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ENVIRONMENTAL MONITORING AND ANALYSIS OF QUALITY OF THE NIZHNY NOVGOROD REGION GROUND WATER WITH THE DEVELOPMENT OF THE TECHNOLOGIES OF THEIR FILTRATION

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Abstract: This article provides the analysis of the quality of underground water sources of the Nizhny Novgorod region, the necessity of improving of the iron removal and demanganation installations for groundwater treatment. The research data to find the most effective method for iron removal and water demanganation using experimental pilot installation in natural conditions are presented.

Key words: purification, iron, manganese, contamination, water treatment

Introduction

The problem of providing population of Russia with high quality drinking water is considered to be one of the most important as it directly affects the public health and cardinally determines the degree of environmental and epidemiological security of entire regions.

The right of citizens of Russia to a favorable habitat, for priority water use, satisfaction of physiological and domestic water requirements is fixed by the legislation of the Russian Federation.

Every second citizen of the Russian Federation has to use for drinking purpose the water not appropriate for hygienic requirements due to a number of indicators, and nearly one-third of the population uses decentralized water supply sources without adequate water treatment, the population of some regions suffer from a lack of drinking water and a lack of good sanitation living conditions connected with it. Russia lags behind developed countries in life expectancy and increased mortality (especially children), to some extent linked to the consumption of contaminated water.

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Historically more than 70% of water for drinking water supply is taken from surface water bodies. It was accessible, cheaper; in general, it is more expedient. However, it should be borne in mind that the present situation has changed. Anthropogenic load has more impact on surface water sources. Due to the deterioration of water quality in surface water sources more attention is paid to groundwater.

In accordance with the target program "The provision of drinking water in Russia" at the federal level, priority measures are implemented to improve drinking water supply in areas with hard water situation; regional programs of water supply in the regions of the Russian Federation are developed.

The strategic direction in water use is to increase use of groundwater for drinking purposes. Part of ground water serving as a source of drinking water supply depends on the availability of water resources and the quality of the latter.

The chemical composition of fresh groundwater is defined by many natural factors; the most important of them is physical and chemical interaction of water with rocks of different composition and structure of the path from the power supply to the places of consumption. In addition, the qualitative composition of the water can be formed by mixing various underground streams, and also depends on the infiltration of precipitation, nature of soil and vegetation, biochemical interaction with the filter medium, the quantity and quality of the water filled in and taken as a sample. Therefore, when seeking and exploring groundwater for water supply hydrochemical conditions and composition of the waters are always studied in detail.

The Nizhny Novgorod region has significant reserves of fresh groundwater. Small and medium-sized towns, residential townships of the Nizhny Novgorod region, as well as individual building objects (houses, farmsteads and farms) located on its territory, use underground water sources for drinking water supply use. Underground water has a higher degree of protection from pollution than surface water but they are still susceptible to anthropogenic pollution.

Natural features of the Nizhny Novgorod region contributed to the formation of the significant diversity of the chemical composition of groundwater. Substandard water formed under the influence of natural geochemical processes is widespread. Many years' practice of using underground water sources in different regions of the country showed that the exaggerated viewing of more favorable chemical composition of the groundwater, and generally accepted

technical solutions of water treatment do not always let to ensure satisfactory potable water supply.

In some parts of the Nizhny Novgorod region the excess of the maximum allowable concentration of elements in the chemical parameters are registered:

- water hardness is increased in 11 regions (Buturlinsky, Vadsky, Vachsky, Dalnekonstantinovsky, Kstovsky, Lukoyanovsky, Lyskovsky, Pavlovsky, Pilninsky, Sergachsky, Sosnovsky);
- there are 4 areas where the iron content is increased (Bogorodskiy, Lukoyanovsky, Sechenovsky, Shatkovsky);
- there are 6 areas with the increased iron and manganese content (Balakhninsky, Borsky, Volodarsky, Vyksunsky, Gorodetsky, Navashinsky);
- there are 2 districts where fluorine content is increased (Tonshaevsky, Shakhunsky);
- there are 3 districts where one can find the increased content of boron in (Vetluzsky, Voskresensky, Pilninsky).

The use of poor-quality drinking water leads to outbreaks of infectious diseases. Consumption of water that does not meet hygiene standards of quality determines the unfavorable sanitary and epidemiological situation of many rural areas of the Nizhny Novgorod region.

However, the high content of individual standardized components should not be an obstacle to the use of groundwater due to with the use of different methods of water treatment water quality can be brought up to the required standards. Providing the population of Nizhny Novgorod region with drinking water of quality standard is one of the priorities to maintain health, improve the business environment and the living standards of the population.

The search for the effective and efficient methods and means of reliable supply of drinking water has become an important part of the modern science studying water. For the Russian Federation it is the governmental problem, the success of solution of which depends on the effectiveness of joint efforts of relevant federal, territorial organizations and business entities. The difference of quality of groundwater of the Nizhny Novgorod region used for drinking water supply necessitates the use of different technological schemes for cleaning of them before serving the consumer, and in particular this applies to iron removal and demanganation technologies.

Iron is a vital element of the human and animals' bodies but its high content is extremely harmful to health. Ingested iron concentrates mainly in the blood.

During long-term intake of iron the oversaturation of liver with ferritin occurs – an excess of it is stored in the liver in the form of colloidal iron oxide (hemosiderin) that destroys the cells of the liver. According to Sanitary norms and rules total iron content in drinking water should not exceed 0.3 mg/L (Sanitary Norms and Rules 2.1.4.1074-01).

Manganese is a biologically essential element for the living body; it is involved in various vital processes such as lipid and carbohydrate metabolism, generation of the connective tissue to catalyze redox reactions, etc. Permanently increased manganese content in the water contributes to its accumulation in the body and leads to manganism, when a serious, irreversible disruption of the functional state of the central nervous system occurs. Permissible manganese content in drinking water is 0.1 mg/L (Sanitary Norms and Rules 2.1.4.1074-01).

Taking into account the information on polytropic adverse effect of iron and manganese consumed in relatively small doses with drinking water it is necessary to conduct iron removal and demanganation of the water and control the content of iron and manganese in drinking water in order not only to meet the aesthetic needs but also to eliminate the possibility of threat to public health.

Predominant form of iron in groundwater is iron bicarbonate (II) which is stable only in the presence of significant amounts of carbon dioxide and in the absence of dissolved oxygen. Along with it iron is found in the form of sulfide, carbonate and iron sulphate (II), complex compounds with humate and fulvic acids. In groundwater manganese is preferably in the form of bicarbonate, manganese (II) well soluble in water.

Groundwater manganese almost always can be found with iron due paragenetical connection between these elements. Therefore the process of iron removal is closely linked to the demanganation process.

Survey of methods of removal of iron and manganese **Singular survey** **Plural surveys**

Nowadays there are a lot of technologies of iron and manganese removal from groundwater. They are mostly based on physical-chemical processes of oxidation of Mn and Fe ions followed by formation and separation of insoluble phases by filtration through a granular layer of loading or ejecting elements during adsorption by chemisorption through controlling such factors as pH, redox potential, temperature, dissolved oxygen and free carbon dioxide, iron-bacteria.

Obviously it's necessary to give attention to the existing problem of demanganation and iron removal of natural waters. To do this the activities on optimization of existing processes and water treatment technologies need to be realized and new directions in addressing these urgent problems also need to be developed.

Nowadays the method of simplified aeration and filtration is the most widely used. The advantages of this method are the ease of implementation and the stability of the quality of purified water.

The choice of iron removal technology depends on the nature of Fe and Mn compounds (the mineral and organic, the degree of oxidation, the percentage of free carbonic acid, hydrogen sulfide, pH level, carbonate hardness, alkalinity, redox reaction, temperature and mineralization. Each method has advantages and limitations. The combinations of methods is the best alternative, its effectiveness must be proved by laboratory inspection and checked by pilot plants.

The method of simplified aeration is used both in gravity and pressure form depending on the performance of the installation. This method is applied when the content of iron in feed water is up to 10 mg/l (including ferrous iron I not less than 70%), hydrogen sulfide 0.5 mg/L, pH – not less than 6.7, oxidation not more than 6–7 mg/L, O₂ and alkalinity is more $(1 + (\text{Fe}^{2+})/28)$. Other indicators of applicability of this method are the conditions under which the oxidation-reduction potential of the water after a simplified aeration will not be less than +100 mV, and the stability index of water $I > +0.05$. Otherwise the water after iron removal is unstable and during its transportation through metal pipes it will be enriched with iron the second time.

The method of simplified aeration is based on characteristics of water containing bivalent iron and dissolved oxygen while being filtrated through the granular layer to liberate iron on the top of the granules forming the catalytic film consisted of ions and oxides of bi- and trivalent iron. This film intensifies actively the process of oxidation and iron liberation from the water. The process of iron removal from the water at loading covered with the layer is heterogeneous autocatalytic process which results in continuous renewing of the film as a catalyst directly during the filtration process.

This method does not require oxidation of ferrous iron to ferric iron and transferring it to hydroxide, and therefore it eliminates the need for expensive apparatus aeration constructions. Simplified aeration is done by using simple tools to spout water from a low altitude in a pocket or a center channel filter, or by blowing air into the water to be treated. In the gravitational version the

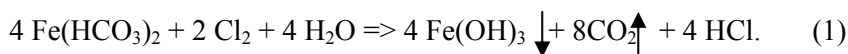
oxygen enrichment of water by spraying it in the air is very intense. Thus when a droplet of water falls from a height of 0.5 m the dissolved oxygen content reaches 5 mg/L. However, it is known that for oxidation of 1 mg of iron (II) 0.143 mg of oxygen is consumed. However, in practice to remove carbon dioxide the greater aeration height is usually given. Under the pressure to enrich water with oxygen, the air is introduced into the conduit (or mixer) before pressure filter in an amount of 1.5–2 liters per 1 g of iron (II). No special aeration equipment or other contact tank needed simplify maintenance and reduce the cost of treatment.

However, the use of this method is difficult at high concentrations of iron and manganese in raw water and also in the presence of ground water humic substances and other organic compounds.

Under chlorine action the destruction of humates and other organic compounds of iron are destructed and change their form to inorganic trivalent iron which are easily hydrolyzed with the deposit which is extracted by filtering:

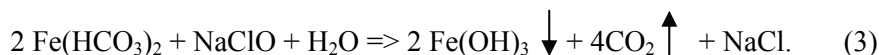
Application of oxidizing reagents followed by filtration through granular feeding. Reagents-oxidants, primarily chlorine to disinfect and remove iron are used in Russia since the beginning of the 20th century.

Under the influence of chlorine humates and other organic compounds of iron are destroyed and transit into the form of inorganic salts of ferric iron which are easily hydrolyzed with precipitate which is extracted by filtration:



According to stoichiometry's data (by equation of reaction) for oxidation of 1 mg of Fe^{2+} 0.64 mg of chlorine is consumed. The alkalinity is reduced to 0.018 meq/L. The dosage of chlorine depending on the iron content can be 5–20 g of water per m^3 by contact of at least 30 min (for the oxidation of not only iron but also for reliable disinfection. Thus hygienic requirements allow free chlorine content in drinking water to 0.5 mg/L and related – to 1.2 mg/L.

As an alternative to chlorination the water treatment with sodium hypochlorite solution is currently used.



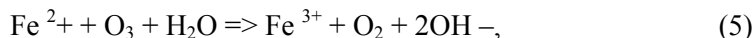
In the process of iron oxidation with sodium hypochlorite no acidification happens. In addition, sodium hypochlorite solution is alkaline which is favorable for filtration.

During water treatment process the oxidation reaction of potassium permanganate followed by iron hydrolysis proceeds according to the equation:



According to stoichiometry (by equation of reaction) for the oxidation of 1 mg Fe^{2+} 0.71 mg of potassium permanganate is consumed. The alkalinity is reduced to 0.036 meq/L. In order to handle the complex water and to save expensive potassium permanganate it may be used in combination with sodium hypochlorite.

Ozone is one of the most powerful oxidants. Compared to the use of chlorine ozone use is more efficient for oxidation of divalent iron and manganese. Ferrous iron Fe^{2+} is oxidized to ferric iron Fe^{3+} which in its turn is hydrolyzed to $\text{Fe}(\text{OH})_3$. Ferric hydrate is deposited in water and can be removed later at clarification or filtration:



To remove the organic compounds of iron the ozone dose is taken as the content of ferrous iron in the source water. Residual ozone concentration in potable water should be 0.1–0.3 mg/L. The contact of ozone with the water depends on the composition of water, ozone mixer design and typically is 5–20 min. It should be noted that ozone is highly toxic and can affect the respiratory system. The equipment is still quite costly, and the process is characterized by a relatively high consumption of electricity.

The biological method is based on the use of microorganisms. The iron bacteria are the most widely spread among them. These bacteria convert ferrous iron (Fe^{2+}) in the oxide (rust Fe^{3+}). Bacteria themselves are not dangerous for human health but their waste products are toxic.

The modern biotechnologies are based on usage of catalytic film generated on sand-gravel filling or other similar finely-porous material, for example the column of activated coconut coal, different synthetic materials, and also the ability of these iron bacteria to provide complex chemical reaction process

without any expenditure of energy and the use of reagents. These processes are natural and are based on biological consistency with natural laws.

Copious evolution of iron bacteria is observed in water with an iron content from 10 to 30 mg/L, however, the experience shows that their development is possible even when the iron concentration is less one hundred times. The only condition is keeping of low medium acidity with oxygen supply from the air at least in negligible quantity.

The final stage of the biological iron removal is sorption purification for the detention of waste products of iron bacteria and the final disinfection of water by germicidal rays. In spite of all its merits (e.g., ecological compatibility) and prospects, bioremediation has only one drawback – relatively low speed of the process. This, particularly, means that to provide larger productivity larger dimensions of capacitive structures are needed.

Ion exchange as a method of water treatment is known for a long time and is mainly used for water softening. Earlier natural resins (sulfonated coal, zeolites) were used. However, with the advent of synthetic ion-exchange resins, the efficiency of ion exchange for water treatment has increased dramatically. From the standpoint of removing iron from water the important is that cation exchangers are capable of removing not only the water calcium and magnesium ions but other divalent metal, and hence the dissolved ferrous iron.

The advantage of ion exchange is the fact that it is "not afraid" of the faithful companion of iron - manganese, greatly complicating the work of systems based on the methods of oxidation. The main advantage of ion exchange is that of the dissolved iron and manganese may be removed from the water.

However, in practice, the application of cation exchange resins for iron is very complicated. This is due to the following reasons: firstly, the use of cation exchange resins is useful where there is also a problem with water hardness, since the iron is removed from the water along with rigidity. Where the rigidity state is relatively good, the use of cation exchange resins is irrational, and secondly, ion exchange resins are very critical to the presence of ferric iron in the water which fills the resin in and is washed away from it with difficulty. That's why the presence of not only oxidized iron but also dissolved oxygen and other oxidizing agents which may lead to its formation is not desirable. These factors require more frequent regeneration which increases the consumption of salt. Thirdly, the presence of organic substances in water (including organic iron) can lead to a rapid "overgrowing" of organic resin by film which simultaneously serves as a breeding ground for bacteria.

For household and commercial – industrial systems the catalytic method of the removal of iron and manganese is widely used. The oxidation reaction occurs on the surface of the granules of special filter media having the properties of the catalyst (accelerator of chemical reaction of oxidation). The most widely spread in modern water treatment are filter media based on manganese dioxide (MnO_2).

Iron and manganese in the presence of manganese dioxide are rapidly oxidized and subsided at the surface of the filter medium granules. Subsequently, most of the oxidized iron is washed into the drainage backwash. Thus, a catalytic granular layer is simultaneously a filter medium. All the catalytic material can be divided into two main groups: 1) catalytic materials in which the restoration of catalytic ability occurs via backwash, 2) catalytic materials in which the catalytic recovery ability is performed using a solution of potassium permanganate $KMnO_4$.

To improve the process of oxidation additional chemical oxidants may be added to the water. The most common is potassium permanganate $KMnO_4$ ("permanganic") since it doesn't only activate the oxidation reaction but also compensates the "washout" of manganese granule from the surface of filter medium i.e. regenerates it. Both batch and continuous regeneration are used. To intensify the process sodium hypochlorite, hydrogen peroxide, ozone may be also added to the water (for the downloads of the first group) (Kuberis, Gorbachov, 2008).

This method has several disadvantages: first, the catalytic loading is ineffective against organic iron. Moreover, any water present in the form of organic iron on the surface of the granules of filter material eventually formed organic film, the insulating catalyst – manganese dioxide from the water. Thus the whole catalyst of the filter filling is reduced to zero, and secondly, systems of this type are ineffective in cases where the iron content in the water is greater than 10–15 mg/L, which is not uncommon. The presence of manganese in the water only aggravates the situation.

Taking into account the above mentioned, it means that the problem of iron removal and demanganation of groundwater is not fully resolved, and requires an integrated approach in each case. Therefore, an important challenge facing professionals is to improve the technology and the intensification of iron removal and demanganation of groundwater using catalytic materials.

The use of catalytic filter material of foreign and domestic production for the purification of drinking water from iron and manganese makes it pay special

attention to the study of properties of the filter and the justification of the use of different types of catalytic materials.

Methodology and results

The study of catalytic properties of materials offers the prospect of finding optimal solutions for intensification of iron removal and demanganation of groundwater, as well as the development of highly hygienic technological schemes adapted to specific local conditions for small local water users.

To determine the most efficient and economical method for the particular water it is necessary to make an experimental iron removal and demanganation in a pilot plant directly at the source.

At developing the technology for the removal of iron and manganese the combined iron removal and demanganation method by the use of simplified aeration, alkalizing, oxidizers and filtration through granular catalyst loading was taken as a basis. Source water quality does not meet the requirements of Sanitary norms and rules 2.1.4.1074-01 “Drinking water. Hygienic requirements to the quality of water of centralized system of water supply. Quality Control.” regarding the content of iron 48.7 MAC (14.6 mg/L), manganese up to 22 MAC (2.2 mg/L), oxidability up to 2 MAC (10.4 mgO₂/L). Turbidity exceeded MAC 5 times (up to 7.4 mg/L) color – 3.3 times (up to 66 deg.) hydrogen sulfide content presented in the water – 4.5 mg/L, ammonia – 3.2 mg/L (2PDK). Active reaction pH – 6.6.

The essence of this combination of methods is to remove the excess of carbon dioxide and hydrogen sulfide at water aeration during which the air is enriched with oxygen. However alkalization increases alkalinity and pH of the water creating favorable conditions for the oxidation of iron and manganese. The final disintegration of complex compounds of iron (II) and manganese (II) and their oxidation is achieved by inputting a strong oxidant into the water to be treated (chlorine, ozone, potassium permanganate, etc.). Compounds of ferrous and ferric iron and manganese are extracted from the water by filtering it through the granular catalyst feeding. (Kuberis, 2007).

The developed technology provides water treatment with sodium hypochlorite solution (an alternative to chlorination). In the process of iron oxidation with sodium hypochlorite there's no acidification. In addition, sodium hypochlorite solution is alkaline which is favorable for filtration.

In the developed technology water treatment with sodium hypochlorite solution (an alternative to chlorination) is provided. In the process of iron oxidation with sodium hypochlorite no acidification happens. In addition, sodium hypochlorite is alkaline which is favorable for filtering.

The pilot plant included: strainer – tank aerator – low-pressure compressor – a complex dosing of caustic soda – the second lift pumps – a complex dosing sodium hypochlorite – pressure contact chamber – iron removing pressure filter – filter of fine cleaning – tank of clean water – pumps of a third lift, and also stop and control valves and instrumentation. Pilot Plant Performance is 0.5 m³/h. The scheme of the pilot plant is shown at Figure 1.

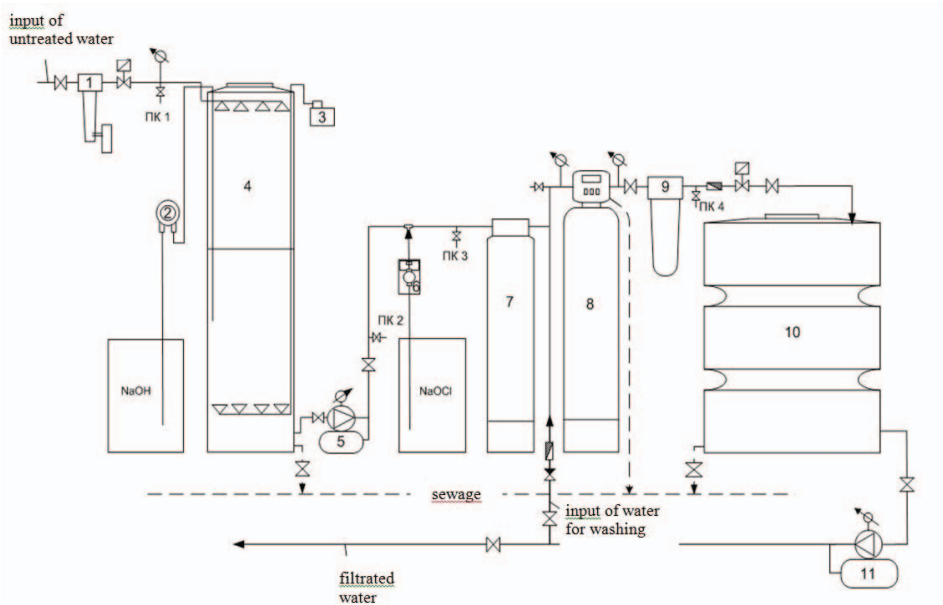


Figure 1. Scheme of the pilot installation: 1. screen filter of the primary purification; 2. pump-dosator of alkaline; 3. aeration compressor; 4. tank-aerator; 5. pump installation of the second lift; 6. pump dosator of sodium chloride; 7. catalyst case; 8. deironing filter; 9. fine filter; 10. pure water tank; 11. pump installation of the third lift.

Tank aerator of 550 mm diameter, 1920 mm height is equipped with a spray machine – crevice tool. It is divided into two parts: the aeration zone (height – 900 mm) and settling portion (height – 1000 mm). As part of the settling tank aerator provides air flow from the low pressure compressor in an amount of 45 L/min ($Q_{air}/Q_{water} = 5$), and entering an alkalizing agent (sodium hydroxide) using a peristaltic dosing pump. Slop tank-aerator is designed for contact time –

20–25 min. Tank is equipped with an aerator valve for emptying and disposal of accumulated sediment. Tank-aerator is equipped with float level control, managing the work of submersible pumping (second lift) pumps, compressor and metering pump.

The pressure contact chamber which diameter is 203 mm and height is 1115 mm is made of food grade plastics. Contact time of the camera is designed for contact with the chlorine treated water – 4–5 minutes. At the bottom of the pipe drain valve is provided for emptying the chamber.

Reagent (caustic-sodium hydroxide solution (NaOH) and chlorine (NaOCl)) includes in its composition: solution-filling tanks (2 pcs.) with volume of 100 liters, peristaltic dosing pump is operating in automatic mode in accordance with the alarm float switch, solenoid diaphragm metering pump operating in automatic mode, the signal pulse water meter.

The concentration of the working solution of sodium hypochlorite was 8%. The concentration of the working solution of caustic soda was 10%. The initial dose for sodium hypochlorite (GOST 11086-76 brand A) was 8 mg/L (as active Cl); sodium hydroxide (GOST 2263-79) – 100 mg/L. Optimal dosages of reagents were determined in experiments in a pilot plant. According to studies the graphs presented in Figures 2–3 have been made.

Deironing pressure filter which is 370 mm in diameter and 1650 mm height is formed of food grade plastics. A mixture of catalytic filter materials – "Sorbent AS + MS" in the ratio of 50/50% was used as the filter media.

Filtration rate fixed – 5 m/h. Material of iron removing filter and filtration rate have been selected based on the results of previous laboratory studies. Breakup: Sorbent AC – 0.7–1.5 mm; Sorbent MC – 0.5–1.0 mm. Coefficient of filling heterogeneity: Sorbent AC – 1.6–1.8 mm; Sorbent MC – 1.6–1.8 mm. The height of the filtering layer – 1000 mm (500 mm + 500). Drainage layer of gravel – 200 mm. The filter comprises 90 L of a filter material and 14 kg of gravel (2–4 mm).

Experimental studies have shown that if sodium hypochlorite dose is 100 mg/l positive results were not achieved. The content of iron and manganese in the filtrate practically in all the experiments exceeds the MPC. Residual chlorine is not determined. With increasing of dose of sodium hypochlorite till 125 mg/L in the last series of experiments (sodium hydroxide dose of 160 mg/L) showed reduction of iron and manganese to the maximum allowable concentration. At a

dose of sodium hydroxide 140 mg/L manganese content in the filtrate exceeds the maximum allowable concentration.

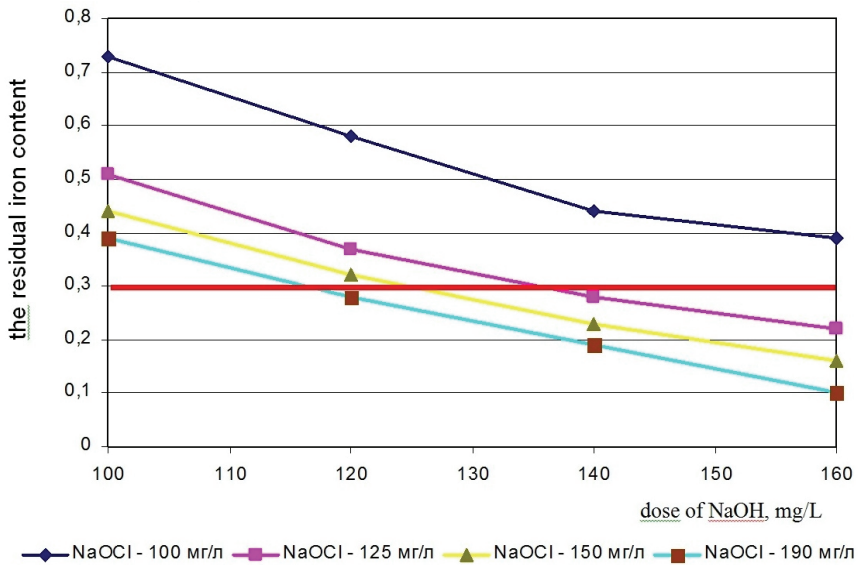


Figure 2. Changing of the residual iron content depending on the dose of sodium hydroxide and sodium hypochlorite

When sodium hydroxide dose is 100 and 120 mg/L, iron and manganese content exceeds the maximum allowable concentration. Residual chlorine (in all series of experiments) was 0.4–0.45 mg/L.

With increasing doses of sodium hypochlorite to 150 mg/L in the last two series of experiments (the dose of sodium hydroxide being 140 and 160 mg/L) showed reduction of iron and manganese to the MACs. At a dose of 100 sodium hydroxide and 120 mg /L of iron and manganese content in the filtrate was not significantly higher than the MAC. Residual chlorine (in all series of experiments) was 0.7–0.75 mg/L.

Increasing the dose of sodium hypochlorite to 190 mg/L in the last two series of experiments (sodium hydroxide dose – 140 mg and 160 mg/L) showed a significant reduction of iron and manganese to the maximum allowable concentration. At a dose of 100 mg of sodium hydroxide and 120 mg/L the iron and manganese content in the filtrate doesn't significantly exceed the maximum allowable concentration. Residual chlorine (in all series of experiments) was 1.13–1.15 mg/L.

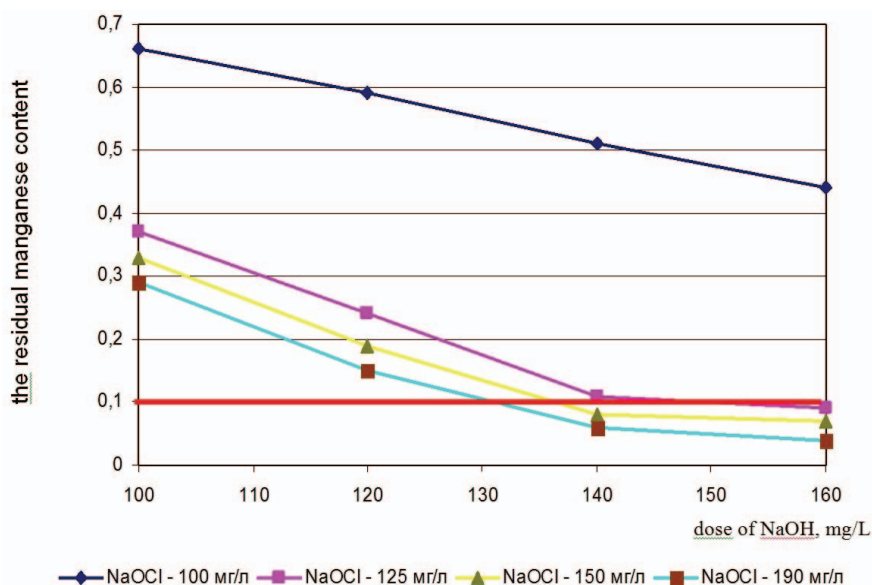


Figure 3. Changing the content of manganese in the residual dose of sodium hydroxide and sodium hypochlorite

In all series of experiments when a sodium hydroxide dose was 160 mg/L of water pH was 9.0–9.3 which exceeds the requirements of the MPC.

Conclusion

As a result of experiments made at a pilot plant it was found that the combined use of sodium hydroxide (NaOH) and sodium hypochlorite (NaOCl) at doses of 140 mg/L and 150 mg/L provides the desired degree of purification of ground water of iron (0.23 mg/L), manganese (0.08 mg/L). The pH and residual chlorine were 8.7 and 0.75 mg/L.

According to the experimental researches the technology of removal of high concentrations of iron and manganese in small-sized apparatus with using a catalytic filter material that provides effective cleaning underground water has been developed (Kuberis, 2010).

Basing on the experimental research findings the technology of high content of iron and manganese removal small-sized plant with using a catalytic filter material which effective cleaning underground water is developed (Kuberis, 2010).

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Five-year experience of operating the proposed technology of iron removal and demanganation show its efficiency and reliability in working in recommended mode while ensuring the required quality of water.

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