

THE IMPACT OF POTASSIUM MANGANATE (VII) ON THE EFFECTIVENESS OF COAGULATION IN THE REMOVAL OF IRON AND MANGANESE FROM GROUNDWATER WITH AN INCREASED CONTENT OF ORGANIC SUBSTANCES

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Abstract

The article presents the results of studies concerning the impact of the method of Fe(II) ion oxidation (dissolved oxygen and potassium manganate (VII)) on the effectiveness of coagulation in the removal of iron and manganese from groundwater with an increased content of organic substances. The efficiencies of two coagulants were compared: aluminium sulphate (VI) and polyaluminium chloride (Floklor 1.2A). Among the used methods of iron (II) oxidation, the best effects have been achieved by potassium manganate (VII) because one of the oxidation products was manganese oxide (IV) precipitating from water. Better results in purifying the water were obtained with the use of a prehydrolysed coagulant Floklor 1.2 A than aluminium sulphate (VI).

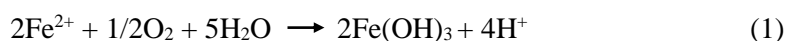
Keywords: groundwater, iron, manganese, organic substances, potassium manganate (VII), coagulation

1. INTRODUCTION

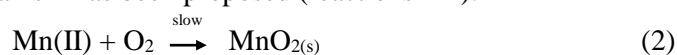
Traditional air oxidation in water treatment is used for the removal of divalent forms of iron and manganese for public health purposes. These ions are frequently present in groundwaters with low oxygen content [22, 28, 29].

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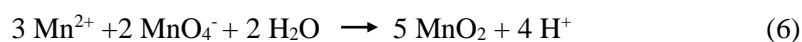
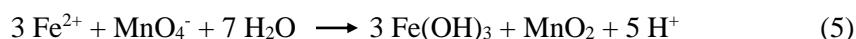
Ferrous iron removal from groundwater is based on transforming soluble ferrous iron (by oxidation) into insoluble ferric iron compounds. Iron is oxidized with oxygen (reaction 1) or other oxidants (potassium manganate (VII), ozone, chlorine), and with the use of bacteria that oxidise iron and manganese.



For bivalent manganese, autocatalysis plays an important role in oxygenation, and multiple-step mechanism has been proposed (reactions 2-4):

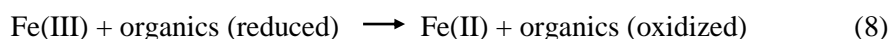
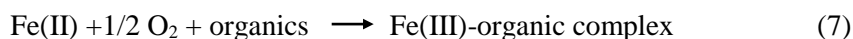


In some cases potassium manganate (VII) is used as an oxidant (reactions 5-6).



This step is very efficient separation if a coagulant is added [1, 29].

One of the problems with iron removal from groundwater is organic matters as they have a negative impact on the process of iron removal. Organic substances in water may interfere with the iron removal process, forming stable complexes with iron, both Fe(II) and Fe(III) and manganese. Albrektiene with colleagues [2] states that manganese doesn't form complex compounds with any fraction of organic matters. The Fe(III)-organic complexes are stronger and more stable as compared to Fe(II)-organic complexes. Considerable research has been conducted to explain the chemical nature of the complexed iron. Soil scientists have been quite consistent in stating that organics which cause these complexes or peptized small metallic colloids are of relatively high molecular weight (greater than 2000), whereas water chemists generally concede that low molecular weight organics are responsible for holding iron [19, 20, 24-26, 30, 31]. Jobin and Ghosh [10] established that in the presence of organic matter, Fe(II)-Fe(III) redox couple acts as a catalyst for the oxidation of the organic matter. In such systems, both complexation of Fe(II) and reduction of Fe(III) by organic matter are possible (reactions 7-9):





The relative rates of reactions 7 and 8 dictate the eventual oxidation of Fe(II) in the presence of organics. In such cases, the ferrous-ferric system acts as an electron transfer catalyst for the oxidation of organic material. Depending on the relative rates of Fe(II) oxygenation and Fe(III) reduction by organic material, substantial retardation of the over-all oxidation of Fe(II) by organic material is possible. The higher the pH, the more the oxidation process of the Fe(II)-organic complex is retarded. For example, at pH 8, decrease in rate constant by a factor of 10 results in doubling the half-life of the Fe(II)-organic complex with respect to oxidation [8, 19]. Organic compounds lengthen the time oxidization of Fe(II) to Fe(III) and decrease its effectiveness, and oxidization products are colloidal in character [1]. They also proved that the presence of organic compounds in water makes the removal of iron much more difficult. Therefore, several various attempts have been made in order to intensify removal of iron present in combinations with organic ligands. One of the recommended methods here is the application of strong oxidisers such as chlorine, potassium manganate (VII), ozone and hydrogen peroxide instead of aeration. The inclusion of the oxidation process into the water treatment technological system is justifiable (safe) only when the oxidizing agents used do not cause the occurrence of inorganic and organic oxidation by-products harmful to human health, and products of incomplete oxidation of organic substances are removed from the water [13, 14, 17]. The use of potassium manganate (VII) should be right for oxidizing Fe(II) occurring in compounds with organic substances because the oxidation process is additionally aided by the adsorptive and catalytic properties of the precipitated manganese oxide (IV) [11, 12, 18, 21]. There is very little information about the kinetics of oxidizing Fe(II) by means of potassium manganate (VII). It is believed that the reactions take place quickly with $\text{pH} > 7$. In an environment with neutral reaction, which is usual in natural water, potassium manganate (VII), is reduced to manganese oxide (IV). The hydrated colloidal manganese oxide (IV) which appears has a negative charge with $\text{pH} < 8$, and a positive charge with $\text{pH} \geq 8$ and a vast specific surface with good sorptive characteristics [3, 4, 7, 18]. The use of potassium manganate (VII), is particularly favourable if there is Mn(II) in the water being purified, because apart from iron (III) hydroxide a product of the reaction is manganese oxide (IV) which catalyses the oxidization of Fe(II) and Mn(II). The catalytic oxidization of Fe(II) and Mn(II) ions makes it possible to decrease the dose of potassium manganate (VII), below stoichiometric amounts if there are not any other reduced substances [7, 18, 27] in the water being purified. Another method suggested for treating colour waters contaminated with organic substances and

iron compounds is the coagulation process [5, 6, 9, 15-18]. It was proved that aluminium coagulants were more efficient in treatment of such water than iron salts, due to the fact that products of aluminium hydrolysis, unlike iron, do not make coloured complexes with organic substances. The total efficiency of coagulation process depends on: destabilization of colloidal pollutants and neutralization of anion charges by cation products of coagulant hydrolysis, ion exchange, surface complexing, adsorption on hydroxide particles of metals of coagulants applied and coprecipitation. The role of the mechanisms referred to above depends on pH reaction of the treated water, a quantity and charge of the products of coagulant hydrolysis as well as the initial concentration and form of organic substances [17, 32]. In recent years, prehydrolyzed aluminium coagulants, e.g. polyaluminium chloride and polyaluminium sulphate have been developed and researched. It has also been found that polymerised polyaluminium chlorides are more effective at removing organic substances and reducing turbidity than aluminium sulphate (VI) [5, 23, 14-17]. Better effectiveness of prehydrolysed aluminium coagulants is caused by the presence of polymeric aluminium complexes with large positive charges, which effectively destabilise and bridge the coloured organic pollution being removed [6]. The article presents the results of studies concerning the impact of the method of Fe(II) ion oxidation (dissolved oxygen and potassium manganate (VII)) on the effectiveness of coagulation in the removal of iron and manganese from groundwater with an increased content of organic substances.

2. MATERIALS AND METHODS

The subject of research was groundwater from Quaternary formations characterized by a high concentration of total iron – amounting to 5.80 mgFe/dm³, increased colour (B) – up to 38 mgPt/dm³ and turbidity (M) up to 21.1 NTU, and also an increased amount of organic substances (TOC from 3.60 to 6.40 mgC/dm³), and manganese (up to 0.60 mgMn/dm³). Iron (II) and iron (III) compounds were present in the water. The concentration of dissolved oxygen changed within the range of 0.50 to 0.60 mgO₂/dm³, and the pH value within the range of 6.84 to 7.37 (Tab. 1). These characteristics of the water were the cause of the unfavourable extent of the removal of iron compounds from it in the conventional system of underground water treatment. The turbidity and intensity of apparent and real colours of water proved the occurrence of iron in the form of colloidal and dissolved bonds with organic compounds, for the removal of which the coagulation process was used. The effectiveness of voluminal coagulation in groundwater treatment, after aeration and chemical oxidation by potassium manganate (VII) was determined in the studies. The raw water was aerated with compressed air for a period of time of 15- minutes,

obtaining a dissolved oxygen concentration at a level of about 10 mgO₂/dm³, that is, about 100 % water saturation with oxygen. Also, chemically pure solution of potassium manganate (VII) was used to oxidise the Fe(II) ions; the oxidation time amounted to 5 min, and the dose of oxidising agent was determined in view of the concentration of Fe(II) at an amount equal to 100% of the stoichiometric demand. The coagulation was performed in water samples with a volume of 1-dm³, applying 1-minute of rapid stirring at an intensity of 250-rev./min and 25-minute of flocculation with a stirring intensity of 30-rev./min.

Table 1. Groundwater quality indicators

Indicator	Unit	Value		
		Minimum	Average	Maximum
pH	-	6.84	-	7.37
Dissolved Oxygen	mgO ₂ /dm ³	0.50	0.55	0.60
Colour	mgPt/dm ³	19	26	38
Turbidity	NTU	1.73	4.50	21.1
Iron total	mgFe/dm ³	3.30	4.00	5.80
Iron (II)	mgFe/dm ³	2.80	3.40	4.00
Iron(III)	mgFe/dm ³	0.50	0.70	1.80
Manganese	mgMn/dm ³	0.20	0.29	0.60
TOC	mgC/dm ³	3.60	4.73	6.40

Aluminium sulphate (VI) (SAL) and polyaluminium chloride (Floklor 1.2A) dosed in the form of water solutions were used as coagulants. The doses of coagulants were expressed as mgAl/dm³. Six different doses of coagulants were selected 1, 2, 3, 4, 5 and 6 mgAl/dm³. After the process of coagulation, the water samples underwent 1 hour of sedimentation. The physical-chemical composition of both the raw groundwater as well as treated water was determined according to the International Standard methods. The pH of the raw water and the purified water was determined with an WTW Multi Line P4 with an combination pH electrode with temperature corrections. The dissolved oxygen of the raw water and the purified water was determined with an WTW Multi 3410 SET 4 with an optical oxygen sensor FDOR 925 (Germany). Turbidity was measured using the Hach 2100N Turbidimeter - the nephelometric method. The colour (according to Pt scale), total iron, iron (II) concentrations were determined with the Dr 3900 (HACH Lange) spectrophotometer. Iron (II) was measured using the 1,10 phenanthroline method. Total iron was measured using the same method except that a reducing reagent was also included in a reagent powder pillow to convert Fe(III) to Fe(II). The colour was indicated in accordance with ISO 7887-Method C [33]. Quartz

cuvette with path length of light 50 mm was used. It was determined after filtration of the water sample through a membrane filter of pore size 0.45 μm . Colour of the sample was calculated using following equation: $C = A_{410}/ad$ [mgPt/dm^3]. Where C is true color of the sample, A_{410} -absorbance of the sample at $\lambda = 410$ nm, a -specific absorption coefficient of the calibration solution of potassium hexachloroplatine and cobalt chloride [$\text{mm}^{-1}(\text{mgPt}/\text{dm}^3)^{-1}$], d -optical pathlength (mm) [33]. The TOC was measured with a Shimadzu TOC analyzer. Manganese concentrations were determined with the atomic absorption method (Analyst 700, Perkin Elmer Company, US).

3. RESULTS AND DISCUSSION

Efficacy of water treatment by oxidation and coagulation processes

The effects of the removal of pollutants from the water after oxidisation with potassium manganate (VII) showed that the precipitating manganese (IV) oxide increased the effectiveness of the removal of pollutants from the water. Comparison of the effects of coagulation in the treatment of aerated water and water after oxidisation with potassium manganate (VII) showed that the application of potassium manganate (VII) as an oxidising agent was much better than oxidisation with compressed air regards to the degree of the reduction in colour, turbidity, concentration of iron, manganese and organic substances (Figs 1, 2 and 5).

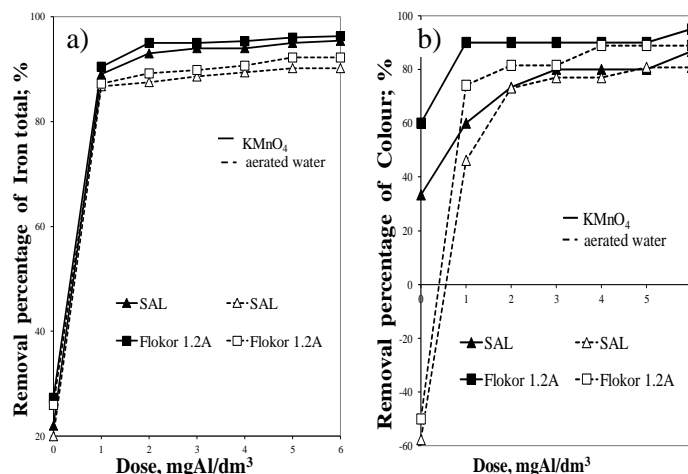


Fig. 1. Comparison of the effectiveness of coagulation in the reduction of the total iron concentration (a) and colour (b) in water after oxidisation with KMnO_4 or after aeration

Application of potassium manganate (VII) as an oxidizer, as compared with dissolved oxygen, was also beneficial because in the treated water pH did not grow. As a result of aeration the pH value of the treated water reached ≥ 8 which may have caused a complete dissociation of organic substances, and at the same time may have created conditions for forming of colourful hardly sedimenting ferro-organic combinations. The lack of increase in the pH in water after oxidation with potassium manganate (VII) ($\text{pH} = 7$ (KMnO_4)), $\text{pH} = 8$ (O_2)) and additionally precipitated manganese (IV) oxide was especially favourable in terms of the removal of the organic substances (Fig 2b).

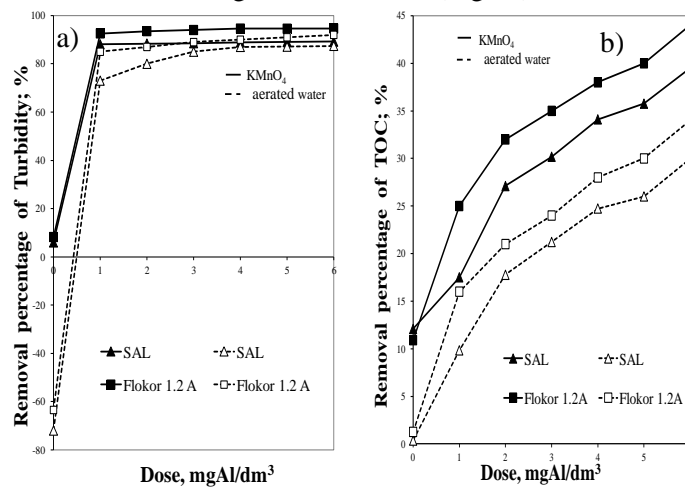


Fig. 2. Comparison of the effectiveness of coagulation in reducing the turbidity (a) and concentration of total organic carbon (b) in water after oxidation with KMnO_4 or after aeration

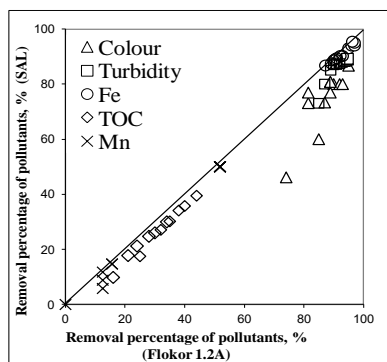


Fig. 3. The impact of the type of coagulants (SAL, Flokor 1.2A) on the effects of the removal of pollutants in the coagulation process after oxidation with KMnO_4 or after aeration

According to Ficek and Vella [7] application of potassium manganate (VII) to drinking water treatment systems can help solve a number of problems. It is a versatile oxidant that will react with organic compounds easily breaking carbon-carbon double bonds and oxidizing functional groups. The comparison of the usefulness of the studied coagulants in the removal of pollutants from water after oxidation of Fe(II) with potassium (VII) manganate, just as in the case of the treatment of aerated waters, confirmed the greatest effectiveness of coagulant which was prehydrolyzed with Flokor 1.2A (Figs 1-3).

Also, the concentration of the residual aluminium was determined in the studies. It was ascertained that also in this aspect, polyaluminium chloride was more useful than aluminium sulphate (VI). The lowest aluminium concentrations (0.28-0.22 mgAl/dm³) were found in water after oxidation with potassium (VII) manganate and coagulation with coagulant Flokor 1.2A ($D_c=1-6\text{mgAl/dm}^3$) (Fig. 4).

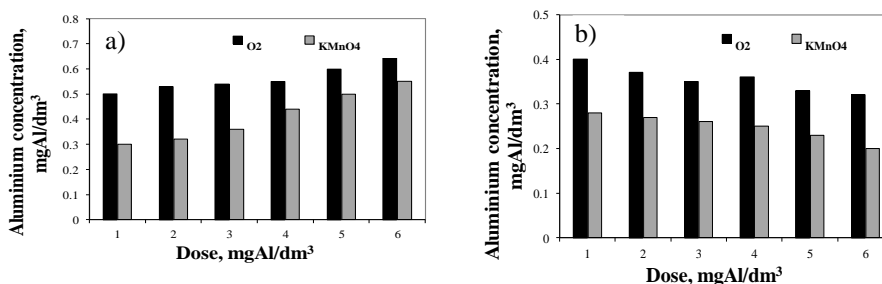


Fig. 4. The impact of the type and dose of coagulant and the oxidising agent on the change of aluminium concentration: a) SAL, b) Flokor 1.2A

Also according to Edzwald et al. [6] substitution of aluminium sulphate (VI) by polyaluminium chlorides reduced the concentration of aluminium residual in water after coagulation. Nowacka and Włodarczyk-Makula [23] and Edzwald et al. [6] believe that polyaluminium chlorides are more effective than aluminium sulphate (VI). The higher efficiency of polyaluminium chloride than aluminium sulphate (VI) is attributable to the fact that the solutions of polyaluminium chloride contain (in addition to aluminium monomers) large quantities of polycationic products generated during prehydrolysis of aluminium, which effectively neutralize the negative charges of the colloids [5,16,17]. The degree of the removal of manganese from water samples in which potassium (VII) manganate was applied as the oxidising agent was constant and did not depend on the dose of the coagulant. On the other hand, in the case of preceding the coagulation with aeration the effectiveness of the removal of manganese was reduced together with an increase in the dose of the studied coagulants (Fig. 5).

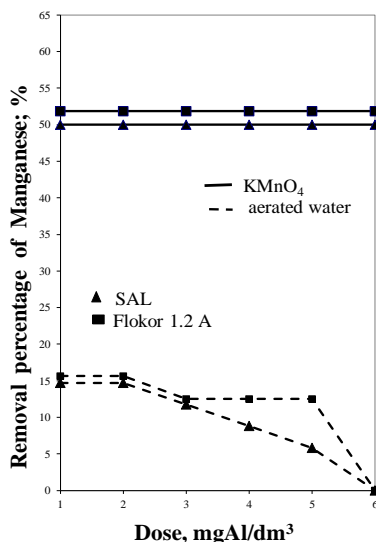


Fig. 5. The impact of the type and dose of coagulant and the oxidising agent used on the effectiveness of the removal of manganese from water

Analysis of the results of the studies presented in Table 2 showed that irrespective of the oxidising agent used, an increase in the effectiveness of coagulation and sedimentation in the removal of Fe(III) resulted in an increase in the degree of the removal of TOC, which proves the removal of Fe(III) together with the organic substances, and thus, the occurrence of iron in iron-organic bonds in water.

Table 2. Parameters of linear regression between the amount of TOC removed and the amount of Fe(III) removed in the coagulation process

Type of oxidant	Type of coagulant	Linear regression equation	Coefficient of determination (R ²)
O ₂	Flokor 1.2 A	$\eta\text{TOC} = 1.319 \eta \text{Fe(III)} - 98.101$	0.9501
	SAL	$\eta\text{TOC} = 1.812 \eta \text{Fe(III)} - 147.12$	0.9465
KMnO ₄	Flokor 1.2A	$\eta\text{TOC} = 1.850 \eta \text{Fe(III)} - 121.40$	0.9688
	SAL	$\eta\text{TOC} = 1.506 \eta \text{Fe(III)} - 88.805$	0.9457

In the water samples in which O₂ or KMnO₄ were used for oxidisation of Fe(II), no relationship between the effectiveness of the removal of manganese and TOC in the coagulation process. This confirms the opinion of Albrektiene [2] that manganese doesn't form colloids complexes with fraction of organic matters. The amount of colour substances removed from Fe(III) ($\Delta B/\Delta \text{Fe(III)}$) was

greater in the water samples after oxidization with KMnO_4 than after aeration and they increased together with a dosed of coagulants (Table 3).

Table 3. Influence of the type and dose of coagulant and the type of oxidizing agent on the amount of colour pollutant (mg) removed from 1 mg of Fe(III) ($\Delta B/\Delta \text{Fe(III)}$)

Type of oxidant	Type of coagulant	$\Delta B/\Delta \text{Fe(III)}$					
		Dose, mgAl/dm^3					
		1	2	3	4	5	6
O_2	Floklor 1.2 A	9.18	8.87	8.70	8.57	8.26	8.45
	SAL	4.80	6.88	7.19	7.19	7.64	7.66
KMnO_4	Floklor 1.2A	9.52	9.82	9.99	10.13	10.20	10.25
	SAL	9.37	9.02	8.69	8.51	8.39	8.50

4. CONCLUSIONS

The test results analysis leads to the following conclusions:

1. The effectiveness of the process of coagulation for the removal of iron, manganese and organic substances from underground water depended on the type of oxidizing agent used and the type of coagulant.
2. Better results in purifying the water were obtained with the use of a prehydrolysed coagulant Floklor 1.2 A than sulphate (VI) of aluminium. The highest concentration of the aluminium residual was found in samples subject to coagulation with aluminium sulphate (VI).
3. Independently of the type of coagulants used and the type of oxidizing agent, iron compounds were removed together with organic substances, which proved that they are present in water in the form of colloidal iron-organic compounds. No relationship between the effectiveness of the removal of manganese and organic substances in the coagulation process.
4. A greater efficiency in removing iron, manganese and organic substances in oxidation and coagulation processes was achieved when applying potassium manganate (VII) as an oxidizer, because one of the oxidation products was manganese oxide (IV) precipitating from water.
5. The experimental results suggested that manganese oxide (IV) may adsorb naturally occurring organic materials through surface bonding to form bigger particulates, thus increasing the floc density and improving the removal of organic particulates and the inorganic particles such as iron and manganese.
6. The degree of the removal of manganese from water samples in which potassium (VII) manganate was applied as the oxidising agent was constant and did not depend on the dose of the coagulant but the removal of manganese depended on the amount of precipitated manganese oxide (IV).

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WPŁYW MNANGANIANU (VII) POTASU NA SKUTECZNOŚĆ KOAGULACJI W USUWANIU ZWIĄZKÓW ŻELAZA I MANGANU Z WODY PODZIEMNEJ O PODWYŻSZONEJ ZAWARTOŚCI SUBSTANCJI ORGANICZNYCH

Streszczenie

Skuteczności usuwania żelaza i manganu z wody podziemnej o podwyższonej zawartości substancji organicznych zależała od rodzaju stosowanego utleniacza (tlen rozpuszczony lub manganian (VII) potasu) oraz dawki i rodzaju koagulantu (siarczan (VI) glinu lub chlorek poliglinu). Spośród stosowanych sposobów utleniania żelaza (II) przed procesem koagulacji lepsze efekty z uwagi na usuwanie żelaza, manganu i substancji organicznych zapewniło utlenianie manganianem (VII) potasu niż napowietrzanie. Skuteczność badanych koagulantów w usuwaniu zanieczyszczeń zwiększała się wraz ze wzrostem ich dawki, a lepsze efekty oczyszczania wody podziemnej zapewnił koagulant wstępnie zhydrolizowany Flokor 1,2A niż siarczan (VI) glinu.

Słowa kluczowe: wody podziemne, żelazo, mangan, substancje organiczne, manganian (VII) potasu, koagulacja

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