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Revolutionizing membrane Performance: Repurposing drinking water bottle waste for enhanced chromium removal in wastewater treatment

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Highlights

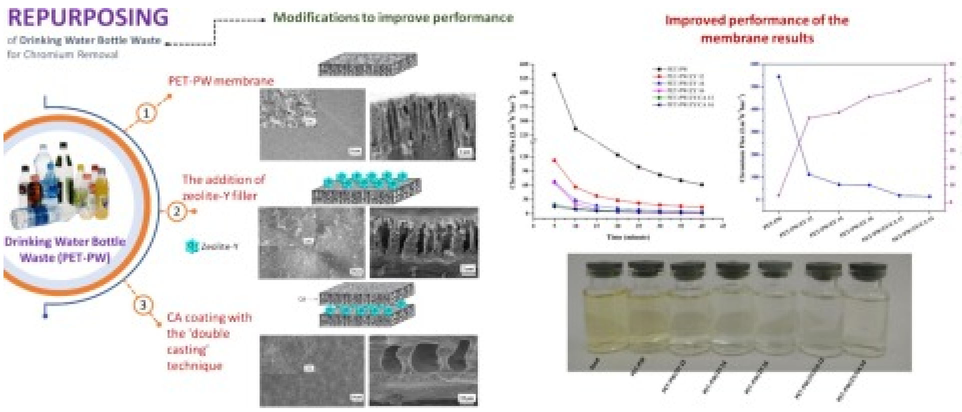
- PET-PW membrane from drinking water bottles successfully fabricated & modified.
- Zeolite-Y filler boosted chromium (VI) ion rejection.
- Zeolite-Y filler changed membrane morphology from finger-like to sponge-like.
- The 'double casting' technique for CA coating boosts PET-PW/ZY membrane rejection.

- The PET-PW/ZY/CA 14 membrane performs best chromium (VI) ion removal.

Abstract

Wastewater from the electroplating industry contains chromium (VI) ions, which can cause lung cancer, nose cancer, DNA transcription issues, kidney failure, liver damage, and digestive tract disorders. Developing membrane technology that effectively removes chromium ions at a low cost is crucial to addressing this problem. This research utilizes plastic waste from drinking water bottles of polyethylene terephthalate (PET-PW) to create a low-cost and environmentally friendly membrane material to address this problem. Modifications were made by adding zeolite-Y (ZY) filler and cellulose acetate (CA) membrane coating to enhance its performance in removing chromium (VI) ions. The research results showed that adding ZY and CA coating increased the porosity value of the PET-PW membrane from 75% to 82% and 83%, respectively. These modifications changed the surface pores and cross-section of the membrane to become relatively smaller and more sponge-like than the PET-PW membrane. The CA coating on the PET-PW/ZY membrane produces a membrane with two layers, where the top layer has larger finger-like pores than the bottom layer. The performance of the ZY-modified PET-PW membrane increased the rejection of chromium (VI) ions from 4% to 48%. The chromium flux produced by the PET-PW/ZY membrane is 120 $Lm^{-2}h^{-1}bar^{-1}$. The CA coating increased the water flux from 50 $Lm^{-2}h^{-1}bar^{-1}$ to 200 $Lm^{-2}h^{-1}bar^{-1}$. This CA coating also increased chromium (VI) ion rejection from 60% to 70%. Thus, this study demonstrates a very perspective and cheap way of pollution abuse regarding chromium VI, while utilizing plastic waste to create an environment-friendly solution.

Graphical abstract



Introduction

Pollution caused by heavy metals such as chromium in wastewater from the electroplating industry is hazardous for human health and the environment. Chromium is of particular concern due to its high toxicity, which can cause lung cancer [1], nose cancer [2], DNA transcription [3], liver harm [4], kidney failure [5], gastrointestinal disorders [6], and its potential to accumulate in the food chain [7]. Electroplating industry wastewater contains Cr(VI) ions in the form of bichromates ($HCrO_4^-$), dichromates ($Cr_2O_7^{2-}$), and chromates (CrO_4^{2-}) [8]. The chromium ion concentration in the effluent from the electroplating industry was reported to be 100 ppm, which exceeds the maximum threshold (0.05 ppm according to the National Primary Drinking Water Regulations for industrial wastewater) [9]. There is a need for a well-performing and low-cost technology to reduce chromium ion levels in electroplating industry wastewater.

Membrane technology is reported to have smaller energy consumption, low operating pressure, and high removal efficiency [10] compared to chemical precipitation [11], adsorption [12], electrocoagulation [13], and coagulation-flocculation [14] for chromium (VI) ion removal. Ultrafiltration membrane is easy to manufacture and modify to produce good performance [10], [15], [16]. For wastewater treatment, the ultrafiltration (UF) process with a pore size range of 1–100 nm can be used to separate macromolecules, organic compounds, viruses, and bacteria at relatively small pressures (2–10 bar) [17]. According to reports, UF membranes featuring finer pores (0.2 µm) have demonstrated the capability to decrease wastewater turbidity by 99.62%, eliminate suspended solids by 99.99%, and capture particles with an average diameter of 1.39 µm within wastewater [18]. UF membranes with less than 100 nm pores can produce 97% sulfate rejection, 89% Chemical

Oxygen Demand (COD), and 95% turbidity [19]. UF membranes were reported to produce 89% [20] and 95.2% [21] rejection of chromium ions. However, the performance of ultrafiltration membranes is affected by the materials used. The search for cheap and well-performing materials in the world of membranes is still a challenge and continues to be carried out today by industry and researchers.

On the other hand, pollution caused by plastic waste from drinking water bottles increases yearly. From 2010 to 2020, various countries worldwide produced 381.73 million tons of plastic [22], which has since continued to rise, reaching 400.3 million tons or more [23]. The Asian continent accounts for approximately 86% of plastic waste discharged into the environment [24]. According to the Association of Olefin Aromatic and Plastic Industries, plastics from food and beverage packaging dominate environmental pollution, comprising 65% of plastic waste. Among these, polyethylene terephthalate (PET) plastic holds the largest market share in the packaging sector (41%), surpassing polypropylene (5%), polyethylene (35%), and polysulfone (19%) [25]. Using PET water bottle waste to create useful or economically valuable materials is reported as an effective step compared to burning or landfilling [26], [27]. Therefore, using PET to produce low-cost and environmentally friendly polymeric materials is an appropriate solution to address environmental pollution from plastic waste in drinking water bottles. The elastic nature of PET plastic, its ability to form thin films, and its ease of dissolution in organic solvents are key factors contributing to its suitability for polymer membranes. Additionally, the presence of hydrophilic functional groups (–CO, –COO, and −O-) on PET enhances the hydrophilicity of the membrane, thereby improving the performance of polymer membranes, particularly in terms of high permeability and selectivity in the chromium ion separation process [28].

Modifying membranes made from PET plastic waste from drinking water bottles (PET-PW) is necessary to enhance their performance. Adding fillers can significantly improve the properties and performance of polymer membranes during the ultrafiltration process of wastewater treatment. Zeolite-Y (ZY) is one such filler that is easy to synthesize and has been reported to enhance membrane performance, with a water flux of 11,760 $Lm^{-2}h^{-1}bar^{-1}$ [29], surpassing MOF (water flux 39.2 $Lm^{-2}h^{-1}bar^{-1}$) [30], silica (water flux 50 $Lm^{-2}h^{-1}bar^{-1}$) [31], and carbon (water flux 400 $Lm^{-2}h^{-1}bar^{-1}$) [32]. The choice of zeolites is based on their properties, which include uniformly sized pores composed of tetrahedral aluminosilicates with oxygen atoms at the corners of their geometric patterns, ensuring high thermal and chemical stability. The nucleophilic functional groups on zeolites (OH, Si-O-Al, Al-O-Al, O-Si-O) facilitate effective interaction with positively charged chromium ions present in wastewater [33], [34]. Studies have shown that zeolite Na-A modified polyethersulfone (PES) membranes can achieve 98% rejection of chromium (III) ions [35]. Furthermore, researchers have reported that ZY-modified PES membranes can achieve a rejection rate of 99.2% for removing cesium-137 ions from liquid radioactive waste [36]. However, the use of ZY fillers in PET-PW membranes has not been previously documented. This modification is anticipated to enhance chromium ions' rejection rate during wastewater treatment from the electroplating industry.

Further modification is necessary to enhance the performance of ZY-modified PET-PW membranes for maximizing the removal of chromium (VI) ions. One promising option is the incorporation of other polymers known for their effectiveness in chromium ion removal. Cellulose acetate (CA) membranes, for instance, have been reported to achieve higher rejection rates (98%) [37] compared to other polymers such as polyetherimide (90%) [38], polyethersulfone (92%) [39], polyacrylic acid (20%) [38], and chitosan (90%) [40] in heavy metal removal. Polymer blending is a common technique for creating high-performance membranes by combining two polymers [41], [42], [43]. For example, blending CA with sulfonated poly(ether ether ketone) (CA/SPEEK) membranes yielded 82% chromium ion rejection [44]. Similarly, blending CA with chitosan resulted in 83.40% chromium ion rejection [45]. However, the experimental blending of CA with PET-PW polymers poses a challenge due to the inability to produce a homogeneous dope solution. To address this challenge, researchers face the task of developing a “double casting” technique, where the CA dope solution is printed on top of the PET-PW/ZY membrane. This technique, utilized in cases where two polymers are difficult to blend entirely during membrane manufacturing [46], [47], [48], has yet to be applied to PET-PW/ZY membranes with CA.

Therefore, in this study, the utilization of plastic waste from drinking water bottles made from polyethylene terephthalate for the production of membranes aimed at removing chromium (VI) ions from wastewater in the electroplating industry was conducted. Modifications, adding ZY filler and CA coating using the “double casting” technique, were employed to enhance the membrane's chromium (VI) ion removal performance. This study utilized phase inversion via the immersion precipitation method with the non-solvent-induced phase separation (NIPS) technique to fabricate all membranes.

Section snippets

Chemical reagents and materials

Membrane materials were obtained from polyethylene terephthalate drinking water bottle waste (PET-PW). Ethanol (99%, Merck, C_2H_5OH), phenol (99%, Merck, C_6H_6O), sodium dichromate (99%, Merck, $K_2Cr_2O_7$), cellulose acetate (CA) (99%, Sigma-Aldrich,

$[C_6H_7O_2(OCOCH_3)_3]_x$), glacial acetic acid (99%, Sigma-Aldrich, CH_3COOH), 1,5-Diphenylcarbazide (98%, Merck, $((C_6H_5)NHNH)_2CO$), phosphoric acid (85%, Merck, H_3PO_4), and acetone (99%, Merck, C_3H_6O) were obtained from Germani. Zeolite-NaY was…

Porosity and thickness

Porosity indicates the ratio of pore volume to total membrane volume [73]. Porosity affects the membrane's performance because it correlates with the number of particles capable of passing through the membrane pores. Table 2 compares the porosity values of PET-PW membranes before and after modification by adding ZY filler and coating with CA. ZY can increase the porosity value of the PET-PW membrane by 5–7%. The porous structure of ZY is responsible for augmenting the number of pores in the…

Conclusions

This study successfully made membranes from plastic waste from drinking water bottles made from polyethylene terephthalate (PET-PW), modified ZY (PET-PW/ZY), and modified CA (PET-PW/ZY/CA). The PET-PW/ZY 12 membrane increased the rejection of chromium (VI) ions from 4% to 48%. Increasing the concentration of ZY added to the PET-PW membrane increased the removal of chromium ions up to 60%. PET-PW/ZY 12 membrane had a greater chromium flux (120 $Lm^{-2}h^{-1}bar^{-1}$) than PET-PW/ZY 14 (75 $Lm^{-2}h^{-1}bar^{-1}$) …

CRediT authorship contribution statement

Badrut Tamam Ibnu Ali: Writing – review & editing, Writing – original draft, Visualization, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Hanifah Syifa Azzahra Bay:** Methodology, Formal analysis. **Hens Saputra:** Writing – review & editing. **Semuel Pati Senda:** Writing – review & editing. **Nurul Widiastuti:** Supervision, Methodology.…

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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