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Original Research Article

IMPACT OF POLYELECTROLYTES ON THE EFFECTIVENESS OF TREATMENT OF GROUNDWATER WITH INCREASED NATURAL ORGANIC MATTER CONTENT

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Abstract

The article discusses effectiveness of treatment of groundwater with increased natural organic matter content with the use of organic polyelectrolytes. The effects of water treatments were determined by the ionic character of the polyelectrolyte and its dose. Due to the amount of removed general ferric and coloured matters a greater usefulness of anionic and non-ionic polyelectrolytes was shown, while due to decreased turbidity and TOC, cationic flocculants proved more useful. Using the Praestol 2540 semi-anionic polyelectrolyte as the substance aiding the coagulation process decreased the effectiveness of groundwater treatment, especially in terms of the removal of iron and organic substances when using the PIX-112 coagulating agent.

Keywords: groundwater, natural organic matter, polyelectrolytes: cationic, anionic, non-ionic, coagulation

1. INTRODUCTION

Polyelectrolytes, i.e. high molecular organic polymers, have been used in water treatment since 1950. We distinguish between natural and synthetic

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polyelectrolytes. Natural polyelectrolytes are usually produced from starch, while the synthetic kind is produced as a result of polymerisation of organic monomers with unsaturated bindings [5]. Due to the type of ionogenic groups, polyelectrolytes are divided into: non-ionic, anionic and cationic. Polyelectrolyte's charge is produced as a result of dissociation of ionogenic groups of acidic or alkaline character. Molecule of a polyelectrolyte can have hundreds or even thousands of dissociating groups since their degree of polymerisation can be significant. Capacity for dissociation depends on the structure of the polyelectrolyte, pH of the solution, and the content of ionogenic groups. As a result of dissociation, anion and polycation are created in cationic polyelectrolytes water, and cation and polyanion in anionic polyelectrolytes water. Anionic and non-ionic polymers are used as flocculation enhancers, while cationic polymers are also used as independent coagulants [3, 4, 7, 14]. The purpose of using polyelectrolytes as coagulation enhancers is to reduce the dosage of basic coagulant, accelerate the flocculation process, produce flakes of good sedimentation properties, and reduce the negative impact of low temperatures on the effectiveness of pollution removal in the coagulation process [3, 4, 5, 7]. Currently, there are several thousands organic polyelectrolytes produced worldwide, among which those which obtained the hygienic approval can be used for the treatment of water intended for human consumption. There are, however, countries such as France and Japan where using synthetic polyelectrolytes in water treatment is forbidden due to the risk of chlorinated organic compounds creation arising from the reaction of chloride with polymers remaining in the water, and the creation of monomers in the treated water. The working mechanism of polyelectrolytes is not fully known and explained. It is claimed that polyelectrolytes cause destabilisation of removed colloids as a result of charge neutralisation, and by acting as a bridging agent and a cross-linker cause agglomeration of microflocs, and thus the emergence of solid flocs of good sedimentation properties [5, 7]. Literature analysis [3-5] indicates that during coagulation of hydrophobic colloids, dominates the adsorption of polyelectrolytes on their surface, while in case of hydrophilic colloids, key role is played by mechanisms of destabilisation and precipitation of removed pollution. According to Edzwald [1], aluminium salts coagulation with the use of cationic polyelectrolytes is the most effective way to remove humus substances from water, while the polymers used can react mechanically, chemically, and cause destabilisation of removed colloids as a result of charge neutralisation. Mechanical reaction consists in confounding the coagulating particles with long chains of cationic, anionic and non-ionic polymers, which causes the accelerated creation of large floccule suspensions. Neutralisation of the colloids' charge with the use of organic polymers, similarly as in the case of coagulants, consists in decreasing the electrokinetic potential of colloids being removed [3, 4, 7, 16, 17].

Chemical reactions are credited with the lowest impact on the course and effectiveness of the process. For the proper progress of coagulation, apart from ensuring appropriate coagulant or flocculent, it is very important to use optimal dosage thereof, ensuring maximal decrease of the absolute value of electrokinetic potential. It is claimed that dosages of polyelectrolytes should be from 0.1 to 1% of the basic coagulant dose [3-5]. With optimal amount of cationic flocculants, there are polymer groups created from the adsorbed colloidal particles capable of rapid sedimentation. With the amount lesser than optimal, not all colloidal particles will be bound, while with a larger amount occurs the decrease of the effects of pollution removal and a stable colloidal system of a positive surface charge is created in case of using cationic polyelectrolytes. It was also shown that the effectiveness of the coagulation process is affected not only by dosage of the polyelectrolyte, but also by the order of adding in to the water in relation to the basic coagulant. Most often it is added to the water with a delay from 1 to 3 minutes in relation to the basic coagulant, and also at the flocculation stage or simultaneously with the primary coagulant. In recent years, there have also been studies conducted on the usefulness of the so-called blends in water treatment, i.e. mixtures of non-organic coagulant and organic polymers [5]. In Poland, water supply system uses groundwater and surface water. Water supply in most villages and small towns is based on groundwater resources. Only some plants source groundwater with physical and chemical composition meeting the standards of water intended for human consumption [5, 20]. Components which disqualify sourced groundwater are mainly ferric, manganese, aggressive carbon dioxide, and in many cases also natural organic matter (NOM), usually including humus substances i.e. complex heterocyclic acid polymers [5, 15-19, 22, 23]. Application of processes traditionally used for purification of groundwater, i.e. aeration, flocculation, sedimentation, rapid filtration with filter bed for removing manganese, and disinfection, are not sufficient for treatment of water containing organic substances and ferric compounds. As a result of water aeration, agglomerates of ferrous hydroxide (III) with good sedimentation properties are not produced because the natural organic matter present in the water creates water-soluble coloured bonds of a colloidal character with ions and ferric compounds [2, 21]. Due to the fact that in the studies previously conducted by the author [6-13] ferric compounds (III) created as a result of aeration agglomerated with difficulty due to the presence of organic substances, this study attempted to intensify flocculation by using polyelectrolytes.

2. MATERIALS AND METHODS

The purpose of the study was to determine the effectiveness of polyelectrolytes as independent coagulants and as enhancers in the process of coagulation with aluminium (VI) sulphate or ferric (III) sulphate (VI) in treatment of groundwater. of increased organic substances content, considering also the removal of iron compounds. The subject of the study was the groundwater from quaternary aquifer characterised by variable level of organic substances (TOC 5.00 ÷ 6.50 mgC/dm^3), variable content of iron compounds (5.84 ÷ 7.11 mgFe/dm^3), increased colour intensity (16 ÷ 20 mgPt/dm^3) and turbidity (3.11 ÷ 12.7 NTU). The water was characterised by the content of Fe(II) compounds from 2.51 to 5.84 mgFe(II)/dm^3 and Fe(III) from 1.22 to 3.33 mgFe(III)/dm^3. Raw water was aerated with compressed air in time span of 15 minutes (t_a) achieving the dissolved oxygen concentration of 10 mgO$_2$/dm^3 i.e. approx. 100 % oxygen saturation. Water aeration caused the increase of turbidity, colour and pH of water by one unit. Aeration, sedimentation and filtration with a "worked-in" sand filter bed layered with oxides of iron and manganese at the speed of 5m/h did not provide the required level of the decrease of colour (25 mgPt/dm^3), turbidity (24 NTU), concentration of iron (4.0 mgFe/dm^3), manganese (0.80 mgMn/dm^3) and organic substances (6.50 mgC/dm^3). For that reason, in order to intensify the agglomeration of ferric compounds, after the aeration the following 1% aqueous solutions of polyelectrolytes were dosed into the water: cationic anionic, non-ionic, and their doses (D_p) were altered within the range of 0.1 to 0.5 mg/dm^3. After the intense mixing of water with polyelectrolytes, 25 minutes of flocculation was applied with mixing intensity of 30 rotations/minute. Afterwards, water samples were subjected to sedimentation for 2h and filtration through a soft filter. Table 1 shows the characteristic of polyelectrolytes tested, among which only Praestol-2540 and Praestol-2515 hold the certificate from the National Institute of Hygiene. The following were used as coagulants: aluminium sulphate (VI) - SAL and iron sulphate (VI) under the trade name of PIX-112, dosed in standard water solutions. Dose of coagulants (D_c) was expressed in mgAl/dm^3 or mgFe/dm^3 and dosed in the amount of 1.78 mgAl(Fe)/dm^3. Dose of coagulants was determined on the basis of previously conducted research [6-13]. Apart from primary coagulants, also the solution of semi-anionic polyelectrolyte (Praestol 2540) of 1000 mg/dm^3 concentration was dosed in the amount of D_p = 0.15 or 0.25 mg/dm^3, in advance or with delay in relation to the time of coagulant dosing. Coagulation was conducted in water samples of 1 dm^3 capacity, using 1 minute of rapid mixing with intensity of 250 rotations/min and 25 minutes of slow mixing with the intensity of 30 rotations/min. After the coagulation, samples were subjected to 2h of sedimentation, some experimental series were additionally subjected to filtration through a soft filter. Physical and chemical composition of raw and treated water was determined in accordance with the existing PN.

Table 1. The characteristic of polyelectrolytes used [24]

Polyelectrolyte type						
Praestol 611 BC	Praestol 650 BC	Praestol 624 BC	Praestol 2515	Praestol 2540	Superflock A-110	N-100
Ionic character						
Weakly cationic	Moderately cationic	Strongly cationic	Weakly anionic	Moderately anionic	Weakly anionic	Non-ionic
Viscosity, mPa.s						
35	65	30	400	600	180	26
pH						
7.0	7.0	6.5	7.0- 8.0	7.0-8.0	7.4	6.4
Molecular weight, kD						
6 mln	6 mln	6 mln	14 mln	14 mln	11 mln	11 mln

In the article, the measure of effectiveness for water treatment (η) was assumed to be the ratio of decrease of the values of studied water quality indicators to the reduction required for achieving values allowable in water indented for human consumption, expressed in percentage [20]. The decrease of indicator's value (ηX) was assumed to be the treatment effect resultant from aeration, polyelectrolyte coagulation and sedimentation of post-coagulation suspensions, as well as as a results of aeration, coagulation (SAL or PIX-112) enhanced with the Praestol-2540 polyelectrolyte and sedimentation of post-coagulation suspensions.

3. RESULTS AND DISCUSSION

3.1. Impact of a type of polyelectrolyte on the effectiveness of groundwater treatment

Results of the experiment on the effect of a type of polyelectrolyte (D_p=0.5 mg/dm^3) on the effectiveness of groundwater treatment in the process of aeration and 2h of sedimentation is presented in table 2. It has been shown that all organic polymers (characteristic of which is presented in table 1) regardless of their ionic character, by causing agglomeration improve sedimentation qualities of solid particles present in the water after aeration.

Table 2. Comparison of the amount of pollution removed in 2h of sedimentation with the application of different polyelectrolytes (D_p = 0.5 mg/dm^3)

Indicator, Unit	Polyelectrolyte type						
	Praestol 611 BC	Praestol 650 BC	Praestol 624 BC	Praestol 2515	Superflock A-110	Praestol 2540	N-100
	Weakly cationic	Moderately cationic	Strongly cationic	Weakly anionic	Moderately anionic	Weakly anionic	Non-ionic
	Reduction of the indicator ΔX, mg/dm^3 *						
Colour, mgPt/dm^3	24	25	23	33	40	56	40
Turbidity, NTU	22.2	21.2	21.0	16.3	17.5	19.0	17.1
Iron total, mgFe/dm^3	2.04	2.06	2.02	2.09	2.14	2.90	2.18
TOC, mgC/dm^3	0.25	0.23	0.47	0.20	0.13	0.19	0.11

** - in comparison to the water sample after aeration, flocculation and 2h of sedimentation without a polyelectrolyte*

Considering the amount of pollution removed (Tab. 2), in terms of reduction of total iron concentration the anionic polyelectrolytes and non-ionic polymer proved more useful, while in terms of reduction of turbidity and TOC cationic polyelectrolytes were more effective. In case of removing organic substances, strongly cationic polyelectrolyte was the most effective. Water samples treated with anionic and non-ionic polyelectrolytes after 2h of sedimentation showed more than a twofold increase of turbidity and colour compared to raw water. Additional filtration of water samples through a soft filter paper increased the degree of elimination of iron compounds and reduction of turbidity (Fig. 1). The contribution of filtration to the reduction of total iron concentration was comparable for all polyelectrolytes tested, while in terms of turbidity reduction it was greater in case of sample with added anionic and non-ionic polyelectrolytes (Fig. 1). Despite the fact that filtration of water samples through soft filter paper increased the effectiveness of removing iron and reduction of turbidity, the required values of these indicators for water intended for human consumption were not achieved. The lowest concentration of iron was 0.25 mgFe/dm^3 (moderately cationic polyelectrolyte 650 BC), and the lowest turbidity was 2.28 NTU (weakly cationic polyelectrolyte 611 BC).

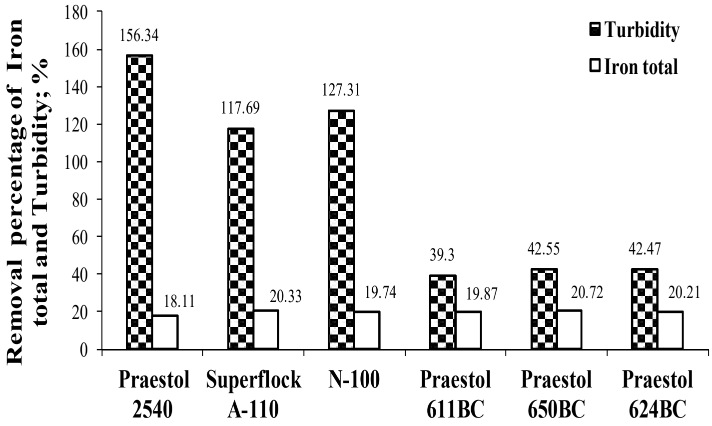

Fig. 1. Reduction of turbidity and concentration of total iron in the filtration process

Sufficient reduction of colour was ensured by all of the tested cationic polyelectrolytes. Anionic and non-ionic polyelectrolytes despite enhancing the effects of colour reduction after aeration, have not ensured a sufficient reduction of its intensity. Taking into account the fact of the increase of iron concentration and colour and turbidity of water after 2h of sedimentation and additional filtration through filter paper (compared to raw water), the agglomerating effect of anionic and non-ionic polyelectrolytes was unsatisfactory.

3.2. The effect of the Praestol 2540 moderately anionic polyelectrolyte dose on the effectiveness of groundwater treatment

Due to the fact that among the selected polyelectrolytes with the attestation of Polish Institute of Hygiene, the moderately anionic polyelectrolyte Praestol 2540 ensured better effects of removing pollution than the weakly anionic Praestol 2515, the impact of the dose of Praestol 2540 on the water treatment effects was determined (Fig. 2). The effectiveness of reducing turbidity and colour, and removing iron compounds increased along with the increase of the dose of polyelectrolyte from 0.1 to 0.4 mg/dm^3. At the same time, in the range of doses from 0.1 to 0.3 mg/dm^3, the increase of effects of removing iron was very little and reached only around 1.60 %. Increasing the dose to 0.5 mg/dm^3 caused reduction of the degree of removal of iron, turbidity and colour, while the small decrease of effectiveness in removing organic compounds was determined already for the dose of 0.4 mg/dm^3 (Fig. 2).

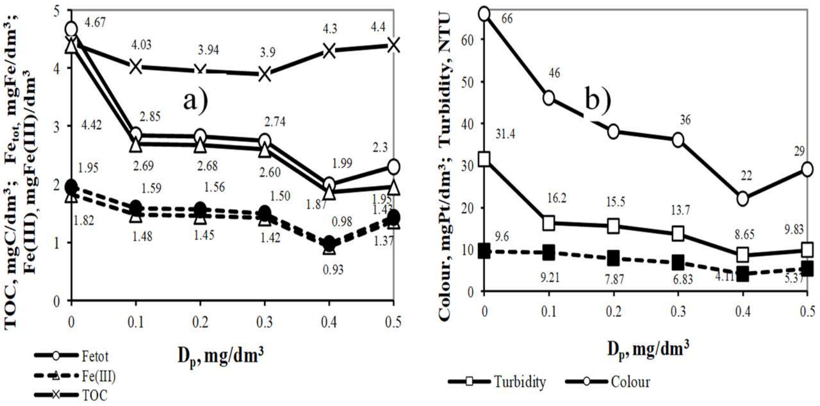

Fig. 2. The effect of a dose of polyelectrolyte (D_p) Praestol 2540 on:
TOC and iron concentration (a) and turbidity and colour of water (b)
(—— after 2h of sedimentation, - - - - after 2h of sedimentation and filtration)

Introduction filtration process into the treatment system improved the effectiveness of removing iron compounds and decreasing turbidity. However, even the most effective dose of polyelectrolyte of 0.4 mg/dm^3, has not ensured the concentration of iron of ≤ 0.2 mgFe/dm^3, and in case of turbidity and colour, despite their decreasing in comparison to a sample without polyelectrolyte, an increase of their value in relation to raw water was determined.

3.3. The effect of polyelectrolyte dosing on the effectiveness of the coagulation process

The analysis of test results presented in table 3 showed that enhancing the coagulation process with moderately anionic polyelectrolyte (Praestol 2540) dosed in advance in relation to the primary coagulant (SAL or PIX-112), has decreased the effectiveness of removing organic substances, particularly significantly in case of using the ferric coagulant PIX-112.

Table 3. The impact of a dose of the moderately anionic polyelectrolyte Praestol 2540 (D_p) on the effectiveness of lowering (η) TOC, iron content and the intensity of water colour (D_c=1.78 mgAl(Fe)/dm^3, t_a=15min) in the process of coagulation and sedimentation

Removal effectiveness (η), %	SAL			PIX -112		
	D_p, mg/dm^3					
	0	0.15	0.25	0	0.15	0.25
TOC	26.89	22.41	24.76	25.47	16.51	12.50
Fe_{tot}	83.40	80.17	88.75	79.15	75.11	69.15
Colour	28.57	21.43	50.00	-14.29	-21.43	-42.86

Together with the increase of the dose of polyelectrolyte enhancing the coagulation process with the PIX-112 coagulant, the effectiveness of removing TOC decreased, as well as the effectiveness of removing iron compounds and actual colour.This suggest that the polyelectrolyte has probably partially remained in the water creating coloured and difficult to sediment bonds with iron ions introduced into the water together with the iron coagulant. In case of coagulation with aluminium sulphate -SAL, the polyelectrolyte has again not increased the effectiveness of organic pollution removal, however, using this flocculent in the dose of 0.25 mg/dm^3 has improved the effects of removing iron and coloured pollution from the water. Despite the above mentioned effects, enhancing coagulation with polyelectrolyte and additional filtration of water samples through filter paper has not ensured a degree of removal of iron compounds and reduction of turbidity sufficient for water intended for human consumption.

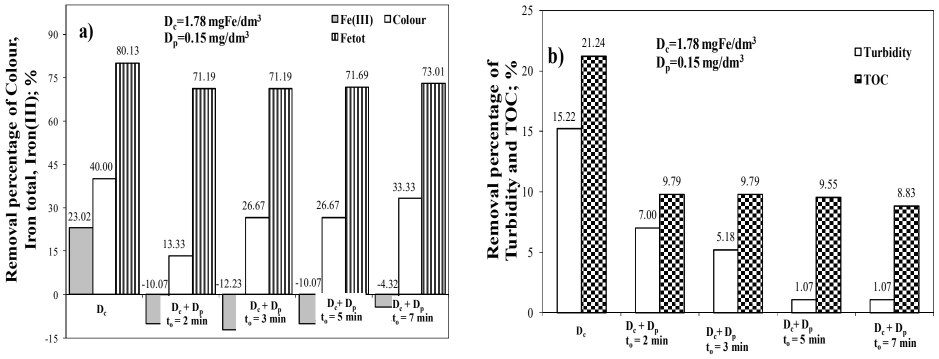

Fig. 3. The effect of dosing the Praestol 2540 polyelectrolyte during slow mixing on the effectiveness of: removing iron compounds and reducing colouring (a) and removing organic substances and reducing turbidity of water (b) in coagulation and 2h of sedimentation (t_o - delay of dosing of polyelectrolyte in relation to the primary coagulant)

Figure 3 shows the results of tests concerning the effect of dosing the moderately anionic polyelectrolyte (Praestol 2540) during flocculation on the effectiveness of groundwater treatment. Moderately anionic polyelectrolyte in the amount of (D_P) 0.15 mg/dm^3 was dosed with delay in relation to the iron coagulant PIX-112 (D_c=1.78 mgFe/dm^3) with total time (t_o) from 2 to 7 min. The analysis of results showed that polyelectrolyte has not improved agglomeration and sedimentation qualities of the pollution being removed, but conversely, has decreased the effects of coagulation with iron sulphate and sedimentation of post-coagulation suspensions. This could have been caused by the insufficient mixing of the polyelectrolyte with the water being treated. One cannot dismiss its stabilising effect on iron compounds (mainly Fe(III)), which was shown by the increase of iron concentration (III) in relation to iron concentration determined in the treated water without polyelectrolyte. A significant, yet insufficient improvement of the degree of reduction of turbidity, concentration of total iron and Fe(III) was ensured by additional filtration of water samples, effectiveness of which is shown in table 4.

Table 4.The effects of time (t_o) of dosing polyelectrolyte (D_p=0.15 mg/dm^3) on the effectiveness of filtration ($\Delta\eta_f$) in reducing turbidity and concentrations of iron and ferric (III)

Filtration effectiveness %	PIX -112, D_c = 1.78 mgFe/dm^3				
	D_p= 0	t_o, min			
		2	3	5	7
$\Delta\eta_f$ Fe_{tot}	14.9	22.85	23.01	23.67	23.84
$\Delta\eta_f$ Fe(III)	61.15	89.93	93.52	98.56	94.97
$\Delta\eta_f$ Turbidity	52.36	57.69	67.73	73.82	80.82

4. CONCLUSIONS

The analysis of test results showed that:

1. Tested organic polymers were insufficient in improving the effects of agglomeration of removed pollution, and their effect of sedimentation qualities of suspensions depended on dose and ionic character of organic polymers.
2. Due to the amount of removed total iron and coloured pollution anionic and non-ionic polyelectrolytes proved more useful, while in terms of reduction of turbidity cationic polyelectrolytes were more useful. Tested polyelectrolytes were least effective in improving the effectiveness of removing organic substances and total iron. In case of removing organic substances, strongly cationic polyelectrolyte was the most effective.
3. Using too large of a dose of moderately anionic polyelectrolytes Praestol 2540 (D_p= 0.5 mg/dm^3) decreased the effectiveness of water treatment because the surplus of organic polymer stabilised iron (III) compounds present in the water, creating with them coloured iron-organic bonds.
4. Using moderately anionic polyelectrolyte Praestol 2540 as a coagulation enhancer decreased the effectiveness of groundwater treatment, especially in case of using the iron coagulant PIX-112, which confirms its stabilising effect towards the ions of iron present not only in the water being treated, but also introduced to it with the coagulant.

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WPŁYW POLIELEKTROLITÓW NA SKUTECZNOŚĆ OCZYSZCZANIA WODY PODZIEMNEJ O PODWYŻSZONEJ ZAWARTOŚCI NATURALNEJ MATERII ORGANICZNEJ

Streszczenie

W artykule omówiono skuteczność oczyszczania wody podziemnej o podwyższonej zawartości naturalnej materii organicznej z zastosowaniem polielektrolitów organicznych. O efektach oczyszczania wody decydował charakter jonowy polielektrolitu oraz jego dawka. Ze względu na ilość usuniętego żelaza ogólnego oraz związków barwnych wykazano większą przydatność polielektrolitów anionowych i niejonowego, a z uwagi na obniżenie mętności i OWO flokulantów kationowych. Zastosowanie polielektrolitu średnioanionowego Praestol 2540, jako substancji wspomagającej proces koagulacji, zmniejszyło skuteczność oczyszczania wody podziemnej, a zwłaszcza usuwania żelaza i naturalnej materii organicznej, w przypadku stosowania koagulantu żelazowego PIX-112.

Słowa kluczowe: wody podziemne, naturalna materia organiczna, polielektrolity: kationowe, anionowe, niejonowe, koagulacja

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