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# Application of Artificial Intelligent Approaches for the Efficiency and Energy Consumption of a Novel Sonocatalyst

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**Original Article**

## Abstract

The sonocatalytic activity of nano-sized ZnO powder was studied via the degradation of the Direct Blue 71 azo dye. The nano-sized ZnO powder that was selected was the same as that which was synthesized and characterized in our previous study. The influences of six operational parameters including the initial pH, the initial concentration, the dose of sonocatalyst, the ultrasound frequency, the ultrasound power, and the process time were investigated on the basis of process efficiency and energy consumption. The design of experiments was applied and the experiments were conducted according to the design. The experiments were carried out in a batch reactor. The experimentally obtained dye removal percent (DR%) and the energy consumption per mass ranged from 0.03–100 and 0.19–1273 (wh/g), respectively. The data were used for modelling by using reduced quadratic multiple regression models and the artificial neural network (ANN). Multi-objective optimization of DR% and EPM was applied by using the genetic algorithm (GA) over the outperformed ANN models. The mineralization was studied using total organic carbon analysis. The study indicated promising results in the application of both the novel sonocatalyst and the Artificial Intelligent Approaches.

**KEYWORDS:** Sonocatalytic Degradation; Nano-ZnO; Direct Blue 71; Dye Removal; Design of Experiments; Artificial neural network; Genetic algorithm.

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## Introduction

Advanced oxidation processes (AOPs) have been developed and identified by many researchers to remove organic compounds from wastewater. Semiconductor photo-catalysis among AOPs is considered an important destructive technique. Among them, nano sized ZnO has been widely used as a photo-catalyst due to its high efficiency, non-toxic nature, and low cost.[1]However, this has some disadvantages. First, the cost of inducing the semiconductor catalyst with ultraviolet light is high. Second, the photo-catalyst has low efficiency in the presence of low translucent organic wastewaters. Third, a large number of aromatic compounds in some organic material categories, especially dyes, sometimes make customary photo-catalytic degradation methods ineffective for decolourization and mineralization. The final limiting factor is the low utilization efficiency of ultraviolet light due to the UV-screening effect of catalysts.[2]

In recent times, the sonocatalytic method has been regarded as a novel technology for treating wastewater. In this technology, low power ultrasonic irradiation is used as the exciting source to take the place of ultraviolet light irradiation. The sonocatalytic degradation

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of some organic pollutants in aqueous solution using $TiO_2$ or ZnO powder has been reported previously.[3, 4]

In this process, some parameters affect the process efficiency and energy consumption including the initial solution pH ($pH_0$), the initial concentration of pollutant ($C_0$), the dose of nano-sized catalyst ($D_{SC}$), the ultrasound irradiation frequency ($Fr_{SC}$), the ultrasound irradiation power ($P_{SC}$), and the treatment time ($t_{SC}$). The efficiency of the process may be increased by the optimization of these factors.[1, 5]

The aim of this study is to evaluate the efficiency of DR% and EPM for a solution of DB71during the SC process using synthesized nano-sized ZnO, followed by the optimization of the SC process in order to maximize DR% and to minimize the EPM with regard to the effective operational factors using GA based on the RQRM and the ANN models.

## Materials and Methods

Water type-one and distilled water were produced by a TKA Smart2Pure ultra-pure water production system (Thermo Electron LED GmbH, Germany). The DB71 ($C_{40}H_{23}N_7Na_4O_{13}S_4$) was purchased from Alvan Sabet Co., Iran. All other the reagents were purchased from Merck, Germany. The nano-sized ZnO powder that we selected was synthesized from and was characterized in our previous study.[6]

Some 200ml portions of the DB 71 synthetic effluent were prepared to investigate the sonocatalytic activity of nano-sized ZnO powder. The nano-sized ZnO powder was mixed with DB71 solutions. Then, the suspension was dispersed by vigorous stirring to make a homogeneous mixture of nano-sized ZnO particles. Dispersion was made in a dark place to prevent the initiation of probable photo-catalytic processes. Afterwards, this suspension was injected into an ultrasound apparatus and irradiated. In each run, the parameters consisting of $C_0$, $pH_0$, $Fr_{SC}$, $P_{SC}$, and $D_{SC}$ were set to desired values, and then, the SC was started. The SC and the dispersion were performed in a dark place. For each of the six levels of $t_{SC}$, 5 ml of the sample was extracted at the same position of the SC reactor using a 5-ml pipette. Each sample was centrifuged (10 min at 5000 rpm) alongside reference samples at $t_{SC}$=0. The DR% was calculated for the decanted solution.[7]

## Results and Discussion

The brief of the characterization information that was presented in the previous study[6] is presented here. The particle size distribution analysis showed that the particles are uniform with an average size of 888 nm. The SEM analysis shows that the nano-sized ZnO powders are agglomerated with a size around 800 nm, which is in agreement with the particle size distribution analysis results. The results of zeta potential analysis in the water media showed a zeta potential equal to 8.59 mv, which belonged to low stable particles within the water media. The XRD and EDX analysis results showed up to 99.6% purity with less than 0.4% Cu impurity.[8] The UV-Vis spectra were used for energy gap determination. Based on the UV-Vis spectra analysis, the energy gaps were calculated as 3.41 and 3.39 ev in water and ethanol, respectively.

**Sonocatalysis Process**

Figures 1 and 2 show the whole 108 experimental DR% and the EPM data, respectively. Based on the figures, the nano-sized ZnO powder is a promising sonocatalyst. In addition, the figures illustrate that different levels of experimental parameters result in different DR% and EPM.

**Mineralization studies using TOC and UV-Vis**

The decay of total organic compound(TOC) reflects the extent of mineralization in an organic compound; the reduction percent of TOC was studied for dye samples ($C_0$=120 mg/L, $D_{SC}$=1.9 g/L, P=91 w, pH=1.9, Fr=37 kHz) as a function of irradiation time. The control sample was considered for the removal of a possible trace of acetate from the TOC in nano-sized zinc oxide. It was observed that the TOC reduction percentage in the main sample lies between 223 and 155 in a period of 5 and 60 minutes, respectively; the TOC reduction percentage is 160 and 174 for the control samples when the TOC of 120 mg/l DB71 was 50 (Figure 3). Both the spectroscopy and the TOC analyses showed that at $t_{SC}$ = 20 minutes, the degradation and colour fading was complete. This means that the results of the TOC analysis are in accordance with the spectroscopy analysis. In the TOC analysis, the reduction of the TOC was continued even after

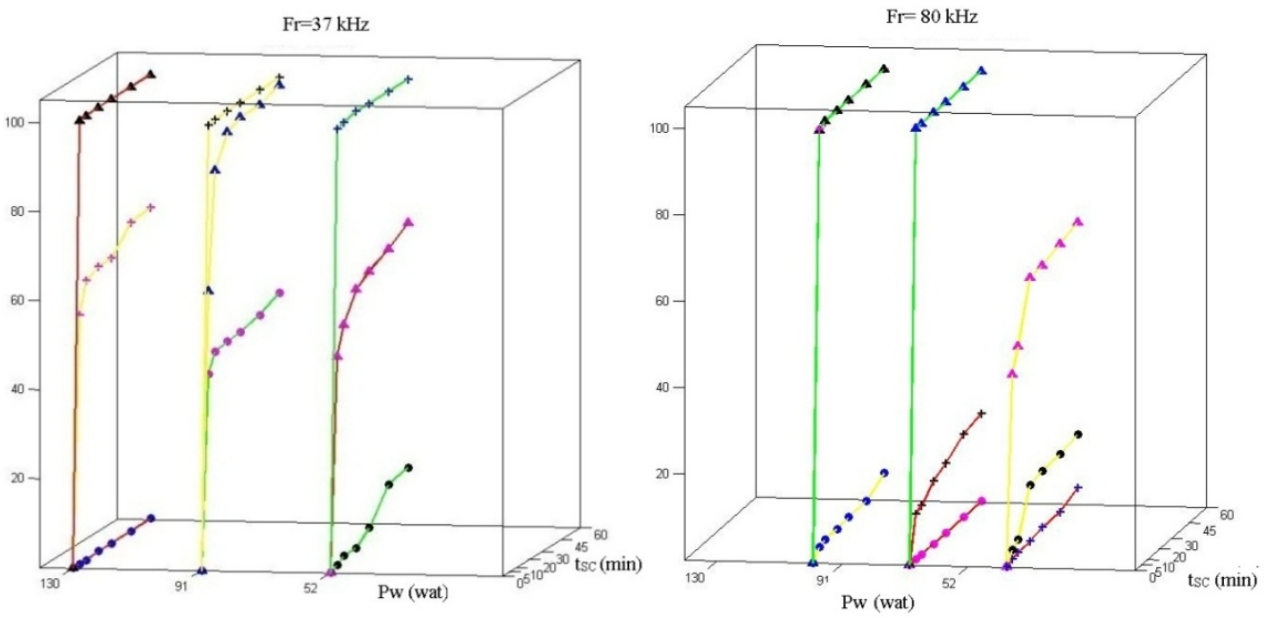


**Figure 1. DR% of 18 complete runs (green line (C0=20 mg/Lit), yellow line (C0=120 mg/Lit), red line (C0=220 mg/Lit)/ circle marker (DSC=0.1 g/Lit), plus sign marker (DSC=1 g/Lit), triangle marker (DSC=1.9 g/Lit)/ black marker (pH=3), magenta marker (pH=7), and blue marker (pH=10)).**

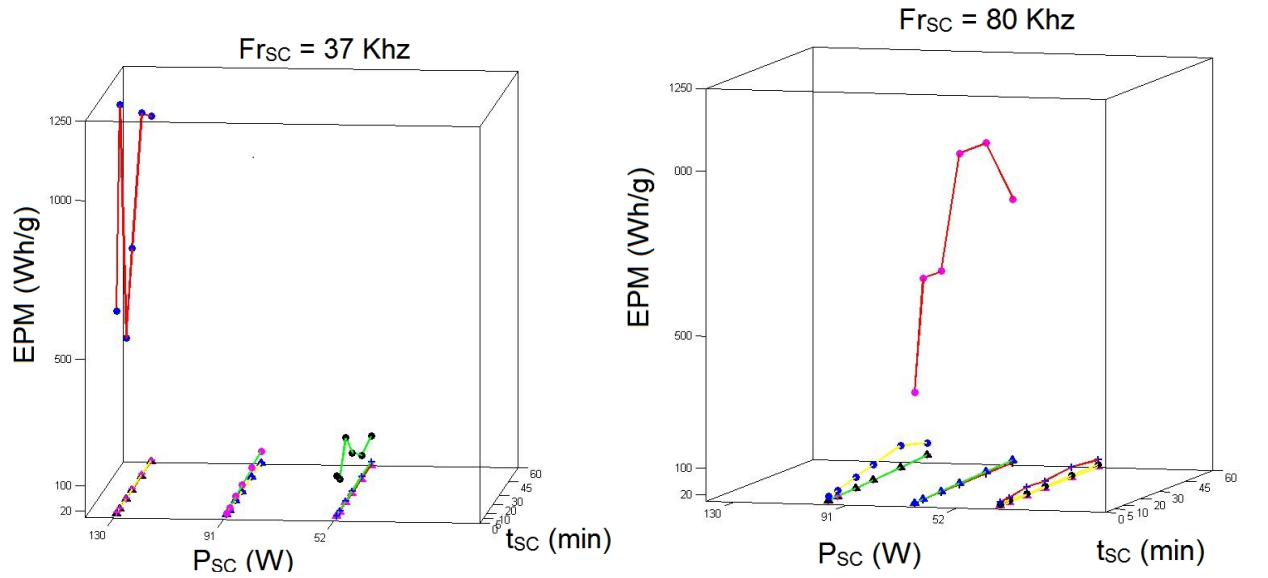


**Figure 2. EPM of 18 complete runs (green line (C0=20 mg/Lit), yellow line (C0=120 mg/Lit), red line (C0=220 mg/Lit)/ circle marker (DSC=0.1 g/Lit), plus sign marker (DSC=1 g/Lit), triangle marker (DSC=1.9 g/Lit)/ black marker (pH=3), magenta marker (pH=7), and blue marker (pH=10)).**

the TOC received the control sample TOC. This means that the sonocatalytic process degraded organic materials such as the residual acetate and this reduction lasted for an hour. The increase of the TOC in the control sample is due to the ultrasonic irradiation that stimulates the release of residual organic materials like acetate from the surface and the pores of nano-particles. The same results for the TOC and spectroscopy also mean that the dye removal is equal to mineralization in this study.

Figure 3 shows the UV–Vis spectra of DB71 under the same TOC experiment conditions. It was found that the characteristic absorption peaks of DB71 solutions declined under ultrasonic irradiation in the presence of nano-sized ZnO powder. This indicates that the DB71 has mostly decomposed and disappeared gradually in the aqueous solution.

**ANN model**

The best ANN model was separately constructed for each DR% and EPM because of different data sets. In the case of DR%, one hidden layer with 8 neurons and a learning rate of 0.13 were applied. The EPM model was constructed based on 5 neurons for only one hidden layer. The ‘tansig’ transfer function

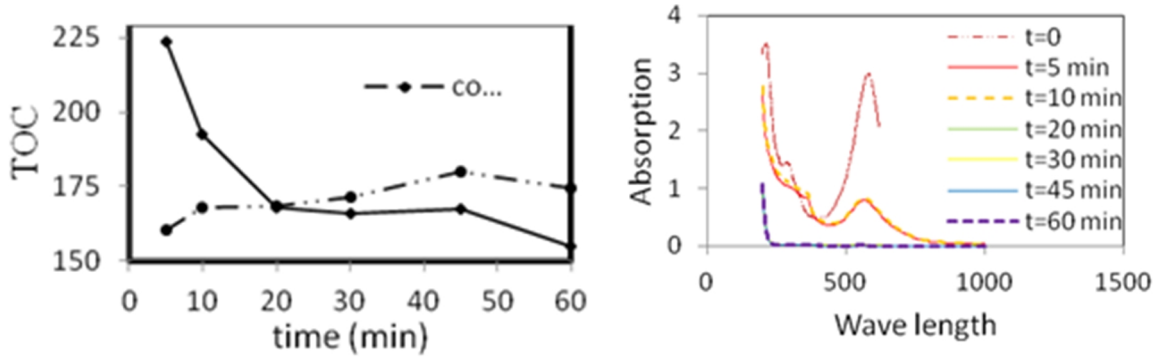


Figure 3. Sample of TOC and absorption removal during SC for the main and the control samples.

was selected for input and the hidden layer, and 'purelin' function for the output in both models.[9] Once the networks were trained, the weights and biases of each neuron and layer were saved in the ANN model. Then, they were used to estimate the test set. The (6:8:1) ANN for DR% and the (6:5:1) ANN for EPM were trained using the 75 and the 64 data of the train set for DR% and EPM models, respectively, by the backpropagation algorithm. Table 1 presents the statistical parameters of the ANN models for DR% and EPM, respectively.[10, 11]

Finally, the ANN models would be used in the optimization procedure by GA.

**GA multi-objective optimization**

Multi objective optimization process results in a Pareto-optimal solution set. Each point on the Pareto set is associated with a set of decision variables. Furthermore, the input decision variables corresponding to each of the Pareto optimal solutions were tabulated in Table 2. Interpretation of Figures 1–2 and Table 2 were used to discriminate the effect of each investigated parameter.[12]

**Influences of solution pH on the DR% and the EPM**

It is generally well known that the pH values markedly influence the degradation of organic pollutants in the photo-catalytic degradation reaction. Thus, the effects of pH values were studied at three levels of ultrasonic degradation in the range of 3–10. As can be seen from Table 2, the optimum value of $pH_0$ in all Pareto-optimal solutions is in the range of 3–5. In general, the point of zero charge (PZC) of nano-ZnO powder is around pH = 9. Above this pH, the surface of nano-sized ZnO particles is negatively charged, while below this pH, it is positively charged.

Table 1. Statistical characteristics of both ANN models for DR% and EPM

| Model | DR% | | | EPM (wh) | | |
|---|---|---|---|---|---|---|
| Data set | Train | Validation | Test | Train | Validation | Test |
| $R^2$ | 1 | 0.98 | 0.98 | 0.99 | 0.92 | 0.90 |
| $Q^2$ | 1 | 0.98 | 0.98 | 0.92 | 0.91 | 0.90 |
| RMSE | 1.6 | 5.8 | 5.2 | 72 | 83 | 12 |

Some ionizable compounds such as DB71 (negatively charged) after ionization can be easily adsorbed on the positively charged surface of nano-sized particles below pH = 9, which is propitious to the degradation of DB71. In basic mediums, the adsorption ability of nano-sized ZnO powder decreases as DB71 has negative charge.[1]

Effect of $C_0$ on the DR% and EPM As presented in Table 2, the optimum value of $C_0$ is two values (50 and 206 mg/L). In Table 2, for both cases with 50 mg/L concentration, a DR% of more than 98% was achieved, whereas for the other 26 cases, more concentration of $C_0$ caused lower DR% until it reached 1%. The two following reasons, perhaps, cause the decrease of DR%. Firstly, the adsorption amounts of nano-sized ZnO powder attain saturation for superfluous DB71. Secondly, the mutual screens among the DB71 molecules also increase along with the rising concentration of DB71.[5]

Influences of $Fr_{SC}$ on the DR% and EPM

The influence of ultrasonic frequency on the DR% of DB71 was studied using two different frequencies, 37 and 80 kHz. The optimum value of $Fr_{SC}$ was almost 50–70 kHz (Table 2).[5,13]

Influences of a dose of sonocatalyst on efficiency and energy consumption Table 2,

energy consumption. The experiments performed with different $D_{SC}$ showed that the sonocatalytic degradation trend of DB71 tempestuously increases with the increase of nano-sized ZnO powder up to 1.0 g/L. The optimum value of $D_{SC}$ was almost 1.3–1.8 g/L (Table 2). More $D_{SC}$ caused mutual screens among nano-sized ZnO particles, which resulted in the decrease of sonocatalytic activity of the ZnO nano-sized powder .[14]

Influences of ultrasonic radiation power on efficiency and energy consumption Table 2 shows that more ultrasonic radiation power brings about greater process efficiency. It is clear that more ultrasonic power provides more hydroxyl radicals and thermal cavities as a motive in the process. In addition, more ultrasonic power makes the sonocatalyst more dispersed and stable in the solution. Based on the optimization process results, the optimum values for $P_{SC}$ was about 95 watts in all conditions. However, more ultrasonic power caused more degradation, but it also caused greater energy consumption. The optimum of the parameters was found to be about 95 watts by the genetic algorithm.

Table 2. Variables values corresponding to each of the Pareto-optimal solution

| solutions | $pH_0$ | $C_0$ | $Fr_{SC}$ | $D_{SC}$ | $P_{SC}$ | $t_{SC}$ | DR% | EPM | solutions | $pH_0$ | $C_0$ | $Fr_{SC}$ | $D_{SC}$ | $P_{SC}$ | $t_{SC}$ | DR% | EPM |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | 3.2 | 207 | 51 | 1.7 | 94 | 2.6 | 8 | 0.19 | 15 | 3.2 | 207 | 51 | 1.7 | 94 | 3.2 | 13 | 0.19 |
| 2 | 3.2 | 207 | 64 | 1.9 | 96 | 2.6 | 64 | 0.19 | 16 | 3.2 | 207 | 63 | 1.9 | 96 | 1.5 | 30 | 0.19 |
| 3 | 3.3 | 206 | 64 | 1.9 | 96 | 6.0 | 88 | 0.19 | 17 | 3.2 | 207 | 64 | 1.9 | 96 | 2.6 | 60 | 0.19 |
| 4 | 3.2 | 207 | 70 | 1.9 | 96 | 3.8 | 92 | 0.19 | 18 | 3.2 | 207 | 64 | 1.9 | 96 | 3.8 | 78 | 0.19 |
| 5 | 3.2 | 207 | 51 | 1.7 | 94 | 3.2 | 13 | 0.19 | 19 | 3.2 | 207 | 51 | 1.7 | 94 | 1.5 | 2 | 0.19 |
| 6 | 3.1 | 207 | 51 | 1.8 | 94 | 0.2 | 0 | 0.19 | 20 | 3.2 | 207 | 64 | 1.9 | 96 | 4.4 | 82 | 0.19 |
| 7 | 4.8 | 51 | 50 | 1.3 | 96 | 5.9 | 100 | 0.19 | 21 | 4.8 | 51 | 51 | 1.7 | 94 | 5.9 | 99 | 0.19 |
| 8 | 3.2 | 207 | 70 | 1.9 | 96 | 5.9 | 95 | 0.19 | 22 | 3.2 | 207 | 63 | 1.9 | 96 | 3.1 | 70 | 0.19 |
| 9 | 3.2 | 207 | 64 | 1.9 | 96 | 4.4 | 82 | 0.19 | 23 | 3.2 | 207 | 51 | 1.7 | 94 | 2.3 | 6 | 0.19 |
| 10 | 3.2 | 207 | 64 | 1.9 | 96 | 1.3 | 26 | 0.19 | 24 | 3.2 | 207 | 64 | 1.9 | 96 | 2.6 | 61 | 0.19 |
| 11 | 3.2 | 207 | 64 | 1.9 | 96 | 5.2 | 86 | 0.19 | 25 | 3.1 | 207 | 51 | 1.7 | 94 | 3.8 | 18 | 0.19 |
| 12 | 3.3 | 206 | 64 | 1.9 | 96 | 3.1 | 72 | 0.19 | 26 | 3.3 | 207 | 70 | 1.9 | 96 | 3.8 | 92 | 0.19 |
| 13 | 3.2 | 207 | 64 | 1.9 | 96 | 2.3 | 55 | 0.19 | 27 | 3.2 | 207 | 64 | 1.9 | 96 | 2.3 | 57 | 0.19 |
| 14 | 3.2 | 207 | 63 | 1.9 | 96 | 1.3 | 26 | 0.19 | 28 | 3.2 | 207 | 64 | 1.9 | 96 | 1.5 | 32 | 0.19 |

Influences of processing time on efficiency and energy consumption

Figures 1 and 2 demonstrate the influences of process time on the process efficiency and energy consumption. Across the whole experiment, the process efficiency increased rapidly by an increase in process time in the early minutes, and then, levelled off in less than 10 minutes. This indicates the fast kinetic of the sonocatalytic process as a promising process for the removal of organic materials from wastewaters. It is recognized that more process time causes more oxidative products, which causes more degradation of organic materials.

## Conclusion

The aqueous solutions of DB71 can be decoloured, degraded, and mineralized by the sonocatalytic technique in the presence of simple synthesized nano-sized ZnO powder. This study demonstrates that the considered operational parameters have a significant influence on the process efficiency. The artificial intelligent systems were successfully applied for multi-factorial modelling, the optimization of process efficiency and energy consumption. The multi-objective optimal solution was found as $pH_0$ 3–5, $C_0$ 50 and 220 mg/L, $D_{SC}$ 1.3–1.9 g/L, $Fr_{SC}$50–70 kHz, and $P_{SC}$96 W. This study indicated promising results regarding the application of both the novel sonocatalyst and the Artificial Intelligent Approaches.

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## References

1. Wang J, Jiang Z, Zhang Z, Xie Y, Wang X, Xing Z, et al. Sonocatalytic degradation of acid red B and rhodamine B catalyzed by nano-sized ZnO powder under ultrasonic irradiation. Ultrasonics sonochemistry. 2008;15(5):768-774.
2. Wang J, Guo B, Zhang X, Zhang Z, Han J, Wu J. Sonocatalytic degradation of methyl orange

in the presence of $TiO_2$ catalysts and catalytic activity comparison of rutile and anatase. Ultrasonics sonochemistry. 2005;12(5):331-337.
2. Turner C, Van Hook A. The effect of ultrasonic irradiation on the formation of colloidal sulfur and ice. Journal of colloid science. 1950;5(4):315-316.
3. Mahvi AH, Maleki A. Photosonochemical degradation of phenol in water. Desalination and Water Treatment. 2010;20(1-3):197-202.
4. Wang J, Jiang Z, Zhang L, Kang P, Xie Y, Lv Y, et al. Sonocatalytic degradation of some dyestuffs and comparison of catalytic activities of nano-sized $TiO_2$, nano-sized ZnO and composite $TiO_2$/ZnO powders under ultrasonic irradiation. Ultrasonics sonochemistry. 2009;16(2):225-231.
5. Daraei H, Maleki A, Mahvi AH, Zandsalimi Y, Alaei L, Gharibi F. Synthesis of ZnO nano-sono-catalyst for degradation of reactive dye focusing on energy consumption: operational parameters influence, modeling, and optimization. Desalination and Water Treatment. 2014;52(34-36):6745-6755.
6. Maleki A, Safari M, Shahmoradi B, Zandsalimi Y, Daraei H, Gharibi F. Photocatalytic degradation of humic substances in aqueous solution using Cu-doped ZnO nanoparticles under natural sunlight irradiation. Environmental Science and Pollution Research. 2015;22(21):16875-16880.
7. Srikanth CK, Jeevanandam P. Effect of anion on the homogeneous precipitation of precursors and their thermal decomposition to zinc oxide. J Alloy Compd. 2009;486(1-2):677-684.
8. Hatami T, Rahimi M, Daraei H, Heidaryan E, Alsairafi AA. PRSV equation of state parameter modeling through artificial neural network and adaptive network-based fuzzy inference system. Korean Journal of Chemical Engineering. 2012:1-11.
9. Maleki A, Daraei H, Khodaei F, Bayazid-Aghdam K, Rezaee R, Naghizadeh A. Investigation of potato peel-based bio-sorbent efficiency in reactive dye removal: Artificial neural network modeling and genetic algorithms optimization. Journal of Advances in Environmental Health Research. 2013;1(1):21-28.
10. Maleki A, Daraei H, Shahmoradi B, Razee S, Ghobadi N. Electrocoagulation efficiency and energy consumption probing by artificial intelligent approaches. Desalination and Water Treatment. 2014;52(13-15):2400-2411.
11. Hatami T, Rahimi M, Daraei H, Heidaryan E, Alsairafi AA. PRSV equation of state parameter modeling through artificial neural network and adaptive network-based fuzzy inference system. Korean Journal of Chemical Engineering. 2012;29(5):657-667.
12. Cheng Z, Quan X, Xiong Y, Yang L, Huang Y. Synergistic degradation of methyl orange in an ultrasound intensified photocatalytic reactor. Ultrasonics sonochemistry. 2012;19(5):1027-1032.
13. Wang J, Sun W, Zhang Z, Zhang X, Li R, Ma T, et al. Sonocatalytic degradation of methyl parathion in the presence of micron-sized and nano-sized rutile titanium dioxide catalysts and comparison of their sonocatalytic abilities. Journal of Molecular Catalysis A: Chemical. 2007;272(1):84-90.
14. Eren Z, Ince NH. Sonolytic and sonocatalytic degradation of azo dyes by low and high frequency ultrasound. Journal of hazardous materials. 2010;177(1):1019-1024.