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Synthetic Textile Wastewater Treatment using Potassium Ferrate(VI) – Application of Taguchi Method for Optimisation of Experiment

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Abstract

The article attempts to assess the usefulness of the Taguchi method to optimise the purification process of synthetic textile wastewater (pH 6.7 - 7.2, Conductivity = 6.71 - 6.84 mS/cm, Salinity = 3581 - 3648 mg NaCl/l, Colour = 560 - 4710 mg Pt/l, COD = 2220 - 2290 mg O₂/l, TOC = 394 - 551 mg/l) using K₂FeO₄. The research was conducted using 3 types of wastewater containing anionic detergent (sodium lauryl sulfate, 100 mg/l) and differing only in the concentration of azo dye Acid Green 16 (AG 16). Technical K₂FeO₄ was used as an oxidiser, which was subjected to physico-chemical analysis (purity, UV-VIS spectrum, surface characteristics and chemical composition using SEM and EDX methods). For planning and optimising the wastewater treatment process, the Taguchi method was used for four input parameters: pH (2, 7, 12), reaction time (10, 30, 50 min), AG 16 concentrations (20, 120, 220 mg/l) and K₂FeO₄ concentrations (25, 125, 225 mg/l), for which 9 experiments were performed in accordance with the plan adopted. Test result analysis allowed to indicate the optimal values for individual input parameters (pH 2, time = 50 min, AG 16 = 20 mg/l, K₂FeO₄ = 125 mg/l). Under these conditions, visual discoloration of wastewater was obtained (AG 16 = 0.4 mg/l, \\$98%), colour removal (66 mg Pt/l, \\$8%) and DOC (249 mg/l, \\$37%).

Key words: textile wastewater, potassium ferrate(VI), Acid Green 16, Taguchi method.

[5, 6]. The diversity of chemical substances present in wastewater and the low susceptibility of many of them (including dyes) to biodegradation cause many technological problems during the treatment of dyeing wastewater [7]. Physical (sedimentation, filtration, flotation), chemical (chlorination, ozonation, neutralization, coagulation) and biological (activated sludge, aerobic and anaerobic digestion) processes are used for the purification of the wastewater, most often purification methods which combine several processes. Textile wastewater treatment is usually carried out in stages in which certain groups of pollutants are removed, with increasingly sophisticated technologies being used. Generally these processes include three stages: primary treatment (removal of suspended solids, oil, grit etc.), secondary treatment (removal of BOD, colour, oil and phenols) and tertiary treatment (use of electrodialysis, ion exchange, reverse osmosis for the final removal of chemical pollution, purification of the wastewater or the reuse of water) [8]. Classic wastewater treatment processes (neutralisation, coagulation and flocculation) are characterised by low effectiveness and do not allow to achieve complete removal (degradation) of dyes and other organic compounds. In order to solve these problems, Advanced Oxidation Processes (AOPs) are used, which use free radicals generated by various techniques. These methods proved

to be effective in the purification of many types of industrial wastewater [9, 10], including dye wastewater [11-14]. A very promising oxidant that is being used to treat industrial wastewater is K₂FeO₄, which contains Fe(VI) and thus has strong oxidising properties ($E^{o} = 2.20 \text{ V}$ in acidic media and $E^{o} = 0.72 \text{ V}$ in basic media). As a result of contact with impurities present in wastewater, Fe(VI) ions are reduced to Fe(III) ions, which causes the precipitation of Fe(OH)₃ in an aqueous environment. Therefore K₂FeO₄ can simultaneously act as an oxidiser and coagulate, and thus be an alternative to current methods of water and wastewater treatment [15-17]. Potassium ferrate(VI) was used to remove azo dye Orange II (removal of 62% of colour after 30 min) [18], Active Brillant Red X-3B (removal of 99% of colour, 42% of COD and 9% of TOC) [19] and for wastewater treatment containing azo and anthraquinone dyes (removal of 87% of colour) [20]. One of the conditions for obtaining high efficiency of wastewater treatment is to conduct the process under optimal conditions. Methods of planning and optimising experiments, including the Taguchi method, are used for this purpose. This method is a multi-parameter statistical optimisation technique that uses a small number of experiments to identify and optimise parameters and obtain the desired response [21, 22]. The Taguchi method was used, among others, to op-

Introduction

The textile industry produces wastewater that contains a number of soluble and insoluble substances, including unreacted dyes and other substances used at particular stages of the production process. The effective use of dyes in the production process is sometimes less than 60%. The remaining part, along with other substances, gets into the wastewater, which increases the values of BOD, COD, TDS (total dissolved solids) and others parameters. An additional problem is the low susceptibility of dyes to biodegradation [1-4]. Technological processes of textile production are characterised by a significant consumption of dyes, detergents and many other substances, as a result of which the consequent wastewater is characterised by a very high pollution degree

timise the electrocoagulation process and chemical coagulation of dye wastewater [23, 24], as well as to optimise the course of a Fenton reaction [25]. The aim of the research presented was to use the Taguchi method to optimise the process of removing Acid Green 16 dye (AG 16) from synthetic wastewater with the use of K₂FeO₄. Experimental verification of the process efficiency was performed using the parameters indicated by the Taguchi method as optimal (pH, reaction time, AG 16 concentration, K₂FeO₄ concentration), for which the concentration of AG 16, colour and DOC concentration in purified wastewater was determined.

Materials

In tests, technical grade K₂FeO₄ (Envifer, Nano Iron, s.r.o., Czech Republic) containing 40.2% K₂FeO₄ (CAS 13718-66-6, 198.04 g/mol) was used. Three samples of synthetic textile wastewater (Table 1) were prepared based on literature data, which present the composition of real textile wastewater with a total dye concentration of 20, 120 and 220 mg/l [26]. To prepare synthetic textile wastewater, Acid Green 16 (AG 16, triarylmethane class, CAS 3369-56-0, C.I. 44025, 616.73 g/mol, C₃₁H₃₃N₂NaO₆S₂, Boruta-Zachem S.A., Poland) was used, which is harmful to aquatic life, with long lasting effects [27, 28]. AG 16 is also a typical leather dye, and its colour and structure strongly depend on pH [29]. In the research presented AG 16 was used without further purification. Moreover sodium lauryl sulfate (Sigma-Aldrich, Germany), starch ((C₆H₁₀O₅)_n), acetic acid (CH₃COOH, 99.5%), sucrose (C₁₂H₂₂O₁₁), sodium hydroxide (NaOH), sulfuric acid (H₂SO₄, 98%), sodium carbonate (Na₂CO₃) and sodium chloride (NaCl) (Chempur, Poland) were used, which were dissolved in demineralised water with a total hardness of 0.5 °dH. Sodium sulfite (Na₂SO₃) 0.5 mol/l) was used to quench the K₂FeO₄ oxidation reaction. For pH correction, 25% H₂SO₄ and NaOH solutions were applied. Analytical grade reagents and distilled water were used in tests, with the exception of demineralised water, used for the preparation of wastewater samples.

Analytical methods

The pH value, conductivity and salinity were determined using inolab® pH/Ion/

Cond 750 and an SenTix® 81 electrode (WTW, Germany) [30]. The colour (by 405 nm) was determined using a PF-11 spectrophotometer (Macherey-Nagel, Germany) after filtering the sample (0.45 µm) [31]. COD was determined by the dichromate method using sealed tubes and the PF-11 spectrophotometer [32]. DOC was determined after filtering the sample (0.45 µm) using a Nanocolor® TOC 60 and the PF-11 spectrophotometer (Macherey-Nagel, Germany). Determination of DOC was carried out in two steps which included the disposing of inorganic carbon (NaHSO₄, 500 r.p.m., 10 min), decomposition of organic carbon (Na₂S₂O₈, 150 °C, 120 min), and detection of carbon dioxide formed by means of an indicator (Thymol Blue, sodium salt solution). The concentration of AG 16 in synthetic and purified wastewater was determined using a standard curve $(\lambda = 640 \text{ nm}, R^2 = 0.9997)$ and a UV-VIS spectrophotometer (Cary® 50 UV-VIS, Varian Inc., Australia). The concentration of K₂FeO₄ was determined using the chromite titration method based on oxidation in the strongly alkaline environment of ions CrO₄ by FeO₄ ions with the creation of Fe(OH)₃, CrO $_4^{2-}$ and OH -. Dichromate ions were titrated with a standard solution of ferrous ions. The K₂FeO₄ content in % in Envirer was calculated as *Equation (1)*:

$$= \frac{C_{Fe(II)} \cdot V_{Fe(II)} \cdot M_{K2FeO4}}{3000 \cdot m_{sample}}$$
(1)

where, C_{Fe(II)} and V_{Fe(II)} are the concentration and volume of standard ferrous ammonium sulphate solution (0.085 mol/l and ml, respectively), M_{K2FeO4} is 198.04 g/mol, and m_{sample} represents the weight of the sample in g [33]. The UV-VIS spectrum (180 -1100 nm, 0.035 g of Envifer product in 3 mol/l KOH) and content of K₂FeO₄ in technical grade K₂FeO₄ (by 505 nm) were determined using a UV-VIS spectrophotometer (Cary® 50 UV-VIS, Varian Inc., Australia). In order to determine the concentration of K₂FeO₄ in Envifer, the modified method set out in [34] was also used. A specific amount of the product was dissolved in distilled water, up to 100 ml in a volumetric flask, filtered (0.45 µm) into a quartz cell (the light path of the quartz cell was 10 mm), and the absorbance was measured at 505 nm. In this case, the K₂FeO₄ content in % in Envirence was calculated as *Equation (2)*:

% of K₂FeO₄ =
$$= \frac{A \cdot 0.1 \cdot M_{K_2FeO_4} \cdot 100\%}{1070 \cdot m_{sample}}$$
(2)

where, A is the absorbance at 505 nm, $M_{\mbox{\scriptsize K}_{2}\mbox{\scriptsize FeO}_{4}} - 198.04$ g/mol, 1070 – the mo-

Table 1. Chemical composition of synthetic textile wastewater [26].

Materials used	Concentration, mg/l	Function
Acid Green 16	20, 120, 220	Coloring agent
Sodium lauryl sulphate, SLS	100	Scouring agent
Sucrose, C ₁₂ H ₂₂ O ₁₁	600	Sizing agent
Starch, (C ₆ H ₁₀ O ₅) _n	1000	Sizing agent
Sodium hydroxide, NaOH	175	Hydrolysis
Sulfuric acid, H ₂ SO ₄	300	pH regulator
Sodium carbonate, Na ₂ CO ₃	500	Fixing agent
Sodium chloride, NaCl	3000	Fixing agent
Acetic acid, CH ₃ COOH	200	Sizing agent

Table 2. Physicochemical parameters of synthetic textile wastewater. *the reading accuracy of the analytical balance was adopted, ± 1 mg.

Parameter	Physicochemical parameters of wastewater (Acid Green 16 stock soln., mg/l) (value ± expanded uncertainty)			
	20	120	220	
pH	6.7 ± 0.1	6.9 ± 0.1	7.2 ± 0.1	
Conductivity, µS/cm	6710 ± 336	6790 ± 340	6840 ± 342	
Salinity, mg NaCl/l	3581 ± 179	3628 ± 181	3648 ± 182	
Colour, mg Pt/I	560 ± 112	2540 ± 508	4710 ± 942	
Concentration of Acid Green, mg/l	20 ± 1*	120 ± 1*	220 ± 1*	
COD, mg O ₂ /l	2230 ± 335	2270 ± 341	2295 ± 344	
DOC, mg/l	394 ± 51	433 ± 65	551 ± 83	

Table 3. Experimental conditions (factors and levels of an orthogonal array) and results for synthetic textile wastewater. *the Envifer dose for pure K_2FeO_4 was calculated, **in optimal conditions: pH 2.0, time = 50 min, AG 16 = 20 mg/l, K_2FeO_4 = 125 mg/l, ***slightly yellow.

		E	xperimental conditions	Experimental results			
Run	Factor 1	Factor 2	Factor 3	Factor 4	Physico-chemical parameters of wastewater (value ± expanded uncertainty)		
	pН	Time, min	Concentration of AG 16	Concentration of K ₂ FeO ₄ *	AG 16, mg/l	Colour mg Pt/I	DOC, mg/l
1	2.0	10	20	25	1.7 ± 0.4	250 ± 50***	289 ± 43
2	2.0	30	120	125	21.6 ± 4.3	1240 ± 248	307 ± 46
3	2.0	50	220	225	66.5 ± 13.3	2860 ± 572	331 ± 50
4	7.0	10	120	225	24.3 ± 4.9	2340 ± 468	284 ± 43
5	7.0	30	220	25	211.1 ± 42.2	4840 ± 968	378 ± 57
6	7.0	50	20	125	1.2 ± 0.3	640 ± 128***	276 ± 41
7	12.0	10	220	125	105.3 ± 21.1	3720 ± 744	307 ± 46
8	12.0	30	20	225	11.3 ± 2.3	1570 ± 314	268 ± 40
9	12.0	50	120	25	46.4 ± 9.3	1530 ± 306	291 ± 44
10**	2.0	50	20	125	0.4 ± 0.1	66 ± 13	249 ± 37

lar absorbance coefficient – M-1cm-1, and m_{sample} represents the weight of the sample in g. K₂FeO₄ morphology studies were performed using the SEM/EDX method with an EVO MA 15 microscope (Carl Zeiss, Germany) equipped with a Quantax X-ray dispersion energy spectrometer (Bruker, USA). Sample preparation for SEM analysis included vacuum evaporation of conductive material in the form of an Au layer of 10-9 m thickness. Tests were performed using a deep vacuum (2.67×10-4 Pa), while EDX analysis was carried out at an electron beam acceleration voltage of 20 keV.

Conditions of experiments

The research was carried out as planned (*Table 3*), at a constant temperature of 18 ± 1 °C, in beakers containing 500 ml of the wastewater under study, which was mixed using a magnetic stirrer (200 r.p.m., MS11, Wigo, Poland). Up to 500 ml of wastewater with AG 16 = 20,

120 or 220 mg/l and Envifer (calculated as pure K₂FeO₄) was added, pH was adjusted with 25% NaOH and H₂SO₄ solutions and the reaction established for a set time. 0.5 mol/l Na₂SO₃ was added to stop the oxidation reaction. In a filtered wastewater sample (0.45 µm), the concentration of AG 16, colour and DOC was determined. The concentration of AG 16 was assumed as the main criterion for the efficiency of the oxidation process, while the colour of wastewater and the concentration of DOC were assumed as the auxiliary criterion. Optimisation of the AG 16 removal process was carried out using the Taguchi method and Statistica 13 software (StatSoft, Poland). The influence of four input parameters (pH, time, AG 16 concentration and K₂FeO₄ concentration) on the concentration of AG 16 in the wastewater treated was analysed. The mixing speed (200 r.p.m.) and temperature $(18 \pm 1 \, {}^{\circ}\text{C})$ were assumed as constant values, whereas the range of values for

each of the input parameters (Table 3) was selected on the basis of literature data [19, 20, 34]. Finally the subsequent ranges for the pH (2, 7, 12), reaction time (10, 30, 50 min), AG 16 concentrations (20, 120, 220 mg/l) and K₂FeO₄ concentrations (25, 125, 225 mg/l) were adopted. As a result of experimental planning, a research plan was devised containing 9 experiments (4 parameters, with 3 numerical values of each parameter), which were implemented in accordance with the plan adopted. The tests were repeated three times. The results of experimental tests were subjected to statistical analysis (ANOVA), the expected S/N ratio calculated, the dependence of the mean values of the criterion function (η) on the values of the input parameters presented on a graph, and experimental verification carried out.



In the initial stage of the research, qualitative and quantitative analysis were used in the study of a commercial product (Envifer) containing K₂FeO₄. Determination of K₂FeO₄ content was carried out using two analytical methods (chromite titration method and spectrophotometric method). The test results obtained were characterised by high compliance and indicated that Envifer contained 40.2% and 40.0% K₂FeO₄, respectively. The mean value (40.1%) was used for further testing. The UV-VIS spectrum studied in the 180 - 1100 nm range (*Figure 1*) visualised the three peak absorption characteristic of Fe(VI) (510 and 785 nm, and the absorption shoulder at 570 nm) and two minimum absorption peaks (400 and 678 nm). Other authors for K₂FeO₄ (purity 84 and 98.7%) received similar values of maximum (505 and 510 nm, 570

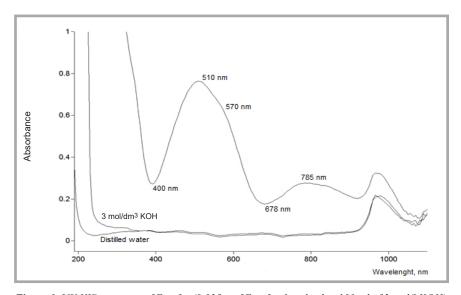


Figure 1. UV-VIS spectrum of Envifer (0.035 g of Envifer dissolved in 100 ml of 3 mol/l KOH).

and 785 nm) and minimum absorption peaks (390 and 400 nm, 675 and 678 nm) [34, 35]. On the basis of studies using the chromite titration and spectrophotometric methods, the Fe(VI) content was found to be 11.3%. The composition analysis (EDX) results showed that the commercial product contained $47.31 \pm 1.50\%$ K, $15.00 \pm 0.45\%$ Fe and $37.69 \pm 5.20\%$ O (Figure 2). The percentage composition of K₂FeO₄ calculated on the basis of the molar mass (198.04 g/mol) indicates that K₂FeO₄ contains 39.5% K, 28.2% Fe and 32.3% O, which means that the components of the commercial product, in addition to K₂FeO₄, also constitute a certain amount of impurities, most likely in the form of K₂O and Fe compounds such as K₂FeO₄ and KFeO₂. The SEM analysis carried out (Figure 3) showed the heterogeneous crystal structure of the product tested, characterised by the presence of imperfect crystals of various sizes. They have some features of K₂FeO₄ crystals (plump, columnar, cone-shape growth), as observed by other authors; however, due to the admixtures present in the product, these features were disturbed during the formation of the crystalline structure [35]. Synthetic wastes used in the study (Table 2), depending on the concentration of AG 16 (20, 120 and 220 mg/l), were characterised by different values of the physicochemical parameters determined. The largest variation concerned the colour of the wastewater (560, 2540 and 4710 mg Pt/l, respectively). The COD value was 2230, 2270 and 2295 mg O₂/l, respectively, while the TOC content was 394, 433 and 551 mg/l. As a result of 9 experiments, according to the plan adopted (Table 3), it was found that the lowest concentrations of AG 16 in the wastewater treated were obtained in experiments 1 and 6 (1.7 and 1.2 mg/l, respectively), and the highest in experiments 5 and 7 (211.1 and 105.3 mg/l, respectively). The smallest values of wastewater colour were obtained in experiments 1 and 6 (250 and 640 mg Pt/l, respectively). In these cases, despite the slight final concentration of AG 16 (1.7 and 1.2 mg/l, respectively), no complete visual discolouration of wastewater, which was characterised by a slightly yellowish colour, was obtained. The final concentrations of AG 16 attained were also associated with changes in DOC values of the wastewater treated; however, in the case of this parameter, they changed to a small extent. The lowest DOC values were obtained in experiments 6 and 8 (276 and 268 mg/l, respec-

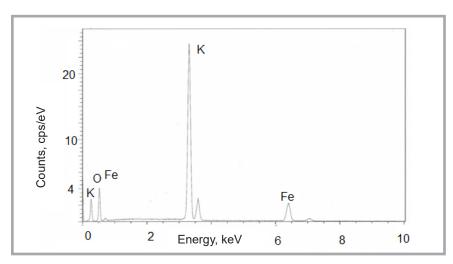


Figure 2. EDX analysis of Envifer.

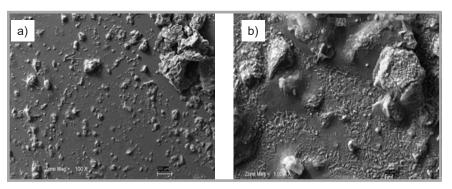


Figure 3. SEM analysis of Envilor $(a - 100 \times, b - 1000 \times)$.

Table 4. Analysis of the experiment using Statistica 13 (ANOVA). SS - predicted residual error sum of squares, MS - mean square error, F - statistics, df - number of degrees of freedom.

Efect/Factor	Analysis of variance, Mean = -26.531, Sigma = 14.6215					
Elect/Factor	SS	df	MS	F	р	
Repetition	0.017	2	0.009	0.29	0.755	
pН	380.643	2	190.322	6266.81		
Time	322.177	2	161.088	5304.24		
Concentration of AG 16	4685.562	2	2342.781	77141.94	< 0.05	
Concentration of K ₂ FeO ₄	169.576	2	84.788	2791.85		
Rest	0.486	16	0.030	_	_	

Table 5. S/N ratio in optimal conditions.

Effect/Factor	S/N ratio predicted in optimal conditions, Mean = -26.531, Sigma = 14.622			
	Parameter value	Significance of the effect	Standard error	
Repetition	3	0.0357		
pH	2.0	3.8880		
Time	50	2.6630		
Concentration of AG 16	20	17.3224	0.0581	
Concentration of K ₂ FeO ₄	125	3.5422		
Expected S/N ratio	-	0.9204	_	

tively). In some cases (experiments 2 and 4), despite comparable concentrations of AG 16 (21.6 and 24.3 mg/l) and DOC (307 and 284 mg/l), a significant differ-

ence in wastewater colour was observed (1240 and 2340 mg Pt/l, respectively). This was probably due to the formation of dye degradation intermediates

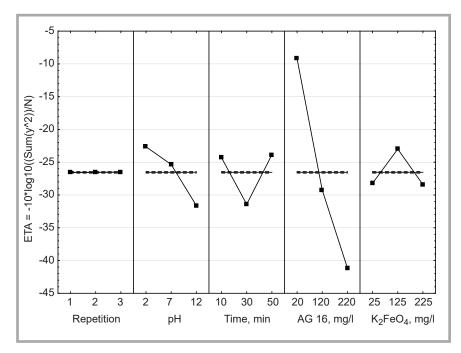


Figure 4. Plot of ETA (η) values versus input parameter values for synthetic textile wastewater.

at different pH values (2 and 7), which affected the colour value determined at 405 nm. The Taguchi method, also used by other authors to optimise dye removal processes, was used to optimise AG 16 removal conditions [36]. The results of the experiments were initially evaluated by means of analysis of variance (ANOVA), thanks to which the influence of each parameter on the variance of results in relation to the total variance of all parameters was examined (Table 4). The analysis conducted showed that the "repeat" parameter was statistically insignificant, which meant that the results obtained did not differ significantly in subsequent replicates of the adopted plan of 9 experiments (p > 0.05). The other parameters (pH, time, and concentration of AG 16 & K₂FeO₄) have been classified as statistically significant, and therefore one can infer their influence on the value of criterion function η. In the Taguchi method, process efficiency was assessed to minimise process variability in response to N factors (noise factors), while simultaneously maximising variability in response to S factors (signal factors). Both of the above criteria were analysed together by maximising the function of criterion $\eta = S/N$. In the investigation process, the criterion "smaller is better" was applied, due to the deliberate reduction of AG 16 concentration in the wastewater treated according to the criterion function (3):

$$\eta = -10 \cdot \log_{10}(\frac{1}{n} \sum_{i=1}^{n} y_i^2)$$
 (3)

where, i – number of measurements, η – S/N factor, n – number of measurements for a specific process, and y - feature measured [37]. The S/N ratio expected in optimal conditions (pH 2, time – 50 min, concentration of AG 16 = 20 mg/l, concentration of $K_2FeO_4 = 125 \text{ mg/l}$) is presented in Table 5. The analysis conducted highlighted that the least influence on the effectiveness of AG 16 removal in the case of the synthetic wastewater tested was shown by the process time, followed by the concentration of K₂FeO₄ and the pH value. The value of the final concentration of AG 16 in the wastewater treated was largely dependent on the initial concentration of dye. Due to the exclusion of the possibility of interaction between individual input parameters, owing to the use of orthogonal arrays during the planning of experiments, and based on the results obtained, one can infer the exclusive influence of individual parameters on the criterion function n [35]. Figure 4 is presented in the form of a graphical value of the criterion function η in dependence on all input parameters. The criterion function η reaches the maximum value for the following input parameters: pH 2, time - 50 min, concentration of AG 16 = 20 mg/l, and concentration of $K_2FeO_4 = 125$ mg/l. For the assumed optimal parameters, a verification experiment was carried

out, which resulted in purified wastewater characterised by a low concentration of AG 16 (0.4 mg/l), a low colour value (66 mg Pt/l), and a DOC concentration of 249 mg/l (experiment 10, *Table 3*). Under these conditions, the values of physicochemical parameters obtained were the lowest as compared to those obtained at the stage of implementing the test plan (experiments 1 – 9). In addition, the visual discoloration of wastewater and reduction of AG 16, colour and DOC values were by 98, 88 and 37%, respectively.

Conclusions

- Application of the Taguchi method allowed to identify parameters of the AG 16 dye removal process from synthetic wastewater for optimal initial conditions (AG 16 concentration 20 mg/l, pH 2, K₂FeO₄ concentration 125 mg/l, time 50 min). Under these conditions wastewater characterised by a low concentration of AG 16 (0.4 mg/l) as well as a reduced concentration of DOC (249 mg/l) and colour (66 mg Pt/l) was obtained, which corresponded to a removal efficiency of 98% (AG 16), 37% (DOC) and 88% (colour).
- Application of the Taguchi method enabled quantitative classification of the impact of individual parameters on the efficiency of the removal process of AG 16 from the wastewater treated (time < concentration of K₂FeO₄ < pH < concentration of AG 16).
- The Taguchi method allowed to optimise the process with a small number of experiments and calculations, the results of which were presented graphically and were easy to interpret.
 - The Taguchi method, however, enforces the adoption of "rigid" ranges of concentrations of individual pollutants removed from the wastewater, which in technological practice can be a difficult task, especially in the case of wastewater treatment with high variability in quality. In the case where a lower concentration value than the one currently determined in the wastewater is indicated as optimal, the efficiency of the process will probably be lower than it would appear from the parameters obtained as a result of the Taguchi method. This problem can be minimised if it is possible to obtain a relatively constant composition of wastewater flowing

- into the treatment plant as a result of, for example, retention reservoirs.
- In practical terms, in wastewater treatment technology, the Taguchi method can play an auxiliary role as a tool for initial optimisation of the wastewater treatment process, allowing identification of the most important process parameters and the significance of their impact on the efficiency of treatment.

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