the estimation of catalytic activities of lanthanide oxides in the oxidation of butane.

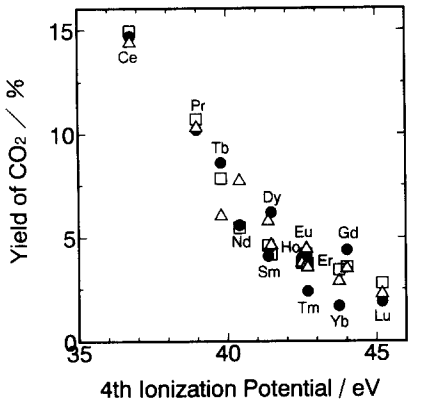

Fig. 4. Comparison of estimated catalytic activities of lanthanide oxides (open squares for Eq. (3) and open triangles for Eq. (5)) with experimentally observed values (closed circles).

consisted of a single unit representing the numerical value of the yield of CO_2. As for the input layer, four series of input data were tested, as shown by Eqs. (2) to (5).

$$Y_{CO_2}=f(Z_i, CN_i, r_i, X_i, \delta o_i, \Delta Hf_i) \quad (2)$$

$$Y_{CO_2}=f(Z_i, CN_i, r_i, X_i, \delta o_i, \Delta Hf_i, I_4) \quad (3)$$

$$Y_{CO_2}=f(r_i, \Delta Hf_i) \quad (4)$$

$$Y_{CO_2}=f(r_i, I_4) \quad (5)$$

In the first series, the input data were identical to those used for the estimation of acid strengths of mixed oxides. The results of estimation by the leave-one-out test were, however, far from the observed values. In the second series, the fourth ionization potential (I_4) was added to the input data, as shown by Eq. (3). Fig. 4 shows the yield of CO_2 estimated by the leave-one-out test as a function of the fourth ionization potential as well as the yield obtained experimentally. As shown, the estimated yield is well correlated with the fourth ionization potential and agrees with the observed values within probable experimental error. In the third series, only ionic radius (r) and heat of formation of the oxide (ΔHf_i) were included in the input data. The estimated yield were far from the observed values, as in the first series. The use of the fourth ionization potential in place of the heat of formation in the fourth series gave good estimates, as shown in Fig. 4. The estimated yields agree well with the experimental values, confirming that the fourth ionization potential is a major factor controlling the catalytic activity of lanthanide oxides in the oxidation of butane.

These results indicate that a neural network can predict the catalytic activity, only when the appropriate controlling factors are included in the input data.

5.2. *Selectivity in the oxidative dehydrogenation of ethylbenzene*

The possibility of estimating the selectivities to various products was examined by taking the oxidative dehydrogenation of ethylbenzene on a series of promoted SnO_2 catalysts as an example. This system is chosen, because an expert system, INCAP [6], has been applied to estimating styrene selectivity in this reaction [8b], which enables us to compare the ability of neural network with an expert systems approach.

A neural network with the construction shown in Fig. 5 was applied for the estimation of selectivities: the output data were numerical values of the selectivities of styrene (ST), benzaldehyde (BA), benzene + toluene (B + T), CO, and CO_2. The input data consist mainly of those used for the estimation of acid strengths of mixed oxides, because the acid property is considered to play an important role in the oxidative dehydrogenation

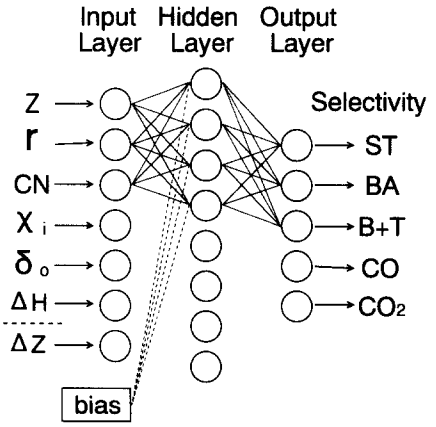

Fig. 5. Construction of the neural network used for the estimation of selectivities to various products in the oxidative dehydrogenation of ethylbenzene.

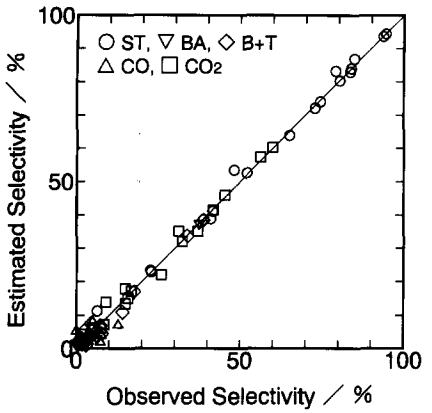

Fig. 6. Comparison of selectivities to various products in the oxidative dehydrogenation of ethylbenzene estimated by the neural network with experimentally observed selectivities.

of ethylbenzene [19]. The presence of untypical valence (ΔZ) was tentatively added to the input data.

In Fig. 6, the selectivities to five groups of products on eighteen promoted and unpromoted catalysts are compared with those obtained experimentally. As shown, the accuracy of estimation is satisfactory: the averaged absolute error in the estimated selectivity is only 1.5%. The largest absolute error in the estimated selectivity for each product was 5.3% for styrene on SnO_2–ZrO_2, 5.2% for benzaldehyde on SnO_2–WO_3, 3.3% for benzene + toluene on SnO_2–ZrO_2, 5.7% for CO on SnO_2–ZrO_2, and 5.0% for CO_2 on SnO_2–Nb_2O_5. These relatively large errors, however, do not seem serious in comparison with reasonable experimental error. The largest absolute error in the average of five products was 3.9% on SnO_2–ZrO_2. This value, however, is close to an average error obtained experimentally on two SnO_2–ZrO_2 catalysts prepared by different methods, 2.8% [20]. Thus, it may be concluded that the selectivity can be predicted by the neural network with an accuracy close to experimental reproducibility.

We previously reported that the catalytic activity and styrene selectivity on the same series of catalysts could be estimated fairly well by using an expert system, INCAP [8b]. The absolute error in styrene selectivity estimated by INCAP was about 20%, with some serious exceptions. The averaged absolute error in the present case was only 1.5%, as mentioned above. Thus, the neural network gives much better estimates than INCAP, demonstrating the strong capability of neural networks for such estimations or predictions.

6. Applicability of neural network

In all of the above-mentioned examples, the results estimated through the leave-one-out test are in good agreement with those observed experimentally, indicating a strong capability for interpolation. In the first example, the acid strengths of mixed oxides containing unknown constituents can be estimated within reasonable error, indicating significant extrapolation ability. As for the estimates of catalytic performance, only the effect of single component catalysts or single promoters has been examined. However, it seems possible to estimate directly the performance of multi-component catalysts, because it has been shown in the first example that synergistic effects can be estimated.

Another possible application of the neural network is to estimate factors controlling results such as the acid strengths and catalytic performances, which would help the researcher to find a way, by using his own human brain, to improve catalysts. One of the methods is a leave-out input data: The effect of each set of input data can be estimated by eliminating one of the input data from the training set. A preliminary example is given in the estimation of catalytic activities of lanthanide oxides in butane oxidation. In this case, it was found that the fourth ionization potential is a key controlling factor. Another possible method would be an analysis of the trained network or partial differentiation of the trained network with respect to each set of input data [21].

Similar estimations could possibly be conducted by using an expert system, but only if one has enough knowledge of the system. It should be noted, however, that the construction of an expert system, especially the acquisition of the needed knowledge, requires much effort and labor by an

expert. The use of a neural network also entails some efforts such as in the selection of possible factors for the input data and the representation of the output data. These tasks, however, are much less important than those required for the expert system. Further, as shown in the last example, the neural network gives much better estimates than the expert system.

Another important feature of the neural network is that the network can be easily modified by introducing newly obtained results even after the initial training. If one conducts catalytic tests on a number of catalysts, the neural network will be able to predict better catalysts by using the results of the catalytic tests as output data. The results of the testing of recommended catalysts can be easily introduced into the training set to give precise prediction or to modify the prediction, depending on the accuracy of the initial prediction. The repetition of this procedure will effectively reduce the number of trial-and-error experimental cycles in the development of catalysts.

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