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STUDY OF THE EFFECT OF MEDIUM PH ON THE KINETICS OF THE WATER DEMANGANATION PROCESS

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ABSTRACT

During the investigation of manganese ions oxidation with the oxygen of the air in an aqueous medium it was established that pH has a significant effect on the process. Studies have shown that within 24 hours of settling the solutions with manganese ions concentrations in the range between 1.0 to 30.0 mg dm^{-3} under the condition of free contact of solution surface with the oxygen of the air, the efficiency of the oxidation was quite insignificant and was only 10 - 20 %. However, in the case of increasing the pH to 8.5 - 9.5 and passing the air through the solution, an increase in the removal efficiency of manganese ions was achieved, which depended on ions initial concentrations. At pH = 9.5 and a manganese ions concentration in the solution of 1.0 mg dm^{-3}, the degree of removal reached 35.0 %, and at concentrations of ions of 15.0 and 30 mg dm^{-3}, it was possible to achieve removal of 24.0 and 26.7 %, respectively. Under the same conditions, manganese ions minimum concentration was 0.65 mg dm^{-3}.

Keywords: *removal of manganese, water treatment, low-waste technology.*

INTRODUCTION

Providing quality water to the population and industry is an important problem of our time [1]. Intensive anthropogenic activity, the influence of various industries leads to a sharp deterioration in water quality [2, 3]. Ingress of heavy metals into surface waters has a negative impact on ecosystems and public health [4, 5].

Manganese is one of the substances that pollute water bodies. This metal is included in the list of basic indicators of water quality according to the requirements of sanitary standards of Ukraine (DSanPin) [6], World Health Organization (Guidelines for drinking-water quality) [7], United States of America (USEPA) [8], European Union (Directive (EU) 2020/2184) [9], included in the priority list of pollutants of water bodies recommended for systematic control (Decision of the European Parliament and the Council of the EU № 2455/2001 / EU).

Manganese enters surface water as a result of leaching from soils and minerals, as well as with sewage from manganese iron-processing plants, metallurgical plants and other industries [10]. In natural waters, manganese can be present in various forms: dissolved, colloidal and suspended [11].

Removal of manganese from water is the goal of many water supply enterprises. Manganese is a necessary biogenic element in normal processes in living organisms as well as in humans [12]. Despite this, its content in water more than 0.1 mg dm^{-3} can be dangerous to health [13] and lead to its bioaccumulation [14].

Improving the efficiency of treatment of mineralized wastewater by creating new and improving existing methods of water treatment technologies will improve the condition of surface water with an ever-increasing anthropogenic load [15, 16]. Today, a significant number of effective methods for extracting heavy metals from water have been developed [17 - 20]. However, the problem of purifying water from manganese ions is quite complex. Despite the large volume of published research results in this area [21, 22], described technological processes [23, 24], including catalytic methods for purifying water from Mn^{2+} ions [25 - 28], the problem largely remains unresolved. This is because the extraction of manganese ions from water is sorption [29 - 31], ion-exchange [32], membrane [33, 34], methods is not effective enough. Especially in the presence of hardness ions, the concentration of which in natural waters is usually much higher than the concentration of manganese and iron ions. This leads to a significant decrease in the capacity of sorbents and ion-exchange materials for manganese and iron ions, as well as to a decrease in the selectivity of membranes for ions of these metals. Moreover, calcium compounds, during filtration, deposited on the surface of the membrane lead to a sharp reduction in their life and a decrease in productivity. Iron (III) compounds formed during water purification poison sorbents and ionites, blocking their pores, thereby reducing their sorption and exchange capacity.

The choice of Mn removal method is very much impacted by overall water chemistry and co-contaminants [35, 36]. When choosing a cleaning method, it is important to pre-determine its economic feasibility [37]. It should be noted that sorption, ion-exchange, and especially membrane methods are quite expensive with limited productivity [38, 39]. As a result, methods for oxidizing water from manganese ions are more promising. The simplest method is the chemical oxidation of dissolved Mn (II) to Mn (IV), and then its separation as a solid phase from the solution using clarification and filtration processes [40, 41]. It is advisable to dispose of the formed sediments as part of building materials [42 - 46]. Insufficient research on possible ways to dispose of the formed sediments leads to the need for these studies.

RESULTS AND DISCUSSION

Materials

Model solutions prepared on tap and distilled water with different initial concentrations of manganese ions (from 1.0 to 30.0 mg dm^{-3}) were used in this study. Sediments obtained during water demanganation were used as additives in cement production.

Removal of manganese (II) from water by oxidation

The study of manganese ion oxidation process in the aqueous medium was carried out on model solutions with a sample volume of 0.5 dm^3. Residual iron concentrations were determined every hour during the first 6 h.

To investigate the effects of oxygen a porous aerator with a bubbling intensity of 0.5 dm^3 min^{-1} was used for the oxidation of manganese ions. Under static conditions, solutions with a volume of 0.5 dm^3 with concentrations of manganese ions from 1.0 to 30.0 mg dm^{-3} were blown with air for 6 hours, determining the residual manganese content every hour of bubbling.

To study the effect of the pH of the medium on the rate of manganese ions oxidation, model solutions with ions concentration in tap and distilled water 1.0, 15.0 and 30.0 mg dm^{-3}, respectively, were used. The pH of solutions was adjusted from 7.50 to 10.00 and left for 6 h for precipitation. The recording of the change of manganese concentrations was done every 60 min and the efficiency, Z of the process was calculated:

$$Z = \frac{C_0 - C_f}{C_0} \cdot 100, \% \qquad (1)$$

C_0 and C_f - starting and final concentration of manganese, mg dm^{-3}.

The experiments were carried out according to full factorial design 2^2 (Table 1). The variables investigated were pH of the solution (X_1) and time of treatment (X_2).

The efficiency of the process (Y_2) was evaluated. A second-order polynomial were used to describe the process:

$$Y=b_0 + b_1X_1 + b_2X_2 + b_3X_1X_2 + b_4X_1^2 + b_5X_2^2 \qquad (2)$$

where Y is a response factor, b_i are regression coefficients. MINITAB 17 software were used for the calculations.

During the research were used manganese sulfate

Table 1. 2^2-factorial design for manganese ions removal.

Factor	Low level (-1)	High level (1)
pH X_1, %	8.5	10.5
Time X_2, h	2	6

solutions prepared on distilled and tap water with concentrations of 1.0 - 30.0 mg dm^{-3}. The reaction of the medium in model solutions on distilled water was at pH = ~ 5 at the beginning of the process and reached ~ 6.0 after a day. In tap water, the pH increased from 7.50 to 8.20 after a day of settling the solutions. During the 24 h of settling the solutions in free contact with air, no significant oxidation and removal of manganese ions was observed.

For the model solutions on distilled water with initial manganese ions concentrations at the level of 1.0 - 5.0 mg dm^{-3}, a slight decrease in the concentration was observed.

In solutions prepared on tap water, oxidation efficiency increases with increasing initial concentration of manganese ions. Obviously, this is due to the higher pH level in tap water. In addition, the reaction rate always increases with increasing concentrations of the starting components. In this case, with constant aeration of water and a very low rate of binding of oxygen by manganese, its concentration in water was approximately constant. Therefore, the process speed was determined mainly by the initial concentration of manganese.

To assess the influence of the pH of the medium on the processes of manganese oxidation in an aqueous medium, were used manganese solutions prepared in distilled and tap water with initial concentrations with different pH from 7.50 to 10.00. In solutions prepared on distilled water already at pH ≥ 7.50 happens faster oxidation and extraction of manganese ions. At the same time difference in Mn^{2+} concentration after 24 h in the solution with pH = 7.50 was only 0.15 mg dm^{-3} higher than the concentration in the solution with pH = 10.00. In terms of recovery, the difference reached 18 %. In general, the maximum degree of purification of water from manganese ions in distilled water did not exceed 34 %. In solutions prepared on tap water, the maximum recovery was 53 % at pH = 10.00. The results are presented in summary Table 2.

At pH ≤ 9 there was no oxidation of manganese

Table 2. The effect of pH on the change in manganese concentration over time.

pH	C (Mn^{2+}), mg dm^{-3}	Standing time, h					
		1	2	3	4	6	24
7.50	1.0	1.00	1.00	1.00	1.00	1.00	1.00
8.00	1.0	1.00	1.00	1.00	1.00	1.00	1.00
8.50	1.0	1.00	1.00	1.00	1.00	1.00	1.00
9.00	1.0	1.00	1.00	1.00	1.00	1.00	0.90
9.50	1.0	0.90	0.80	0.70	0.65	0.65	0.55
10.00	1.0	0.85	0.65	0.60	0.50	0.50	0.50
7.50	15.0	15.00	15.00	15.0	15.00	15.00	15.0
8.00	15.0	15.00	15.00	15.0	15.00	15.00	15.00
8.50	15.0	15.00	15.00	15.0	15.00	15.00	14.90
9.00	15.0	15.00	15.00	15.0	15.00	14.90	14.70
9.50	15.0	13.60	11.10	10.10	9.90	9.80	9.80
10.00	15.0	12.30	10.30	9.30	8.70	8.30	7.10
7.50	30.0	30.00	30.00	30.00	30.00	30.00	30.00
8.00	30.0	30.00	30.00	30.00	30.00	30.00	30.00
8.50	30.0	30.00	30.00	30.00	30.00	29.90	29.70
9.00	30.0	30.00	30.00	30.00	29.90	29.80	29.30
9.50	30.0	26.90	23.70	19.40	19.00	18.90	17.10
10.00	30.0	25.70	24.90	16.70	16.40	15.80	12.90

at all, only at pH above 9.5 a certain reduction in the concentration was noticed during oxidation for 4 h. This is explained by the passage of reactions:

$$Mn^{2+} + 2(OH)^- \rightarrow Mn(OH)_2, \quad (3)$$

$$4Mn(OH)_2 + 2O_2 \rightarrow 2Mn_2O_3 + 4H_2O, \quad (4)$$

$$2Mn_2O_3 + 8H_2O \rightarrow 4Mn(OH)_4 \text{ or } MnO_2 \cdot xH_2O \quad (5)$$

According to reactions (3) - (5), various solid products $Mn(OH)_2$, Mn_2O_3 and $Mn(OH)_4$ are formed as a result of oxidation of Mn^{2+}. At pH values in the range from 9.0 to 9.5, the reaction between Mn^{2+} and O_2 gives Mn_2O_3 or MnO_2 oxides as oxidation products. The surface of which can then catalyse the reaction between Mn^{2+} and O_2.

For intensification of manganese oxidation processes aqueous solutions with a manganese ions concentration of 1.0 to 30.0 mg dm^{-3} are adjusted to pH 8.50 - 10.5 intensively blown with air through porous aerators for 6 h (Fig. 1 - 3).

If at pH = 8.50 the maximum oxidation state of manganese reached 24 %, then at pH = 9.00 this

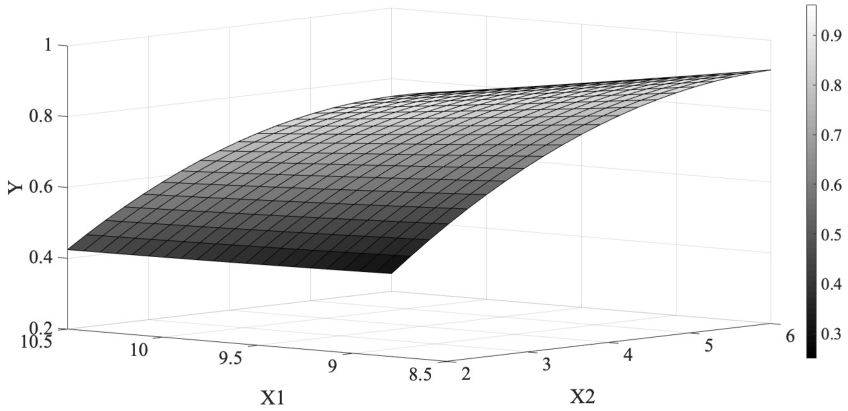

Fig. 1. Dependence of the Mn (II) residual concentration on the time of solution aeration (manganese concentration of 1.0 mg dm^{-3}, intensity of air bubbling 0.5 dm^3 dm^{-3} min^{-1}): y = 6.403 – 1.7647x1 – 0.12321x2 + 0.01622x1x2 + 0.1053x1^2 – – 0.00042234x2^2.

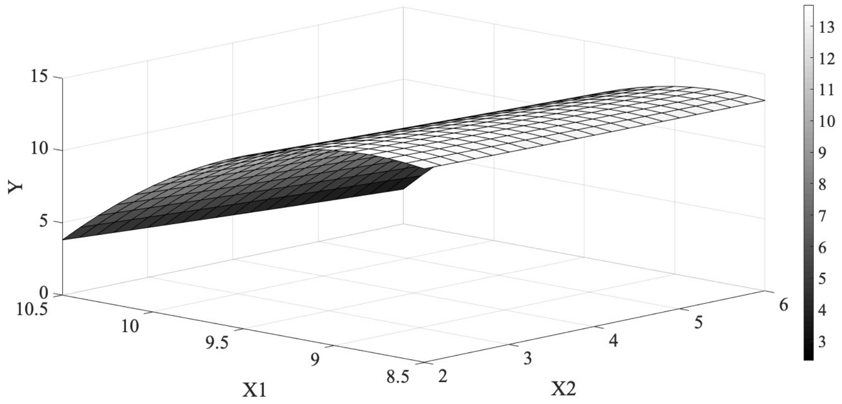

Fig. 2. Dependence of the Mn (II) residual concentration on the time of solution aeration (manganese concentration of 15.0 mg dm^{-3}, intensity of air bubbling 0.5 dm^3 dm^{-3} min^{-1}): y = 246.81 – 59.08x1 – 1.1645x2 + 0.15135x1x2 + 3.346x1^2 – – 0.0081396x2^2.

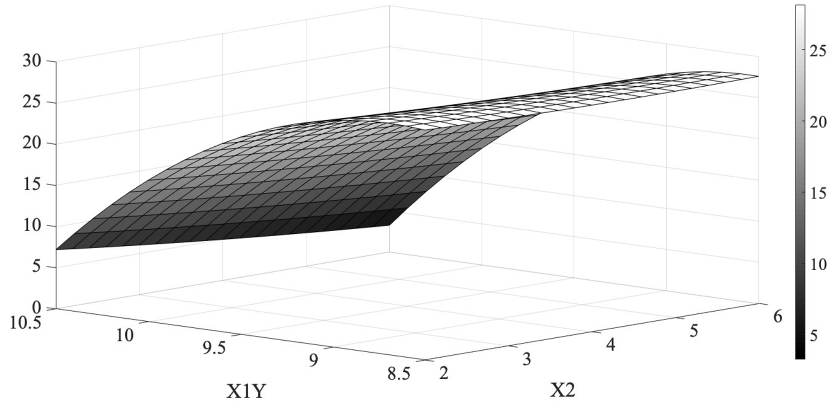

Fig. 3. Dependence of the Mn (II) residual concentration on the time of solution aeration (manganese concentration of 30.0 mg dm^{-3}, intensity of air bubbling 0.5 dm^3 dm^{-3} min^{-1}): y = 462.9 – 112.36x1 – 3.2354x2 + 0.4465x1x2 + 6.4129x1^2 – – 0.059102x2^2.

indicator reached 30 %, and at pH = 9.50 it was at the level of 62.5 %. At the same time, according to pH 8.5 and 9.00, the best indicators for water demanganation were obtained according to manganese concentrations in water 5 mg dm^{-3}. At the concentrations 1 mg dm^{-3}, the efficiency of purification obtained 5 % at pH 8.50 and 15 % at pH 9.00. Obviously, at pH > 9.5, even a slight increase in oxygen concentration affects the efficiency of manganese oxidation. Thus, at a concentration of manganese of 15 and 30 mg dm^{-3}, the degree of its extraction reached 23 %. Aeration of solutions for 24 h did not significantly affect the efficiency of water purification. In all cases, the pH of water decreases depending on the duration of oxidation. For the complete oxidation of manganese to MnO_2, high concentrations of dissolved oxygen in water and the creation of an alkaline environment are necessary, which is not always acceptable when purifying drinking water.

Determination of kinetic parameters of manganese ions oxidation in aqueous medium

The order of the reaction of manganese ions oxidation in water depending on its concentration was determined graphically. As a result, were built dependency plots fore 0 order C = f(t), for 1st order reaction lnC = f(t), for 2nd order reaction 1/C = f(t) and for 3rd $1/C^2 = f(t)$.

To assess the influence of aeration and pH on the process of oxidation of manganese ions in an aqueous medium, rate constants of 0, 1, 2 and 3 orders were calculated according to the procedure. The results are presented as a summary Table 3 to compare their values.

Based on the calculated velocity constants, at pH = 9.50 the numerical value of the constants is several orders of magnitude higher than at pH = 8.50 and 9.00. If we compare the calculated constants of the rate of the manganese oxidation reaction in tap water without adjusting the pH and aeration with the constants obtained because of studies under conditions of increasing the pH of the medium and blowing solutions with air, a natural acceleration of manganese ions removal from water is seen.

Based on the calculated rate constants, at pH = 9.50 the numerical value of the constants is several orders of magnitude higher than at pH = 8.50 and 9.00. If we compare the calculated rate constants of the manganese oxidation reaction in tap water without pH adjustment and aeration with the constants obtained as a result of research under conditions of increased pH of the environment and blowing of solutions with air, then the natural acceleration of the removal of manganese ions from water can be seen. As the initial concentration of manganese in solutions increases, the rate of the oxygen oxidation reaction decreases. Obviously, with a low content of Mn^{2+} ions in the solution, there is enough oxygen, which is why the reaction of the formation of oxidized MnO_2 compounds goes almost to the end. With a higher content of Mn^{2+} ions, the speed of the reaction is inhibited by extraneous ions present in the solution and a lack of soluble oxygen.

Table 3. Calculated constants of the rate of manganese oxidation in tap water by equations for reactions of 0, 1, 2 and 3 orders at different pH of the medium and aeration of the solution.

C (Mn^{2+}), mg dm^{-3}	$\text{К}_{6\ h}$	pH=8.50	pH=9.00	pH=9.50
1.0	К_0, mg dm^{-3} h^{-1}	0.0084	0.0260	0.0584
	К_1, h^{-1}	0.0086	0.0281	0.0719
	К_2, dm ^{3} $mg^{-1}h^{-1}$	0.0089	0.0296	0.0898
	К_3, mg^{-2} dm ^{6} h^{-1}	0.0091	0.0325	0.1138
15.0	К_0, mg dm^{-3} h^{-1}	0.5100	0.4168	0.7000
	К_1, h^{-1}	0.0373	0.0284	0.0458
	К_2, dm ^{3} mg^{-1} h^{-1}	0.0029	0.0018	0.0036
	К_3, mg^{-2} dm ^{6} h^{-1}	$0.0209 \cdot 10^{-2}$	$0.0132 \cdot 10^{2}$	0.0003
30.0	К_0, mg dm^{-3} h^{-1}	0.3333	0.5000	1.3333
	К_1, h^{-1}	0.0115	0.0176	0.0517
	К_2, dm ^{3} mg ^{1} h^{-1}	$0.0397 \cdot 10^{-2}$	0.0006	0.0020
	К_3, mg^{-2} dm ^{6} h^{-1}	$0.0014 \cdot 10^{-2}$	$0.0022 \cdot 10^{2}$	$0.0080 \cdot 10^{-2}$

CONCLUSIONS

It was found effect of aeration on the oxidation rate of manganese ions in tap water with concentrations from 1.0 to 30.0 mg dm^{-3} by pH of 8.50, 9.00 and 9.50 solutions. Defined, that the removal efficiency depended on manganese concentration and pH of the medium. At ions concentration 1 mg dm^{-3}, the highest efficiency of removal was 35.0 % at pH 9.50, and at ions concentrations 15.0 and 30.0 mg dm^{-3} - 24.0 and 26.7 %, respectively. However, simply increasing the water pH and aeration of the water is not sufficient to completely oxidize the manganese in the aqueous medium.

Authors' contributions: M.T.: investigation, writing original draft; I.T.: literary review, methodology, validation; V.H: investigation, data processing; A.T.: investigation, visualization, data curation; M.G: conceptualization, supervision, editing.

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