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ANALYSIS OF THE EFFICIENCY OF THE APPLICATION OF NATURAL COAGULANTS

The object of the study is the process of purifying natural waters using coagulants of natural and industrial origin. The subject of the study is the analysis of the effectiveness of coagulants of natural and industrial origin.

Today, there is great interest in finding alternative methods of water purification that would be more economical and environmentally friendly. The availability of clean and safe water is especially important during times of martial law. Coagulants of natural origin can be an affordable and effective means of water purification and disinfection. One of the important advantages is availability, which indicates the possibility of becoming an alternative to chemical coagulants. Thus, there is a need to study the possibility of using natural coagulants both on an industrial scale and in emergency conditions.

This study presents a characterization of existing coagulants of natural origin, an analysis of the effectiveness of their use, comparison with chemical analogues, as well as an analysis of the effectiveness of using Moringa oleifera as a natural coagulant.

During the work, two coagulants were synthesized from red mud from aluminium production. A comparative analysis of coagulants synthesized from red mud, Moringa oleifera, aquatone and aluminium sulphate was carried out. All reagents were used to remove turbidity and other impurities. The studies were carried out 3 times in different seasons of the year. All results were compared to determine the most effective coagulant and its dose.

As a result, it was found that all of these coagulants can be used and reduce water treatment costs at local water treatment plants. The natural coagulant can be used in areas where there is no access to a central water supply, especially in war zones.

Keywords: *coagulation, natural coagulant, Moringa oleifera, water treatment, turbidity, synthesized coagulants.*

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1. Introduction

Nowadays, the issue of people's access to safe and clean water is acute. Drinking water is a vital resource for all people, so the issue of its quality is one of the main problems in the world.

The usual sources of drinking water are groundwater, surface water and rainwater. Most often, groundwater is the main source of drinking water, if it does not contain a large amount of mineral substances. Groundwater is usually extracted through wells or boreholes. Surface water needs to be cleaned for safe human consumption. Surface waters are almost always polluted by human and animal waste products. Rainwater can be collected in large tanks or smaller containers. However, rainwater collected in dirty or unclean containers also needs treatment to make it safe for consumption, as discussed in the paper [1].

Coagulation is among the most applied processes for water and wastewater treatment. Water industries globally consider coagulation/flocculation is one of the major treatment units used to improve overall treatment efficiency and cost effectiveness for water and wastewater treatment. And then fundamental and applied studies have never been ceased although the modern coagulation has been applied for water treatment since the early 1900s.

Aluminum salts are the most common used synthetic coagulants in water and wastewater treatment all over the world. However, recent studies by several workers have raised doubts about the advisability of introducing aluminum into the environment. Ferric salts and synthetic polymers have been used as alternatives but with limited success due to the fact that their impact on living beings is not fully investigated. Besides, many developing countries can hardly afford the costs of imported chemicals for water and wastewater treatment.

Natural coagulants of vegetable and mineral origin were in use in water and wastewater treatment before the advent of synthetic chemicals like aluminum and ferric salts. Previous studies however, have not determined whether such natural coagulants are economically and environmentally more acceptable than chemical coagulants. Since recently there has been more interest in the subject of natural coagulants, especially to alleviate problems of water and wastewater treatment in developing countries and to reuse some by-products.

The possibility of using *Moringa oleifera* powder as a coagulant for water purification was considered in [2–5].

In these works, authors indicate that Moringa effectively purifies surface waters of turbidity and cyanobacteria. They also note that one potential disadvantage of using certain substances is that they may contribute to increased levels of organic matter in treated water. Disinfecting this water with chlorine can result in the formation of trihalomethanes, which are known to be carcinogenic. Using *Moringa* for water purification may be of concern due to the organic load it may add to the water. The presence of albumin and globulin protein fractions in moringa increases its coagulant potential, making it a more effective water purification option. This process minimizes any negative impact on the water, making *Moringa oleifera* a promising solution for water purification.

The study of the qualitative and quantitative aspects of the metabolite profile of *Moringa oleifera* leaves and seeds, as well as the assessment of the antimicrobial activity of their polar and apolar extracts was presented in the research [6–8]. In addition, the authors determined that apolar extracts from seeds showed a significant concentration-dependent antimicrobial activity against *Staphylococcus aureus* and *Staphylococcus epidermidis*, (4 mg/mL reduced the viability up to 50 %) that was associated to the content of specific fatty acids. In their results they noted that the advantages of an integrated approach for the identification of plant metabolites and its use in association with biological tests to recognize the compounds responsible for bioactivity without compounds purification.

In a review of plant coagulant sources [9], processes, efficiencies, and relevant coagulation mechanisms for water and wastewater treatment were considered. These coagulants are, in general, used as point-of-use technology in less-developed communities since they are relatively cost-effective compared to chemical coagulants, can be easily processed in usable form and biodegradable. These natural coagulants, when used for treatment of waters with low-to-medium turbidity range (50–500 NTU), are comparable to their chemical counterparts in terms of treatment efficiency. Their application for industrial wastewater treatment is still at their infancy, though they are technically promising as coagulant for dyeing effluent as afforded by Yoshida intermolecular interactions. These natural coagulants function by means of adsorption mechanism followed by charge neutralization or polymeric bridging effect. Frequently studied plant-based coagulants include nirmali seeds (*Strychnos potatorum*), *Moringa oleifera*, tannin and cactus. Utilization of these coagulants represents important progress in sustainable environmental technology as they are renewable resources and their application is directly related to the improvement of quality of life for underdeveloped communities.

The most common process identified using Moringa oleifera (MO) seeds was coagulation/flocculation, as noted in the paper [10]. The coagulation efficiency in high turbidity waters was similar to chemical coagulants; however, for low turbidity waters it is remarkably reduced. The best parameters are not yet well established. The identification and characterization of the whole range of proteins of MO seed and their amino acid sequences and structure is a field that still requires extensive research. There is still not general agreement in the literature about the active compound of MO seeds responsible for the coagulation-flocculation.

In the study [11] the impact was evaluated of *Moringa oleifera* (MO) seeds and MO seeds with a reduced amount of oil as coagulants in conventional water treatment to remove cyanobacteria from different natural surface waters. Tests were performed for waters with low (5–10 NTU) and high (30–60 NTU) initial turbidity with and without *Microcystis aeruginosa* cells and for MO integral powder, MO oil-extracted with ethanol and with a pressurized technique, and MO extracted with NaCl. The results show that MO oil-extraction is not necessary when using MO seeds as a coagulant in coagulation/flocculation/sedimentation (CF/S). Chlorophyll a and turbidity removals were up to 85 % for high-turbidity waters, with 50 mg·L^{-1} MO obtained, for MO integral powder and for all of the MO extraction methods used. Therefore, for these waters, integral powder (without any extraction) can be used. For low-turbidity waters and using a 1 M saline extraction of the active coagulant compound, chlorophyll a and turbidity removals were approximately 60 %. It was also demonstrated that MO is capable of removing some of the organic matter present in water, namely, aromatic organic matter, between 40 and 50 % when using MO extracted with NaCl. However, dissolved organic matter results showed an increase independent of the type of MO extraction used.

A study [12] was conducted on the effectiveness of *Moringa oleifera* compared with that of aluminium sulphate ($Al_2(SO_4)_3$) and ferric sulphate ($Fe_2(SO_4)_3$), termed alum and ferric respectively. A series of jar tests was undertaken using model water, different raw water sources and hybrid water containing a mixture of both of these types of water. The model water consisted of deionized water spiked with *Escherichia coli* (*E. coli*) at 104 per 100 ml and turbidity (146 NTU) artificially created by kaolin. Results showed that *Moringa oleifera* removed 84 % turbidity and 88 % *E. coli*, whereas alum removed greater than 99 % turbidity and *E. coli*. Low turbidity river water (<5 NTU), with an *E. coli* count of 605 colony forming units (cfu)/100 ml was treated with *M. oleifera* and ferric. Results showed an 82 % and 94 % reduction in *E. coli* for *Moringa oleifera* and ferric respectively. Tests on turbid river water of 45 NTU, with an *E. coli* count of 2650 cfu/100 ml, showed a removal of turbidity of 76 % and *E. coli* reduction of 93 % with *Moringa oleifera*. The equivalent reductions for alum were 91 % and 98 % respectively. Highly colored reservoir water was also spiked with *E. coli* (104 cfu/100 ml) and turbidity (160 NTU) artificially created by kaolin; termed hybrid water. Under these conditions *M. oleifera* removed 83 % color, 97 % turbidity and reduced *E. coli* by 66 %. Corresponding removal values for alum were 88 % color, 99 % turbidity and 89 % *E. coli*, and for ferric were 93 % color, 98 % turbidity and 86 % *E. coli*. Tests on model water, using a secondary treatment stage sand filter showed maximum turbidity removal of 97 % and maximum *E. coli* reduction of 98 % using *Moringa oleifera*, compared with 100 % turbidity and 97 % *E. coli* for alum. Although not as effective as alum or ferric, *Moringa oleifera* showed sufficient removal capability to encourage its use for treatment of turbid waters in developing countries.

The ability of coagulation active proteins from common bean (*Phaseolus vulgaris*) seed for the removal of water turbidity was studied was studied in [13]. Partial purification of protein coagulant was performed by precipitation with ammonium sulphate, dialysis and anion exchange chromatography. Adsorption parameters for ion-exchange process were established using dialysate extract. Results revealed that the highest values of the adsorbed protein were achieved in 50 mmol/L phosphate buffers at pH 7.5 and the maximum adsorption capacity was calculated to be 0.51 mg protein/mL matrix. Partially purified coagulant at initial turbidity 35 NTU expressed the highest value of coagulation activity, 72.3 %, which was almost 22 times higher than those obtained by crude extract considering applied dosages. At the same time, the increase in organic matter that remained in water after coagulation with purified protein coagulant was more than 16 times lower than those with crude extract, relatively to its content in blank.

The availability of coagulants from plants is of great importance, indicating that plant-based coagulants are a possible modern alternative to chemical coagulants.

Technologies using natural, locally available coagulants are environmentally friendly, reduce external dependence on mechanisms and equipment, especially in wartime conditions [14, 15].

Considering the above, there is a need for a more detailed study of the use of natural coagulants. Therefore, *the aim of the research* is to establish the expediency of using natural coagulants in comparison with widely used chemical coagulants.

2. Materials and Methods

In this study, water parameters such as turbidity and pH.

The research was conducted with samples of tap water from the environmental laboratory of Admiral Makarov National University of Shipbuilding and water from the river Ingul (Mykolaiv region, Ukraine).

Extraction of *Moringa oleifera* seeds was used as natural coagulant for water turbidity and pH [16].

A new synthesized coagulant from red mud was prepared. The first sample of this reagent was based on HCl, the second – on H_2SO_4.

The comparative analysis was carried out with natural, synthesized and wild spread commercial chemicals such as aluminum sulphate and aquatone. All coagulants were used in different doses (5, 10, 15, 20, 25, 30 and 50 mg/dm^3) to determine the optimal one.

Turbidity of water was determined according to the procedure specified in GOST 3351-74. Determination of turbidity was performed before and after filtering at different concentrations of coagulants. Simulated samples were based on water with adding kaolin at a concentration of 20 mg/dm^3.

All measurements were carried out on the BECKMAN COULTER DU 520 (USA).

Determination of these parameters was conducted before and after filtration. As filters were used a filter paper «blue tape» and filter siliceous filter.

The pH value was measured using pH-150MI (Ukraine). The measurement range of this device is between 0.00–12.0, and temperature is (–10–100 °C). In accuracy of measurement for pH value is ±0.05, and for temperature is ±2 °C.

3. Results and Discussion

Water from the river Ingul was selected to research coagulative efficiency of natural and synthesized coagulants. Tests were carried out in May and August 2021. All the results are gathered in the Fig. 1.

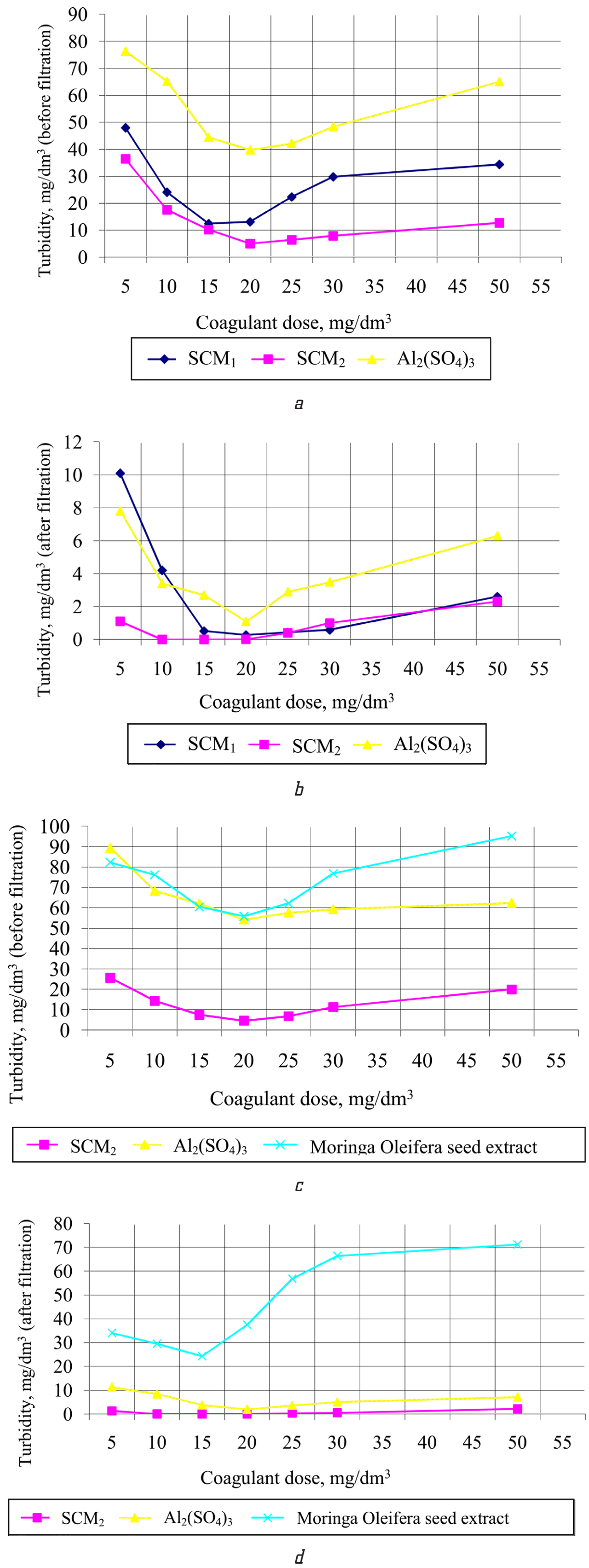

Fig. 1. Water turbidity removal before and after filtration in: *a*, *b* – May 2021; *c*, *d* – August 2021

Fig. 1 shows, that SCM*n* coagulants are more effective in water turbidity removal than aluminum sulphate and natural coagulant. But there's the tendency of turbidity increasing from 25 mg/dm^3 coagulant concentration. Seeds extract isn't effective in turbidity removal at such concentrations. That's why its properties were tested at lower concentration.

According to these results natural coagulant properties were analyzed at concentration 0.1; 0.2; 0.3; 0.5 and 1 mg/dm^3 (Fig. 2).

Consequently, natural coagulant is more effective at concentration till 1 mg/dm^3 as before and after filtration. At 1.0 mg/dm^3 before filtration water parameters of turbidity corresponds to drinking water parameters (lower 20 mg/dm^3). After filtration results are much better, it means that natural coagulant is effective with turbidity removal only at concentrations less than 1 mg/dm^3.

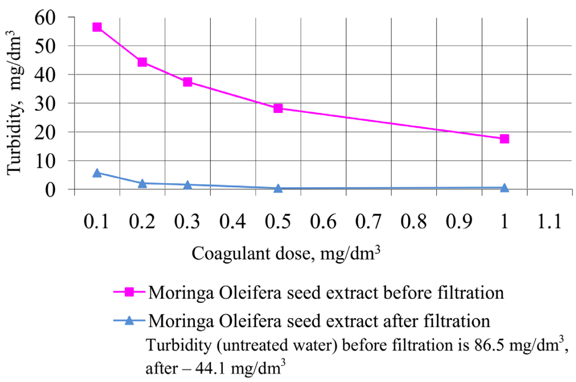

Fig. 2. Water turbidity removal by seeds extract at low concentrations

During the research several tests proved that natural coagulant doesn't have a significant effect on pH. SCM_1 decreases pH below the lowest established control parameter 6.5 at 30 mg/dm^3 and higher concentrations. SCM_2 possibly use the concentration from 5 to 25 mg/dm^3. $Al_2(SO_4)_3$ better to use to correct pH value in water solution.

If compare 2 types SCM_1 and SCM_2 effect on pH, SCM_2 at 5 mg/dm^3 concentration decrease pH value higher than SCM_1 due to the composition of this coagulant (Fig. 3).

pH studies showed that seeds extract doesn't have a significant effect on it, synthesized coagulants are better to use at concentration from 5 to 20 mg/dm^3, $Al_2(SO_4)_3$ decreased pH lower 6.5 at 25 mg/dm^3 in water solution.

Considering that we studied different concentrations of coagulants with the aim of reducing the dose and determining the most effective one, including from an economic point of view. The research was stopped in 2022 and 2023 due to the fact that Mykolaiv was in the war zone and the laboratory complex of the Scientific Research Institute of Ecology and Energy Conservation at the Admiral Makarov National University of Shipbuilding was destroyed. Since the beginning of 2024, part of the laboratories has been restored and research continues in the direction of finding the dose necessary for the disinfection of drinking water in field conditions.

In the future, it is planned to create a tablet form of a plant coagulant for water disinfection in the combat zone. It is also planned to carry out a comparative analysis of this drug with existing ones.

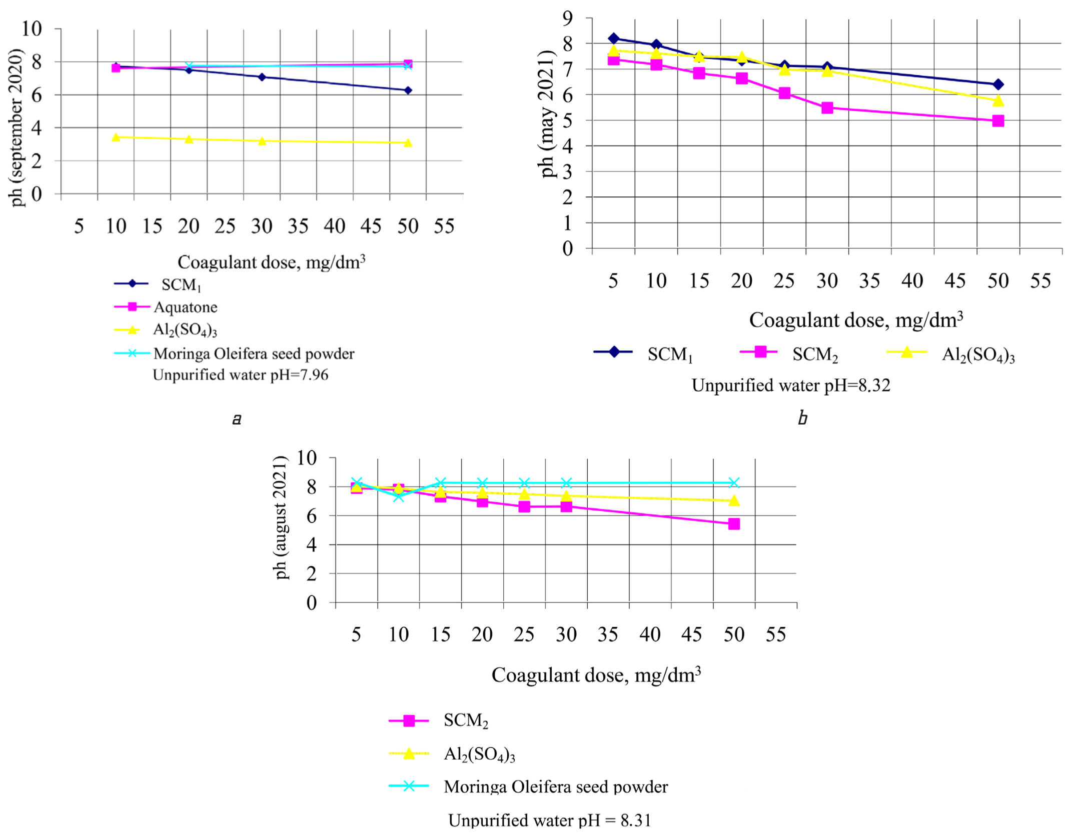

Fig. 3. pH values dependence on coagulant dose:
a – September 2020 (simulated solutions); *b* – May 2021 (river water); *c* – August 2021 (river water)

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4. Conclusions

The water purification efficiency by natural and synthesized coagulants was studied at this work. In May and August 2021, research was conducted with the water of the Ingul River. The water turbidity hardness was tested by adding natural and synthesized coagulants. In average, the study results showed that seeds extract in unacceptable to use at concentration over 1 mg, because even after filtration at 5 mg/dm^3 the water turbidity was increased. After the test with concentration from 0.1 to 1 mg/dm^3 the optimal dose for turbidity and chromaticity removal was determined – 0.5 mg/dm^3.

SCM_1 and SCM_2, coagulants, which were synthesized from a red mud, have similar properties in water contaminants removal, but SCM_2 turned out more effective. SCM_2 decreased turbidity lower 20 mg/dm^3 at 10 mg/dm^3 while SCM_1 at 15 mg/dm^3.

Thereby, synthesized coagulants and natural coagulants are effective in water purification at small doses. Natural coagulant can be used in areas without access to the central water supply, especially in war zones.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this study, including financial, personal, authorship, or any other, that could affect the study and its results presented in this article.

Financing

The study was conducted without financial support.

Data availability

The manuscript has no associated data.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the presented work

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