World Scientific Research Journal
Volume 11 Issue 11, 2025
ISSN: 2472-3703
DOI: 10.6911/WSRJ.202511_11(11).0002

# Research Progress and Prospects on the Adsorption of Ions in Wastewater by Eggshell Membranes

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## Abstract

**With the acceleration of industrialization, wastewater pollution has become an increasingly severe issue, and the search for efficient, low-cost wastewater treatment methods has become a research hotspot. Eggshell membrane, as a biological adsorbent material, has shown great potential in wastewater treatment due to its abundant sources, rich active functional groups, and unique structure. This paper provides a comprehensive review of the current research status of eggshell membrane in wastewater treatment, including preparation and characterization methods, adsorption performance for different ions, adsorption mechanisms, and practical application scenarios, and offers insights into future development directions, aiming to provide references for further research and application of eggshell membrane in the field of wastewater treatment.**

## Keywords

**Eggshell membrane; Wastewater treatment; Adsorption performance; Adsorption mechanism.**

## 1. INTRODUCTION

### 1.1. Current Status and Hazards of Wastewater Pollution

With the acceleration of global industrialization, wastewater discharge volumes continue to rise. Industrial wastewater and domestic sewage often contain large amounts of heavy metal ions (such as copper, lead, cadmium, and mercury) and dye ions (such as methylene blue and Congo red). These pollutants are highly toxic, difficult to degrade, and tend to accumulate in living organisms, posing a serious threat to the ecological environment and human health [1]. Heavy metal ions can enter the human body through the food chain, damaging the nervous system, immune system, and reproductive system, and causing various diseases; dye ions not only affect the aesthetic quality of water bodies but may also contain carcinogenic and teratogenic substances, posing potential hazards to aquatic organisms and human health. Traditional wastewater treatment technologies such as chemical precipitation, ion exchange, and membrane separation can remove pollutants to some extent, but they have issues like high costs, potential secondary pollution, and high energy consumption [2]. Therefore, developing efficient, low-cost, and environmentally friendly wastewater treatment technologies is urgent.

### 1.2. Advantages and Development of Bioadsorbent Materials

Bioadsorbent materials have become a research hotspot in the field of wastewater treatment due to their wide availability, low cost, and biodegradability. These materials can adsorb pollutant ions in wastewater through physical adsorption, chemical complexation, ion exchange, and other mechanisms, thereby achieving wastewater purification [3]. Eggshells, as the primary waste product of the poultry egg processing industry, have a massive global annual production volume. The inner eggshell membrane is a thin yet resilient biological membrane primarily composed of biomacromolecules such as collagen, glycoproteins, and polysaccharides. It is rich in active functional groups such as hydroxyl, carboxyl, and amino groups and possesses a porous network structure, endowing it with excellent adsorption potential [4]. Utilizing eggshell membranes for wastewater treatment not only achieves resource recovery from waste but also reduces wastewater treatment costs, yielding significant environmental and economic benefits. In recent years, research on the application of eggshell membranes in wastewater treatment has increased significantly, becoming a cross-disciplinary frontier in the fields of resource recycling and water pollution control [5].

## 2. RESEARCH PROGRESS ON THE PREPARATION AND CHARACTERIZATION OF EGGSHELL MEMBRANES

### 2.1. Separation and Pretreatment Technologies for Eggshell Membranes

2.1.1. Separation Methods

The separation of eggshell membranes from eggshells is the first step in achieving their adsorption applications. Currently, common separation methods include physical, chemical, and biological methods. Among physical methods, mechanical peeling is simple to operate but has low efficiency, making it suitable only for small-scale experiments; ultrasonic-assisted methods utilize the cavitation effect of ultrasonic waves to efficiently separate eggshell membranes at room temperature with minimal damage to membrane structure, making it a commonly used method in laboratories; pyrolysis can achieve separation, but high temperatures may damage the functional groups on the membrane surface, affecting adsorption performance [6]. Chemical separation primarily involves treating eggshells with acids, bases, or chelating agents. Dilute acids can dissolve calcium carbonate in eggshells to detach the membrane, but may corrode the membrane structure; Alkali solutions can hydrolyze adhesive substances at certain temperatures to achieve efficient separation with minimal damage to the membrane; chelating agents disrupt eggshell structure by complexing calcium ions, offering a gentle separation process but at a higher cost [7]. Biological methods utilize carbonic anhydrase enzymes secreted by microorganisms to decompose eggshells, which are environmentally friendly but have a long processing cycle and have not yet been scaled up for industrial application [8].

2.1.2. Pretreatment Methods

Pretreatment is crucial for enhancing the adsorption performance of eggshell membranes. Cleaning typically uses deionized water or dilute ethanol solutions to remove impurities and residual egg liquid from the membrane surface; drying methods include vacuum drying and freeze-drying, with freeze-drying better preserving the porous structure and improving adsorption performance; grinding can convert eggshell membranes into powder form, increasing the specific surface area and enhancing adsorption efficiency; Chemical modification involves increasing the number of surface-active functional groups through acid-base treatment, graft copolymerization, etc. For example, nitric acid treatment introduces more carboxyl groups, significantly enhancing adsorption capacity for heavy metal ions [9].

### 2.2. Structural and Chemical Characterization of Eggshell Membrane

2.2.1. Microscopic Structural Characterization

Scanning electron microscopy (SEM) revealed that the eggshell membrane exhibits a three-dimensional porous network structure with pore sizes ranging from 1 to 10 μm, providing ample diffusion pathways and adsorption sites for contaminant ions. The surface of fresh eggshell membrane is smooth, and after pretreatment, the pore structure becomes more distinct, resulting in an increased specific surface area. For example, the specific surface area of freeze-dried eggshell membrane can reach 10–15 $m^2/g$, significantly higher than that of naturally air-dried samples [10].

2.2.2. Chemical Functional Group Analysis

Fourier transform infrared spectroscopy (FT-IR) analysis revealed the chemical functional group composition of eggshell membrane. The absorption peak around 3400 $cm^{-1}$ corresponds to the stretching vibrations of hydroxyl and amino groups, the peak at 1650 $cm^{-1}$ represents the amide I band, the peak at 1540 $cm^{-1}$ represents the amide II band, and the peak around 1080 $cm^{-1}$ corresponds to the stretching vibrations of C-O-C in polysaccharide compounds. These functional groups serve as key sites for adsorbing pollutant ions, and shifts or changes in the intensity of characteristic peaks after adsorption experiments can reflect the adsorption mechanism [11].

2.2.3. Specific Surface Area and Pore Size Analysis

Results from the BET surface area and pore size analyzer indicate that the surface area of eggshell membranes typically ranges from 5 to 20 $m^2/g$, with a pore volume of 0.05 to 0.15 $cm^3/g$, primarily consisting of mesopores. Surface area is positively correlated with adsorption capacity; processes such as grinding and expansion can increase surface area and enhance adsorption performance. X-ray photoelectron spectroscopy (XPS) was used to analyze the elemental composition and chemical valence of the eggshell membrane surface. The results showed that the membrane surface primarily contains elements such as C, O, and N, providing a chemical basis for adsorption reactions [12].

# 3. STUDY ON THE ADSORPTION PERFORMANCE OF EGGSHELL MEMBRANE FOR IONS IN WASTEWATER

## 3.1. Adsorption Kinetic Characteristics

Adsorption kinetics studies the relationship between the adsorption rate of pollutants by eggshell membranes and time. Commonly used models include the pseudo-first-order kinetic model, pseudo-second-order kinetic model, and particle internal diffusion model. Studies indicate that the adsorption of heavy metal ions and dye ions by eggshell membranes better conforms to the pseudo-second-order kinetic model, with a correlation coefficient $R^2$ typically above 0.99, suggesting that chemical adsorption is the rate-limiting step [13]. Taking copper ion adsorption as an example, in a solution with an initial concentration of 100 mg/L, eggshell membrane reaches adsorption equilibrium in 60 minutes, with an equilibrium adsorption capacity of approximately 25 mg/g. The adsorption rate increases with rising initial concentration, and higher temperatures shorten the equilibrium time, indicating that adsorption is an endothermic reaction [14]. For dye ions such as methylene blue, due to their large molecular size and slow diffusion rate, the equilibrium time requires 120–180 minutes, but it still conforms to the pseudo-second-order kinetic model. The intra-particle diffusion model shows that the adsorption process is divided into two stages: boundary layer diffusion and intra-particle diffusion. Intra-particle diffusion is the primary rate-limiting step, and the smaller the particle size, the faster the intra-particle diffusion rate and the shorter the equilibrium time [15].

## 3.2. Adsorption Isotherm Characteristics

Adsorption isotherms describe the relationship between the amount of pollutant adsorbed by the adsorbent at equilibrium and the concentration of the pollutant in the solution. Commonly used models include the Langmuir model and the Freundlich model. The adsorption of heavy metal ions by eggshell membranes better conforms to the Langmuir model, indicating that adsorption primarily occurs in a single molecular layer with a saturated adsorption capacity. The maximum adsorption capacity for different heavy metal ions varies due to differences in ionic chemical properties and affinity with membrane surface functional groups. For example, the maximum adsorption capacity for lead ions can reach 50–80 mg/g, for copper ions 20–30 mg/g, and for cadmium ions 15–25 mg/g. For dye ions, both the Langmuir and Freundlich models fit well, with the maximum adsorption capacity for methylene blue being approximately 30–50 mg/g and for Congo red approximately 25–40 mg/g. The empirical constant 1/n in the Freundlich model reflects adsorption strength. The 1/n values for eggshell membranes range from 0.2 to 0.4 for most ions, indicating strong adsorption capacity [16]. Increasing temperature increases the maximum adsorption capacity in the Langmuir model, consistent with the conclusion that the adsorption process is an endothermic reaction [17].

### 3.3. Key Factors Affecting Adsorption Performance

The pH value of the solution significantly affects adsorption performance by altering the ionization state of functional groups on the eggshell membrane surface and the existence form of pollutant ions. For heavy metal ions, under acidic conditions, the functional groups on the membrane surface become protonated, leading to electrostatic repulsion with metal cations, resulting in low adsorption capacity. As pH increases, the complexation and ion exchange interactions between functional groups and metal ions strengthen, increasing adsorption capacity. However, at excessively high pH levels, metal ions tend to form hydroxide precipitates, interfering with adsorption. Therefore, the optimal pH range for heavy metal ion adsorption is typically 4–6 [18]. For dye ions, the effect of pH varies depending on the charge characteristics. Cationic dyes exhibit increased adsorption capacity with rising pH under acidic conditions, while anionic dyes perform better under alkaline conditions. Temperature increases typically enhance adsorption capacity, but above 60°C, proteins in the eggshell membrane may denature, leading to reduced adsorption capacity [19]. The initial ion concentration and adsorbent dosage also affect adsorption efficiency. Adsorption capacity increases with rising initial concentration until saturation, while adsorption rate decreases with increasing initial concentration but increases with higher adsorbent dosage. In practical applications, the dosage should be optimized based on the wastewater concentration. Coexisting ions compete with target ions for adsorption sites, leading to a decrease in adsorption capacity. However, eggshell membranes still exhibit selective adsorption capacity for target ions, with selectivity related to the ion's charge density and complexation constant [20].

## 4. INVESTIGATION OF THE MECHANISM OF EGGSHELL MEMBRANE ADSORPTION OF IONS IN WASTEWATER

### 4.1. Physical Adsorption Mechanism

Physical adsorption is achieved through van der Waals forces, electrostatic attraction, and pore retention. The porous structure of the eggshell membrane provides conditions for pore retention. SEM observations revealed that pollutants accumulate in the pores after adsorption, and specific surface area analysis also confirmed that pollutant molecules entering the pores cause a slight decrease in specific surface area and pore volume. Electrostatic attraction plays a significant role in the adsorption of ionic pollutants. The surface charge properties of eggshell membranes vary with pH, enabling the adsorption of pollutants with different charges through electrostatic attraction. Zeta potential measurements provide direct evidence for the electrostatic adsorption mechanism [21]. Van der Waals forces, as ubiquitous intermolecular forces, assist in physical adsorption, particularly for non-polar or weakly polar pollutant molecules. Physical adsorption forces are relatively weak and typically reversible; rinsing with clean water can remove some of the adsorbed pollutants [22].

### 4.2. Chemical Adsorption Mechanism

Chemical adsorption is the primary mechanism by which eggshell membranes adsorb pollutant ions, including complexation reactions, ion exchange, and redox reactions. Complexation reactions are the core mechanism for the adsorption of heavy metal ions. Functional groups such as hydroxyl, carboxyl, and amino groups in eggshell membranes can form stable complexes with metal ions. FT-IR and XPS analyses have confirmed this process [23]. Ion exchange reactions occur between exchangeable ions on the membrane surface and heavy metal ions in wastewater. The proportional relationship between changes in the concentration of exchangeable ions in the solution and the amount of heavy metal ions adsorbed indicates the presence of ion exchange [24]. For ions with redox activity, such as $Cr^{6+}$, the reductive substances in eggshell membranes can reduce them to $Cr^{3+}$, which are then adsorbed through complexation. XPS analysis confirmed the occurrence of redox reactions [25].

### 4.3. Synergistic Effects of Adsorption Mechanisms

In actual adsorption processes, physical adsorption and chemical adsorption act synergistically. Heavy metal ions are first adsorbed onto the membrane surface via electrostatic forces, followed by complexation or ion exchange reactions. The pore structure provides pathways for ion diffusion. Chemical adsorption is the rate-limiting step, while physical adsorption can accelerate the initial adsorption rate. Different factors influence adsorption mechanisms in varying ways: pH primarily affects chemical adsorption, temperature primarily affects the reaction rate of chemical adsorption, and specific surface area primarily affects physical adsorption capacity. Comparing eggshell membranes with different pretreatments revealed that chemical modification increases the contribution of chemical adsorption, while physical treatment enhances the proportion of physical adsorption, providing insights for optimizing adsorption mechanisms [26].

## 5. APPLICATION OF EGGSHELL MEMBRANES IN ACTUAL WASTEWATER TREATMENT

### 5.1. Evaluation of Actual Wastewater Treatment Effectiveness

Experiments on the treatment of industrial wastewater and domestic sewage indicate that eggshell membranes have good application potential in actual wastewater treatment. When treating copper-containing electroplating wastewater, under optimal conditions, the removal rate of $Cu^{2+}$ can reach 85%–92%, with the $Cu^{2+}$ concentration in the treated water reduced to below national emission standards. When treating dyeing wastewater, the removal rate of methylene blue can reach 80%–90%, and it can also reduce the color and COD values of the wastewater. Compared to simulated wastewater, the adsorption capacity of actual wastewater is slightly reduced, which is attributed to the competition for adsorption sites by coexisting ions and organic matter in the actual wastewater. When treating domestic wastewater, eggshell membranes achieve a total heavy metal removal rate of 70%–80% and also exhibit some adsorption effects on nutrients such as phosphorus and nitrogen. Dynamic adsorption experiments show that eggshell membrane-filled columns can treat 50–80 bed volumes of actual wastewater, with stable effluent quality, enabling continuous treatment [27].

### 5.2. Regeneration and Reuse of Eggshell Membranes

Regenerating saturated eggshell membranes is key to reducing treatment costs. Common regeneration methods include acid elution, alkali elution, and salt elution. For eggshell membranes adsorbed with heavy metal ions, acid or nitric acid elution can achieve a regeneration rate of 70–80%, and after five regeneration cycles, the adsorption capacity remains at 50–60% of the initial capacity. For dye ions, alkaline solution elution is more effective, achieving a regeneration rate of 65–75%. Salt elution removes pollutants through ion exchange, with a slightly lower regeneration rate than acid-base elution, but it causes less damage to the eggshell membrane, thereby extending its service life [28].

## 6. CONCLUSIONS AND OUTLOOK

### 6.1. Research Summary

Eggshell membrane, as a bioadsorbent material with unique structural and chemical properties, shows great potential for application in wastewater treatment. Through appropriate separation and pretreatment methods, its adsorption performance can be significantly enhanced. Studies on the adsorption of heavy metal ions and dye ions indicate that eggshell membrane exhibits rapid adsorption rates, high adsorption capacity, and certain selectivity, with the adsorption process conforming to the pseudo-second-order kinetic model and Langmuir or Freundlich adsorption isotherms. The synergistic interaction between physical and chemical adsorption mechanisms determines its adsorption performance, and factors such as solution pH, temperature, initial ion concentration, adsorbent dosage, and coexisting ions significantly influence adsorption efficiency. In practical wastewater treatment, eggshell membranes demonstrate good removal efficiency for pollutants in industrial wastewater and domestic sewage, and possess certain regenerative and reusable properties [29].

### 6.2. Future Research Directions

Although eggshell membranes have made some progress in wastewater treatment, many issues remain to be further studied. Future research can be conducted in the following areas: first, further investigate modification methods for eggshell membranes to develop more efficient and environmentally friendly modification technologies, thereby enhancing their adsorption performance and selectivity; Second, strengthen research on adsorption mechanisms, combining advanced characterization techniques to deeply reveal the synergistic mechanisms of physical and chemical adsorption under different conditions, providing a theoretical foundation for optimizing the adsorption process; Third, conduct application studies of eggshell membranes in complex wastewater systems, investigating their adsorption performance and competitive adsorption mechanisms when multiple pollutants coexist, to enhance their adaptability in actual wastewater treatment; Fourth, explore the combined application of eggshell membranes with other wastewater treatment technologies, such as biological treatment technologies and membrane separation technologies, to form efficient composite treatment processes; Fifth, focus on the large-scale preparation and application of eggshell membrane adsorbent materials, address engineering and technical issues in practical applications, reduce costs, and promote their industrialization process [30]. It is believed that with the continuous deepening of research, eggshell membranes will play a greater role in the field of wastewater treatment, providing new effective solutions to water pollution issues.

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