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Integrated Particle Size Classification and Baromembrane Filtration for Fouling Control in Natural Water Treatment

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Abstract

This article presents the results of theoretical and experimental studies on the mechanisms of natural water contamination via nanofiltration under turbulent flow conditions. A particle size classifier for natural waters has been developed that effectively describes the composition of the water; based on this, appropriate baromembrane filtration processes can be selected to ensure the gradual removal of particles of various sizes. The particle size classifier for natural waters serves as a tool for elucidating the mechanisms of nanofiltration fouling. Filtration of natural waters and model solutions was conducted under turbulent flow conditions (Re > 4000) using a laboratory cross-flow module and polyamide membranes with varying selectivity. It was found that the particle size distribution (PSD) of the deposit formed on the membrane surface systematically changes with membrane selectivity. A membrane with 80% selectivity retained larger particles ($d_{50} \approx$ 5–15 μm). A membrane with 96% selectivity retained smaller colloids ($d_{50} \approx$ 0.5–2 μm), increasing the fraction of 10–500 nm particles. Direct nanofiltration of highly turbid solutions (FTU 10–50) leads to rapid membrane fouling and a significant reduction in flux (>70%). Therefore, it is recommended to perform preliminary micro- and ultrafiltration of highly turbid solutions, reducing turbidity to FTU 2, followed by nanofiltration, which ensures the production of sterile water from heavily contaminated natural water. It was found that as membrane selectivity increases, the particle size distribution (PSD) shifts toward smaller colloids (10–500 nm), leading to the formation of a more compact precipitate. The results confirm that the particle size classifier in natural waters is an effective tool for studying contamination mechanisms and optimizing nanofiltration systems.

Keywords: nanofiltration; membrane fouling; turbulent flow; classifier

Introduction

The deterioration in drinking water quality and the introduction of stricter standards have increased the demand for advanced drinking water treatment technologies, such as microfiltration, ultrafiltration and nanofiltration [1–4]. Nanofiltration membranes are sensitive to water contaminated with colloidal, organic, inorganic and biological particles, leading to membrane fouling, reduced flow, increased transmembrane pressure, higher energy consumption and reduced system efficiency [5–7].

The aim of this study is to conduct a theoretical and experimental investigation of the tangential baromembrane filtration process for industrial-scale flows and domestic water supply, taking into account turbulent flow; and to develop a suitable classification system based on dissolved and suspended particles present in natural waters.

Materials and Methods

Membranes with pore sizes of 5 µm, 1 µm, 0.45 µm and 0.1 µm were used for filtration. At each stage, the retentates were collected, weighed and characterised using elemental analysis, microscopy, IR spectroscopy, light scattering and zeta potential measurement. Model solutions were prepared and used: 0.5% NaCl, 0.4% $MgSO_4$, 0.3% $CaCl_2$, 0.15% LiCl, the turbidity of which was adjusted within the range of FTU-1, FTU-3, FTU-5, FTU-10 to FTU-50. Membranes and equipment: micro-, ultra- and nanofiltration polyamide membranes with selectivities of 80%, 90% and 96% were used. Filtration of natural waters was carried out under turbulent conditions using a laboratory cross-flow module with a Reynolds number Re > 4000. Operating conditions: transmembrane pressure: 5–10 bar; cross-flow velocity: 0.5–2.5 m/s; high-turbulence model solutions FTU-10, FTU-20, FTU-40 and FTU-50 were pre-treated by microfiltration (0.2–0.45 µm), ultrafiltration (0.1 µm) and nanofiltration (0.01 µm).

Results and conclusions

Particle size plays a decisive role in membrane surface fouling during nanofiltration: nanoparticles and colloids can penetrate the pores or be adsorbed within them, whilst larger particles settle on the surface and contribute to the formation of a fouling layer. Organic matter (NOM fractions), particularly polysaccharides and humic substances, interacts with ions (hydrated K, Cl, Ca, Mg and SO_4 ions), which further exacerbates fouling. Based on theoretical and experimental studies, a particle size classifier for natural waters has been developed.

Table 1. Classifier of particle size

Category	Size range (nm)	Examples
Dissolved substances	<1 nm	Hydrated ions K, Cl, Ca, Mg, SO_4, Simple sugars, amino acids
Colloids	1nm-1000nm	Viruses (20-30nm), humic and fulvic acids Proteins, polysaccharides (fibrous) (1-10nm), Natural nanoparticles: iron and silicon oxide (1-100nm); nanoplastics (1 to 1000nm) and microplastics (1000nm to 5000000nm).
Suspended solids	➢ 1000 nm	bacteria (1000nm-10000nm), sediment (2000-50000 nm), clay (500-2000nm), sand over 2000nm

Based on the classifier developed, a suitable baromembrane process was selected for the treatment of natural waters of varying turbidity. For the filtration of low-turbidity water, in which the ionic fraction (<1 nm) predominates, nanofiltration was selected, whilst water containing colloidal and suspended particles was pre-treated by micro- and ultrafiltration. Pre-treatment of heavily contaminated water using microfiltration/ultrafiltration is a critical step in the nanofiltration process, as it reduces the colloidal and solid load on the membrane, minimises pore blockage and sludge formation, and improves the stability of the nanofiltration flow and the service life of the membrane. Furthermore, pre-treatment reduces the rate of fouling, simplifies chemical cleaning cycles and ensures energy-efficient operation of the system. The integration of microfiltration/ultrafiltration processes is particularly effective when the water contains high concentrations of colloids, organic macromolecules, microbiological particles and suspended solids [8-10].

The particle size distribution (PSD) of the sediment formed on the membrane surface varied systematically depending on the membrane's selectivity. On a membrane with 80% selectivity, larger particles ($d_{50} \approx 5$–15 μm) predominated in the deposit. A membrane with 96% selectivity retained smaller colloids ($d_{50} \approx 0.5$–2 μm, increasing in the 10–500 nm fraction). Particles ranging in size from 0.3–100 nm were deposited on the membrane surface with higher selectivity, forming finer but more stable precipitates.

Scanning probe microscopy revealed that the membrane surface was covered with gel-like, sticky contaminants (Fig. 1, Fig.2, Fig.3, Fig.4), distributed in a layer of uneven thickness. The portion of the contamination in direct contact with the membrane surface and pores could not be removed mechanically, as evidenced by the brown residue remaining on the membrane after physical cleaning. Direct nanofiltration of solutions with high turbidity (FTU 10–50) resulted in a loss of more than 70% of the pure water flow within 30 minutes (and the formation of a thick deposit (d_{50} >20 μm) on the membrane surface. Pre-treatment of these solutions by microfiltration/ultrafiltration reduced turbidity to <2 FTU, restoring the clear water flux by more than 95%.

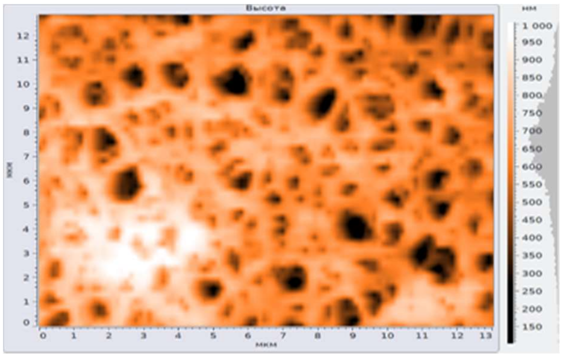

Fig.1 clean membrane

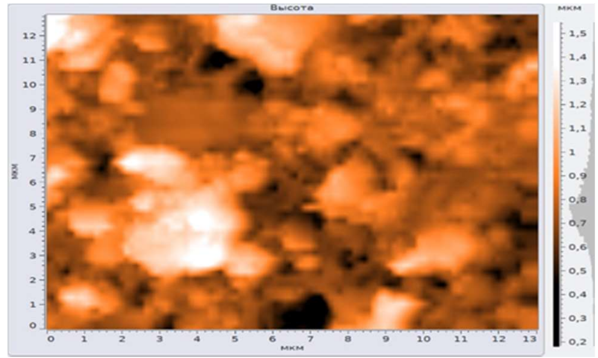

Fig.2 2D-image of sediment formed during filtration of FTU1 turbidity water

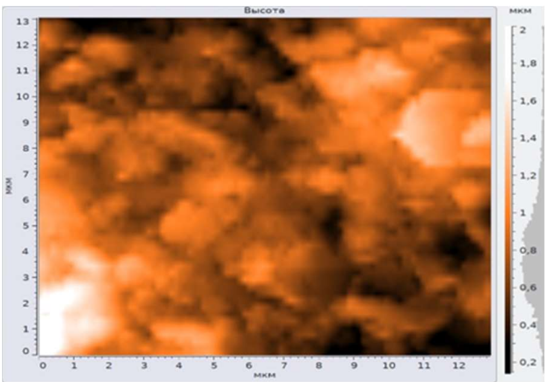

Fig. 3 2D-image of sediment formed during filtration of FTU5 turbidity water

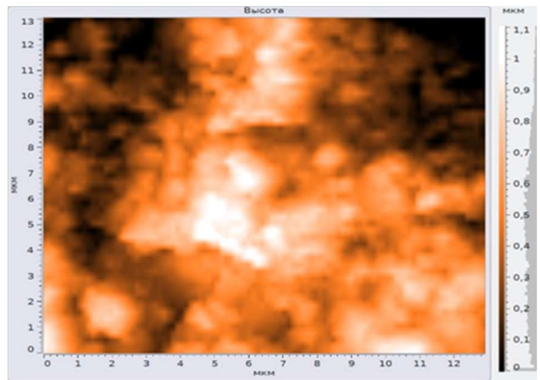

Fig.4 2D-image of sediment formed during filtration of FTU10 turbidity water

During the first two hours of filtration, particles 1–50 nm in size cause pore constriction, resulting in a 45% reduction in flux. Particles 50–500 nm in size block intermediate pores and moderately reduce the pure water flux. Particles 500 nm–2 μm in size form a sediment layer on the membrane surface, which is the smallest initial reduction in flux.

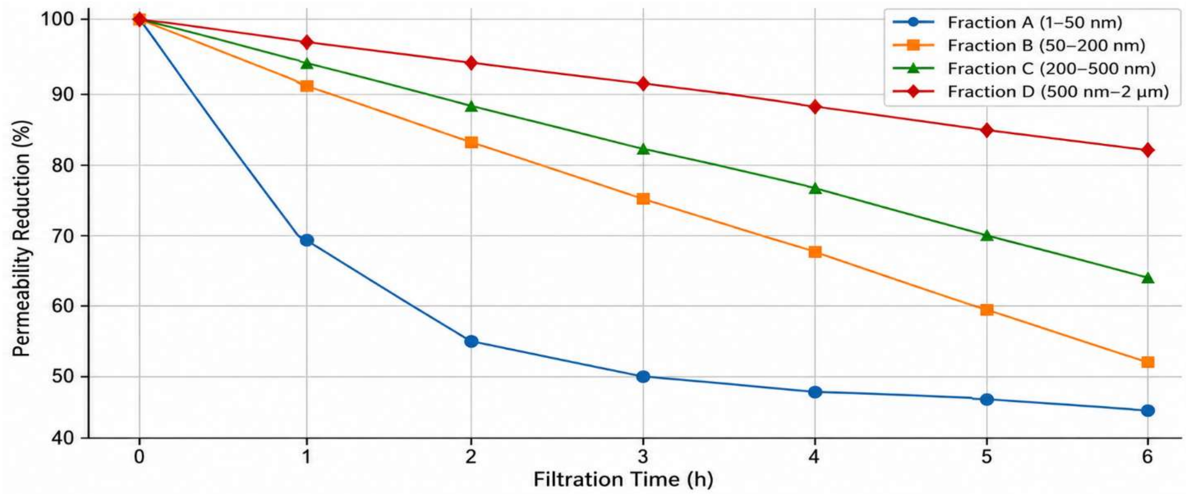

Fig.5 Reduced filter consumption for particles of different sizes

Thus, the fouling mechanism during the filtration of natural water containing particles ranging in size from 10 nm to 1 µm involves the formation of a dense gel layer that blocks the entrance to the pores, leading to the most undesirable type of fouling due to the irreversible loss of membrane selectivity. Particles smaller than 1 nm crystallise on the membrane surface and, due to increased concentration, may detach from the membrane surface. Dissolved macromolecules (NOM) ranging in size from 1 to 10 nm are adsorbed within the pores and on the surface of the membrane matrix, leading to pore narrowing and a change in the membrane's surface charge. Fine colloids adhere to the walls of the internal polyamide pores, whilst larger particles form a porous matrix that compacts under high operating pressure.

The particle-size-based classifier developed enables the rational selection of a specific baromembrane process for the filtration of natural waters with varying degrees of contamination. Natural waters containing particles smaller than 1 nm (dissolved substances, hydrated K and Cl ions (0.7–1 nm), simple sugars) constitute the ionic fraction and are directly subjected to nanofiltration. For waters containing colloids of 1–1000 nm in size, ultra- and nanofiltration are recommended, whilst for suspended colloids of 1 µm in size, microfiltration is recommended. The results obtained show that the efficiency of nanofiltration is determined by three main factors: membrane selectivity, particle size distribution and hydrodynamic conditions. Pre-treatment by microfiltration/ultrafiltration is essential for highly turbid waters, as it reduces the colloidal load and improves the stability of the nanofiltration process. The developed particle size classifier is an effective tool for selecting and optimising baromembrane processes. The results obtained enable the design of nanofiltration systems to be predicted and the treatment of complex waters to be managed.

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ინტეგრირებული მიდგომა ბუნებრივი წყლების დამუშავებაში დაბინძურების კონტროლისთვის

გიორგი ბიბილეიშვილი, მზია კეჟერაშვილი, ზაზა ჯავაშვილი, ნანა გოგესაშვილი, ლიანა ყუფარაძე, ელენე კაკაბაძე

საქართველოს ტექნიკური უნივერსიტეტის მემბრანული ტექნოლოგიების საიჟინრო ინსტიტუტი

რეზიუმე

ნაშრომში წარმოდგენილია ბუნებრივი წყლების ნანოფილტრაციის დაბინძურების მექანიზმების თეორიული და ექსპერიმენტული კვლევის შედეგები ტურბულენტური ნაკადის პირობებში. შემუშავებული იქნა ბუნებრივ წყლებში არსებული ნაწილაკების ზომის კლასიფიკატორი, რომელიც ეფექტურად აღწერს წყლის შედგენილობას, რომლის მიხედვით შესაძლებელია ფილტრაციისათვის შეირჩეს შესაბამისი ბარომემბრანული პროცესები და უზრუნველყოს სხვადასხვა ზომის ნაწილაკების ეტაპობრივი მოცილება. ბუნებრივ წყლებში ნაწილაკების ზომის კლასიფიკატორი წარმოადგენს ინსტრუმენტს ნანოფილტრაციული დაბინძურების მექანიზმების გასარკვევად. ბუნებრივი წყლების და მოდელური ხსნარების ფილტრაცია ხორციელდებოდა ტურბულენტურ რეჟიმის პირობებში Re>4000 ლაბორატორიული კროს-ფლოუს მოდულის გამოყენებით სხვადასხვა სელექტიურობის მქონე პოლიამიდური მემბრანების გამოყენებით. დადგენილი იქნა, რომ მემბრანის ზედაპირზე წარმოქმნილი ნალექის ნაწილაკების ზომის განაწილება სისტემატურად იცვლებოდა მემბრანის სელექტიურობასთან ერთად. 80% სელექტიურობის მემბრანაზე ნალექი დომინირებული იყო უფრო დიდი ნაწილაკებით ($d_{50} \approx 5–15$ მკმ). 96% სელექტიურობის მემბრანამ შეინარჩუნა უფრო წვრილი კოლოიდები ($d_{50} \approx 0,5–2$ მკმ, 10–500 ნმ ფრაქციის ზრდა). მაღალი სიმღვრივის (FTU-10–50) ხსნარების პირდაპირი ნანოფილტრაცია იწვევს მემბრანის ზედაპირის სწრაფ დაბინძურებას და ნაკადის მნიშვნელოვან შემცირებას (>70%). ამიტომ რეკომენდირებულია მაღალი სიმღვრივის ხსნარების წინასწარი მიკრო- და ულტრაფილტრაცია, რაც ამცირებს სიმღვრივეს FTU2-მდე და შემდეგ ეტაპზე ნანოფილტრაცია, რაც უზრუნველყოფს მაღალი დაბინძურების მქონე ბუნებრივი წყლებიდან სტერილური წყლის მიღებას. დადგინდა, რომ მემბრანის სელექტიურობის ზრდასთან ერთად ნალექის ნაწილაკების ზომის განაწილება (PSD) გადადის უფრო წვრილი კოლოიდების მიმართულებით (10–500 ნმ), რაც იწვევს უფრო კომპაქტური ნალექის ფორმირებას. მიღებული შედეგები ადასტურებს, რომ ბუნებრივ წყლებში ნაწილაკების ზომის კლასიფიკატორი წარმოადგენს ეფექტურ ინსტრუმენტს დაბინძურების მექანიზმების კვლევისათვის და ნანოფილტრაციული სისტემების ოპტიმიზაციისთვის.

საკვანძო სიტყვები: ნანოფილტრაცია; მემბრანის დაბინძურება; ტურბულენტური ნაკადი; კლასიფიკატორი