Evaluation of Optimal Conditions for the Application of Capillary Materials for the Purpose of Water Deironing

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ABSTRACT
Iron is one of the most common components in water that adversely affect humans, other living organisms and parameters of water bodies. Therefore, during using and consuming natural waters, and discharging sewage into surface water bodies, it is necessary to remove iron compounds from the aquatic environment. The use of capillary materials in water purification processes is a promising area of research. Experimental data proved the high efficiency of capillary materials application, providing higher efficiency of iron ions removal from model solutions for real water sources of different origin compared with the traditional method of settling. The main advantage of the application of materials with capillary properties is the simplicity of implementation of the method and there is no need to use electricity and any additional reagents. The effect of various factors on the process of water deironing with the application of capillary materials was studied. It was found out that the density has a little effect on the process at an iron concentration range from 1 to 5 mg/dm3. The increase in competing ions content intensifies the process of divalent ferrous ions transition to trivalent. During the deironing of artesian water, the degree of iron removal exceeded 90 %, while the residual concentration was below the MPC.

Keywords: heavy metals, natural waters, capillary filter, removal of iron, mineralization, capillary filter density

INTRODUCTION
The discharge of harmful substances of technogenic origin leads to its accumulation in significant quantities resulting in disturbances of natural geochemical substances cycles which cause the pollution of water sources (Buzylko et al. 2018, Naidu et al. 2019, Kolesnyk et al. 2020). Heavy metals are the most common highly toxic pollutants in water bodies (Malik et al. 2019, Vardhan et al. 2019, Sorochkina et al. 2018). A peculiar feature of heavy metal ions is that they do not break down under natural conditions, but only their shape changes and accumulation in various components of the ecosystem takes place resulting in their damages (Tchounwou et al. 2012; Fazzo et al. 2017, Halysh et al. 2020a). Low-waste complex technologies for wastewater treatment from heavy metal ions will prevent their accumulation in surface water (Trus et al. 2019b, Trus et al. 2020c, Skiba et al. 2018).

Analysis of literature data shows that for the removal of different heavy metal ions from water various methods can be applied, for instance, membrane (nanofiltration (Trus et al. 2020b, Al-Rashidi et al. 2013, Zheng et al. 2018) and ultrafiltration (Ambiando et al. 2017, Kasim et al. 2017)), electrochemical (Trokhymenko et al. 2020, Chen et al. 2018, Kim et al. 2013), ion exchange (Hu and Boyer 2018, Dąbrowski et al.
Removal of iron from water is a rather difficult and unsolved problem (Ellis et al. 2000, Trus et al. 2020a). As a rule, sources of water supply are surface and groundwater, the qualitative and quantitative indicators of which are determined by the natural conditions of origin and by the level of anthropogenic load (Buzyllo et al. 2020). Water quality, depending on the purpose of use, is regulated by state standards and by other documents. In recent decades, almost all surface waters of Ukraine have been intensively polluted due to increasing anthropogenic load because of the discharge of insufficiently treated water, the inflow of surface agricultural lands, industrial and urban areas, and so on. The quality of river water, especially at the sources, meets the requirements of I – II quality class from an ecological point of view. However, some regions of Ukraine are characterized by high iron content in natural waters. It was established that drinking water contributes to the accumulation of iron in the human body. The water of many groundwater aquifers in Ukraine have a high iron content, which ranges from 0.5 to 30 mg/dm³ and even more, but more often up to 5 mg/dm³ (Shevchenko et al. 2013, Dolina et al. 2016).

Prolonged consumption of water with high content of iron leads to numerous diseases, for example, liver disease, increases the risk of heart attacks, adversely affects the central nervous system and reproductive function (Duruibe et al. 2007; Jaishankar et al. 2014). High content of iron ions gives water a brownish color, unpleasant taste, odor, causes overgrowth of water supply networks, and it is a reason for the defects in the textile, food, paper and cosmetics industries. Specific bacteria and growths can affect pipes and heat exchangers at the presence of iron reducing the cross section. Analysis of sludge from heat exchangers indicates the presence of iron, calcium and magnesium. That is why iron excess in drinking water is a dangerous impurity (Chowdhury et al. 2016, Kumar et al. 2012, Khashij Ma et al. 2016).

Thus, the high content of iron in surface and groundwater requires mandatory deironing, i.e. it is necessary to take a complex of measures to reduce the iron content. Today the deironing of water is an important economic, environmental and social problem that has scientific, theoretical and practical significance. This is one of the priority areas for providing the population with quality water in Ukraine.

The method of water deironing can be chosen depending on the chemical composition of water, the degree of deironing, the productivity of the station, technological tests (Khadse et al. 2015). Deironing of surface water can be carried out by reagent methods. In this case, different oxidants, such as chlorine and potassium permanganate, lime, soda are added into the source water (Aziz et al. 2020, van Genuchten and Ahmad 2020, Vries et al. 2017).

Reagent-free methods are most often used for groundwater deironing because of its simplicity and cheapness. The process consists in the fact that in the aeration device the water is saturated with oxygen, while carbonic acid is partially removed, iron is partially oxidized. Then the water is settled in tanks and filtered on filters, where the formed flakes of iron hydroxide are removed. Only if the process of deironing by the reagent-free method is not satisfied, then reagent method is applied. In this case, different oxidants, such as chlorine, potassium permanganate, lime, soda are added into the source water.

Despite the effectiveness of the mentioned methods, there are many disadvantages such as the generation of harmful sludge, high reagent consumption, high operational cost, complexity and high energy consumption.

The latest promising technology is the use of filters made of capillary-porous materials. Such filters consist of a housing, a porous filter material, pneumatic cylinders and a tray for collecting purified water. The operation of this filter involves several stages. First, wastewater or slurry is get into the filter housing between the capillary-porous material. The compressing of filter housing, cause the absorbtion of water by capillaries of the porous material. The compressing of filter housing, cause the absorbtion of water by capillaries of the porous material. As a result, the purified water through the perforated partitions inside the housing is collected in a special tray, and the separated solid particles in the form of sludge are removed (Fadel 2014).

The capillary effect in combination with other processes has become quite widely used in various fields. Certain methods of chemical analysis are based on the phenomenon of capillarity, many industrial and household equipment contain capillary tubes or materials. But, at the same time, there are no technologies that would
consider the use of the capillary effect as the main technological process in water purification or water treatment. Therefore, research in this direction is quite promising.

The study of the processes of iron ions removal from waters of different origins is an urgent problem. The process of water deironing with the application of capillary-porous materials can be affected by different factors. To evaluate the effectiveness of the use of materials with capillary properties it is necessary to study the influence of filter density and the presence of competing ions on the efficiency of iron ions removal.

**MATERIALS AND METHODS**

**Materials**

Non-sterile gauze material cutted into pieces was used as a capillary material. Tape length and width of the material were 25 cm and 3 cm, respectively. To prepare the model solutions of the natural waters, chemicals such as Fe^{2+}, NaCl were used. Artesian water from the Kyiv region (Coordinates: 50°26′55″N 30°27′33″E) was used to determine the efficiency of the proposed method. The parameters of the artesian water was the following: C (Fe^{3+}) = 1.9 mg/dm³, hardness = 4.5 mg-eq/dm³, C (Ca^{2+}) = 2.9 mg-eq/dm³, C (Mg^{2+}) = 1.6 mg-eq/dm³, alkalinity = 3.9 mg-eq/dm³, mineralization = 405 mg/dm³, pH = 7.9.

**Influence of capillary filter density on iron removal processes**

Capillary filters with different number of filter layers were immersed in 100 cm³ flasks with a prepared model solution of Fe^{2+} with different concentrations. One end of the filter was immersed in the solution, the other was removed outside the flask. Due to the difference in the levels of the liquid phase at the ends of the filter, the solution dripped into another flask. Samples of the liquid filtered by capillary filters were taken for further analysis for the determination of residual iron ions concentration. The ambient temperature was 25 °C.

Spectrophotometric method for iron ions determination (Li et al. 2013) was used to estimate its initial and equilibrium concentrations in solution. Water solutions were filtered through capillary filters according to a statistical 2²-factorial design as shown in Table 1. The variables studied were filter density (X₁) and iron ions initial concentration (X₂). The concentration of Fe^{3+} in solution after passing through the filter (Y) was investigated. The model of the process is presented as a second-order polynomial that can help to study the individual and combined effects of the variables:

\[ Y = b_0 + b_1X_1 + b_2X_2 + b_3X_1X_2 + b_4X_{12} + b_5X_{22} \]

where: Y is a response factor, \( b_0, b_1, b_2, b_3, b_4, b_5 \) are regression coefficients.

To calculate the coefficients of the second-order polynomial the multiple regression analysis was used. The analysis of variance was done with MINITAB 17 software and MATLAB software was used to construct the response.

**Influence of mineralization on the transition of Fe^{2+} ions to Fe^{3+} ions**

To study the effect of competing ions presence on the deironing of water of different origin, a series of experiments were performed using NaCl solutions with the salt concentration of 1 mg/dm³, 2 mg/dm³ and 5 mg/dm³. Model solutions of iron ions with concentrations of 1 mg/dm³, 2 mg/dm³, 5 mg/dm³ were prepared.

**Efficiency of iron ions removal**

For the removal of iron ions from artesian water in laboratory conditions the capillary material was used as a filter. The density of capillary materials was 45 sheets. The study was performed at 25 °C. The efficiency (Z) of iron ions removal was calculated by the formula:

\[ Z = \frac{C_0 - C_f}{C_0} \times 100\% \]

where: \( C_0 \) – initial concentration, mg/dm³, \( C_f \) – final concentration, mg/dm³.

**Table 1.** 2²-factorial design for water treatment from iron ions

<table>
<thead>
<tr>
<th>Factor</th>
<th>Levels</th>
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<tbody>
<tr>
<td></td>
<td>(-1)</td>
</tr>
<tr>
<td>Filter density, sheets</td>
<td>5</td>
</tr>
<tr>
<td>Iron ions initial concentration, mg/dm³</td>
<td>1</td>
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</tbody>
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All experiments were performed three times and the average value is given.

RESULTS AND DISCUSSION

Influence of capillary filter density on deironing processes

For effective removal of iron ions from water, it is important to know the process parameters at which the maximum value of this indicator will be reached. At the first stage, the influence of the density of capillary materials on the efficiency of water deironing was the primary task for investigation. The results of the research at different densities of the filter material and initial Fe$^{2+}$ concentration in solution are shown in Table 2.

The presented results show that the density of capillary materials in the deironing of water with different initial concentrations of iron ions significantly affects the process. At an initial iron ions concentration of 1 mg/dm$^3$, a residual concentration of Fe$^{3+}$ less than the MPC was achieved. The increase in density of capillary materials from 5 to 45 sheets leads to a decrease in the residual iron ions concentration. For comparison, additional experiments were performed using 50 filter sheets. However, in this case the efficiency of the process was the same: during water deironing with the initial iron ions concentration of 1 mg/dm$^3$ throughout 50 sheets the residual concentration of Fe$^{3+}$ was 0.17 mg/dm$^3$; for the solution with the initial iron ions concentration 5 mg/dm$^3$ the residual concentration of Fe$^{3+}$ 1.67 mg/dm$^3$ was achieved.

Based on the experimental data, a regression equation was obtained that adequately describes the process of water deironing:

$$y = -0.87 + 0.012x_1 + 0.979x_2 - 0.011x_1x_2 - 0.0004x_1^2 - 0.015x_2^2$$

Figure 1 shows 3D surfaces for predicting the residual Fe$^{3+}$ concentration based on the obtained regression equations.

The data indicate that the maximum value of iron ions removal can be achieved with the application of 45 sheets of filtering material. It can be concluded that this density of filter material is optimal. All further studies are performed using 45 sheets of filter material.

Influence of mineralization on the transition of Fe$^{2+}$ ions to Fe$^{3+}$

Mineralization of groundwater and surface water has seasonal fluctuations, in spring and autumn an increase in the content of pollutants in the water is observed. Therefore, it is important to determine the effect of various ions on the efficiency of the oxidation of Fe$^{2+}$ to Fe$^{3+}$ and its transition into hydroxide, followed by its precipitation and removal from water. Water deironing is significantly dependent on competing ions. The effect of mineralization on this process is shown in Figure 2.

At an initial iron ions concentration 1 mg/dm$^3$ with an increase in NaCl concentration from 1 to 5 mg/dm$^3$, a decrease in the residual iron concentration to 0.3–0.4 mg/dm$^3$ is observed.

An increase in NaCl concentration from 1 to 5 mg/dm$^3$ at initial iron ions concentration 2 mg/dm$^3$ has a more significant effect on its residual concentration, which decreases to ~ 0.2 mg/dm$^3$. Deironing of the solution with an initial iron concentration of 5 mg/dm$^3$ at a NaCl concentration

<table>
<thead>
<tr>
<th></th>
<th>X1</th>
<th>X2</th>
<th>Y</th>
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<tbody>
<tr>
<td>X1</td>
<td>5</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>X2</td>
<td>1</td>
<td>3</td>
<td>5</td>
</tr>
<tr>
<td>Y</td>
<td>0.87</td>
<td>2.60</td>
<td>4.20</td>
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Figure 1. 3D surfaces for predicting the residual Fe$^{3+}$ concentration
of 5 mg/dm³ managed to reduce the residual iron concentration to ~ 0.9 mg/dm³.

The presence of competing ions significantly affects the transition of Fe²⁺ to Fe³⁺. In the presence of NaCl in solution, iron ions form a number of compounds and associates. From the obtained results it can be concluded that with increasing iron content in the solution an increase in the influence of other ions on the deironing process is observed. On the other hand, the increase in salt content leads to the intensification of the process of conversion of Fe²⁺ to Fe³⁺.

Analyzing the results of the research, we can conclude about the high efficiency of the use of capillary materials in the processes of deironing of waters of various origins. The use of capillary materials can be an alternative to such methods of reducing the iron content as aeration or precipitation. The oxidation efficiency of iron ions on capillary materials is much higher compared to precipitation.

**Efficiency of iron ions removal**

The application of the proposed capillary filtration process, it became possible to reduce the iron ions concentration up to 0.17 mg/dm³ (iron removal efficiency Z = 91.05 %), which meets the requirements for purified water.

**CONCLUSIONS**

1. Capillary filtration today is a promising area of iron removal in water and wastewater treatment.

2. The density of capillary materials in the deironing of waters with different initial concentrations of iron significantly affect the process. The content of competing ions significantly affects the transition of Fe²⁺ to Fe³⁺. With increase in salt content, the process of transition of Fe²⁺ to Fe³⁺ is quite significantly intensified.

3. The developed method of water deironing has economic and energy advantages, which indicates the feasibility of its application for iron ions removal from waters of different origins.

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