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NEURAL MODELING AND OPTIMIZATION OF A MECHANICAL-CHEMICAL TREATMENT APPLIED FOR SOME INDUSTRIAL EFFLUENTS. A ROUMANIAN CASE STUDY

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Abstract. The paper proposes an artificial neural network (ANN) model of multilayers perceptron type (MLP3:10:1) adapted for mechanical-chemical treatment system of an industrial effluent (*i.e.* coagulation-flocculation - sedimentation applied for an industrial effluent produced in a manufacturing plant of bricks and other ceramic products). This model of multiple inputs-one single output considers three input variables (independent variables) like the temperature (z_1), dose of polyelectrolyte (z_2) and agitation time (z_3) and one single output variable (dependent variable) as the removal of turbidity (Y_1) or colour (Y_2). Consequently, the proposed ANN model is optimized and also tested for some data from outside of the training experimental field. The optimal removal of turbidity (91.7%) is performed working at a temperature of 20°C, with a polyelectrolyte dose of 20 mg/L, for 30 min of agitation at 50 rpm, and in the case of optimal colour removal (92.2%) by working at a temperature of 26°C, with a polyelectrolyte dose of 15 mg/L, for no more than 30 min of agitation at 50 rpm, respectively.

Keywords: artificial neural network (ANN), industrial wastewater treatment, multilayers perceptron model (MLP).

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Introduction

The central concept of an industrial effluent treatment is to save water and also to reduce at zero or minimize the polluting effects of treated effluent in the discharging point in both sewer system and natural receptor, or for other purposes, such as recycling or onsite reuse. Therefore, the modern concept is moving away from the classic 'end-of-pipe' technology towards 'decentralized effluent treatment processes', 'process integrated water management' and ultimately, in a number of possible cases, to 'fresh water-free processes'. Frequently, it becomes more economic and favourable to treat some industrial effluents by specialized processes which make possible to reuse one or more streams (*i.e.* treated effluents) and to save fresh water [1].

In this context, some developments were performed to combine production processes and wastewater treatment often named 'process-integrated water management' (*i.e.* sustainable water use, sustainable industrial water use, or cleaner production) following mainly two different wastewater treatment concepts as: (*1*) separation of pollutants from water/wastewater, especially solid matters, and (*2*) partial or complete mineralization of polluting species, especially dissolved ones. Collection, treatment and disposal are three basic components of any industrial wastewater management system, where collection component is kept as minimal as possible, the highest importance being focus mainly on necessary treatment and disposal of treated wastewater [2]. The selection of appropriate industrial wastewater treatment must consider the life cycle cost of such a system including design, construction, operation, maintenance, repair and replacement. The most cost-effective industrial wastewater treatment system is a decentralized system (onsite or cluster) based on primary and secondary treatment steps (*e.g.*, at least a conventional mechanical-chemical, or mechanical-biological treatment system), but many systems currently in use do not provide a treatment level that is needed to protect public health and receiving environment [3].

A cost effective treatment system for an industrial productive company can be based on one-single, or two treatment steps such as mixed coagulation-flocculation followed by sedimentation and/or rapid filtration applied with minimal costs in existing treatment units that are operating at optimal parameters for high treatment performance and no disposal risks.

The integrated process water management is proposing also optimization of existing, or new processes with the aim of saving water, materials and energy together with consideration of the best way for a balance of economical success, environmental protection and social acceptance. This requires firstly process modeling and after optimization [1]. For an efficient optimization of the industrial effluent treatment process it is requiring adequate advanced modeling, simulation and optimization using modern, available and ease adaptable procedures.

In actual times, some studies using artificial neural networks (ANNs) in modeling of biological wastewater treatment processes have been published, providing some alternative approaches [4-11]. Based on the obtained results, the researchers were appreciated the great potential of ANNs as tools for the prediction of water resources' quality, or assurance of significant remediation efficiencies in terms of permanent and adequate values of the main monitoring quality indicators (physical, chemical and biological/microbiological ones), or hydraulic loads for health and protection of natural aquatic receptors.

Meenakhsipriya *et al.* [12] performed the application of artificial neural network techniques to estimate the pH value for an effluent treatment process. Thus, ANN had the ability to identify the non-linear dynamical systems from the input-output data. An important requirement of this type of application was the robustness of the system against erroneous sensor measurements. A simulation model of system pH for common effluent treatment plant (CETP) was developed. A novel off-line and on-line training scheme for the neural network was developed by error back propagation training algorithm to model accurately the system pH for CETP. For this purpose, a simple feed forward, back propagation neural network, with only one hidden layer, and sigmoidal activation functions was used [12].

The ANN models with multiple inputs - single output have been developed [13], and used in analyzing how wastewater quality indicators such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), and suspended solids (SS) are affecting each other. It was concluded from the reported works [10-11] that ANNs can be efficiently applied to analyze the complex, non-linear and dynamic data which can make ANNs a valid tool to study biological and/or physical-chemical processes, but also the hybrid or mixed ones.

A Wiener-Laguerre model with artificial neural network, as its nonlinear static part, was employed to describe the dynamic behaviour of a sequencing batch reactor (SBR) used for the treatment of a dye-containing wastewater [14]. The results from this study revealed that the developed model is accurate and efficacious in prediction of COD and BOD of the dye-containing wastewater treated by SBR. The proposed modelling approach can be applied to other industrial wastewater treatment systems to predict effluent characteristics.

The reference literature concludes that neuronal modelling (ANN modelling) and optimization through simulation of industrial effluent treatments, such as an industrial mechanical-chemical effluent treatment, can be useful in modernization of existing wastewater treatment plant (WWTP).

Consequently, it is proposed in this original paper the presentation of an artificial neural network model of multilayers perceptron (MLP) type adapted for mechanical-chemical treatment system of an industrial effluent produced by a manufacturing unit of bricks and other ceramics (*i.e.* coagulation/flocculation - sedimentation) for evaluation of its treatment efficiency in terms of turbidity (Y_1) and colour (Y_2) removals (single output) considering three input variables (temperature, polymeric flocculant dose and agitation time), and its optimization for a few data from outside of training/testing experimental field of ANN model (overfitting and overtraining data).

Experimental

Materials

All chemicals used for industrial effluent treatment process and analyses of different quality indicators of non-treated and/or treated effluent were of analytical purity, being purchased from Romanian chemical manufacturing companies (CHEMICAL Co., Iasi, Romania and/or NORDIC INVEST Co., Cluj Napoca, Romania) [1].

Ferric sulphate, aqueous stock solution of 1 g/L, was used as coagulation agent, without or with addition of bentonite (0.5 g/L) (commonly without bentonite addition). As flocculation agent was tested a polymer, like PONILIT GT-2 anionic polyelectrolyte, the sodium salt of copolymer based on maleic acid and vinyl acetate patented by 'P.Poni' Institute of Macromolecular Chemistry, Iasi, Romania (1981), which was soluble in water and used in effluent treatment at a stock concentration of 0.5% (w/v) [15-16].

This anionic polyelectrolyte has been firstly produced by the Chemical Enterprise of Falticeni and purchased by CHEMICA Company, Bucharest, having the following properties: amber colour, specific smell, pH of 6.5–8, content of active product in solution of 33–36% (w/w), density of 1.18–1.21 g/cm^3, water soluble, viscosity at 20±1°C of 150–1800 cP, average molecular weight of $2 \cdot 10^5$–$3 \cdot 10^6$ g/mol, no corrosive or toxic effects [15].

Analysis methods

There were used internationally approved standard analysis methods for all quality indicators of treated or un-treated industrial effluents (*i.e.* simulated synthetic wastewaters and/or real final effluents from a bricks' manufacturing plant).

Colour determination. It was expressed by absorbance measurements at three wavelengths (436, 525, and 620 nm), obligatorily being absorbance at 436 nm, or Hazen units (HU) (*i.e.* 50 HU corresponds to an absorbance of 0.069 at 456 nm) [15-16].

Turbidity determination. It was directly measured at DRELL 2000 spectrophotometer (HACH Company) in formazine units (FTU), at 450 nm, using distilled water as blank, in accordance with test program #750 [15-16].

pH determination. It was measured directly at HACH One Laboratory pH meter.

Other determinations of studied quality indicators. The concentrations of chlorides, hardness, conductivity, suspended solids, or chemical oxygen demand were determined using internationally approved standards and reference materials, especially spectrophotometer-based analysis methods, adapted for the specific test programs, and reagent kits of DRELL 2000 spectrophotometer, HACH Company [17].

Treatment methodology of industrial effluent

The industrial effluent treatment technology was considered a mechanical-chemical process based on different steps like coagulation-flocculation (chemical step), achieved with coagulation agent (ferric sulphate solution, 7.5 mg Fe^{3+}/L, without or with optional addition of indigene bentonite, 0.5 g/L), and flocculation agent (Ponilit GT-2 anionic polyelectrolyte, 0.05-1.0 mg/L) under agitation (50 rpm) for no more than 45 min (usually, the testing agitation range was varied between 20-45 min), and after settlement of min 30 min (mechanical step) without agitation [15-16] (a batch experimental system is used). The supernatant was then analyzed in terms of turbidity, colour and other quality indicators in order to appreciate the treatment performance.

The removal of turbidity and colour is calculated with Eq.(1):

$$Y\,(\%) = \frac{C_i - C_f}{C_i} \cdot 100 \qquad (1)$$

where, Y - removal degree (for turbidity - Y_2, or for colour - Y_1), in %;
C_i - initial value of quality indicator, turbidity or colour, in FTU or HU;
C_f - final value of quality indicator, final turbidity or colour, in FTU or HU.

Neural modelling and optimization

ANN is a highly simplified model of decision-making and predictive process, which imitates the function of a human brain in processing information to understand the input–output relationships, as to be finally able to transform inputs into meaningful outputs [4-5]. The obtained knowledge is used then in prediction of response values.

In this study, a multilayers' perceptron neural network was utilized [18], consisting of identical neurons, interconnected and organized in layers, such as the outputs in one layer become the inputs in the subsequent layer. Data flow via the layer input passes through one or more hidden layers and exits finally via the layer output, being known generically as a feed-forward network. The hidden and output nodes of an MLP network contain an appropriate activation function, which calculates the node's output from the weighted input signals [18], such as a type of sigmoid or tangential hyperbolic functions, which is a family of S- or T-shaped functions.

The first step in the development of ANN model for the studied industrial effluent treatment consists of selection of ANN type and its optimal configuration (*i.e.* number of layers, number of processing elements as neurons in the layers, and transfer or activation functions). The number of elements in the input and output layers are equal to the number of input and output variables (responses) of the system to be studied, respectively. Therefore, it was considered for the studied industrial effluent treatment three input variables (temperature-z_1, polyelectrolyte dose-z_2 and agitation time-z_3), and one output variable of two types (Y_1-turbidity and Y_2-color removal). There are no strict rules to determine the optimal number of neurons in the hidden layer; this is normally done by a trial and error approach, with the initial number of neurons set equal to half the sum of the number of input and output neurons as a recommended starting point [18-19].

The second step consists of training the configured ANN by fitting the model represented by ANN to a set of given experimental data (resulted after laboratory simulated synthetic wastewater treatments in specific given operating conditions). In other words, the error function computed by comparing outputs of the ANN and real outputs of the effluent treatment system should be minimized. In cases where training was performed too long, or where experimental data used in training are rare, the configured ANN may adjust to very specific features of the training data, which have no causal relation to the target function. Therefore, the ANN is departing from the general structure of the target function to learning about the individual cases instead. This problem is well known as over-fitting or over-learning, when the general performance of the neural network (its performance on new data) is poor, even if the performance of the training data is acceptable [20]. To avoid over-fitting, the technology of 'cross verification' is applied. In this case, the available experimental data should be split in two sets: one is used for training (training set) and the other is used for cross verification (validation set), and during the training process, the performance of the validation set is observed as concurrent with the training set.

In other words, the artificial neural network is trained on the training set, and its performance verified using the validation set (real experimental data performed for an industrial effluent treatment system, a wastewater treatment plant of a bricks' manufacturing plant). If the predictive performance of the training set is opposite to that of the validation set, the process of training is stopped to avoid overfitting.

The third step involves testing the performance of the trained ANN model using a set of experimental data independent from the training data, obtained in the simulated and real WWTP, meaning a set of new input and output data (independent input and dependent output data). In this study, the performances of ANNs were also compared with respect to mean squared error (MSE), normalized mean squared error (NMSE), linear correlation coefficient (r), mean absolute error (MAE) and mean absolute percent error (% Error). These performance measures are defined in Eqs.(2-6) [21]:

$$\mathrm{MSE} = \sum_{i=0}^{N} \frac{(d_i - y_i)^2}{N} \tag{2}$$

$$\mathrm{NMSE} = \frac{\mathrm{MSE}}{N\sum_{i=0}^{N} d_i^2 - \left(\sum_{i=0}^{N} d_i\right)^2} \tag{3}$$

$$r = \frac{\dfrac{\sum_i (x_i - \bar{x})(d_i - \bar{d})}{N}}{\sqrt{\dfrac{\sum_i (d_i - \bar{d})^2}{N}}\sqrt{\dfrac{\sum_i (x_i - \bar{x})^2}{N}}} \tag{4}$$

$$\mathrm{MAE} = \frac{\sum_{i=0}^{N} |d_i - y_i|}{N} \tag{5}$$

$$\%\ Error = \frac{\left|\sum_{i=0}^{N} \dfrac{(d_i - y_i)}{d_i}\right|}{N} \cdot 100 \tag{6}$$

where, N is the total of training or testing experiments;
y_i is network output for experiment i;
d_i is desired output for experiment i;
x_i is network output;
$\bar{x}$ is average network output;
d_i is desired output, and $\bar{d}$ is the average desired output.

Once the ANN is trained the weights are then frozen, the testing set is fed into network, and the network output is compared with the desired output [22-26].

Results and discussion

The industrial effluents produced by a productive company, meaning a manufacturing unit of bricks and other ceramic products (as real industrial effluents), or prepared synthetic wastewaters based on the owner formulations related to manufacturing process, are characterized as in Table 1, considering some quality indicators like the total suspended solids, turbidity, conductivity, colour, chemical oxygen demand, total hardness and chlorides content, among others.

The experimental results after the proposed chemical-mechanical treatments (*i.e.* coagulation-flocculation and sedimentation) of synthetic wastewaters, or real effluent samples treated with coagulation agent (ferric sulphate, 7.5 mg Fe^{3+}/L, without or with optional addition of indigene bentonite, 0.5 g/L) and flocculation agent (PONILIT GT-2 anionic polyelectrolyte, 0.05-1.0 mg/L) were processed for training and testing of an artificial neural model (more than 110 experimental data for mechanical-chemical treatment of synthetic wastewaters or real effluents).

Table 1

Principal initial characteristics of studied industrial effluent.

Quality indicator	*Measurement units*	*Synthetic wastewater*	*Real effluent*
pH	-	6.5 - 7.0	6.83 ± 0.25
Conductivity	µS/cm	250 - 270	265 ± 20
Suspended solids	mg/L	95 - 500	112 ± 12
Turbidity	FTU	50 - 250	85 ± 5
COD	mg O_2/L	52.7 - 72.0	64 ± 10
Temporary hardness	°G	3.07 - 3.7	3.27 ± 1.2
Total hardness	°G	14.25 - 14.76	14.53 ± 3.5
Chlorides	mg/L	75.0 - 75.1	75.1 ± 10.5

The recommendation of such treatment system (mechanical-chemical treatment process, consisting of coagulation-flocculation followed by sedimentation) applied for industrial wastewaters produced in a manufacturing company of bricks and other ceramic products was found eligible and efficient, being reported in other authors' works [15-16] in which the experimental variation fields for the principal operating or desired input variables (*e.g.*, operating parameters and some chemicals doses) were found for the highest removals of turbidity (> 90%), colour (>80-90%) and COD (> 40-50%), considered as desired single outputs and calculated by processing the output data based on statistic formulations, or well-defined relationships as Eq.(1).

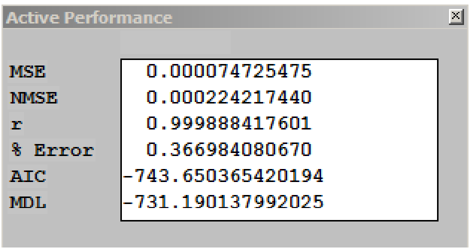
Thus, it was permitted the development of a neural model (*i.e.* neural model with a single hidden layer) by using an artificial neural network of multilayers perceptron type applied in the case of studied industrial effluent treatment, and its validation for real treatment data performed considering inputs from experimental variation field or overfitting and overtraining ones (from outside of experimental variation fields).

The ANN is developed in NeuroSolutions version 6.02 (Trial version). The transfer or activation function used for the hidden layer is of 'tangent hyperbolic axon' type (TANH). Error minimization is performed by using the algorithm Levenberg-Marquardt. It is worked with 100,000 epochs and the threshold of 10^{-10}. There were done 110 series of experiments, 80% for network training, the rest for testing (20%). The artificial neural network model and its validation for both Y_1 (turbidity) and Y_2 (colour) removal functions (real output data) are presented in Table 2 (*ANN Modelling* for Y_1 and Y_2), Figure 1 (*Training performance*), Figure 2 (*Cross validation performance*) and Figure 3 (*Testing performance for 10% of experimental data*).

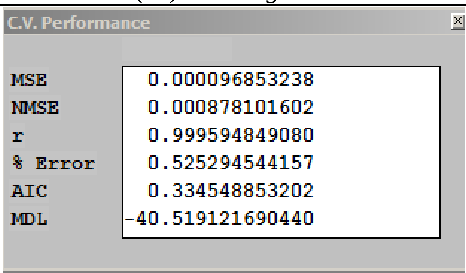
Table 2

Artificial neural network models for Y_1 (turbidity) and Y_2 (colour) removals.

(1) ANN Modeling for Y_1: MLP (3:10:1)

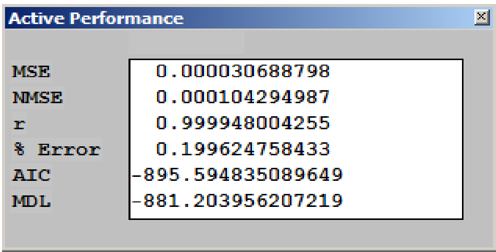


(1a) Training 10%

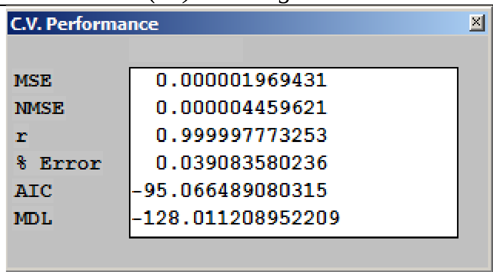


(1b) Cross Validation - CV 10%

(2) ANN Modeling for Y_2: MLP (3:10:1)



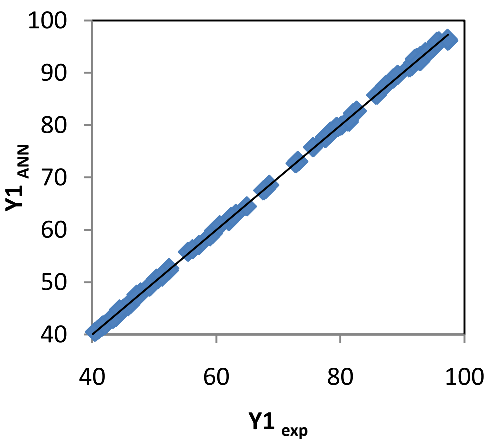
(2a) Training 10%



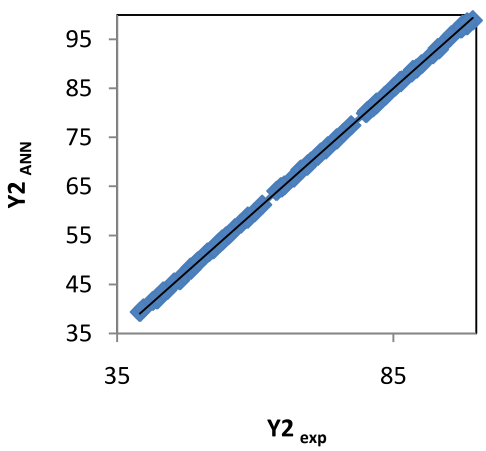
(2b) Cross Validation - CV 10%

For the Y_1 output (turbidity removal), the best neural model is MLP (3:10:1). The choice of model was done based on the performances obtained at the processing steps of training and cross validation such as $MSE_{training}$= 0.747·10^{-3} (mean squared errors for network training); $MSE_{validation}$= 0.969·10^{-3} (mean squared errors for network validation); linear correlation coefficient - $r_{training}$= 0.9999 (for network training) and $r_{validation}$= 0.9996 (for ANN model validation), and average relative error - $eRel_{av., training}$= 0.367% (for artificial network training) and $eRel_{av., validation}$= 0.525% (for ANN model validation).

Neural modelling of Y_2 output (colour removal) by using MLP model (3:10:1) leads to the following performances: $MSE_{training}$= 0.307·10^{-3}; $MSE_{validation}$= 0.002·10^{-3}; linear correlation coefficient - $r_{training}$= 0.9999 and $r_{validation}$= 0.9999; average relative error - $eRel_{av., training}$= 0.1996% and $eRel_{av., validation}$= 0.039%.

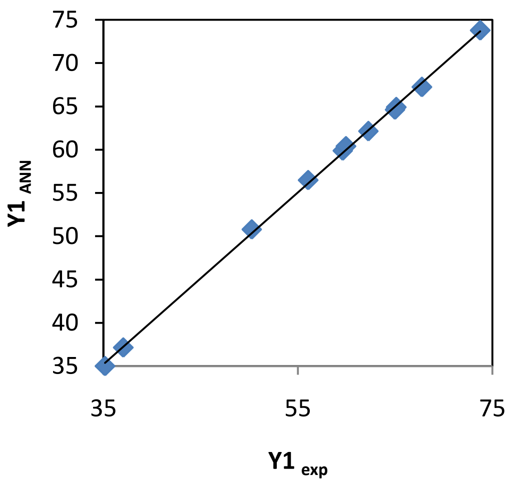


(a) $Y_{1, ANN}$ ($Y_{1, exp}$)

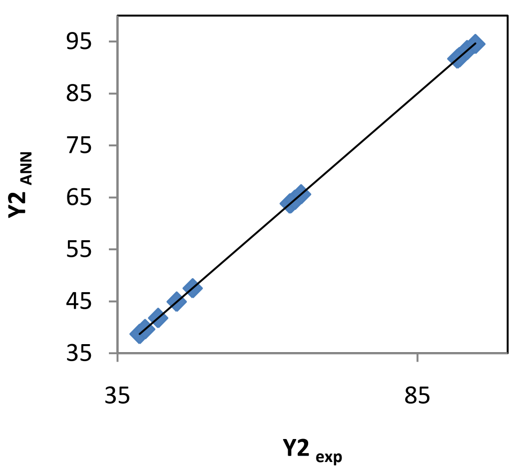


(b) $Y_{2, ANN}$ ($Y_{2, exp}$)

Figure 1. Training performance – Y_{ANN} (Y_{exp})**.**



(a) $Y_{1, ANN}$ ($Y_{1, exp}$)



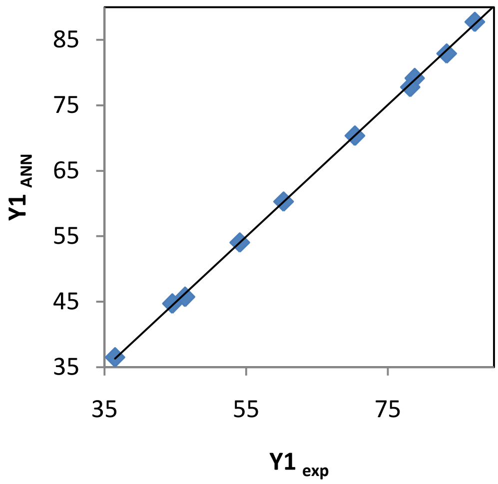
(b) $Y_{2, ANN}$ ($Y_{2, exp}$)

Figure 2. Cross validation performance – Y_{ANN} (Y_{exp})**.**

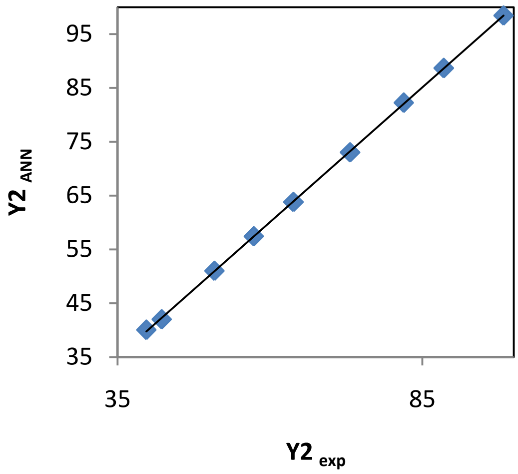
The obtained artificial neural models were applied in case of real Romanian treatment system (industrial scale setup, *i.e.* mechanical-chemical treatment system of industrial wastewaters produced by a manufacturing plant of bricks, Iasi, Romania) considering some input data from outside of experimental field (overfitting data) and obtained output results, in order to find the optimal input and output values in case of the real studied industrial effluent treatment. The effluent treatment optimization using the proposed ANN model leads to results summarized in Table 3.

The proposed optimum of neuronal MLP (3:10:1) model was verified through real experiments being found correspondent for the output values, meaning $Y_{1,\ opt}$ = 91.8% (for turbidity removal) and $Y_{2,\ opt}$ = 92.8% (for colour removal). Transposed in real input values, the optimal operating data (desired input data) are corresponded to a temperature of 18°C (z_1), a polymer dose of 2.0 mg/L (anionic polyelectrolyte PONILIT GT-2) (z_2) and an agitation time of 30 minutes (z_3) for turbidity removal (Y_1), and a temperature of 26 °C (z_1), a polyelectrolyte dose of 1.5 mg/L (z_2) and 30 minutes of agitation (z_3) for colour removal (Y_2), respectively.

The results experimentally verified for the industrial effluent treatment system (decentralized system) applied for industrial effluents of a Romanian manufacturing plant of bricks (Iasi, Romania) are adequate and can drive optimally the industrial effluent treatment in the corresponding operating conditions (as desired input data) for highest efficiencies in terms of turbidity and colour removals (as desired output data).



***(a)** $Y_{1,\ ANN}$ ($Y_{1,\ exp}$)*: **% Error= 0.366984**



***(b)** $Y_{2,\ ANN}$ ($Y_{2,\ exp}$)*: **% Error= 0.199624**

Figure 3. Testing (for 10% of experimental data) performance – $Y_{i,\ ANN}$ ($Y_{i,\ exp}$).

Table 3

Application of ANN model - MLP (3:10:1) for real input data, from outside of experimental field, and optimal output values found.

Y_1 (turbidity removal)				*Y_2 (colour removal)*			
z_1 (T, °C)	*$z_2 \cdot 10^{-1}$ (C_p, mg/L)*	*z_3 (t, min)*	*$Y_{1,\ ANN}$ (%)*	*z_1 (T, °C)*	*$z_2 \cdot 10^{-1}$ (C_p, mg/L)*	*z_3 (t, min)*	*$Y_{2,\ ANN}$ (%)*
5.5	12.5	20	24.9	5.5	12.5	20	32.6
10	5	10	40.6	10	5	10	39.8
10	20	25	51.9	10	20	25	60.5
24	20	10	74.6	24	20	10	82.3
26	15	30	89.9	***26***	***15***	***30***	***92.2***
21.6	10	20	80.5	21.6	10	20	90.9
17	15	20	67.3	17	15	20	71.9
13.5	10	20	56.4	13.5	10	20	58.7
15	10	20	60.9	15	10	20	65.0
18	***20***	***30***	***91.7***	18	20	30	91.1

Conclusions

Series of mechanical-chemical treatment experiments were performed for industrial effluents produced in a manufacturing plant of bricks and other ceramic products (simulated experiments with synthetic industrial effluents prepared at laboratory setup, as well as real industrial effluents at industrial setup) for finding an adequate artificial neural network (ANN) model for high removal of turbidity and colour (more than 80-90%), as desired output data.

Thus, it is proposed the ANN model – MPL 3:10:1 which was tested and validated as performance in statistic terms, being found very good for the industrial effluent treatment system, meaning industrial setup (error % = 0.191-0.364).

The optimal input and output values (desired values) are found for the mechanical-chemical treatment system (*i.e.* pre-treatment before discharging treated effluent in the local urban sewer system, based on coagulation-flocculation followed by sedimentation, applied in the real case of a Romanian manufacturing plant of bricks, Iasi, Romania), together with the experimental results from outside of training experimental field of ANN model (*i.e.* real results performed for input values from outside of experimental variation field of proposed ANN model) in order to achieve the highest colour and turbidity removals as most desired outputs.

The calculated results sustain the application of proposed ANN model for an industrial effluent treatment system (as mechanical-chemical treatment system) because of the very good industrial effluent treatment performance and minimization of any additional treatment cost.

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