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Purification of water contaminated with petroleum and oil products

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Abstract
The process of purification of water contaminated with oil and oil products (kerosene) by the sorption method is considered. As a sorbent, a natural mineral bentonite and a mixture of bentonite and agricultural waste of hazelnut shells were used in a mass ratio of 1:1. Bentonite was activated with 5 and 10% sulfuric acid, thermally treated at 300, 350, 400, and 450^0 degrees (C), and a mixture of bentonite with hazelnut shells were used as a sorbent. The cleaning effect of both sorbents was analyzed separately, comparisons were made, and the effects of acid concentration and temperature on the sorption capacity of the sorbent were studied. It was determined that, 99.5% of oil and 98.8% of kerosene can be removed from water contaminated with petroleum and oil products using an adsorbent from a mixture of bentonite and hazelnut shells, activated with 10% sulfuric acid and thermally treated at 350^0 C. The process is economically and environmentally preferable.

Keywords: oil products, pollution, bentonite, hazelnut shells, thermal treatment, sorption capacity

Introduction

In recent years, water treatment has become one of the most relevant common technological processes. The existing technologies for water purification from organic and inorganic ions are often based on electric discharge methods, membrane technologies, sedimentation, oxidation, coagulation processes, and as a rule, these processes and technologies do not always allow extracting ions top the MAC (Maximum Allowable Concentration) level.

Currently, sorption methods are the most common among various methods of drinking water purification [1, 3]. In terms of reducing the cost of sorption processes used for water treatment, using natural mineral sorbents such as magnetite, chalk, anthracite chips, zeolite, quartz sand, diatomite, and dolomite seems very promising. These natural sorbents are increasingly used due to their low cost and high sorption capacity; therefore, their use in the process of water treatment makes it possible to exclude the stage of adsorbent regeneration [4]. A review of publications on sorption processes has shown that the development and application of new materials with enhanced sorption properties is an urgent task and requires a more detailed study of the sorption mechanism [5, 6].

Recently, the interest in the creation of new environmentally friendly sorbents based on natural clay materials and aluminosilicates has increased significantly. Compared to other inexpensive adsorbents, clays and composite materials based on them have a higher adsorption capacity. Bentonites, montmorillonites, kaolinites, illites, chlorites, and other clay minerals are widely used because of their high specific surface area, chemical and mechanical stability, surface and structural variation, and low cost. Of particular interest are materials with a layered columnar structure obtained from natural clay by modifying it with inorganic polyhydroxy cations [7].

The authors studied the sorption capacity of sorbents based on bentonite modified by the "sol-gel" method in relation to permanganate anions. It has been found that in this case the adsorption isotherms have the form of the Langmuir isotherms. It was determined that in this case the adsorption isotherms have the form of Langmuir isotherms. It is shown that Al-modified sorbents have a higher sorption capacity with respect to permanganate ions, which reaches 14.5 mg/g (for unannealed) and 10.6 mg/g (for annealed). It is much higher than the analogous values for the original bentonite (4.4 mg /g) and Fe-modified sorbents (8.7 mg/g). Moreover, it was determined that sorbents modified on the basis of natural bentonite are finely porous (nanostructured) objects dominated by pores with a size of 1.5-6.0 nm. The value of the specific surface of sorbents depends on the method of modification and the amount of the input modifying component - iron (III) and aluminum ions. The specific surface area of Al-modified sorbents according to the “sol-gel” method reaches 100 m^2/g, which significantly exceeds the value of the original bentonite specific surface [8, 9].

Bentonite clays have been used since prehistoric times to purify wine, oils, and water. They are non-toxic and have an extremely developed surface, which leads to a significant adsorption capacity, i.e., the ability to absorb various substances from solutions. A limiting factor in the widespread use of natural bentonite sorbents for the purification of drinking water and industrial wastewater is the lack of effective granulation technologies, since clay minerals are subject to the effect of peptization in aqueous solution, i.e., dispersion [10].

However, mineral sorbents often do not have the required sorption properties and must be subjected to heat treatment or chemical modification [11, 12]. As a result of modification, the number of active sites on the surface and the maximum sorption capacity, unlike the original mineral increase several times. The paper investigates the modification of Tripoli by creating a hydrophobic layer on the surface of its particles to increase the oil absorption capacity of Tripoli. It [13, 14] shows the modification of the sorbent by introducing ionogenic groups to reduce the hardness of water. The influence of ultrasonic and thermal treatment of Tripoli on the change in its sorption properties was studied; however, these works do not provide an explanation of the mechanism of the sorption process [15–18].

A technology has been developed for obtaining a granular sorbent for water purification from natural bentonite by extrusion followed by firing. It has been established that the thermal treatment of bentonite clays has practically no effect on the mineralogical composition and structure of the sorbents obtained from it, but has a positive effect on the mechanical strength of the granules. It has also been established that the optimal firing temperature of bentonite clay is 550–650 °C, since at lower firing temperatures, sorbent samples do not have sufficient strength, and exposure to higher temperatures leads to a decrease in specific surface area, which negatively affects the sorption properties of granules.

The performed tests for mechanical and chemical resistance showed that the obtained adsorption material complies with the requirements of GOST R 51641-2000: «Filtering granular materials. General specifications». As a result of studies of the sorption properties of granules, their ability to extract suspended particles from water, as well as to purify them from humic substances, has been proven.

Bentonite granules effectively sorb heavy metal ions from water. The sorption capacity for iron is higher than for lead or arsenic ions.

Thus, the developed sorbent based on bentonite can be recommended for use as an independent filter material for iron removal and purification of highly turbid and colored waters, and for adsorption-membrane water treatment systems [19].

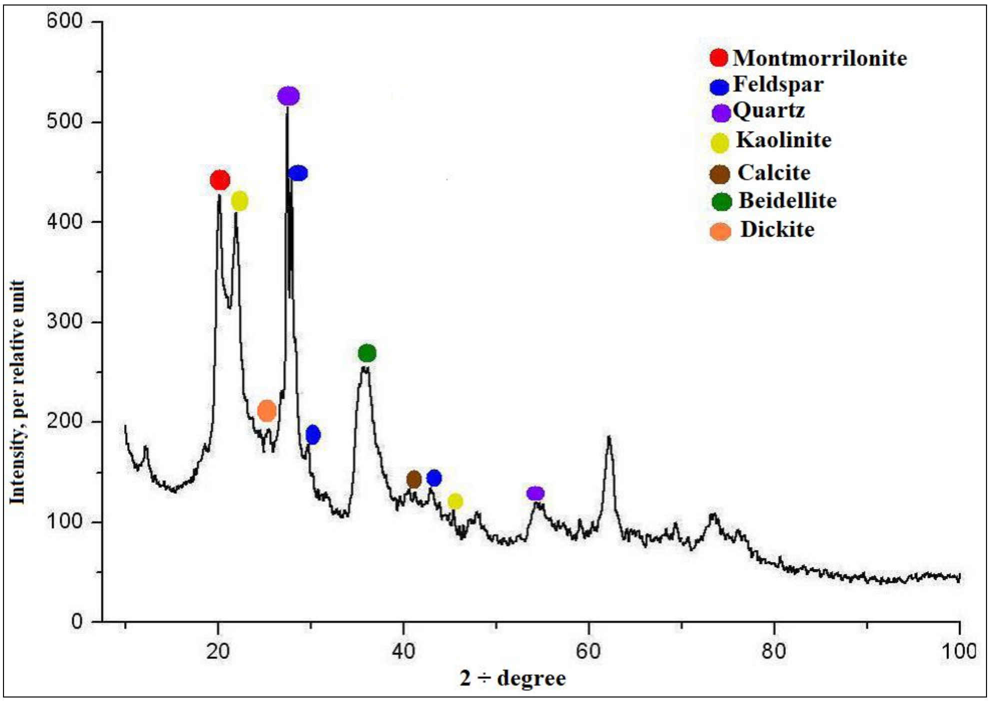

Fig 1: X-ray diffraction pattern of a bentonite sample (t = 600 °C for 1 hour)

Experimental part

The problem of extracting oil products from wastewater has become of great importance, since they suppress the cleaning power of activated sludge in biological water treatment.

The damage caused to the national economy by water pollution with oil products include the deterioration of the organoleptic properties of drinking water, the increase in the cost of water treatment and the replacement of equipment due to increased corrosion, the death of microflora and vegetation in places where oil-bearing effluents are released. Unfortunately, little attention is paid to the issues of efficient and waste-free technology for the treatment of wastewater from industries containing oil and oil products.

To purify water contaminated with oil products, a natural sorbent (bentonite) from the Dash-Salakhly deposit was used. Initially, dispersed particles of bentonite with a size of 200-315 µm were used in the studies. Firstly, we studied the adsorption capacity of bentonite on model of oil and oil products (kerosene) solutions with a concentration of 5 mg/l. Experiments were continued using adsorbents from bentonite and a mixture of bentonite and agricultural waste of hazelnut shells (hazelnut shells at a ratio of 1: 1) activated with sulfuric acid in various concentrations (5 and 10%) and thermo-treated at temperatures of 300, 350, 400, 450 °C. The results of the process of cleaning the prepared model solutions from oil and oil products using bentonite activated with 5% sulfuric acid and calcined at different temperatures are shown in Table 1, and with bentonite activated with 10% sulfuric acid in Table 2.

At the same time, the sorption capacity of chemically and thermally activated sorbents based on bentonite and hazelnut shells was studied.

The sorption capacity is calculated by the ratio of the mass of oil absorbed by the sorbent to the mass of the sorbent itself. The sorption capacity was calculated by the formula:

$$A = \frac{m_n}{m_s}$$

where A is oil intensity, g/g; m_n is the mass of oil sorbed by the sorbent, g; m_s is the mass of the sorbent, g.

The amount of oil and kerosene in water was determined on a Shimadzu GC-2010 plus chromatograph (automatic dispenser). Pre-activated 10% H_2SO_4 and calcined dependency: when bentonite is thermally treated to a temperature of 350^0C, its adsorption capacity increases slightly, however above 400^0C it decreases.

Analysis of results

The use of bentonite according to the experiments of Tables 1 and 2 is less effective than the sorption of oil products on mixed sorbents based on bentonite clay and hazelnut shells (Tables 3 and 4). High efficiency in the purification of oil-containing model solutions is observed in adsorbents with a mixture of bentonite + hazelnut shells (1:1) activated with 10% sulfuric acid and thermally treated at a temperature of 350°C. The degree of purification is 99.5% for oil and 98.8% for kerosene.

Table 1: Treatment of oily wastewater using bentonite activated with 5% sulfuric acid

A Adsorbent	Oil concentration, mg/l	Purification degree, %	Kerosene concentration, mg/l	Purification degree, %
Natural bentonite	1,215	75,7	1,28	74,4
Bentonite (5% H_2SO_4 and 300^0C)	1,05	79,0	1,1	78,0
Bentonite (5% H_2SO_4 and 350^0C)	0,83	83,4	0,85	83,0
Bentonite (5% H_2SO_4 and 400^0C)	0,45	91,0	0,5	90,0
Bentonite (5% H_2SO_4 and 450^0C)	0,65	87,0	0,7	85

Table 2: Treatment of oily wastewater using bentonite activated with 10% sulfuric acid

A Adsorbent	Oil concentration, mg/l	Purification degree, %	Kerosene concentration, mg/l	Purification degree, %
Bentonite (10% H_2SO_4 and 300^0C)	0,68	86,4	0,74	85,2
Bentonite (10% $H_2SO_4$and 350^0C)	0,475	90,5	0,52	89,8
Bentonite (10% $H_2SO_4$and 400^0C)	0,26	94,8	0,285	94,3
Bentonite (10% $H_2SO_4$and 450^0C)	0,415	91,7	0,475	85,2

Table 3: Treatment of oily wastewater using a mixture of bentonite + hazelnut shells (1:1) activated with 5% sulfuric acid

A Adsorbent	Oil concentration, mg/l	Purification degree, %	Kerosene concentration, mg/l	Purification degree, %
Natural bentonite + hazelnut shells	0,575	88,5	0,64	87,2
bentonite + hazelnut shells (5% H_2SO_4 and 300^0C)	0,265	94,7	,51	89,8
bentonite + hazelnut shells (5% H_2SO_4 and 350^0C)	0,225	95,5	0,24	95,2
bentonite + hazelnut shells (5% H_2SO_4 and 400^0C)	0,315	3,7	0,375	92,5
bentonite + hazelnut shells (5% H_2SO_4 and 450^0C)	0,41	91,8	0,44	91,2

Table 4: Treatment of oily wastewater using a mixture of bentonite + hazelnut shells (1: 1) activated with 10% sulfuric acid

Adsorbent	Oil concentration, mg/l	Purification degree, %	Kerosene concentration, mg/l	Purification degree, %
bentonite + hazelnut shells (10% H_2SO_4 and 300^0C)	0,175	96,5	0,26	94,8
bentonite + hazelnut shells (10% H_2SO_4 and 350^0C)	0,035	99,5	0,06	98,8
bentonite + hazelnut shells (10% H_2SO_4 and 400^0C)	0,135	97,3	0,16	96,8
bentonite + hazelnut shells (10% H_2SO_4 and 450^0C)	0,41	91,8	0,44	91,2

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