

STUDIES ON THE EFFICIENCY OF GRUNDWATER TREATMENT PROCESS WITH ADSORPTION ON ACTIVATED ALUMINA

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ABSTRACT

One of inorganic sorbents used in water treatment technology is activated alumina. It is recommended by the European Commission to remove inorganic impurities, such as arsenic, fluoride, selenium and silicates. The adsorbent is usually applied in a granular form, under flow conditions. It can become absorbent material, increasingly used due to the presence of arsenic beside iron and manganese in groundwater intakes. The aim of the study was to evaluate the effectiveness of groundwater treatment in the technological system containing adsorption on activated alumina. The experiment was performed on test model CE 581 manufactured by G.U.N.T. Hamburg, in which four treatment stages can be extracted. The first stage is used in a gravel filter of grain size 1–2 mm, in the second sand filter of grain size 0.4–0.8 mm. The third and fourth phase includes two adsorbers. The first adsorber comprises activated alumina (Al_2O_3) and the other comprises a granular activated carbon. The study was conducted at different speeds of filtration: 5, 10 and 15 m/h. In the raw water samples and the purified water samples after each treatment step the following parameters were determined: pH, O_2 concentration, electrolytic conductivity, SO_4^{2-} concentration, NO_3^- concentration, PO_4^{3-} concentration, Cl concentration, color, turbidity, iron and manganese concentration, COD_{Mn} , total hardness, calcium hardness, magnesium hardness, content of dissolved substances. The conducted research indicates that optimum filtration rate for most pollution is 15 m/h. Moreover, the presence of activated alumina has contributed to increasing the efficiency of nitrate (V) and phosphate (V) ions removal.

Keywords: activated alumina, water treatment, adsorption, popular pollutants

INTRODUCTION

In the adsorption process various materials are used: natural, modified natural or synthetic sorbents, on the surface of which or in the pores and micropores associated components are present in an aqueous solution [Ali 2012]. Typically, the adsorption is used for removal of water soluble organic impurities causing the colour and odour of water, metals, anions. The adsorption process is useful for removing dissolved organic substances, but also for the specific micropollutants, and even contaminants, whose traces may pose a threat to health or are undesirable in drinking water [Kaleta, 2009, Anielak 2015]. The most common adsorbents are activated carbons, but they are not the only recommended materi-

als. For example, the sorbent used to remove the inorganic components present in the water is activated alumina. In a mixture of amorphous alumina varieties obtained by dehydrogenation of aluminum hydroxide $\text{Al}(\text{OH})_3$ at a temperature of 300–600 °C. The adsorbent has a specific surface area equal to 50–300 m^2/g . It exhibits amphoteric properties, and the parameter indicating the nature of the pH, namely a pH of zero charge, is 8.2. In the medium having a pH of less than 8.2 adsorbent material exhibits anion exchange properties, and at pH above 8.2 – cation exchange. For example, the mechanism of removal of arsenate ions from aqueous solutions of this adsorbent is close to the mechanism of removing arsenates in the slightly alkaline anion exchangers [Hering et al. 2004; Bratek et al. 2012].

Activated alumina is much less likely to be used in water purification than activated carbons, however, it is used to remove inorganic contaminants such as fluorides, arsenic, selenium, and silicates. European Commission has allowed activated alumina as an absorbent material for removal of fluoride ions from water. In addition, adsorption on activated alumina has been proposed by the EPA as an effective and inexpensive technology to remove arsenic and selenium from drinking water classified as a BAT [Singh, Pant 2004; EPA 2007; Lin, Wu 2001].

Many authors have tested and examined the efficiency of removal of arsenic and fluoride ions with the activated alumina [Su et al. 2008; Lescano et al. 2015]. Su et al. (2008) have tested activated alumina in the removal from the water of As (V), Se (IV), and V (V), depending on different pH values and the charge of sorbate [Su et al. 2008]. In contrast, the research group led by Naiya [Naiyi et al. 2009] conducted research using samples of industrial wastewater containing Cd (II) and Pb (II), which demonstrated the usefulness of activated alumina for the purifying waste water from these metals. Wang et al. (2009) demonstrated that the activated alumina has a higher adsorption capacity of phosphate ions (V) compared to the other anions, such as sulfate ions (VI) or chloride ions. Wang's research team compared elimination of phosphates (V) on the activated alumina and quartz sand and activated carbon. It has been found that Al_2O_3 is more suitable for removing phosphate (V) because on the surface there are more micropores, thereby has a larger active area of approximately $300m^2/g$ [Wang i in. 2009].

In order to verify the applicability of the activated alumina in the technological treatment of groundwater, it should be checked for the effectiveness of the removal of common contaminants present in the water. The aim of this study was to evaluate the effectiveness of the removal of the main pollutants expressed by parameters such as pH, O_2 concentration, electrolytic conductivity, SO_4^{2-} concentration, NO_3^- concentration, PO_4^{3-} concentration, Cl concentration, color, turbidity, iron and manganese concentration, COD_{Mn} , total hardness, calcium hardness, magnesium hardness, content of dissolved substances of groundwater in the technological system of underground water treatment containing adsorption on activated alumina. The research was also intended to determine the impact of changes in

the speed of water filtration on the effectiveness of the treatment of the common impurities in the tested technological sequence.

MATERIAL AND METHODS

The research was conducted in the laboratory of the Department of Technology Engineering and Environmental Protection Bialystok University of Technology. These included indicators of water pollution after the initial gravel (grain size 1–2 mm) filtration, after filtration through a bed of quartz sand (grain size 0.4–0.8 mm), the process of adsorption on activated alumina (grain size about 1.5 mm) and purified water after adsorption on activated carbon. The studies were carried out in three stages. In the various stages, the speed of filtration differed. The efficiency of removal of various pollutants in a technological process of adsorption on activated alumina using a filtration speed: 5, 10 and 15 m/h was verified. The experiment was performed on test model 581 CE manufactured by G.U.N.T. Hamburg. Figure 1 shows the experimental model.

In the model CE 581 filters and absorbers have an identical structure – are made of transparent tubes made of PMMA. Their dimensions are: outer diameter 200 mm, internal diameter 150 mm, height: about 620 mm. Their upper and lower ends are sealed with the flanges with PVC. Flanges are secured with knurled screws. The bottom flanges are equipped with integrated bearing for holding filtering material (sand and gravel) and adsorbents. The flanges of filters are equipped with faucets for water sampling. The CE 581 device is regulated and controlled by a dedicated software that must be installed on a computer before the start.

The water collected for the purpose of the experiment is the underground water from industrial water intake. It is one of three major groundwater intakes included to the Main Groundwater Reservoir (GZWP No. 218), which is the primary reservoir for drinking water for the agglomeration of Bialystok. The depth of the water intake is about 52.5 m.

In raw water samples and samples after each unitary process, included in the research model CE 581 were determined:

- colour and turbidity measured by using UV-Visible spectrophotometry using a spectrophotometer HACH DR4000V;

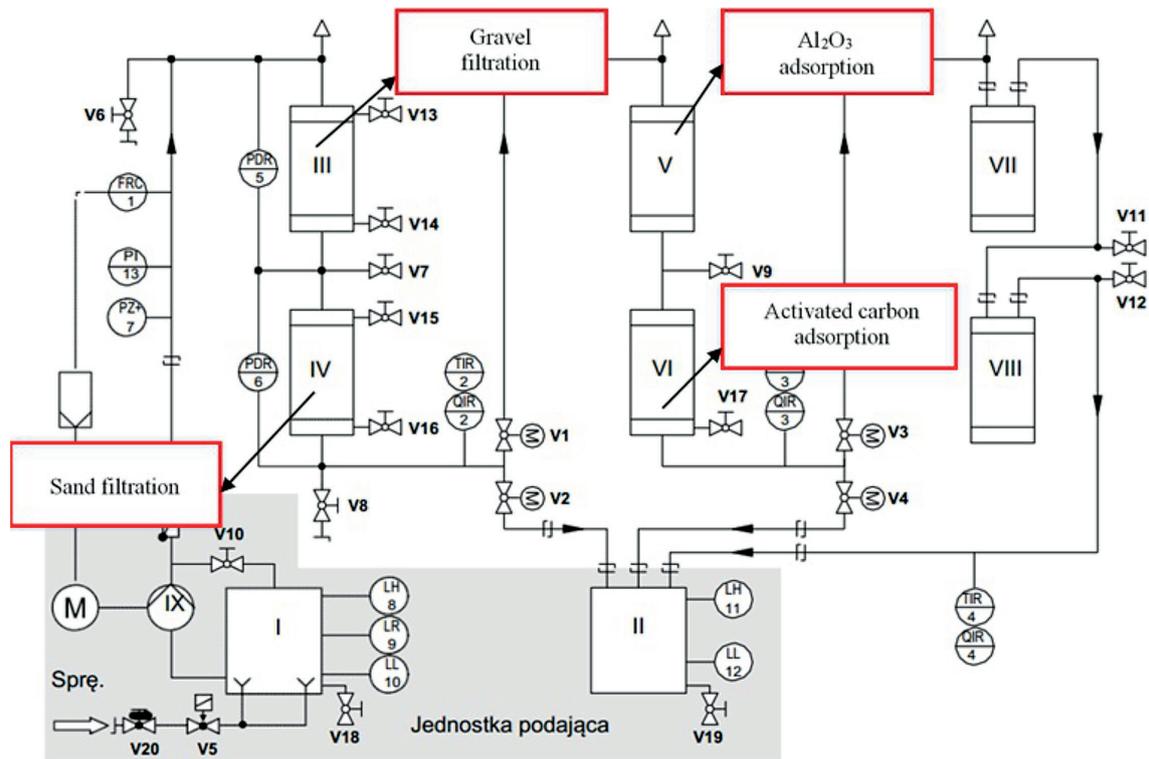


Figure 1. Schematic of CE 581.

- iron and manganese concentration by atomic absorption method using an atomic absorption spectrometer with flame atomization Thermo Scientific iCE3400;
- pH, electrolytic conductivity, redox potential measured by potentiometric method using a multi-parameter WTW inoLab® Multi 9430;
- dissolved oxygen concentration was measured by using oxygen meter HACH Session 156;
- chloride concentration, total hardness, calcium and magnesium hardness measured by using titrimetric method;
- COD_{Mn} was made by using potassium permanganate (VII) in accordance with PN/C-04578.02.

Studies carried out in three test series (three in each repetition), each with a filtration speed. Samples were collected once a day every 24 hours.

RESULTS AND DISCUSSION

The scientific work carried out in the field of water treatment technologies tend to seek more powerful and economic techniques and filtration materials [Nowak 2013]. Most researchers test filtration materials or sorbents against one of the

selected parameters, without taking into account the overall impact on the process of purifying in the whole technological system of water treatment. Therefore, it seemed purposeful to carry out research and assess the usefulness of activated alumina for the treatment of groundwater in the technological system.

The test results are shown in Table 1. The observed changes in such parameters as: pH, O_2 concentration, electrolytic conductivity, SO_4^{2-} concentration, NO_3^- concentration, PO_4^{3-} concentration, Cl⁻ concentration, colour, turbidity, iron and manganese concentration, COD_{Mn} , total hardness, calcium hardness, magnesium hardness, content of dissolved substances. When analyzing the obtained values of individual parameters, it was found that redox potential of water was changing during the purification of water in the technological system under study. The greatest difference was observed in the series in which the water was purified at a speed of 15 m/h, the difference between the redox potential of the raw water and the water after adsorption process on activated carbon was 84 mV. The value of the redox potential provides similar information about the content of reducing substances and oxidizing substances present in the purified water. The decrease of the potential indicates re-

Table 1. Average concentrations of pollutants at any stage of groundwater treatment.

Parameter [unit]	Raw water	Series I				Series II				Series III			
		*1	*2	*3	*4	*1	*2	*3	*4	*1	*2	*3	*4
Filtration rate [m/h]	-	5	5	5	5	10	10	10	10	15	15	15	15
Redox potential [mV]	119	175	187	170	111	191	158	162	87	117	111	100	35
pH	7.9	7.7	7.7	8.4	9.1	7.8	7.8	8	8.6	7.7	7.8	8	8.4
O ₂ [mg/l]	6.1	6.9	7.4	7.2	2.5	7.1	7.2	7.1	2.7	7	7.1	7.1	4.2
Conductivity [μ S/cm]	483	480	480	401	325	484	483	457	361	483	481	467	396
SO ₄ ²⁻ [mg/l]	1.4	0.5	0.8	47.3	22.9	0.5	0.2	16.7	21.9	0	0	8.7	13.4
NO ₃ ⁻ [mg/l]	1.6	1.5	6.7	0.9	1.4	1	6.7	1.1	1.3	3.2	1.9	2.1	0.9
PO ₄ ³⁻ [mg/l]	21.02	7.63	5.86	5.45	4.87	13.35	13.2	6.83	5.76	9.16	8.32	5.75	5.2
Color[mg Pt/l]	67	45	42	24	5	45	37	16	4	41	40	29	6
Fe [mg Fe/l]	0.214	0.115	0.079	0.042	0.06	0.152	0.042	0.054	0.05	0.12	0.044	0.067	0.018
Mn [mg Mn/l]	0.051	0.047	0.039	0.036	0.016	0.048	0.041	0.032	0.0246	0.048	0.044	0.039	0.029
COD _{Mn} [mg O ₂ /l]	4.3	3.9	3.3	2.5	1.9	3.2	3	1.5	0.9	3.8	3.4	2.1	1.2
Total hardness [mg CaCO ₃ /l]	512	488	408	432	400	480	468	432	344	480	472	348	256
Ca hardness [mg Ca/l]	56.11	33.67	40.08	32.06	36.07	37.68	31.26	28.06	24.85	38.48	33.67	32.06	25.65
Mg hardness [mg Mg/l]	28.19	38.88	25.27	33.05	26.73	35.48	37.91	35.48	26.73	34.99	36.94	22.84	15.55
Cl ⁻ [mg/l]	16	16	16	10	8	16	16	14	10	16	12	10	8
Turbidity [NTU]	28.1	5.5	1.78	0.75	0.36	4.8	2.02	0.83	0.27	4.8	2.02	0.83	0.27
Dissolved substances [mg/l]	320.5	217	183	134	35	201	188	143	47	257	199	165	39

*1 – water after gravel filtration, *2 – water after sand filtration

*3 – water after Al₂O₃ adsorption, *4 – water after activated carbon adsorption

ducing the content of oxidizing agents while increasing the content of free electrons that can join the atoms and reduce their degree of oxidation [Szatyłowicz, Skoczko 2016].

Analyzing the changes in the pH of the treated water in the entire technological system (table 1), it was observed that pH increases in the water after purification on Al₂O₃. The largest increase in pH occurred in a series of research at the speed of filtration equal to 5 m/h. In this test series, the pH has increased by 0.70. In the case of other two series, in which the water was purified at 10 and 15 m/h the increase in pH of the water adsorption on Al₂O₃ was much smaller and amounted 0.2 units. The reason for pH increase of purified water after step associated with the activated alumina is an amphoteric nature of the material. When water contacts Al₂O₃ on the surface are formed hydroxides, that raise the pH of water flowing through the adsorbent bed. The highest increase in pH is when using a speed equal to 5 m/h, due to the longer contact time of the water with Al₂O₃ granules forming the adsorbent bed.

The biggest change in oxygen concentration (table 1) in the purified water was observed after the adsorption process on activated carbon. In each test series a significant decrease in dissolved oxy-

gen concentration in the purified water was noted. The largest decrease of 4.7 mg O₂/l was observed at a speed of 5 m/h, while at a speed of 10 m/h a decrease, in comparison to prior process, was equal to 4.4 mg O₂/l. In the case of filtration speed of 15 m/h was observed the smallest decrease in dissolved oxygen concentration, equal to 2.9 mg O₂/l. These changes in oxygen concentration clearly indicate the presence of aerobic microorganisms colonizing deposits of carbon filters, and thus the biological processes involved in the removal of contaminants from water [Zimnoch, Szostak 2006].

Microorganisms inhabiting carbon filters employ organic compounds as nutritional substrates. This allows elimination of pollutants from water by oxidation using dissolved oxygen in the respiratory process and biomass growth of the microorganisms [Nawrocki 2010]. The content of organic substances characterized by COD_{Mn} after filtration through a gravel bed has decreased by 16%, while in the second purification stage – filtration through a sand bed by another 9%. As a result of the third purification step, the process of adsorption on activated alumina has been observed a significant increase in COD_{Mn} removal effect that, whatever the speed of filtration, was approximately 28%. As a final effect of purifica-

tion through a coal-bed, a COD_{Mn} concentration of about 1.3 mg O_2/l has been achieved. Possibly, carbon filters have not been sufficiently developed, it has not produced a suitable amount of micro-organisms decomposing organic materials to further reduce their concentration in the water to be purified. Compared to the two unknown mass filter, X1 and X2, of natural origin, investigated by Skoczko, Szatyłowicz, which were characterized by the effect of the removal of expressed organic substances COD_{Mn} as appropriate: 11–20%, 5–9%, a gravel bed and sand bed remained comparable and granulate of Al_2O_3 was characterized by a much greater effectiveness in removing COD_{Mn} [Skoczko, Szatyłowicz 2016].

In all series of research we observed a slight decrease of the electrolytic conductivity of water, an average of 121 $\mu\text{S}/\text{cm}$ by comparing the raw water to water after treatment on carbon filters. The smallest decrease in conductivity of the water occurred in a series of research at the speed of filtration equal to 15 m/h. When analyzing the purification of water throughout the technological system, it was noticed that the conductivity changed only after purification on Al_2O_3 (table 1), which means that the chemical processes occur during the contact of water with granules of activated alumina.

By observing changes during the purification in the concentration of sulfate (VI) in the analyzed technological system, it was found that after the purification step the concentration of activated alumina has significantly increased (Table 1). The reason for this phenomenon might be the original contamination of granular of alumina during the production of pellets [Szatyłowicz, Skoczko 2016].

The best efficiency of removal of nitrates (V) in the technological system with adsorption on activated alumina was found in the third test series, in which the filtration speed of purified water in the technological system was equal to 15 m/h. The application of the process of adsorption on activated alumina in the technological systems for the purification of groundwater may be an alternative when compared to other methods of removing nitrate (V) ions from the water, such as ion exchange, reverse osmosis, biological denitrification or chemical reduction [Żeglin-Kurbiel, Wójcik 2007]. The efficiency of adsorption of nitrate (V) on activated alumina is determined by numerous micropores on the surface of the granules, which significantly increase the surface area of the absorbent material [Szatyłowicz, Skoczko, 2016; Wang et al. 2009].

In the case of phosphate (V), they have also the high efficiency of reducing the concentration in the water to be purified, on the basis of the analysis of each series of research. The largest decreases in of phosphate ions followed the filtration through a sand and gravel. The concentration of phosphates (V) on average decreased by almost 12 mg/l. The best effect was achieved in a series of test at a filtration speed of 5 m/h, the phosphate concentration decreased from 21.02 mg/l to 5.86 mg/l after filtration through a sand and gravel. However, in the later stages of the water purification in the system, there was a slight decrease in the concentration of phosphate ions after the last step – carbon filters. The presence of the filter adsorption with Al_2O_3 in the system also contributed to the removal of phosphates (V) from the water. Wang et al. (2009) demonstrated that the activated alumina has a high affinity to the adsorption of phosphate anions (V). They found that the quantity and the micropore volume decide about the inner surface of adsorption, and pore size about the dimension of the adsorbed molecules [Wang i in. 2009].

Reducing the color of the water (Table 1, Figure 2) in the system under study in the first stage – filtration through a gravel bed – was on average about 34%. In the second step of purification, the colour of the water has decreased by an average of 7%. Fairly good results were obtained after filtration through an Al_2O_3 adsorbent bed, independent of the speed, the colour of the water was reduced on average by a further 25% as compared to the raw water to be purified. Finally, after the carbon filters, the colour of the water was obtained at about 5 NTU.

In the case of the turbidity (Table 1, Figure 2), practically the same effect of removing was achieved, regardless of the speed of filtration in the system. The greatest reduction of the turbidity of the treated water was obtained in the first purification stage, by filtration through a gravel bed, averaging at about 82% in comparison to the raw water. In further purification steps, i.e. by filtration through a sand bed, Al_2O_3 , and carbon bed, turbidity decreased slightly by a few percent.

The values of the total iron concentration in purified water in a system under study ranged: in raw water 0.214 mg Fe/l, in purified water after the carbon filters from 0.06 to 0.018 mg Fe/l. After the first and second purification stage has been achieved an average of 74% of the effect of removing iron. Sand and gravel in the first two stages of the technological system are deposits

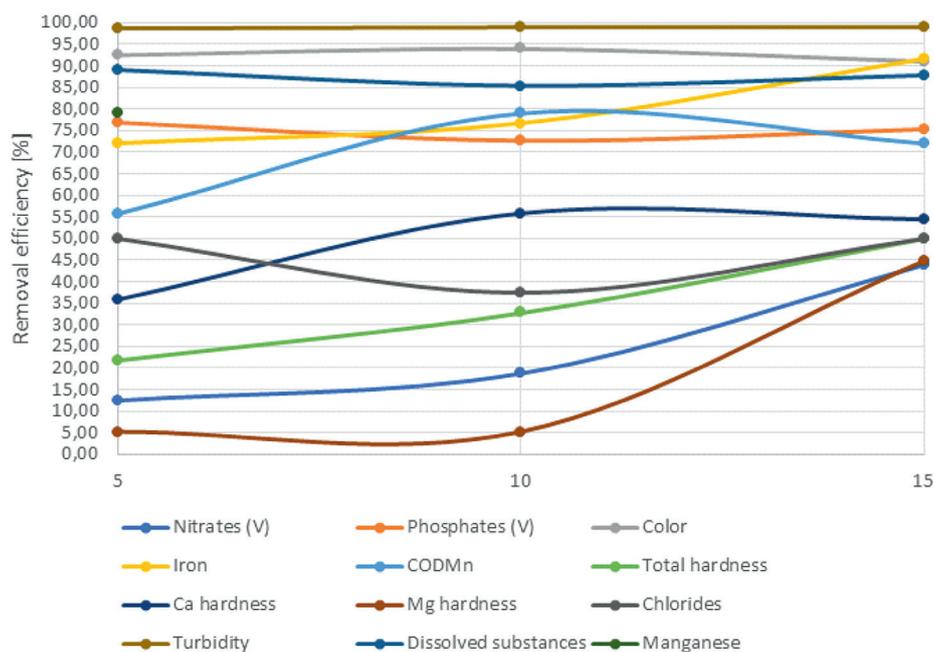


Figure 2. Changes removal efficiency of individual pollutants depending on the speed of filtration.

chemically inactive, so the removal of iron takes place during the oxidation of Fe (II) with oxygen dissolved in water, flocculation or filtration of a suspension of Fe (III), on grain deposits [Jeż-Walkowiak et al. 2011].

The highest efficiency (69%) of manganese removal in the system under study with adsorption on Al_2O_3 was obtained by the filtration speed equal to 5 m/h, while at a speed of 15 m/h the overall effect of the removal in the system was much lower, equal to 41.5%. Nowak's research proved that the manganese removal, better with lower speeds filtration from 5 to 7.5 m/h. Examined the dependence of speed on the manganese concentration in the filtrate from the filter bed such as Birm, G-1, Purolite MZ-10, Defeman [Nowak, 2013].

Perhaps the low result of the elimination of manganese affects iron concentration. The influence of the presence of iron in the water on removal efficiency of Mn is not clear. Relevant here are, inter alia, the concentration and the form in which the iron is present in the water [Nawrocki 2010]. Ability to remove iron and manganese by activated alumina is small, which can be dictated by the low starting concentration of impurities in water. When comparing the efficiency of iron and manganese removal on activated alumina to effect of removal of the impurities through the beds, such as Greensand, Birm, Cristal Right, it can be concluded that Al_2O_3 is not suitable to reduce the concentration of these elements [Skoczko et al.

2015]. One should slightly increase the pH of purified water and re-check the removability of iron and manganese on activated alumina.

Chloride concentration (Table 1) during the purification of water in the analyzed technological system slightly decreased. There has been experienced an average reduction of chloride concentration of 9 mg Cl/l in the water after the carbon filters, in relation to the raw water. By contrast, the total concentration of solutes of all the naturally present in the water cations and anions, derived from organic mineral compounds decreased from an average of 320.5 mg/l in the raw water to about 40 mg/l in the water after purification on a coal bed. The overall efficiency of the test during the process of water purification in relation to dissolved substances was an average of 87% (Table 2).

As a result of water purification, there was also reduction of pollution indicators, such as total hardness, calcium, and magnesium hardness. The average result of the elimination of total hardness was about 35%, magnesium hardness of 48.5% and calcium hardness of 18.4%. When analyzing the efficiencies obtained in each test series with a variable speed of water filtration, it was found that the best results are achieved with a speed of 15 m/h (Table 2, Figure 2).

Considering the efficiency of removal of various pollutants (Figure 1, Table 2) relative to the filtration speed of purified water throughout the

Table 2. Efficiency of removal of individual pollutants.

Filtration rate [m/h]	5	10	15
Parameter [unit]	Removal efficiency [%]		
NO ₃ ⁻ [mg/l]	12.50	18.75	43.75
PO ₄ ³⁻ [mg/l]	76.83	72.60	75.26
Color [mg Pt/l]	92.54	94.03	91.04
Fe [mg Fe/l]	71.96	76.64	91.59
Mn [mg Mn/l]	69.37	51.38	41.50
COD _{Mn} [mg O ₂ /l]	55.81	79.07	72.09
Total hardness [mg CaCO ₃ /l]	21.88	32.81	50.00
Ca hardness [mg Ca/l]	35.71	55.71	54.29
Mg hardness [mg Mg/l]	5.17	5.17	44.83
Cl ⁻ [mg/l]	50.00	37.50	50.00
Turbidity [NTU]	98.72	99.04	99.04
Dissolved substances [mg/l]	89.08	85.34	87.83

technological system, it was noticed that in the case of phosphate (V) (77%), dissolved substances (89%) and manganese (69%), the best result of the elimination was achieved at a speed equal to 5 m/h. However, in the case of pollutants such as colour and concentration of COD_{Mn} greatest percentage of reduction in the concentration was observed at filtration speed of 10 m/h, it amounted to, respectively: 94% and 79%. In the case of other pollutants, best performance has been achieved by filtering of water through the system at a speed of 15 m/h. For some indicators of pollution was observed a significant difference in removal efficiency, depending on the speed of filtration. The greatest difference of about 40% between removal effectiveness, at a speed of 5m/h and 10 m/h, was observed in the case of magnesium hardness. In the case of nitrates (V), in the removal from the purified water, the difference between the filtration speed of 5 m/h and 15 m/h is about 30%, and in the case of Mn, it was about 28%. Minor differences, about 20%, were found by analyzing changes in iron concentration, the total hardness, and the calcium hardness.

Considering the results obtained and the removal of various contaminants in a simple technological system with adsorption on activated alumina at variable speeds filtration was considered as justifiable to assess the operation of this system in terms of the effectiveness of the removal of common pollutants and identifying the optimum speed of filtration of water through the system. According to the recommendations of literature, the filtration speed for removing iron and manganese from groundwater in the high-rate filters should be in the range of 7–10 m/h. Higher speeds should be used in cases where the outline tests were performed, which proved the effect of high

filtration speed of about 10–15 m/h. Moreover, studies of pre-test models in pilot-plant scale allow specifying the design parameters of the filtration process, such as filtration speed and the height of the filter bed, but also the operating parameters, which are related to the length of filter cycles, and the process of rinsing the filters. Obtained from pre-tests filtration speed can determine the size of the filtering surface, which is associated with the amount of investment costs for the necessary fittings, the number of filters or even the size of the building for filters at the water treatment plant [Weber 2012, Dymaczewski et al. 2005].

In summary, activated alumina is used primarily for removing arsenic and fluoride ions from water, whereby there has not been widely used for water treatment [Szatyłowicz, Skoczko 2016], as pollution of groundwater with fluoride ions is absent, and arsenic pollution is small and occurs only in some areas of the country. However, the increase of anthropological contamination may contribute to increased arsenic pollution of water. That was the purpose of these studies, to determine the removal efficiency of other pollutants commonly found in the system, in which one of the adsorption processes on Al₂O₃.

CONCLUSIONS

1. In the analyzed technological system containing adsorption on activated alumina optimal filtration rate equal 15 m/h was achieved for most contaminants. Unfortunately, this filtration rate significantly reduces removal of manganese and phosphate (V) for which the best effect of removing obtained at the speed of 5 m/h.

2. The use of activated alumina sorbent in the test water treatment process system can increase efficiency of nitrate (V) and phosphate (V) removal, and to a lesser extent, enhance the effect of removal: COD_{Mn} , color and turbidity.
3. The efficiency of iron and manganese removal on a bed of activated Al_2O_3 is negligible, and the sorbent is not suitable for effective iron and manganese removal, does not improve the efficacy of the technological system for these parameters.
4. The adsorption process of contaminants on Al_2O_3 should not be used as the final stage of water treatment, because can be released Al ions into the treated water. The test system was used activated carbon for the activated Al_2O_3 , which will remove a potential secondary pollutions of water.

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