Judul Karya Ilmiah (Artikel)		;	Polysulfone Influence on Au Selective Adsorbent Imprinted Membrane Synthesis with Sulfonated Polyeugenol as Functional Polymer		
Jur	nlah Penulis	:	6 Orang Penulis anggota		
Na	ma Penulis	:	Muhammad Cholid Djunaidi, Nor Basid Adiwibawa Prasetya, Arini Khoiriyah, Pardoyo, Abdul Haris, Nabilah Anindita Febriola		
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<u>Prof. Drs. Gunawan, M.Si., Ph.D.</u> NIP. 196408251991031001 Unit kerja: Departemen Kimia FSM Undip Jabatan Fungsional: Guru Besar Bidang Ilmu: Kimia Semarang, 24 Mei 2023

**Reviewer** 2

Dr. Retue Ariadi Lusiana, S.Si., M.Si. NIP. 197012021997022001 Unit kerja: Departemen Kimia FSM Undip Jabatan Fungsional: Lektor Kepala Bidang Ilmu: Kimia

Judul Karya Ilmiah (Artikel)		:	Polysulfone Influence on Au Selective Adsorbent Imprinted Membrane Synthesis with Sulfonated Polyeugenol as Functional Polymer
Jur	nlah Penulis	:	6 Orang Penulis anggota
Na	ma Penulis	:	Muhammad Cholid Djunaidi, Nor Basid Adiwibawa Prasetya, Arini Khoiriyah, Pardoyo, Abdul Haris, Nabilah Anindita Febriola
Ide	ntitas Jurnal Ilmiah		
a.	Nama Jurnal	:	Membranes
b.	Nomor ISSN	:	0022-3093
C.	Volume, No, Bulan, Tahun	:	Vol. 10, issue 12, hal 390-402. Tahun 2020
d.	Penerbit	:	MDPI
e.	DOI artikel (jika ada)	:	https://doi.org/10.3390/membranes10120390
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	Total = (100%)	39,5	39,5	39.5

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Prof. Drs. Gunawan, M.Si., Ph.D. NIP. 196408251991031001 Unit kerja: Departemen Kimia FSM Undip Jabatan Fungsional: Guru Besar Bidang Ilmu: Kimia Semarang, 24 Mei 2023

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d. Kelengkapan unsur dan kualitas penerbit (30%)	12					11,9
Total = (100%)	40					39.5
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- b. Ruang lingkup dan kedalaman pembahasan: Pembahasan artikel secara keseluruhan sudah sesuai dengan topik dan cakupan dari jurnal Membranes. Artikel ini membahas tentang pengaruh dari polisulfone pada imprinted membran berbasis polieugenol tersulfonasi dalam penyerapan selektif ion logam emas. Sebanyak 15 jurnal digunakan sebagai referensi menunjukkan bahwa cakupan dari artikel ini cukup mendalam.
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Semarang, 16 Mei 2023 Reviewer L

Prof. Drs. Gunawan, M.Si., Ph.D. NIP. 196408251991031001

Unit kerja : Departemen Kimia FSM Undip Jabatan Fungsional: Guru Besar Bidang ilmu: Kimia

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- b. Ruang lingkup dan kedalaman pembahasan: Pembahasan paper ini sesuai dengan ruang lingkup Membranes yang berfokus pada pengembangan material membran. Sebanyak 15 jurnal dijadikan referensi menunjukkan bahwa paper ini memiliki pembahasan yang cukup luas dan mendalam. Adanya 11 figures dan 2 tabel menandakan bahwa paper memiliki kedalaman yang cukup baik dalam memberikan informasi. Pembahasan paper ini mampu menawarkan penjelasan tentang adsorpsi ion logam emas menggunakan ionic imprinted membran (IIM) dibandingkan dengan non-ionic imprinted membran (NIM).
- c. Kecukupan dan kemutahiran data/informasi dan metodologi: Penyajian data sangat lengkap dan memberikan informasi terkini melalui gambar reaksi kimia dalam sintesis material aktif membran, gambar SEM, spektra FTIR, serta pengukuran selektivitas. Data yang dijabarkan memberikan informasi yang jelas tentang selektivitas yang baik dari IIM dibandingkan dengan NIM.
- d. Kelengkapan unsur dan kualitas penerbit: Jurnal Membranes diterbitkan oleh MDPI dengan posisi Q2 pada quartile, SJR = 0,49. Similarity index berdasarkan Turnitin adalah 19% sehingga memiliki orisinalitas cukup baik. Tidak ditemukannya kesalahan dalam paper ini menunjukkan proses editorial yang baik. Kuantitas dari referensi yang digunakan juga menunjukkan kelengkapan dan kualitas dari review paper ini.

Semarang, 16 Mei 2023 Reviewer 2

Dr. Retno Ariadi Lusiana, S.Si., M.Si. NIP. 197012021997022001

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#### Abstract

An ionic imprinted membrane (IIM) was synthesized using sulfonated polyeugenol, derived from eugenol, as its functional polymer and polysulfone as its base membrane for the selective adsorption of Au(III). This study aims to determine the adsorption of Au(III) metal ions using IIM compared with the non-imprinted membrane (NIM) and to figure out the membrane selectivity towards Au(III) in mixed solutions of Au/Cd, Au/Cu, and Au/Fe. IIM has a pore size of 0.767 µm while the non-imprinted membrane (NIM) has a pore size of 0.853 µm. The best adsorption result was obtained in the variation of the membrane with the addition of 3.84 g of polysulfone that had pores according to the size of Au. The selectivity results of the Au/Cd mixture solution in NIM and IIM were 17.802 and 36.265. In the mixture of Au/Cu, the NIM and IIM selectivity was 2.386 and 6.886, and in the mixed solution of Au/Fe, the selectivity of NIM and IIM was 0 and 8.489. Thus, the selectivity of IIM towards Au is bigger than NIM.

Keywords: imprinted membrane; sulfonated polyeugenol; polysulfone; gold adsorption; selectivity



Graphical Abstract

#### 1. Introduction

Precious metals such as gold (Au) have high economic value and possess various characteristics compared to other metals. Gold can conduct electricity well, malleable, and can be used in various kinds of ornaments such as coins, jewelry, sculpture, electronic products, and catalysts [1]. As technology develops, gold becomes more broadly applied in industrial process due to its catalytic [2] and corrosion-resistant properties [3]. Industrial processes often leave waste containing useful and valuable metals such as gold [4]. So, waste treatment must be carried out to extract this valuable metal that later on could be used again in the industrial process. Gold concentration in an aquatic environment is low enough that gold determination and recovery require several pre-treatment methods. Due to the low concentration of gold ions and the presence of preconcentration interference, selective metal separation is necessary [5]. There are several techniques for gold recovery such as chemical deposition, ion exchange, adsorption, and membrane filtration [6].

A previous study that has been done regarding Au recovery is by Yuliani who synthesized an ionic imprinted membrane (IIM) which was selective for Au(III) ions using membrane transport techniques, using polyeugenoxy acetyl thiophene methanolate (PEATM) as a functional polymer. The study showed that the transport in a binary mixed metal solution using IIM-PEATM was more selective for transporting Au(III) metal when compared to NIM PEATM. This proves that the presence of the SO<sub>3</sub><sup>-</sup> group in PEATM makes IIM and NIM membranes more selective against Au(III) metal ions which are per the HSAB (Hard Soft Acid Base) theory and it also proves the presence of imprinted ion template in IIM that makes the membranes more selective towards Au(III) [7]. Also, Firlak conducted a study on the recovery of Au(III) ions with imprinted Au(III) hydrogel in water samples. The results of this study indicate that the hydrogel template can selectively adsorb Au(III) nos for high and low concentrations of gold [8].

In this study, sulfonated polyeugenol derived from eugenol will be used as the functional polymer for the synthesis of IIM for Au adsorption. Eugenol is one of the materials that can be used for the separation of metal ions. Eugenol is the starting material for the synthesis of a new compound such as polyeugenol, due to the presence of three functional groups attached to it, namely the allyl, hydroxyl, and methoxy groups, making it a potential functional polymer in synthesis of IIM for selective adsorption [9]. Eugenol is also easily found in Indonesia, it is found in essential oils produced from clove, a very abundant plant in Indonesia. IIM and NIM were synthesized using eugenol derivatives in the form of sulfonated polyeugenol for the adsorption of Au(III) ions. Then, binary metal ions such as Cd, Fe, and Cu were used to determine the membrane selectivity by IIM adsorption to determine the functional polymer effect towards Au selectivity, more specifically, the sulfate group effect in the functional polymer.

#### 2. Materials and Methods

# 2.1. Materials

Eugenol p.a, BF3-diethyl ether, methanol, anhydrous Na2SO4, AIBN (2,2', Azobis(2-methylpropionitrile)), polysulfone and polyethylene glycol were purchased from Sigma-

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by & Kyu Heon Rho, & Youngseung Na, & Taewook Ha and & Dong Kyu Kim Membranes 2020, 10(12), 441; https://doi.org/10.3390/membranes10120441 - 18 Dec 2020 Cited by 8 | Viewed by 3623

Abstract In this study, an electrochemical model was incorporated into a two-phase model using OpenFOAM<sup>®</sup> (London, United Kingdom) to analyze the two-phase flow and electrochemical behaviors in a polymer electrolyte membrane water electrolyzer. The performances of serpentine and parallel designs are compared. The [...] Read more. (This article belongs to the Special Issue Proton Exchange Membrane Water Electrolysis)

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by 🙁 Tsuyoshi Yoda and 🙁 Tomoaki Saito

Membranes 2020, 10(12), 440; https://doi.org/10.3390/membranes10120440 - 18 Dec 2020 Cited by 11 | Viewed by 2486

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Abstract Ethyl caproate (EC) and isoamyl acetate (IA) are key flavor components of sake. Recently, attempts have been made to increase the content of good flavor components, such as EC and IA, in sake brewing. However, the functions of EC and IA in yeast [...] Read more.

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by 🙁 Lwazi Ndlwana, 🝳 Mxolisi M. Motsa and 😫 Bhekie B. Mamba

Membranes 2020, 10(12), 439; https://doi.org/10.3390/membranes10120439 - 18 Dec 2020 Cited by 4 | Viewed by 2215

Abstract Herein we present a two-stage phase inversion method for the preparation of nanocomposite membranes for application in ultra-low-pressure reverse osmosis (ULPRO). The membranes containing DA-stabilized xGnP (xGnP-DA-) were then prepared via dry phase inversion at room temperature, varying the drying time, followed by [...] Read more.

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by Chaeyeon Kim, Chulmin Lee, Soo Wan Kim, Chang Seong Kim and In S. Kim Membranes 2020, 10(12), 438; https://doi.org/10.3390/membranes10120438 - 18 Dec 2020

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Abstract The feasibility of reverse osmosis (RO) for treating coking wastewaters from a steel manufacturing plant, rich in ammonium thiocyanate was assessed. DOW FILMTEC<sup>TM</sup> SW30 membrane performance with synthetic and real thiocyanate-containing solutions was established at the laboratory and (onsite) pilot plant scale. [...] Read more.

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by & Gerardo León, & Asunción María Hidalgo, & Beatriz Miguel and & María Amelia Guzmán Membranes 2020, 10(12), 436; https://doi.org/10.3390/membranes10120436 - 17 Dec 2020 Cited by 9 | Viewed by 1336

Abstract Pertraction of Co(II) through novel supported liquid membranes prepared by ultrasound, using bis-2-ethylhexyl phosphoric acid as carrier, sulfuric acid as stripping agent and a counter-transport mechanism, is studied in this paper. Supported liquid membrane characterization through scanning electron microscopy, energy-dispersive X-ray spectroscopy and [...] Read more. (This article belongs to the Special Issue Membranes for Water and Wastewater Treatment)

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by 😩 Jakob L. Kure, 😫 Camilla B. Andersen, 😩 Thomas E. Rasmussen, 😩 B. Christoffer Lagerholm and 😢 Eva C. Arnspang

Membranes 2020, 10(12), 434; https://doi.org/10.3390/membranes10120434 - 17 Dec 2020 Cited by 4 | Viewed by 1753

Abstract In this study, we explore the use of line FRAP to detect diffusion in synthetic lipid membranes. The study of the dynamics of these membrane lipids can, however, be challenging. The diffusion in two different synthetic membranes consisting of the lipid mixtures 1:1 [...] Read more.

(This article belongs to the Special Issue Dynamics and Nano-Organization in Plasma Membranes)

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# Carbon Nanotubes-Sponge Modified Electro Membrane Bioreactor (EMBR) and Their Prospects for Wastewater Treatment Applications

by P Ali M. Almusawy, Rivad H. Al-Anbari, & Qusay F. Alsalhy and R Arshed Imad Al-Najar Membranes 2020, 10(12), 433; https://doi.org/10.3390/membranes10120433 - 17 Dec 2020 Cited by 6 | Viewed by 1868

Abstract A novel membrane bioreactor system utilizes Multi-Walled Carbon Nanotubes (MWCNTs) coated polyurethane sponge (PUs), an electrical field, and a nanocomposite membrane has been successfully designed to diminish membrane with fouling caused by activated sludge. The classical phase inversion was harnessed to prepare Zinc [...] Read more. (This article belongs to the Special Issue Design and Development of Membrane Bioreactors)

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Transport of Au(III) from HCI Medium across a Liquid Membrane Using R<sub>3</sub>NH<sup>+</sup>CI<sup>−</sup>/Toluene Immobilized on a Microporous Hydrophobic Support: Optimization and Modelling

#### by S Francisco J. Alguacil, S Lorena Alcaraz, Olga R. Largo and Filix A. López Membranes 2020, 10(12), 432; https://doi.org/10.3390/membranes10120432 - 17 Dec 2020

Viewed by 1375

Abstract By the use of the tertiary amine A327 and 1 M HCl solution as precursors, the ionic liquid A327H<sup>+</sup>Cl<sup>-</sup> was generated and used to investigate its performance in the transport of Au(III) from hydrochloric acid medium. The influence of the [...] Read more. (This article belongs to the Special Issue Advances in Supported Liquid Membranes)

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# Theoretical and Experimental Optimization of the Graft Density of Functionalized Anti-Biofouling Surfaces by Cationic Brushes

by & Yijie Ren, & Hongxia Zhou, & Jin Lu, & Sicheng Huang, & Haomiao Zhu and & Li Li Membranes 2020, 10(12), 431; https://doi.org/10.3390/membranes10120431 - 17 Dec 2020 Cited by 1 | Viewed by 1319

Abstract Diseases and complications related to catheter materials are severe problems in biomedical material applications, increasing the infection risk and medical expenses. Therefore, there is an enormous demand for catheter materials with antibacterial and antifouling properties. Considering this, in this work, we developed an [...] Read more. (This article belongs to the Section Membrane Physics and Theory)

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# Open Access Perspective

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# Boron Nitride Nanotube (BNNT) Membranes for Energy and Environmental Applications

by & Numan Yanar, & Eunmok Yang, Hosik Park, Moon Son and Heechul Choi Membranes 2020, 10(12), 430; https://doi.org/10.3390/membranes10120430 - 16 Dec 2020 Cited by 13 | Viewed by 3919

Abstract Owing to their extraordinary thermal, mechanical, optical, and electrical properties, boron nitride nanotubes (BNNTs) have been attracting considerable attention in various scientific fields, making it more promising as a nanomaterial compared to other nanotubes. Recent studies reported that BNNTs exhibit better properties than [...] Read more. (This article belongs to the Special Issue Nanotechnology in Engineered Membranes)

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# Acidic Gases Solubility in Bis(2-Ethylhexyl) Sulfosuccinate Based Ionic Liquids Using the Predictive Thermodynamic Model

by 🔇 Amal Mechergui, 🔇 Alsu I. Akhmetshina, 😩 Olga V. Kazarina, 🧏 Maria E. Atlaskina, 🔇 Anton N. Petukhov and 🔇 Ilya V. Vorotyntsev

Membranes 2020, 10(12), 429; https://doi.org/10.3390/membranes10120429 - 16 Dec 2020 Cited by 2 | Viewed by 1178

Abstract To properly design ionic liquids (ILs) adopted for gases separation uses, a knowledge of ILs thermodynamic properties as well their solubilities with the gases is essential. In the present article, solubilities of  $CO_2$  and  $H_2S$  in bis(2-Ethylhexyl)sulfosuccinate based ILs were [...] Read more.

(This article belongs to the Special Issue Ionic Liquid-based Materials for Membrane Processes)

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# Impact of Inorganic Ions and Organic Matter on the Removal of Trace Organic Contaminants by Combined Direct Contact Membrane Distillation–UV Photolysis

# by Parbab Tufail, William E. Price and S Faisal I. Hai Membranes 2020, 10(12), 428; https://doi.org/10.3390/membranes10120428 - 15 Dec 2020 Cited by 6 | Viewed by 1984

Abstract This study investigated the degradation of five trace organic contaminants (TrOCs) by integrated direct contact membrane distillation (DCMD) and UV photolysis. Specifically, the influence of inorganic ions including halide, nitrate, and carbonate on the performance of the DCMD–UV process was evaluated. TrOC degradation [...] Read more. (This article belongs to the Special Issue CESE-2019: Applications of Membranes for Sustainability)

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# Open Access Editorial

# Special Issue "Advances in Artificial and Biological Membranes: Mechanisms of Ionic Sensitivity, Ion-Sensor Designs, and Applications for Ion Measurement"

by 😫 Andrzej Lewenstam and 😫 Krzysztof Dołowy

Membranes 2020, 10(12), 427; https://doi.org/10.3390/membranes10120427 - 15 Dec 2020 Cited by 3 | Viewed by 1319

Abstract Ion sensors, conventionally known as ion-selective membrane electrodes, were devised 100 years ago with the invention of a pH electrode with a glass membrane (in 1906 Cremer, in 1909 Haber and Klemensiewicz) [...] Full article (This article belongs to the Special Issue Advances in Artificial and Biological Membranes: Mechanisms of Ionic Sensitivity, Ion-Sensor Designs and Applications for Ions Measurement)

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# Synergistic Effect of 2-Acrylamido-2-methyl-1-propanesulfonic Acid on the Enhanced Conductivity for Fuel Cell at Low Temperature

by 😫 Murli Manohar and 😫 Dukjoon Kim

Membranes 2020, 10(12), 426; https://doi.org/10.3390/membranes10120426 - 15 Dec 2020 Cited by 5 | Viewed by 1787

Abstract This present work focused on the aromatic polymer (poly (1,4-phenylene ether-ether-sulfone); SPEES) interconnected/ cross-linked with the aliphatic monomer (2-acrylamido-2-methyl-1-propanesulfonic; AMPS) with the sulfonic group to enhance the conductivity and make it flexible with aliphatic chain of AMPS. Surprisingly, it produced higher conductivity than [...] Read more. (This article belongs to the Special Issue Polymer Electrolyte Membranes)

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# Open Access Review

# Molecular Choreography and Structure of $Ca^{2+}$ Release-Activated $Ca^{2+}$ (CRAC) and $K_{Ca2+}$ Channels and Their Relevance in Disease with Special Focus on Cancer

by & Adéla Tiffner and & Isabella Derler Membranes 2020, 10(12), 425; https://doi.org/10.3390/membranes10120425 - 15 Dec 2020 Cited by 7 | Viewed by 3159

Abstract  $Ca^{2+}$  ions play a variety of roles in the human body as well as within a single cell. Cellular  $Ca^{2+}$  signal transduction processes are governed by  $Ca^{2+}$  sensing and  $Ca^{2+}$  transporting proteins. In this review, we discuss the Ca [...] Read more. (This article belongs to the Special Issue Membrane Channels and Transporters)

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# Erratum: Melnikov, S.; et al. Water Splitting and Transport of lons in Electromembrane System with Bilayer Ion-Exchange Membrane. *Membranes* 2020, *10*, 346

by Stanislav Melnikov, Denis Bondarev, Elena Nosova, Ekaterina Melnikova and Victor Zabolotskiy Membranes 2020, 10(12), 424; https://doi.org/10.3390/membranes10120424 - 15 Dec 2020 Cited by 6 | Viewed by 814

Abstract Due to an error during production, Equations (10), (13)–(20), (23), (24) were unreadable in the published paper [...] Full article

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# In Vivo Comparative Evaluation of Biocompatibility and Biodegradation of Bovine and Porcine Collagen Membranes

by 🞗 Abdu Mansur Dacache Neto, 🤱 Suelen Cristina Sartoretto, 😵 Isabelle Martins Duarte,

🙎 Rodrigo Figueiredo de Brito Resende, 😵 Adriana Terezinha Neves Novellino Alves, 🏟 Carlos Fernando de Almeida Barros Mourão, 😤 Jose Calasans-Maia, 😤 Pietro Montemezzi,

Silson Coutinho Tristão and S Mônica Diuana Calasans-Maia

Membranes 2020, 10(12), 423; https://doi.org/10.3390/membranes10120423 - 15 Dec 2020

Cited by 12 | Viewed by 1887

Abstract Mechanical barriers prevent the invasion of the surrounding soft tissues within the bone defects. This concept is known as Guided Bone Regeneration (GBR). The knowledge about the local tissue reaction and the time of degradation of absorbable membranes favors the correct clinical indication. [...] Read more. (This article belongs to the Special Issue Biocompatible Membranes)

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# Gas Permeation Model of Mixed-Matrix Membranes with Embedded Impermeable Cuboid Nanoparticles

by & Haoyu Wu, Maryam Zamanian, & Boguslaw Kruczek and M Jules Thibault Membranes 2020, 10(12), 422; https://doi.org/10.3390/membranes10120422 - 15 Dec 2020



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# Open Access Article

# LCA of a Membrane Bioreactor Compared to Activated Sludge System for Municipal Wastewater Treatment

#### by & Dimitra C. Banti, & Michail Tsangas, & Petros Samaras and & Antonis Zorpas Membranes 2020, 10(12), 421; https://doi.org/10.3390/membranes10120421 - 14 Dec 2020

Cited by 25 | Viewed by 3146

Abstract Membrane bioreactor (MBR) systems are connected to several advantages compared to the conventional activated sludge (CAS) units. This work aims to the examination of the life cycle environmental impact of an MBR against a CAS unit when treating municipal wastewater with similar influent [...] Read more.

(This article belongs to the Special Issue New Perspectives on Membrane Bioreactors)

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# Effect of PAC on the Behavior of Dynamic Membrane Bioreactor Filtration Layer Based on the Analysis of Mixed Liquid Properties and Model Fitting

by Chunyan Huang, Hongju Liu, Shujuan Meng and Dawei Liang Membranes 2020, 10(12), 420; https://doi.org/10.3390/membranes10120420 - 14 Dec 2020 Cited by 4 | Viewed by 1449

Abstract Recently, dynamic membrane bioreactor (DMBR) has gradually gained the interest of researchers for the development of membrane technology. In this paper, we set up parallel experiments to investigate the effect of powder activated carbon (PAC) on organic matter removal, transmembrane pressure, and filter [...] Read more.

(This article belongs to the Special Issue Membranes for Environmental Applications 2020)

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# Aging of Thin-Film Composite Membranes Based on Crosslinked PTMSP/PEI Loaded with Highly Porous Carbon Nanoparticles of Infrared Pyrolyzed Polyacrylonitrile

by 😮 Danila Bakhtin, 😩 Stepan Bazhenov, 😢 Victoria Polevaya, 🥀 Evgenia Grushevenko, 😤 Sergey Makaev, 😵 Galina Karpacheva, 🕐 Vladimir Volkov and 😩 Alexey Volkov Membranes 2020, 10(12), 419; https://doi.org/10.3390/membranes10120419 - 14 Dec 2020

Cited by 6 | Viewed by 1886

Abstract The mitigation of the physical aging of thin-film composite (TFC) poly[1-trimethylsilyI-1-propyne] (PTMSP) membranes was studied via the simultaneous application of a polymer-selective layer crosslinking and mixed-matrix membrane approach. For the first time, a recently developed highly porous activated carbon material (infrared (IR) pyrolyzed [...] Read more. (This article belongs to the Special Issue Progress in Manufacturing and Applications of Composite Membranes)

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#### Improvement of Component Flux Estimating Model for Pervaporation Processes

by 😣 Botond Szilagyi and 쪻 Andras Jozsef Toth

Membranes 2020, 10(12), 418; https://doi.org/10.3390/membranes10120418 - 13 Dec 2020 Cited by 4 | Viewed by 1764

Abstract Separating non-ideal mixtures by pervaporation (hence PV) is a competitive alternative to most traditional methods, such as distillation, which are based on the vapour-liquid equilibrium (VLE). It must be said, in many cases, accurate VLE data are already well known in the literature. [...] Read more.

(This article belongs to the Special Issue Membrane Separations, Membrane Filtrations, Pervaporation, and Modeling of Membrane Separation)

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# Membrane Biofouling Control by Surface Modification of Quaternary Ammonium Compound Using Atom-Transfer Radical-Polymerization Method with Silica Nanoparticle as Interlayer

by & Lehui Ren, & Meng Ping and & Xingran Zhang Membranes 2020, 10(12), 417; https://doi.org/10.3390/membranes10120417 - 11 Dec 2020 Cited by 6 | Viewed by 1727

Abstract A facile approach to fabricate antibiofouling membrane was developed by grafting quaternary ammonium compounds (QACs) onto polyvinylidene fluoride (PVDF) membrane via surface-initiated activators regenerated by electron transfer atom-transfer radical-polymerization (ARGET ATRP) method. During the modification process, a hydrophilic silica nanoparticle layer was also [...] Read more.

(This article belongs to the Section Membrane Surfaces and Interfaces)

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# Graphene Oxide Incorporated Polysulfone Substrate for Flat Sheet Thin Film Nanocomposite Pressure Retarded Osmosis Membrane

by Siti Nur Amirah Idris, Nora Jullok, Woei Jye Lau, Hui Lin Ong and Cheng-Di Dong Membranes 2020, 10(12), 416; https://doi.org/10.3390/membranes10120416 - 11 Dec 2020 Cited by 14 | Viewed by 2474

Abstract This study focuses on the development of flat sheet thin film nanocomposite (TFN) pressure retarded osmosis (PRO) membranes for the enhancement of osmotic power generation by the incorporation of laboratory-synthesised graphene oxide (GO) into the polysulfone (PSf) polymer matrix. A series of membranes [...] Read more.

(This article belongs to the Special Issue Pervaporation, Vapour Permeation and Membrane Distillation: From Membrane Fabrication to Application)



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Membranes 2020, 10(12), 415; https://doi.org/10.3390/membranes10120415 - 11 Dec 2020 Cited by 7 | Viewed by 2400

Abstract Biofouling is a major concern for numerous reverse osmosis membrane systems. UV pretreatment of the feed stream showed promising results but is still not an established technology as it does not maintain a residual effect. By conducting accelerated biofouling experiments in this study, [...] Read more.

(This article belongs to the Special Issue Enhancing the Efficiency of Membrane Processes for Water Treatment)

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# Development of Chitosan/Starch-Based Forward Osmosis Water Filtration Bags for Emergency Water Supply

by Saiful Saiful, Amurisa Ajrina, Yusuf Wibisono and Amurisa Amurisa Ajrina, Wibisono and Amurisa Membranes 2020, 10(12), 414; https://doi.org/10.3390/membranes10120414 - 11 Dec 2020 Cited by 9 | Viewed by 2667

Abstract A forward osmosis (FO) membrane was developed from a mixture of chitosan and *Dioscorea hispida* starch, cross-linked using glutaraldehyde. The cross-linked chitosan/starch membrane was revealed to have high mechanical properties with an asymmetric structure. The prepared membrane's performance was investigated as an FO [...] Read more. (This article belongs to the Special Issue Membranes for Water and Wastewater Treatment)

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Superglassy Polymers to Treat Natural Gas by Hybrid Membrane/Amine Processes: Can Fillers Help?

by 😤 Ahmed W. Ameen, 🌚 Peter M. Budd and 🚳 Patricia Gorgojo

Membranes 2020, 10(12), 413; https://doi.org/10.3390/membranes10120413 - 10 Dec 2020 Cited by 7 | Viewed by 2496

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Abstract Superglassy polymers have emerged as potential membrane materials for several gas separation applications, including acid gas removal from natural gas. Despite the superior performance shown at laboratory scale, their use at industrial scale is hampered by their large drop in gas permeability over [...] Read more.

(This article belongs to the Special Issue Mixed Matrix Membranes II. From Lab Scale towards Application)

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Characterization and Performance of LbL-Coated Multibore Membranes: Zeta Potential, MWCO, Permeability and Sulfate Rejection

by Saskia Dillmann, Shambhavi Arvind Kaushik, SJakob Stumme and Mathias Ernst Membranes 2020, 10(12), 412; https://doi.org/10.3390/membranes10120412 - 10 Dec 2020 Cited by 6 I Viewed by 2202

Abstract The characterization of membranes is suitable to investigate changes in the membrane properties caused by Layer-by-Layer (LbL) modification. Besides permeability, rejection, and molecular-weight cut-off (MWCO), which give information about the modification of the separation behaviour of the membrane, the zeta potential is capable [...] Read more. (This article belongs to the Special Issue Enhancing the Efficiency of Membrane Processes for Water Treatment)

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Some Critical Remarks about Mathematical Model Used for the Description of Transport Kinetics in Polymer Inclusion Membrane Systems

by 😫 Piotr Szczepański

Membranes 2020, 10(12), 411; https://doi.org/10.3390/membranes10120411 - 10 Dec 2020 Cited by 7 | Viewed by 1488

Abstract Two kinetic models which are applied for the description of metal ion transport in polymer inclusion membrane (PIM) systems are presented and compared. The models were fitted to the real experimental data of Zn(II), Cd(II), Cu(II), and Pb(II) simultaneous transport through PIM with [...] Read more. (This article belongs to the Special Issue Polymer Inclusion Membranes)

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# Effective H<sub>2</sub> Separation through Electroless Pore-Plated Pd Membranes Containing Graphite Lead Barriers

by & David Martinez-Diaz, & Raúl Sanz, Alicia Carrero, & José Antonio Calles and & David Alique Membranes 2020, 10(12), 410; https://doi.org/10.3390/membranes10120410 - 10 Dec 2020 Cited by 4 | Viewed by 1390

Abstract Hydrogen promotion as a clean energy vector could provide an efficient strategy for realizing real decarbonization of the current energy system. Purification steps are usually required in most H<sub>2</sub>-production processes, providing the use of Pd-based membranes, particularly those supported on porous [...] Read more.

(This article belongs to the Special Issue Membrane Reactors for Process Intensification: Recent Advances and Key Applications)

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The Acid–Base Flow Battery: Sustainable Energy Storage via Reversible Water Dissociation with Bipolar Membranes

by 🙎 Ragne Pärnamäe, 🚯 Luigi Gurreri, 🧟 Jan Post, 🧟 Willem Johannes van Egmond, 🙎 Andrea Culcasi.

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Removal of Different Dye Solutions: A Comparison Study Using a Polyamide NF Membrane by Asunción María Hidalgo, A Gerardo León, A María Gómez, María Dolores Murcia, Elisa Gómez and

Sosé Antonio Macario Membranes 2020, 10(12), 408; https://doi.org/10.3390/membranes10120408 - 10 Dec 2020

Cited by 19 | Viewed by 1759

Abstract The removal of organic dyes in aquatic media is, nowadays, a very pressing environmental problem. These dyes usually come from industries, such as textiles, food, and pharmaceuticals, among others, and their harm is produced by preventing the penetration of solar radiation in the [...] Read more.

 $(This \ article \ belongs \ to \ the \ Special \ Issue \ Membranes \ for \ Water \ and \ Wastewater \ Treatment)$ 

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#### Discussion on Water Condensation in Membrane Pores during CO<sub>2</sub> Absorption at High Temperature

by 🎡 Zhe Phak Chan, 😢 Lin Li, 😢 Guodong Kang, 😩 Norfaizah Ab Manan, 🙁 Yiming Cao and 😩 Tonghua Wang Membranes 2020, 10(12), 407; https://doi.org/10.3390/membranes10120407 - 09 Dec 2020

Cited by 1 | Viewed by 1377

Abstract Water condensation is a possible cause of membrane wetting in the operation of membrane contactors, especially under high-temperature conditions. In this study, water condensation in pores of polytetrafluoroethylene (PTFE) hollow fiber membranes was investigated during high-pressure CO<sub>2</sub> absorption around 70 °C. It [...] Read more.

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#### Open Access Review

# Lysosomal Exocytosis: The Extracellular Role of an Intracellular Organelle

by 🚷 Brunella Tancini, 🚷 Sandra Buratta, 🚷 Federica Delo, 🊷 Krizia Sagini, 🏖 Elisabetta Chiaradia, 😰 Roberto Maria Pellegrino, 🍘 Carla Emiliani and 🕿 Lorena Urbanelli Membranes 2020, 10(12), 406; https://doi.org/10.3390/membranes10120406 - 09 Dec 2020

Cited by 37 | Viewed by 6988

Abstract Lysosomes are acidic cell compartments containing a large set of hydrolytic enzymes. These lysosomal hydrolases degrade proteins, lipids, polysaccharides, and nucleic acids into their constituents. Materials to be degraded can reach lysosomes either from inside the cell, by autophagy, or from outside the [...] Read more.

(This article belongs to the Special Issue Membrane Transport and Cytoskeleton Dynamics)

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# Effect of Water and Organic Pollutant in CO<sub>2</sub>/CH<sub>4</sub> Separation Using Hydrophilic and Hydrophobic Composite Membranes

by 🚯 Clara Casado-Coterillo, 😫 Aurora Garea and 🐌 Ángel Irabien

Membranes 2020, 10(12), 405; https://doi.org/10.3390/membranes10120405 - 08 Dec 2020 Cited by 8 | Viewed by 1546

Abstract Membrane technology is a simple and energy-conservative separation option that is considered to be a green alternative for CO<sub>2</sub> capture processes. However, commercially available membranes still face challenges regarding water and chemical resistance. In this study, the effect of water and organic [...] Read more.

(This article belongs to the Special Issue Polymer Membranes for Gas Separation)

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# Spraying of Ultrathin Isoporous Block Copolymer Membranes—A Story about Challenges and Limitations

by & Thomas Bucher, & Juliana Isabel Clodt, & Clarissa Abetz, & Barbara Bajer and & Volkan Filiz Membranes 2020, 10(12), 404; https://doi.org/10.3390/membranes10120404 - 07 Dec 2020 Cited by 3 | Viewed by 1822

Abstract Isoporous membranes can be prepared by a combination of self-assembly of amphiphilic block copolymers and the nonsolvent induced phase separation process. As the general doctor-blade technique suffers from high consumption of expensive block copolymer, other methods to reduce its concentration in the casting [...] Read more.

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A New Design of Tubular Ceramic Membrane Module for Oily Water Treatment: Multiphase Flow Behavior and Performance Evaluation

by 🙎 Guilherme L. Oliveira Neto, 😤 Nívea G. N. Oliveira, 😤 João M. P. Q. Delgado, 😤 Lucas P. C. Nascimento, 😤 Ricardo S. Gomez, 😤 Adriano S. Cabral, 😤 Daniel C. M. Cavalcante, 😤 Vansostenes A. M. Miranda, 😵 Severino R. Farias Neto and 🧟 Antonio G. B. Lima

Membranes 2020, 10(12), 403; https://doi.org/10.3390/membranes10120403 - 07 Dec 2020 Cited by 3 | Viewed by 1437

Abstract Petroleum has been extracted from oil reservoirs using different techniques. This activity is accompanied for a large amount of water and sometimes mixed with gas. This produced water has a high oil concentration and other toxic chemical compounds, thus, it must be treated [...] Read more.

(This article belongs to the Special Issue Transport Processes in Membranes: From Theory and Modeling to Industrial and



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# A "Graft to" Electrospun Zwitterionic Bilayer Membrane for the Separation of Hydraulic Fracturing-Produced Water via Membrane Distillation

by ② Yu-Hsuan Chiao, ② Micah Belle Marie Yap Ang, ③ Yu-Xi Huang, ③ Sandrina Svetlana DePaz, ③ Yung Chang, ③ Jorge Almodovar and ③ S. Ranil Wickramasinghe Membranes 2020, 10(12), 402; https://doi.org/10.3390/membranes10120402 - 07 Dec 2020

Cited by 18 | Viewed by 1870

Abstract Simultaneous fouling and pore wetting of the membrane during membrane distillation (MD) is a major concern. In this work, an electrospun bilayer membrane for enhancing fouling and wetting resistance has been developed for treating hydraulic fractureproduced water (PW) by MD. These PWs can [...] Read more.

(This article belongs to the Special Issue Pervaporation, Vapour Permeation and Membrane Distillation: From Membrane Fabrication to Application)

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#### Rapid Surface Modification of Ultrafiltration Membranes for Enhanced Antifouling Properties

by 😵 Noresah Said, 😵 Ying Siew Khoo, 🍘 Woei Jye Lau, 😤 Mehmet Gürsoy, 😵 Mustafa Karaman, 😵 Teo Ming Ting, 😵 Ebrahim Abouzari-Lotf and 🔇 Ahmad Fauzi Ismail

Membranes 2020, 10(12), 401; https://doi.org/10.3390/membranes10120401 - 07 Dec 2020 Cited by 12 | Viewed by 2229

Abstract In this work, several ultrafiltration (UF) membranes with enhanced antifouling properties were fabricated using a rapid and green surface modification method that was based on the plasma-enhanced chemical vapor deposition (PECVD). Two types of hydrophilic monomers—acrylic acid (AA) and 2-hydroxyethyl methacrylate (HEMA) were, [...] Read more. (This article belongs to the Special Issue Ultrafiltration Polymeric Membranes)

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# Gas Transport in Glassy Polymers

by 😣 Maria Grazia De Angelis and 🌎 Giulio C. Sarti

Membranes 2020, 10(12), 400; https://doi.org/10.3390/membranes10120400 - 07 Dec 2020 Cited by 4 | Viewed by 1105

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Abstract This Special Issue of Membranes provides an updated and comprehensive overview of the state of fundamental knowledge on the fluid sorption and transport in glassy polymers, combining original experimental and modeling works, as well as reviews, prepared by renowned experts [...] Full article

(This article belongs to the Special Issue Gas Transport in Glassy Polymers)

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Treatment of Electroplating Wastewater Using NF pH-Stable Membranes: Characterization and Application

by 🙁 Ignacio Hegoburu, 🙁 Karina Listiarini Zedda and 🥾 Svetlozar Velizarov

Membranes 2020, 10(12), 399; https://doi.org/10.3390/membranes10120399 - 06 Dec 2020 Cited by 5 | Viewed by 1824

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Abstract Industrial adoption of nanofiltration (NF) for treatment of low-pH wastewater is hindered by the limited membrane lifetime at strongly acidic conditions. In this study, the electroplating wastewater (EPWW) filtration performance of a novel pH-stable NF membrane is compared against a commercial NF membrane [...] Read more.

(This article belongs to the Special Issue Membrane-Assisted (Bio)Chemical Process and Technology)

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Performance of Layer-by-Layer-Modified Multibore<sup>®</sup> Ultrafiltration Capillary Membranes for Salt Retention and Removal of Antibiotic Resistance Genes

by 🚏 Robert Niestroj-Pahl, 🆓 Lara Stelmaszyk, 😵 Ibrahim M. A. ElSherbiny, 🕄 Hussein Abuelgasim, 😵 Michaela Krug, 😵 Christian Staaks, 😤 Greta Birkholz, 😤 Harald Horn, 🔗 Tian Li, 😤 Bingzhi Dong, 🔗 Lars Dähne, 🔗 Andreas Tiehm and 😵 Stefan Panglisch

Membranes 2020, 10(12), 398; https://doi.org/10.3390/membranes10120398 - 06 Dec 2020 Cited by 6 | Viewed by 1885

Abstract Polyether sulfone Multibore<sup>®</sup> ultrafiltration membranes were modified using polyelectrolyte multilayers via the layer-by-layer (LbL) technique in order to increase their rejection capabilities towards salts and antibiotic resistance genes. The modified capillary membranes were characterized to exhibit a molecular weight cut-off (at 90% [...] Read more.

(This article belongs to the Special Issue Enhancing the Efficiency of Membrane Processes for Water Treatment)

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Membrane Bioreactor Technology: The Effect of Membrane Filtration on Biogas Potential of the Excess Sludge

by <sup>(2)</sup> Magdalena Zielińska, <sup>(2)</sup> Katarzyna Bernat and <sup>(2)</sup> Wioleta Mikucka Membranes 2020, 10(12), 397; https://doi.org/10.3390/membranes10120397 - 06 Dec 2020

Cited by 4 | Viewed by 1330

Abstract Although the membrane bioreactor technology is gaining increasing interest because of high efficiency of wastewater treatment and reuse, data on the anaerobic transformations of retentate are scarce and divergent. The effects of transmembrane pressure (TMP) in microfiltration (MF) and ultrafiltration (UF) on the [...] Read more. (This article belongs to the Section Membrane Applications)

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Interactions between Beta-2-Glycoprotein-1 and Phospholipid Bilaver—A Molecular Dynamic

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# Open Access Review

# Functions of Ionic Liquids in Preparing Membranes for Liquid Separations: A Review

by 😮 Dayuan Zheng, 😵 Dan Hua, 😵 Yiping Hong, 😵 Abdul-Rauf Ibrahim, 😵 Ayan Yao, 😵 Junyang Pan and

Membranes 2020, 10(12), 395; https://doi.org/10.3390/membranes10120395 - 05 Dec 2020 Cited by 14 | Viewed by 2767

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# Mixed-Matrix Membranes Comprising of Polysulfone and Porous UiO-66, Zeolite 4A, and Their Combination: Preparation, Removal of Humic Acid, and Antifouling Properties

by 😫 Tanzila Anjum, 🏶 Rahma Tamime and 😫 Asim Laeeq Khan

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Abstract High-performance Mixed-Matrix Membranes (MMMs) comprising of two kinds of porous fillers UiO-66 and Zeolite 4A and their combination were fabricated with polysulfone (PSf) polymer matrix. For the very first time, UiO-66 and Zeolite 4A were jointly used as nanofillers in MMMs with the objective [...] Read more.

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Abstract An ionic imprinted membrane (IIM) was synthesized using sulfonated polyeugenol, derived from eugenol, as its functional polymer and polysulfone as its base membrane for the selective adsorption of Au(III). This study aims to determine the adsorption of Au(III) metal ions using IIM compared [...] Read more. (This article belongs to the Section Polymeric Membranes) MDPI

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Abstract The polymer electrolyte based on Dx:Cs:Mg(CH<sub>3</sub>COO)<sub>2</sub>:Ni with three different glycerol concentrations have been prepared. The impedance study has verified that the electrolyte with 42 wt.% of glycerol (A3) has the highest ionic conductivity of 7.71 × 10<sup>-6</sup> S [...] Read more.

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by 😮 Takashi Tachibana, 😵 Tomohisa Yoshioka, 😵 Keizo Nakagawa, 😵 Takuji Shintani, 😵 Eiji Kamio and

Membranes 2020, 10(12), 388; https://doi.org/10.3390/membranes10120388 - 01 Dec 2020 Cited by 3 | Viewed by 1518

Abstract Methyl gallate (MG) and ethyl ferulate (EF) with a benzene ring were separately used as aromatic organic chelating ligands (aOCLs) to prepare two versions of TiO<sub>2</sub>-ZrO<sub>2</sub>-aOCL composite sols via hydrolysis and polycondensation reactions with titanium(IV) isopropoxide (Ti(OC<sub>3</sub>H [...] Read more.

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by 😮 Antonio Montes-Rojas, 🕲 Marlen Ramírez-Orizaga, 😢 Jesús Gerardo Ávila-Rodriguez and

Membranes 2020, 10(12), 387; https://doi.org/10.3390/membranes10120387 - 30 Nov 2020 Viewed by 1088

Abstract One of the intended applications for the modification of ion exchange membranes with polyaniline (PAni) is to use it as a matrix to include chemical species that confer a special property such as resistance to fouling or ion selectivity. In particular, the inclusion [...] Read more.

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Preparation of a Hybrid Membrane from Whey Protein Fibrils and Activated Carbon to Remove Mercury and Chromium from Water

by 🚯 Laura Cristina Ramírez-Rodríguez, 😩 Luis Eduardo Díaz Barrera, 😩 María Ximena Quintanilla-Carvajal, S Didilia Ileana Mendoza-Castillo, S Adrián Bonilla-Petriciolet and Carlos Jiménez-Junca Membranes 2020, 10(12), 386; https://doi.org/10.3390/membranes10120386 - 30 Nov 2020

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Abstract Water contamination by mercury and chromium has a direct effect in human health. A promising technology to remove heavy metals by membrane filtration is the use of hybrid membranes produced with whey protein fibrils (WPF) and activated carbon (AC). In this study, the [...] Read more.

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# by 😵 Ilona Pyszka and 😫 Elzbieta Radzyminska-Lenarcik

Membranes 2020, 10(12), 385; https://doi.org/10.3390/membranes10120385 - 30 Nov 2020 Cited by 11 | Viewed by 1480

Abstract The new polymer inclusion membrane (PIM) with ethylenediamine-bis-acetylacetone (EDAB-acac) matrix was used for the separation of Zn(II) solutions containing non-ferrous metal ions (Co(II), Ni(II) Cu(II), Cd(II)). The effective conditions for carrying out transport studies by PIMs were determined on the basis of solvent [...] Read more.

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# Microstructural Investigation and On-Site Repair of Thin Pd-Ag Alloy Membranes

by 😵 Yuyu Ma, 📚 Chunhua Tang, 😵 Feng Bao, இ Wei Shao, இ Tianying Xu, 😤 Hui Li and 😤 Hengyong Xu Membranes 2020, 10(12), 384; https://doi.org/10.3390/membranes10120384 - 30 Nov 2020 Cited by 7 | Viewed by 1295

Abstract Pd membranes act in an important role in H2 purification and H2 production in membrane reactors. Pd-Ag alloy membranes fabricated by consecutive electroless- and electroplating process on alumina tubes exhibited good stability under stringent heating/cooling cycles at a ramp rate of 10 K/min, [...] Read more.

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#### Abstract

Zinc recovery from spent pickling acids (SPAs) can play an important role in achieving a circular economy in the galvanizing industry. This work evaluates the scale-up of membrane-based solvent extraction technology aimed at the selective separation of zinc from industrial SPAs as a purification step prior to zinc electrowinning (EW). The experiments were carried out at a pilot scale treating SPAs batches of 57 to 91 L in a non-dispersive solvent extraction (NDSX) configuration that simultaneously performed the extraction and backextraction steps. The pilot plant was equipped with four hollow fiber contactors and 80 m<sup>2</sup> of total membrane area, which was approximately 30 times higher than previous bench-scale studies. Tributylphosphate diluted in Shellsol D70 and tap water were used as organic and stripping agents, respectively. Starting with SPAs with high Zn (71.7 ± 4.3 g·L<sup>-1</sup>) and Fe (82.9 ± 5.0 g·L<sup>-1</sup>) content, the NDSX process achieved a stripping phase with 55.7 g Zn·L<sup>-1</sup> and only 3.2 g Fe·L<sup>-1</sup>. Other minor metals were not transferred, providing the purified zinc stripping with better quality for the next EW step. A series of five consecutive pilot-scale experiments showed the reproducibility of results, which is an indicator of the stability of the organic extracted from previous laboratory-scale experiments, allowing us to obtain the design parameter that will enable the leap to the industrial scale. Therefore, the results herein presented demonstrate the NDSX technology in an industrially relevant environment equivalent to TRL 6, which is an essential progress to increase zinc metal resources in the galvanizing sector.

Keywords: hot-dip galvanizing; zinc recovery; spent pickling acid; hollow-fiber membrane contactor; pilot plant; non-dispersive solvent extraction; tributyl phosphate; secondary zinc

#### 1. Introduction

The hot-dip galvanizing (HDG) process is one of the most common methods to prevent steel corrosion by providing steel components with a protective zinc coating [1]. Acid pickling is one of the preliminary HDG stages aimed at removing impurities such as oxides from the steel surface [2,3]. It is also used for dezincing of tools and nonconforming galvanized components. At present, HCl is the most commonly used acid for carbon steel pickling, since it provides optimal surface quality and fast pickling [4,5,6].

However, the management of the spent pickling acids (SPAs) constitutes one of the environmental challenges for the galvanizing industry. SPAs after steel pickling in the HDG plants consist of free HCI, iron, zinc, and chloride ions [7]. The present study concerns residual HCI SPAs from the pickling and dezincing of steel items in HDG plants that contain high concentrations of zinc and iron. A freshly prepared pickling bath typically contains 12–16% HCI, although this concentration is progressively reduced along their use [8]. The pickling bath is considered spent when the acid concentration decreases between 75 and 85% of its initial value, and the metals concentration in solution increases to 150–250 g·L<sup>-1</sup> [6]. It is also worth mentioning that most general galvanizers apply the Kleingarn curve that consists of the periodic replacement of only one part of the pickling bath. This practice allows increasing the pickling rate, reducing the generation of SPAs [4].

Table 1 compiles literature information on the composition of HCI-based SPAs. The iron concentration ranges from 8 to 204 g·L<sup>-1</sup>, the average being 101.6 g·L<sup>-1</sup>. Iron is mostly present as  $Fe^{2+}$ . Zinc concentration varies in a similar range, the average being 95.7 g·L<sup>-1</sup>. The wide variety of zinc and iron concentration is the result of the diverse practices applied by galvanizers, e.g., the remaining acid can be used for stripping the zinc layer from rejected galvanized steel products [9]. In addition to zinc and iron, SPAs may contain a low concentration of other metals such as manganese, lead, aluminum, chromium, cadmium, nickel, copper, and cobalt [10]. Moreover, SPAs contain surfactants, inhibitors, and stabilizers that may hinder the recovery of acid and/or metals [7].





The conventional SPAs treatment consists of residual acid neutralization with lime or some other cheap alkaline agents [7], which is followed by the disposal of the waste metallic sludge in landfills. Solidification/stabilization can be carried out before the disposal to make the contaminants as immobile as possible. Although neutralization is the most economic method, it presents some disadvantages such as excessive sludge production [26], slow metal precipitation kinetics, inefficient metal removal due to poor setting of metal precipitates, leaching of heavy metals to groundwater, and the problem of landfilling [18].

The conventional treatment of SPAs is being substituted by innovative alternatives that can have different objectives: acid recovery, metals recovery, and the conversion of the waste into other products [7]. In this work, we estimate that between 3.5 and 4.9% of the zinc used in the molten zinc bath of the HDG process is lost through the SPAs. Therefore, SPAs valorization could be an additional source of secondary zinc with potential economic benefits [27]. Zinc recovery from SPAs makes sense, since the production of special high-grade (SHG) zinc is a very energy-intensive process with a primary energy demand of 37,500 MJ and a climate change impact of 2600 kg CO<sub>2</sub>- eq. per ton of primary zinc produced [28]. In addition, the zinc price was positioned in November 2020 at around 2200€ per ton, ranging during the last three years between 1650 and 2950€ per ton, thus hightlingiting the industrial interest on its recovery [29].

Technologies that enable acid recovery are spray roasting, evaporation, diffusion dialysis (DD), membrane distillation (MD), electrodialysis (ED), and membrane electrolysis (ME) [30]. Methods enabling both metal and acid recovery are ion exchange (IE)/retardation, crystallization and solvent extraction (SX) [6]. DD, ED, evaporation, precipitation, and spray roasting have been industrially implemented. Moreover, IE has been applied to recover HCl by the Metsep process. Quimigal in Portugal employed SX for the regeneration of SPAs based on the Modified Zincex Process (MZP) that allows the production of SHG zinc. SHG production from secondary sources of zinc, such as Waelz oxides, has been developed using the Zincex Process and MZP by Técnicas Reunidas since 1976, and MZP is still carried out today [31].

Table 2 summarizes previous works focused on the zinc recovery from SPAs at laboratory scale using SX and membrane-based solvent extraction (MBSX), including the

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#### Abstract

Glass manufacturing is an energy-intensive process in which oxy-fuel combustion can offer advantages over the traditional air-blown approach. Examples include the reduction of NO<sub>x</sub> and particulate emissions, improved furnace operations and enhanced heat transfer. This paper presents a one-dimensional mathematical model solving mass, momentum and energy balances for a planar oxygen transport membrane module. The main modelling parameters describing the surface oxygen kinetics and the microstructure morphology of the support are calibrated on experimental data obtained for a 30 µm thick dense  $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3.6}$  (LSCF) membrane layer, supported on a 0.7 mm porous LSCF structure. The model is then used to design and evaluate the performance of an oxygen transport membrane module integrated in a glass melting furnace. Three different oxy-fuel glass furnaces based on oxygen transport membrane and vacuum swing adsorption systems are compared to a reference air-blown unit. The analysis shows that the most efficient membrane-based oxyfuel furnace cuts the energy demand by ~22% as compared to the benchmark air-blown case. A preliminary economic assessment shows that membranes can reduce the overall glass production costs compared to oxyfuel plants based on vacuum swing adsorption technology.

Keywords: oxygen transport membrane; LSCF; perovskite; glass melting; oxy-fuel combustion



#### 1. Introduction

The term glass generically refers to high-viscosity substances, which do not possess enough mobility during solidification. Thus, they do not have sufficient time to form a regular crystal lattice, resulting in an intermediate metastable stage marked by the formation of an amorphous solid, which lacks the geometrical order typical of crystal lattices [1]. Common raw materials for the glass production are silica (SiO<sub>2</sub>), sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), limestone (CaCO<sub>3</sub>) and, dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>). However, the actual composition varies significantly, depending on the glass type. These raw materials are heated in a furnace to high temperatures to form molten glass. The molten glass undergoes further processing to originate the final product [2].

A furnace operates glass melting at very high temperature (>1400–1500 °C) in a confined space surrounded by refractory material. The heat input is generally supplied by burning oil or natural gas in air. Electric booster systems can be installed to supply additional energy to increase the melting capacity. The conventional arrangement of a glass furnace includes a regenerative heat exchanger consisting of two refractory chambers. One chamber is used to store heat from the flue gas, at the same time the other chamber is used to preheat the combustion air. The paths of flue gas and combustion air through the regenerator chambers are interchanged every 15–20 min. The raw material is continuously introduced in the furnace through a feeding port, while molten glass is continuously removed from the furnace [3,4].

The theoretical energy requirement consists of the endothermic heat for the glass reaction, the sensible heat for glass heating and the sensible heat for batch gases (gases from glass reaction). According to the theoretical heat requirement reported by Sardeshpande et al. [5], the soda-lime glass (composed mainly of silicon dioxide (71–75%), sodium oxide (12–16%) and calcium oxide (10–15%)) needs 2671 kJ kg<sup>-1</sup> of glass, if no cullet (i.e., crushed glass ready to be re-melted) is used. On the other hand, the theoretical minimum is 1886 kJ kg<sup>-1</sup> if the furnace operates on 100% cullet. Such a reduction is due to the absence of the reaction heat of the virgin raw material (487 kJ kg<sup>-1</sup>) and the sensible heat of batch gases (298 kJ kg<sup>-1</sup>).

The use of cullet is beneficial since it is cheaper than virgin raw material and allows energy savings. Moreover, the substitution of raw materials by cullet reduces CO<sub>2</sub> process emissions, which are released by the decomposition of carbonates in raw materials. In the industrial practice, external cullet is extensively used only in the container glass and glass wool production [2].

The above mentioned theoretical heat requirements correspond to the lowest thermodynamic limits [5] but even well insulated glass furnaces will be affected by some structural heat losses. In addition, fossil fuel firing is always associated with flue gas production. Although the flue gas heat content can be partly recovered by combustion air and batch preheating, flue gas is released at temperature of at least 200–250 °C. Thus, the minimum achievable energy consumption level is unavoidably greater than the one reported by Sardeshpande et al. [5]. Energy benchmarking for glass furnaces has been thoroughly discussed by Beerkens [6,7,8]. Referring to the most recent paper [6], Figure 1 shows the ranking of the energy efficiency of 168 container glass furnaces, starting from the lowest specific energy consumption (most energy efficient) up to the highest specific energy consumption. The primary energy values are normalized for the case of 50% recycling cullet in the batch and account for the contribution energy required for electric power generation in case of electric boosting and oxygen production. According to the data in Figure 1, the minimum achievable energy consumption level for normal batch would be around 3.8 MJ kg<sup>-1</sup>, which reduces to around 2.5 MJ kg<sup>-1</sup> for 100% cullet.

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#### Abstract

In this study, an electrochemical model was incorporated into a two-phase model using OpenFOAM<sup>®</sup> (London, United Kingdom) to analyze the two-phase flow and electrochemical behaviors in a polymer electrolyte membrane water electrolyzer. The performances of serpentine and parallel designs are compared. The current density and overpotential distribution are analyzed, and the volume fractions of oxygen and hydrogen velocity are studied to verify their influence on the current density. The current density decreases sharply when oxygen accumulates in the porous transport layer. Therefore, the current density increased sharply by 3000 A/m<sup>2</sup> at an operating current density of 10,000 A/m<sup>2</sup>. Maldistribution of the overpotential is also observed. Second, we analyze the behaviors according to the current density. At a low current density, most of the oxygen flows out of the electrolyzer. Therefore, the decrease in performance is low. However, the current density is maldistributed when it is high, which results in decreased performance. The current density increases aburptly by 12,000 A/m<sup>2</sup>. Finally, the performances of the parallel and serpentine channels are analyzed. At a high current density, the performance of the serpentine channel is higher than that of the parallel channel by 0.016 V.

Keywords: polymer electrolyte membrane water electrolyzer; two-phase flow; electrochemical reaction; performance analysis; performance comparison of flow field

#### 1. Introduction

With the increased importance of green energy, many researchers are interested in the use of hydrogen as an energy transport material [1,2]. Due to the fact that the present price of hydrogen in the Republic of Korea is USD 7.17/kg, it is not economical to convert the country's energy base to hydrogen [3]. Therefore, it is necessary to reduce the price of hydrogen through a new technical development. The polymer electrolyte membrane water electrolyzer (PEMWE) has attracted attention as a potential hydrogen production tool. The PEMWE can produce hydrogen with high purity (99%) [4] and operate at high current densities [5]. Therefore, it is suitable for producing large amounts of hydrogen [6]. However, when the current density becomes excessively high, the efficiency of the PEMWE can decrease, since the generated oxygen bubbles disturb the contact between the reactant and catalyst [7,8]. Hence, it is necessary to analyze the phenomenon in the PEMWE and identify a suitable flow field to eliminate the generated oxygen effectively, as well as to uniformly distribute the reactant.

Many researchers have attempted to experimentally analyze the performances of the PEMWE and PEMWE systems. A few researchers performed experiments to examine the reactivity of the water decomposition reaction for different catalysts. They observed the existence of an exchange current density of the hydrogen reduction reaction (HRR) and oxygen oxidation reaction (OOR). For a platinum catalyst, the exchange current densities for the HRR and OOR are reported to be  $1.61 \times 10^{-1}$ – $1.61 \times 10^{-2}$  A/m<sup>2</sup> [9,10] and  $1.61 \times 10^{-9}$ – $1.61 \times 10^{-6}$  A/m<sup>2</sup> [10,11], respectively. When an iridium—platinum catalyst was used for the reaction, the exchange current density of the ORR increased to  $1.61 \times 10^{-4}$  A/m<sup>2</sup> [10,12].

Simultaneously, many researchers have attempted to analyze the influence of two-phase flow on the performance of the PEMWE. A few researchers measured the volume fraction and pressure loss of the water-air flow in capillary tubes [13]. They proposed an empirical equation to calculate the pressure loss using a capillary tube diameter. Other researchers conducted experiments to analyze the two-phase flow in a porous transport layer (PTL) [14]. They verified the relationship between the electrode porosity, bubble diameter of gas, and performance of the PEMWE. The performance increased with the increasing electrode porosity. However, there was no significant effect when the porosity exceeded 0.5. Other researchers have used the transparent cell to observe the two-phase flow in an electrolyzer [15]. They verified that bubbles were converted into slug flow when the superficial gas velocity exceeded  $8 \times 10^{-2}$  m/s for an operating current density of 700 A/m<sup>2</sup>. However, the existing studies observed only the two-phase flow without considering the electrochemical reaction, and it was also insufficient for predicting the performance using the flow field design of the PEMWE.

Numerical analyses have been performed to analyze the two-phase flow of various channel designs. A few researchers observed that the pressure drop increased with the increasing oxygen generation in a parallel flow field [16]. In this study, they assumed that oxygen gas was produced uniformly at the interface between the PTL and membrane, to replicate the electrochemical reaction in the membrane. Other researchers have analyzed the two-phase flow of a circular and interdigitated flow field of the PEMWE to verify the difference in the distribution of the oxygen-water mixture, according to the particle size for this numerical model [17]. They verified that the maximum gas volume fraction increased from 0.4 to 0.45 for a particle diameter of 40 µm. The thermal and electrochemical reactions in the PEMWE have also been analyzed [18]. These studies verified that the local temperature increased by 20.2 °C owing to the oxygen bubble in the PTL at an operating current density of 1500 A/m<sup>2</sup>. Although the relationship between the effort many previous studies, this did not adequately explain the relationship between the two-phase flow in the anode and the electrochemical reaction in the membrane. It is necessary to develop a numerical model to analyze the interaction between the electrochemical reaction between the

In this study, we developed a transient model of PEMWE to analyze the two-phase flow and electrochemical phenomenon of various channel designs. We applied the Euler-Euler two-phase model and the Butler-Volmer equation as the electrochemical model for the PEMWE. First, the proposed model was validated using experimental data. Then, the current density and overpotential distributions were analyzed to understand the performance of the PEMWE. The current density and overpotential in the membrane were analyzed, and the performance was studied according to the operating current density. Finally, the performances of the serpentine and parallel channels were compared. This study contributes to the development of a numerical model and optimizes the flow field of the PEMWE.

#### 2. Methodology

The PEMWE comprises a membrane electrode assembly (MEA), gasket, bipolar plate, and current collector. The MEA of the PEMWE comprises the PTL for the anode and cathode, a catalyst, and a membrane. Liquid water flows into the anode, and the oxygen-water mixture flows out of it. However, no fluid flows into the cathode, and hydrogen flows out of it. The standard voltage of the reaction is 1.23 V, and this reaction is given by Equation (1) [19]:

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# SCOPE

Membranes (ISSN 2077-0375) is an international, peer-reviewed, open access journal. The journal covers the broad aspects of the science and technology of both biological and non-biological membranes, including membrane dynamics and the preparation and characterization of membranes and their applications in water, environment, energy, and food industries. Membranes provides a forum for publishing papers that advance our understanding of membrane structure, performance, processes, and applications covering membrane chemistry, physics, engineering, and biology. The journal publishes original reviews, research articles, and short communications. Authors are encouraged to publish their experimental and theoretical results in as much detail as possible. Thus, there is no restriction on the length of the papers. Full experimental, theoretical, and computational details must be provided so that the results can be reproduced.

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# P prof. Ernő Zádor 2 months ago

Dear Madam/Sir,

I would like to ask why is it that Membranes list its rank as Q1 (in Polymer science) but You do not? However both You and Membranes list the journal's rank also as Q2 (Chemical Engineering (miscellaneous)).

Regards, Ernő Zádor

reply



# Melanie Ortiz 2 months ago

SCImago Team

Dear Prof. Ernő,

Thank you for contacting us. Could you please expand a little bit on your comment? Best Regards, SCImago Team

# Georgian Technical University 3 years ago

We Greetings to you! wish you health and creative success! The scientific group of the Engineering Institute of Membrane Technologies of the Georgian Technical University wants to cooperate with you and publish an article in the. Please write us all the necessary conditions for the publication of the article, price and send us an e-mail at

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