
Volume 24 Number 1 January 2008 ISSN 0109-5641
ELSEVIER
dental
materials
Journal for Oral and Craniofacial Biomaterials Sciences
Available online at
ScienceDirect
www.sciencedirect.com
Official Publication of the Academy of Dental Materials

DENTAL MATERIALS 24 (2008) 18–27

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Estimation of chemical resistance of dental ceramics by neural network

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ARTICLE INFO

Article history:
Received 23 January 2006
Received in revised form
11 January 2007
Accepted 19 January 2007

Keywords:
Dental ceramics
Neural network
Chemical resistance

ABSTRACT

Objectives. The purpose of this research was to determine the mass concentrations of ions eluted from dental ceramic after an exposure to hydrochloric acid and, drawing on those results, to develop a *feedforward backpropagation* neural network (NN).

Materials and methods. Four dental ceramics were selected for this study. The experimental measurement was conducted after 1, 2, 3, 6 and 12 months of exposure to hydrochloric acid. The results of the 1, 2, 6 and 12 months of immersion were used for training a 13-13-5 model of NN. For evaluating NN efficiency, the regression analysis of input variables obtained by the experiment and output variables provided by the trained network was used.

Results. The measured data from the 3-month acid exposure and data obtained by the neural network estimation were compared.

High correlation coefficient (*R*) and low *normalized root mean square error* (NRMSE) between the measured and estimated output values were observed.

Conclusions. It could be concluded that the artificial neural network has a great potential as an additional method in investigating the properties of dental materials.

1. Introduction

Dental materials have to satisfy strict criteria because of their long therapeutic durability in the oral cavity. One of the most important properties of all restorative dental materials is their chemical resistance. Chemical resistance or chemical durability depends on the structure and composition of the material, laboratory conditions, and environment, which is in this case the oral cavity.

There are several methods for testing the chemical resistance of restorative materials. ISO [1] and ADA [2] standards are usually recommended. Both methods use a 4% acetic acid as a solution medium for faster degradation of dental ceramics. The goal of these methods is to find out the weight loss of ceramic samples after an exposure to the mentioned acid. There are also methods that test chemical resistance of ceramic in more detail, in different media, for a longer period, etc. [3–11]. However, technical literature does not mention any method which could predict the amount of chemical degradation of ceramic material after the measuring interval and at all points during the interval. For this reason a neural network is developed. A neural network is a computer simulation of the behavior of a material based on the experimental research of its properties. This method has been used for testing materials in mechanical engineering [12]. However, neural networks are rarely used for testing dental materials and never for evaluating the chemical resistance of dental ceramics [13,14]. The purpose of this research was to determine the mass concen-

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doi:10.1016/j.dental.2007.01.008

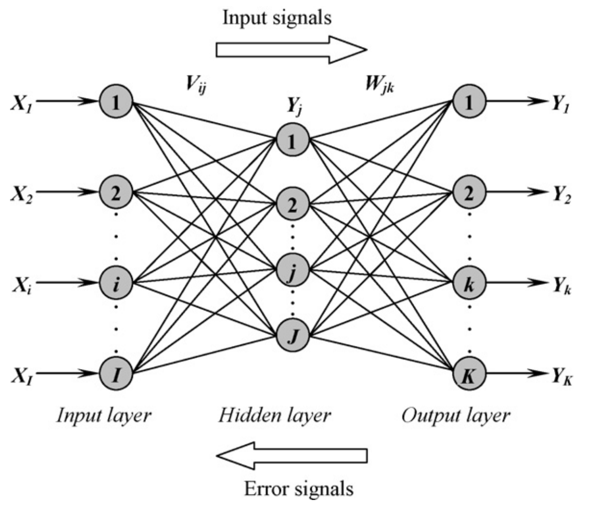

Fig. 1 – Principle of the *feedforward backpropagation* training algorithm.

trations of ions eluted from a dental ceramic after an exposure to hydrochloric acid and, drawing on those results, to develop a *feedforward backpropagation* neural network.

2. Artificial neural network

Artificial neural networks (ANN) are inspired by the biological neural system and its ability to learn through example. Instead of following a group of well-defined rules specified by the user, neural networks learn through intrinsic rules obtained from presented samples. The most commonly used ANN architecture is the multilayer *backpropagation neural network*. Backpropagation was created by generalizing the *Widrow-Hoff* learning rule to multiple-layer networks and nonlinear differentiable transfer functions [15]. Input vectors and the corresponding target vectors are used to train the network until it can approximate a function, associate input vectors with specific output vectors. Standard backpropagation is a gradient descent algorithm, as is the *Widrow-Hoff* learning rule, in which the network weights are moved along the negative of the gradient of the performance function. The term *backpropagation* refers to the manner in which the gradient is computed for nonlinear multilayer networks. *Backpropagation* neural networks often have one or more hidden layers of sigmoid neurons followed by an output layer of linear neurons. Multiple layers of neurons with nonlinear transfer functions allow the network to learn nonlinear and linear relationships between input and output vectors. There are numerous variations of the basic algorithm that are based on other standard optimization techniques, such as conjugate gradient and Newton methods [15]. The one used in this paper is the *feedforward backpropagation* training algorithm designed to minimize the *mean square error* (MSE) between the actual (estimation) output (a, A) and the desired (target) output (d, T). Fig. 1 shows the principle of the *feedforward backpropagation* training algorithm. The basic learning algorithm can be summarized as follows:

Step 1. Set the initial values of weights V_{ij} and W_{jk}.

Step 2. Compute the outputs of all neurons layer-by-layer, starting with the input layer as shown below:

$$\mathrm{net}_j = \sum_{i=1}^{I} V_{ij}X_i, \quad j = 1, 2, \ldots, J-1, \quad i = 1, 2 \ldots, I \tag{1}$$

$$Y_j = f(\mathrm{net}_j) \tag{2}$$

$$\mathrm{net}_k = \sum_{j=1}^{J} W_{jk}Y_j, \quad j = 1, 2, \ldots, J-1, \quad k = 1, 2, \ldots, K \tag{3}$$

$$Y_k = f(\mathrm{net}_k) \tag{4}$$

where V_{ij} is the weight between the input layer and the hidden layer, W_{jk} the weight between the hidden layer and the output layer, X_i the input signals (value of chemical composition), i the number of neurons of the input layer, I the number of inputs of neuron j in the hidden layer, Y_j the output of the hidden neurons, j the number of neurons of the hidden layer, J the number of inputs of neuron k in the output layer. Y_k the output signals (mass of eluted ions per gram of samples), and k is the number of neurons of the output layer. In the case of sigmoidal transfer function of the hidden layer, the following equation applies:

$$f(x) = \frac{2}{1+e^{-x}} - 1 \tag{5}$$

Step 3. Compute system error E:

$$E = \frac{1}{2}\sum_{k=1}^{K}(d_k - a_k)^2 \tag{6}$$

where K represents the total number of patterns, d_k the desired outputs (experimental values) and a_k the actual outputs.

Step 4. If E is small enough or learning iteration is too big, stop learning.

Step 5. Compute learning errors for every neuron layer-by-layer:

$$\delta_k = (d_k - a_k)f'(\mathrm{net}_k), \quad k = 1, 2, \ldots, K \tag{7}$$

$$\delta_k = \sum_{k=1}^{K} W_{jk}\delta_k f'(\mathrm{net}_j), \quad j = 1, 2, \ldots, J-1, \quad k = 1, 2, \ldots, K \tag{8}$$

Step 6. Update weights along negative gradient of E:

$$W_{jk}(n+1) = W_{jk}(n) + l_r\delta_k Y_j + \alpha(W_{ij}(n) - W_{ij}(n-1)) \tag{9}$$

$$V_{ji}(n+1) = V_{ji}(n) + l_r\delta_j X_i + \alpha(V_{ji}(n) - V_{ji}(n-1)) \tag{10}$$

where l_r is the learning rate, α the momentum, and n is the current iteration step.

Step 7. Repeat from Step 2.

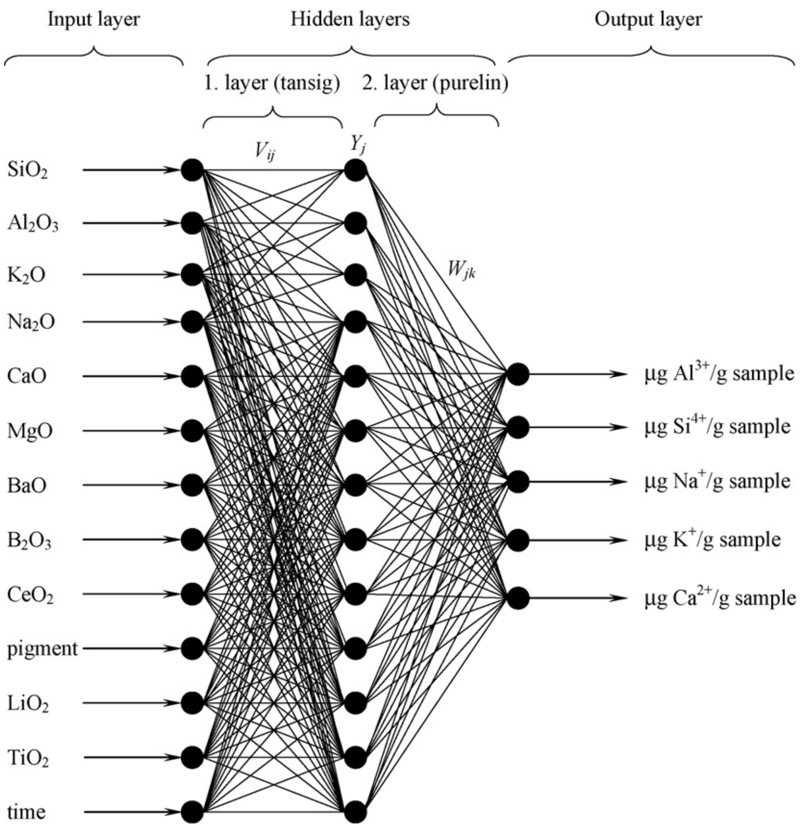

Fig. 2 – Schematic drawing of the model of the used neural network.

3. Material and methods

Four dental ceramics were selected for this study: feldspathic ceramic (IPS-Classic, Ivoclar-Vivadent, Schaan, Liechtenstein), hydrothermal ceramic (Duceragold, Ducere Dental, Rosbach, Germany) and two glass ceramics with different composition (IPS-Empress for staining and layering technique, Ivoclar-Vivadent, Schaan, Liechtenstein). The selection of the ceramic materials was based on their mutual compositional differences. One specimen of each ceramic material was fabricated using a Plexiglas mold (10 mm × 10 mm × 2 mm). The ceramic materials were prepared strictly following their producers' instructions. A creamy mixture of feldspathic ceramic or hydrothermal ceramic was put in the mold and thoroughly condensed. Then the mold was removed and the specimens left on top of a platinum foil and baked. Wax patterns were used for heat-pressed ceramics. The surfaces were ground using 500- and 1000-grit (18 μm) SiC-paper on a disc rotating at 150 rpm, and then glazed.

The samples were cleaned in an ultrasonic bath (Ultra Sonic Bath Model 1510 DTH, Electron Microscopy Sciences) and left to dry for 4 h at 150 ± 5 °C (Sterilizer, Instrumentaria, Zagreb, Croatia). The samples were then placed in a plastic flask (PP, 25.0 ml) filled with 10^{-3} mol dm^{-3} HCl at 50 °C. The mass concentrations of eluted Na^{+}, K^{+} and Ca^{2+} ions were determined using the ATOMIC ABSORPTION SPECTROPHOTOMETER (AAS, AA-6800, SHIMADZU, Kyoto, Japan) and the mass concentrations of Si^{4+} and Al^{3+} ions using the SPECTROPHOTOMETER UV/VIS (COLEMAN 55, PERKIN ELMER, Norwalk, USA). The measurements were conducted after 1, 2, 3, 6 and 12 months of immersion.

4. Model of the neural network

The work included experimenting with a two-layer (13-13-5) *feedforward backpropagation* neural network, whose simplified model is shown in Fig. 2. The input layer is made up by the data on the chemical composition of a dental ceramic (SiO_2, Al_2O_3, K_2O, Na_2O, CaO, MgO, BaO, B_2O_3, CeO_2, pigment, LiO_2, TiO_2) and on the time of exposure (time), and the output layer

Table 1 – Training parameters of neural network

Parameter	Value
Performance goal	0.0001
Learning rate	0.01
Ratio to increase learning rate	1.05
Ratio to decrease learning rate	0.5
Maximum performance increase	1.04
Minimum performance gradient	1e−10
Momentum constant	0.9
Number of layers	2
Number of neurons	13-13-5
Transfer functions	Tansig & purelin
Training function	Traingdx
Number of epochs to train	15,000

Table 2 – Data sets for training (non-shaded) and testing (shaded) of the neural network

Samp.	INPUT DATA: Mean of chemical composition, % (SiO_2; Al_2O_3; K_2O; Na_2O; CaO; MgO; BaO; B_2O_3; CeO_2; pig.; Li_2O; TiO_2)	time, months	OUTPUT DATA: Mass of eluted ions per gram of samples: μg Al^{3+}/g sample	μg Si^{4+}/g sample	μg Na^+/g sample	μg K^+/g sample	μg Ca^{2+}/g sample
1.	65.40; 14.10; 12.85; 5.75; 1.35; 0.95; 0.00; 0.00; 0.00; 0.00; 0.25; 0.00	1	6.50	134.00	102.40	81.51	46.44
		2	12.55	270.00	179.16	99.37	65.04
		3	19.51	371.97	270.76	118.78	80.25
		6	23.80	393.00	283.81	123.93	90.66
		12	28.69	423.78	286.48	125.94	92.35
2.	65.00; 12.50; 12.50; 12.50; 0.10; 0.00; 0.10; 0.00; 0.00; 0.00; 0.00; 0.00	1	29.80	165.00	247.71	121.25	276.36
		2	59.30	319.00	388.11	188.14	513.07
		3	90.24	551.06	500.46	227.98	739.41
		6	142.08	660.49	548.61	268.24	819.77
		12	191.41	781.10	590.30	308.50	900.13
3.	61.00; 19.00; 12.00; 5.00; 1.50; 0.75; 0.00; 0.50; 0.50; 0.25; 0.00; 0.25	1	7.46	77.09	57.57	19.46	8.29
		2	15.05	134.62	89.22	33.68	18.12
		3	23.37	275.39	142.62	53.12	37.67
		6	37.52	301.65	158.89	62.25	47.54
		12	52.78	328.64	175.16	66.37	48.52
4.	61.00; 19.00; 12.00; 5.00; 1.50; 0.95; 0.75; 0.55; 0.50; 1.25; 0.00; 0.25	1	72.86	174.47	144.64	79.81	21.06
		2	148.35	344.98	197.35	182.84	41.63
		3	212.17	529.93	238.86	262.63	57.51
		6	283.69	621.36	258.89	339.32	77.32
		12	366.60	724.10	277.01	412.90	78.69

Key: sample 1, feldspathic ceramic; sample 2, hydrothermal ceramic; sample 3, glass ceramic for staining; sample 4, glass ceramic for layering.

Table 3 – Comparison of the measured data (ME) and data estimated by the neural network (NN)

Samp.	time, months	μg Al^{3+}/g sample ME	NN	Δ	Δ %	μg Si^{4+}/g sample ME	NN	Δ	Δ %	μg Na^+/g sample ME	NN	Δ	Δ %	μg K^+/g sample ME	NN	Δ	Δ %	μg Ca^{2+}/g sample ME	NN	Δ	Δ %
1.	1	6.50	8.31	1.81	21.78	134.00	147.11	13.11	8.91	102.40	98.20	-4.20	-4.27	81.51	81.61	0.10	0.12	46.44	38.84	-7.60	-19.56
	2	12.55	9.75	-2.80	-28.71	270.00	252.35	-17.65	-6.99	179.16	184.59	5.43	2.94	99.37	98.57	-0.80	-0.81	65.04	77.15	12.11	15.69
	3	19.51	13.52	-5.99	-44.30	371.97	319.07	-52.90	-16.57	270.76	234.84	-35.92	-15.29	118.78	110.14	-8.64	-7.84	80.25	90.47	10.22	11.29
	6	23.80	25.13	1.33	5.29	393.00	398.20	5.20	1.30	283.81	282.41	-1.40	-0.49	123.93	125.25	1.32	1.05	90.66	82.18	-8.48	-10.31
	12	28.69	28.26	-0.43	-1.52	423.78	423.18	-0.60	-0.14	286.48	286.63	0.15	0.05	125.94	125.20	-0.74	-0.59	92.35	96.79	4.44	4.58
2.	1	29.80	27.92	-1.88	-6.73	165.00	150.24	-14.76	-9.82	247.71	252.32	4.61	1.82	121.25	122.16	0.91	0.74	276.36	285.22	8.86	3.10
	2	59.30	61.72	2.42	3.92	319.00	339.61	20.61	6.06	388.11	381.59	-6.52	-1.70	188.14	186.34	-1.80	-0.96	513.07	499.47	-13.60	-2.72
	3	90.24	93.35	2.93	3.13	551.06	481.21	-69.85	-14.51	500.46	467.96	-32.50	-6.94	227.98	235.18	7.20	3.06	739.41	656.69	-82.72	-12.59
	6	142.08	141.45	-0.63	-0.44	660.49	652.93	-7.56	-1.15	548.61	550.98	2.37	0.43	268.24	269.15	0.91	0.33	819.77	826.96	7.19	0.86
	12	191.41	191.44	0.03	0.17	781.10	782.63	1.53	0.19	590.30	589.79	-0.51	-0.08	308.50	308.33	-0.17	-0.05	900.13	897.10	-3.03	-0.33
3.	1	7.46	5.84	-1.62	-27.73	77.09	69.39	-7.70	-11.09	57.57	57.93	0.36	0.62	19.46	19.38	-0.08	-0.41	8.29	12.34	4.05	32.82
	2	15.05	17.39	2.34	13.45	134.62	144.02	9.40	6.52	89.22	89.57	0.35	0.39	33.68	34.41	0.73	2.12	18.12	13.51	-4.61	-34.12
	3	23.37	24.47	1.10	4.49	275.39	203.17	-72.22	-35.54	142.62	115.26	-27.36	-23.73	53.12	45.12	-8.00	-17.73	37.67	21.06	-16.61	-78.86
	6	37.52	36.51	-1.01	-2.76	301.65	300.92	-0.73	-0.24	158.89	157.61	-1.28	-0.81	62.25	61.21	-1.04	-1.69	47.54	46.70	-0.70	-1.49
	12	52.78	53.15	0.37	0.69	328.64	327.75	-0.89	-0.27	175.16	175.73	0.57	0.32	66.37	66.89	0.52	0.77	48.52	49.71	1.19	2.39
4.	1	72.86	75.65	2.79	3.64	174.47	179.15	4.68	2.61	144.64	143.84	-0.80	-0.55	79.81	83.80	3.99	4.76	21.06	10.71	-10.35	-96.63
	2	148.35	144.34	-4.01	-2.77	344.98	340.69	-4.29	-1.25	197.35	197.25	-0.10	-0.05	182.84	176.44	-6.40	-3.62	41.63	57.16	15.53	27.16
	3	212.17	196.46	-15.71	-7.99	529.93	455.77	-74.16	-16.27	238.86	230.83	-8.03	-3.47	262.63	243.04	-19.59	-8.06	57.51	77.43	19.92	25.72
	6	283.69	285.87	2.18	0.76	621.36	619.91	-1.45	-0.23	258.89	260.30	1.41	0.54	339.32	343.24	3.92	1.14	77.32	68.43	-8.89	-12.99
	12	366.60	365.74	-0.86	-0.23	724.10	725.38	1.28	0.17	277.01	276.29	-0.72	-0.26	412.90	411.51	-1.39	-0.33	78.69	82.53	3.84	4.65

Key: ME, values obtained by measure; NN, values obtained by neural network; Δ = NN − ME; Δ (%) = ((NN − ME)/NN) × 100; shaded rows, data sets for testing of the neural network.

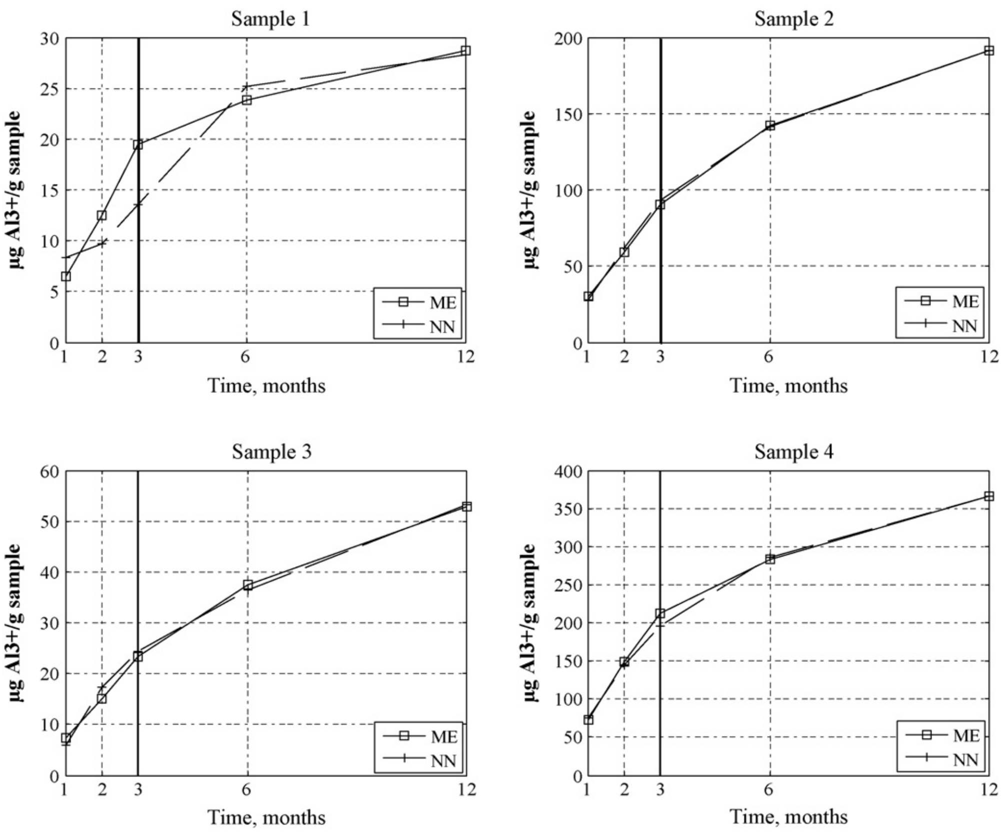

Fig. 3 – Comparison of the measured data (ME) and data estimated by the neural network (NN) for Al^{3+} for the complete immersion period.

is made up by the data on mass concentrations of ions (Al^{3+}, Si^{4+}, Na^{+}, K^{+}, Ca^{2+}) eluted from dental ceramics. The chosen model with 13 input neurons, 13 neurons in the hidden layer, and 5 output neurons is the result of the author's experimenting and practical experiences obtained during work [12,16]. For modelling, MathWorks, Neural Network Toolbox, Release 4.0.1 was used [17] according to whose rules hidden layers of the network were marked. As the transfer function of the first hidden layer, the function of sigmoidal type (tansig) was applied, and of the second hidden layer – the function of linear type (purelin). As a training function, *traingdx* – gradient descent with momentum and the adaptive learning rate backpropagation function was applied. Training parameters of the network are shown in Table 1. Because of relatively small numbers of input training data, it was not possible to define a separate group for validation of the network during the training process and to apply some of the methods (e.g. *early stopping*) for improving generalization of the network. Input–output data for training the network are shown in Table 2. Data obtained during 1, 2, 6, and 12 months of the experiment were included in the network training process. Data obtained during the 3rd month of the experiment were excluded from the training process, and used for network testing.

For the estimation of performance of the learning algorithm in solving the specified task, *performance index* was defined. Performance index enabled comparison of the applied neural network algorithm with the other learning algorithms. The most frequent performance index is the *normalized root mean square error*—NRMSE [15]:

$$\mathrm{NRMSE} = \frac{\sqrt{\sum_{n=1}^{N}(d_n - a_n)^2/N}}{\sigma d_n} \tag{11}$$

$$\sigma_{d_n} = \sqrt{\frac{1}{N}\sum_{n=1}^{N}(d_n - \bar{d})^2} \tag{12}$$

$$\bar{d} = \frac{1}{N}\sum_{n=1}^{N} d_n \tag{13}$$

where N is the total number of patterns, d_n the desired (target, T) outputs, a_n the actual (estimation, A) outputs and σ_{d_n} is the standard deviation.

5. Results

Using experimental data, the *feedforward backpropagation* neural network for estimation of the wear resistance of dental ceramics was modelled (Fig. 2).

The results of the 1, 2, 6 and 12 months of immersion were used for training a 13-13-5 model of neural network and the results of the 3-month immersion period were used for its testing (Table 2).

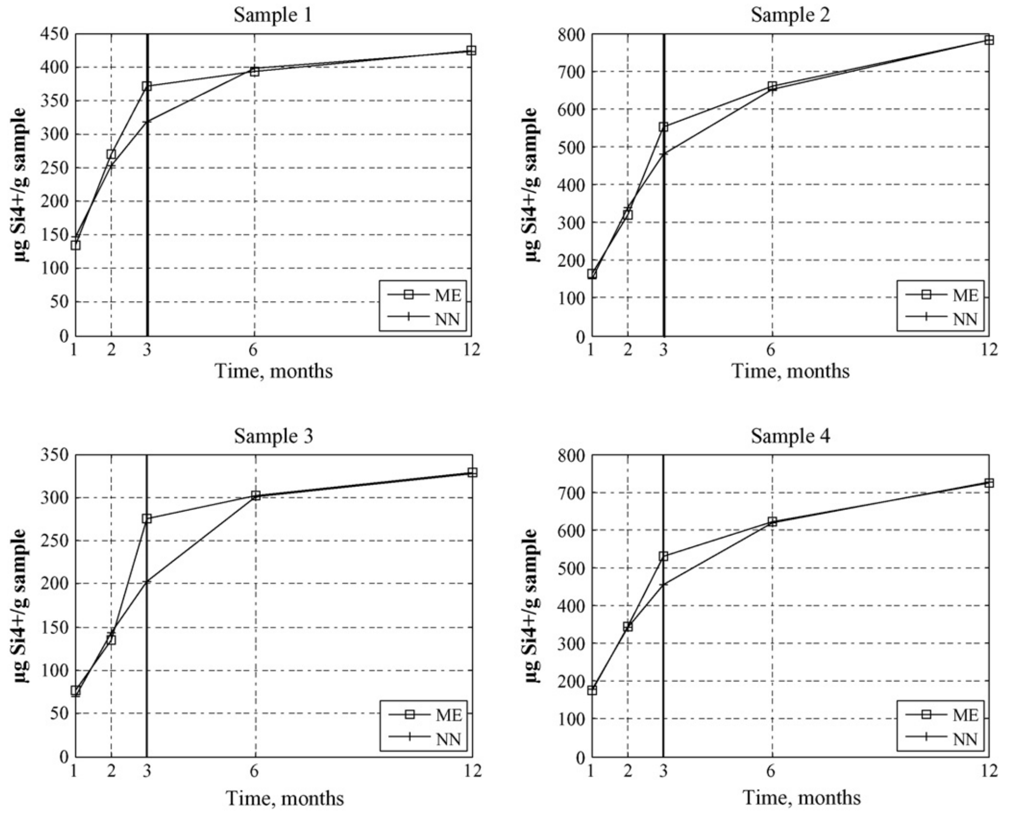

Fig. 4 – Comparison of the measured data (ME) and data estimated by the neural network (NN) for Si^{4+} for the complete immersion period.

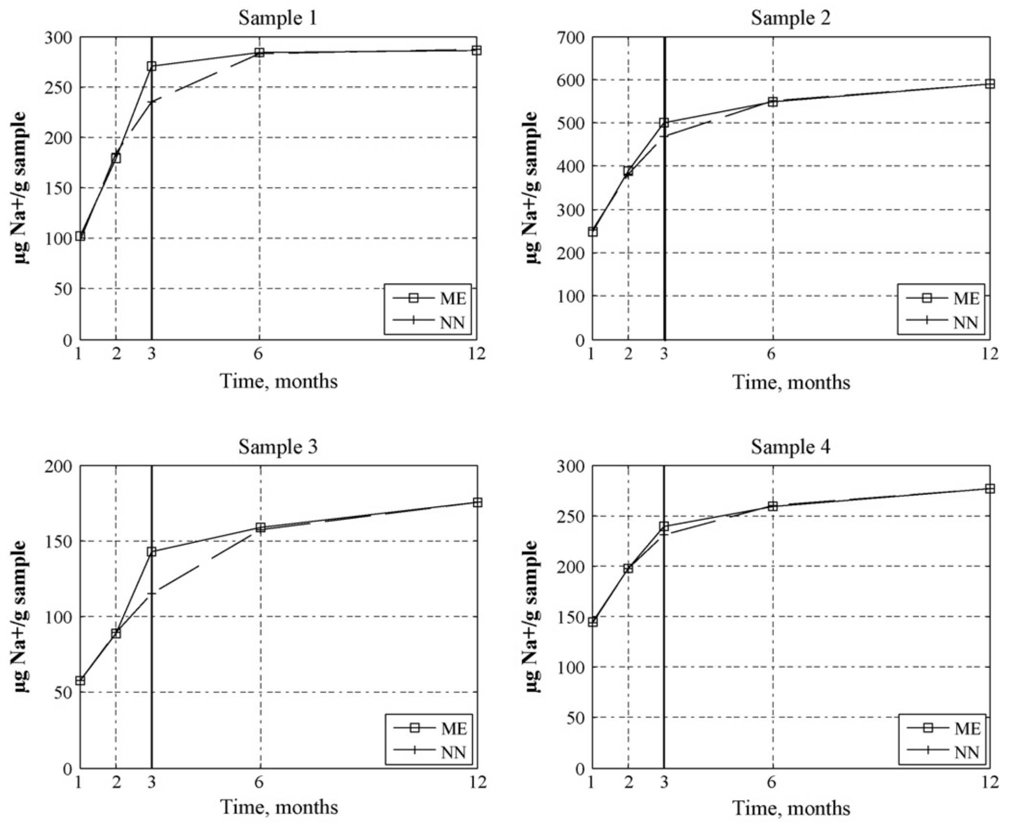

Fig. 5 – Comparison of the measured data (ME) and data estimated by the neural network (NN) for Na^{+} for the complete immersion period.

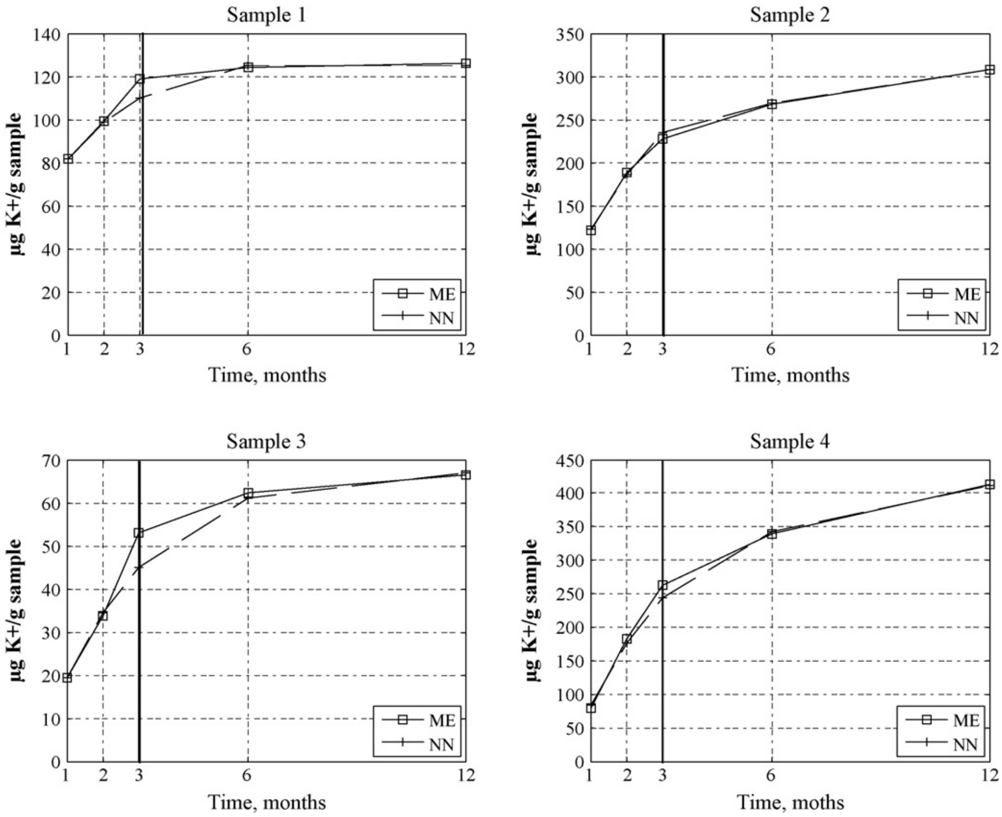

Fig. 6 – Comparison of the measured data (ME) and data estimated by the neural network (NN) for K^+ for the complete immersion period.

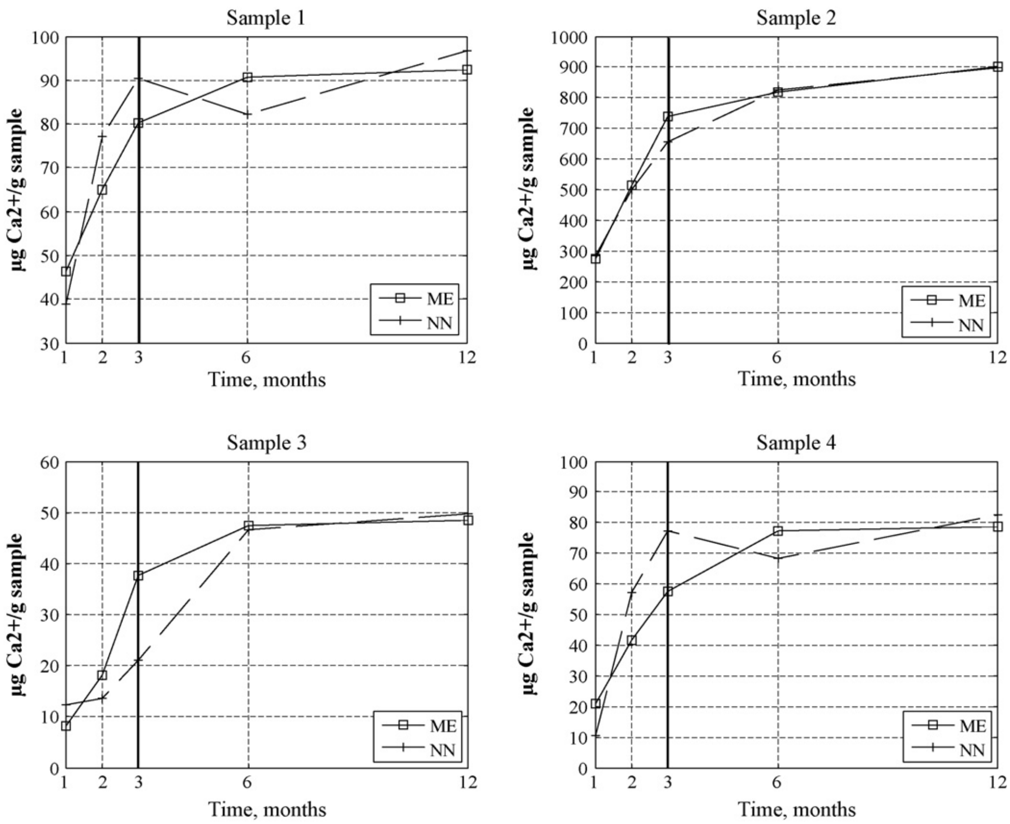

Fig. 7 – Comparison of the measured data (ME) and data estimated by the neural network (NN) for Ca^{2+} for the complete immersion period.

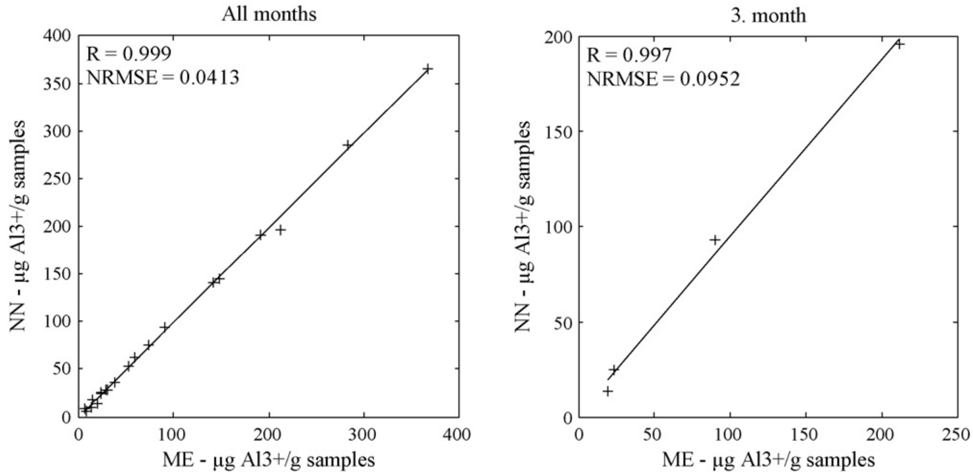

Fig. 8 – Correlation coefficient (R) and normalized root mean square error (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for Al^{3+} for the complete immersion period and the 3-month immersion period.

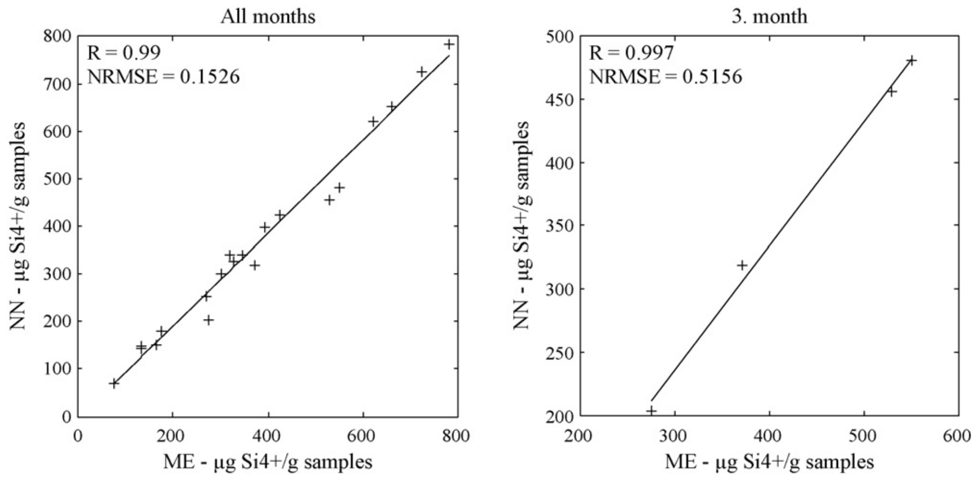

Fig. 9 – Correlation coefficient (R) and normalized root mean square error (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for Si^{4+} for the complete immersion period and the 3-month immersion period.

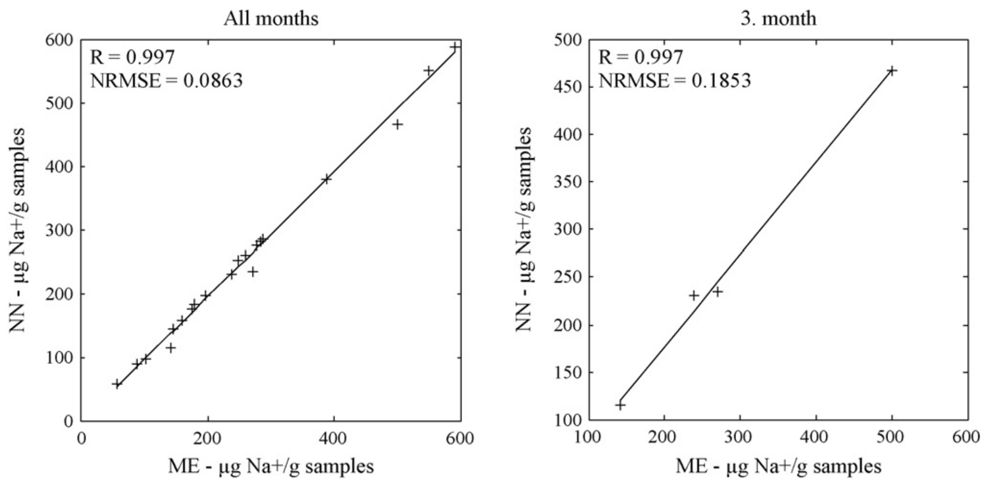

Fig. 10 – Correlation coefficient (R) and normalized root mean square error (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for Na^{+} for the complete immersion period and the 3-month immersion period.

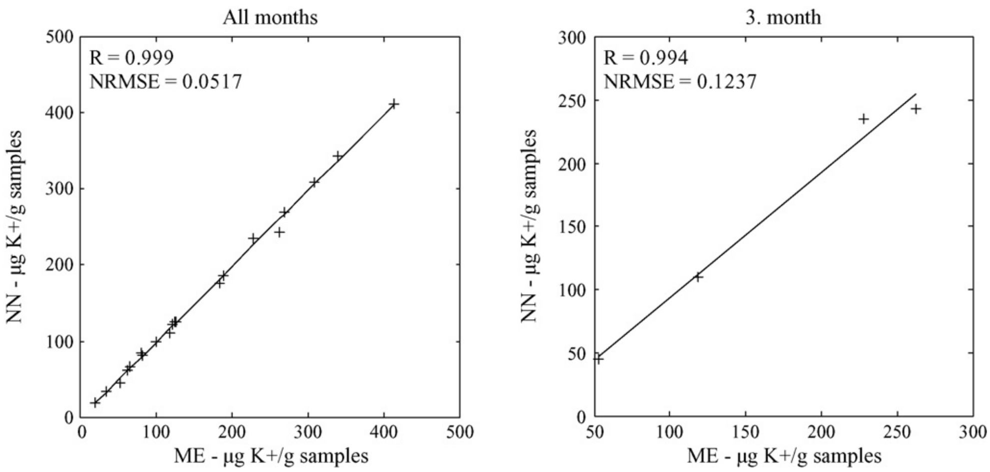

Fig. 11 – Correlation coefficient (R) and normalized root mean square error (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for K^+ for the complete immersion period and the 3-month immersion period.

Numerical comparisons of measured values (ME), mass of eluted ions per gram of samples and the values obtained by estimation of the neural network are shown in Table 3.

Figs. 3–7 show the graphic comparison of the measured data (ME) and data estimated by the neural network (NN), both for all ions and for the 3-month immersion period.

The correlation coefficient (R) and *normalized root mean square error* (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for eluted ions for the complete immersion period and for the 3-month immersion period are shown in Figs. 8–12.

6. Discussion

Hydrochloric acid was used for testing the chemical durability of dental ceramics. This departs from the standard ISO method [1] which uses acetic acid for testing the chemical durability of dental ceramic because of its frequent use in households. The authors find hydrochloric acid suitable since there are patients with gastric disorders, who have lower pH-values in the oral cavity due to the presence of hydrochloric acid (gastric reflux, regurgitation, and bulimia), similar to Grossman et al. [8]. The duration and the temperature of the experiment also differ from the ISO-Standard. It was desirable to include in this research the longest possible elution of ions from dental ceramics in order to test the long-term predicting possibilities of this method.

The artificial neural network method presented in this study is currently being used in different fields of engineering for testing different materials [12]. The dental ceramic was used because of its chemical inertness [4,5]. The only purpose of experimental data in this study was to train the neural network and to determine its efficiency and limits. The chemical stability of dental ceramic was not evaluated. The results of previous studies [3,18] of the same group of authors allowed a presumption that the ions' elution from dental ceramic in an acid solution was very low. The number of measuring intervals was relatively low. Even then the method of the artificial neural network showed a very accurate prediction of the wear

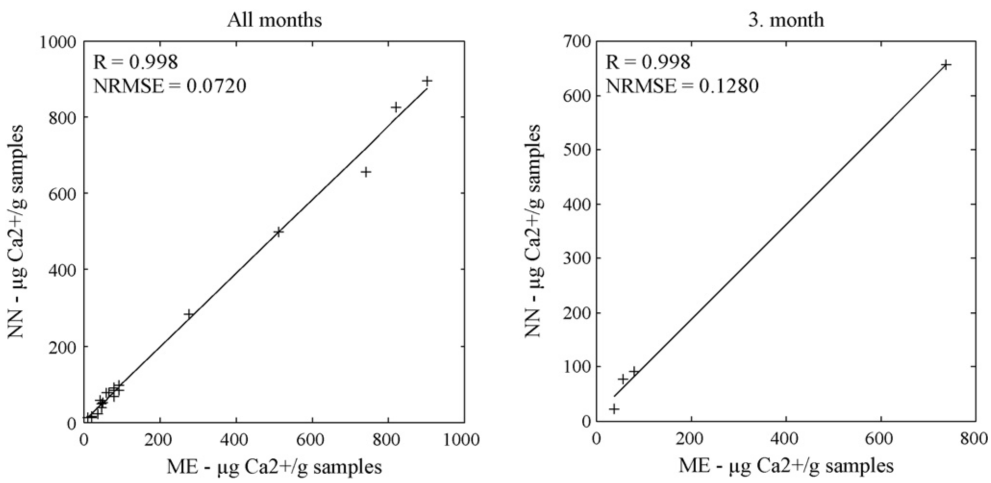

Fig. 12 – Correlation coefficient (R) and normalized root mean square error (NRMSE) of the measured data (ME) and data estimated by the neural network (NN) for Ca^{2+} for the complete immersion period and the 3-month immersion period.

behavior of dental ceramics. A high correlation coefficient (*R*) and a low *normalized root mean square error* (NRMSE) between measured and estimated output values were observed. The single problem was an *underfitting* within the process of neural network training. It was caused by the insufficiency of input data sets for training network. The reason for a small number of input data sets is that the results of investigating the ceramics corrosion were to be used for another investigation which did not require further measurements. However, even that number of measurements showed only minimal differences obtained between measured and estimated mass of eluted ions per gram of dental ceramic sample.

Artificial neural network has a great potential for investigating not only the chemical stability of materials, but also other properties, such as wear resistance, flexural strength, etc., which are changing under the influence of one or more parameters. The value of this method is also in the possibility of predicting future events in correlation with the number of experimental data. The higher number of experimental data allows for more accurate prediction.

It could be concluded that the artificial neural network has great potential as an additional method for investigating the properties of dental materials.

Acknowledgements

This study was supported by the Croatian Ministry of Science, Education and Sports.

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