



Green recovery of hazardous acetonitrile from high-salt chemical wastewater by pervaporation

Yong Wang^{a,*}, Xiang Mei^b, Tengfei Ma^c, Changjing Xue^b, Meidan Wu^a, Min Ji^c, Yuegang Li^c

^a State Key Laboratory of Pollution Control and Resource Reuse, School of the Environmental, Nanjing University, Nanjing, 210023, China

^b College of Biology and the Environmental, Nanjing Forestry University, Nanjing, 210037, China

^c Department of R&D, SinoChem TaiCang Chemical Industrial Park, TaiCang, 215433, China



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ABSTRACT

Acetonitrile in high-salt wastewater is not only a pollutant and a toxic and hazardous problem but also an inefficient use of resources. Acetonitrile in high-salt wastewater is refractory to and difficult for wastewater treatment. In this work, the separation and recovery of hazardous acetonitrile from high-salt aqueous streams by pervaporation were investigated, and the operating process conditions were evaluated. The separation of acetonitrile from wastewater was conducted using a polydimethylsiloxane (PDMS) membrane and was promoted by high concentrations of acetonitrile and high temperatures. The activation energy for the permeation of acetonitrile and water through the membrane indicated that the pervaporation of acetonitrile is more highly temperature dependent than that of water. The maximum permeate flux achieved was 353 g/m²h, and the highest concentrations of acetonitrile in the permeate were found to be greater than 47%. The results also showed that high salt in wastewater had a positive effect on the pervaporation performance and the separation factor. The process provides a paradigm change toward viewing the acetonitrile in wastewater as a resource to be recovered rather than waste to be treated, and thus, clean production can be realized.

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1. Introduction

Acetonitrile is used as a solvent in the fine chemicals industry and the manufacture of pharmaceuticals, including C6-fluoroketone, vitamin A, cortisone and carbon amine drugs. Acetonitrile is also commonly used as a moderately active solvent in the organic synthesis of many typical nitrogen-containing chemical substances, including thiamine and amino acids. The release of acetonitrile into aquatic media is of particular concern since it may be a moderately toxic substance to human beings and can be converted into hydrogen cyanide and acetaldehyde in living organisms (C. Li et al., 2016). Acetonitrile containing wastewater is toxic organic industrial sewage which needs treatment to make it harmless. Furthermore, the presence of acetonitrile in chemical effluents implies that valuable materials are leaking out of economic systems.

Acetonitrile, as a volatile organic compound (VOC), is infinitely

miscible with water and hazardous when discharged into chemical wastewater. Thus, it is required that innovative, cost-effective technology be developed for acetonitrile wastewater treatment. In recent years, biofilm treatment (C. Li et al., 2016; T. Li and Liu, 2008), electrolysis and Fenton oxidation have been used for the degradation of acetonitrile in wastewater. A recombinant bacterium, strain *B. subtilis* N4-pHT01-nit, with biofilm formation and nitrile degradation functions can help degrade acetonitrile in water treatment (C. Li et al., 2016). However, these technologies cannot recover valuable acetonitrile and always necessitate the addition of other reagents, which increase the treatment expenses, thus preventing these technologies from meeting the demands of clean production.

Organophilic pervaporation is a process for removing organics from aqueous solutions to yield an organic concentrate that can be treated for reuse. When the organics/water mixture flows through one side of the membrane, the organics are absorbed and permeated through the membrane driven by a vacuum, and then they are evaporated into the vapour and condensed on the other side of the membrane. The treatment process is promising because it has the

* Corresponding author.

E-mail address: wangyong@nju.edu.cn (Y. Wang).

merits of a small footprint, energy efficiency, environmental benignity and effective recovery of volatile organic components. There is also no emission problems, and it is easy for pervaporation treatment facilities to operate. Pervaporation can be a solution when the organics in the wastewater need to be recovered for their value or when other treatment technologies do work well. Organophilic pervaporation is expected to become a clean technology for the recovery of acetonitrile from high-salt chemical wastewater.

A significant amount of work has been done on VOC wastewater treatment. Because hydrophobic PDMS does not allow water molecules to pass through easily, the water contact angle is much higher ($>100^\circ$) than that of organics (Ramaiah et al., 2013). PDMS has high selectivity and permeability for volatile organic molecules (Liu and Xiao, 2004). The removal of VOCs such as benzene, chloroform and toluene from organics/water mixture using PDMS membranes (Ohshima et al., 2005) and polystyrene-*b*-Polydimethylsiloxane (PSt-*b*-PDMS) membranes by pervaporation has been investigated (Uragami et al., 2016). The pervaporation of styrene/water (Aliabadi et al., 2012) and *n*-butanol/water mixtures by ceramic-supported PDMS membranes (Dong et al., 2014) also was studied. García et al. studied the separation of *n*-butanol/dichloromethane/water mixtures by pervaporation in the presence of sodium chloride (García et al., 2009; García et al., 2013). The extraction of volatile chlorinated hydrocarbons from aqueous solutions was studied using hydrophobic mixed matrix membranes of PDMS supported on a polyvinylidene fluoride (PVDF) substrate (Ramaiah et al., 2013). Santoro et al. revealed that membranes with a PDMS intermediate layer can improve the EtOH/H₂O selectivity in comparison to a pure flat SBS membrane (Santoro et al., 2017). Hao et al. studied phenol removal from water using poly (ether-block-amide) (PEBA 2533) membranes (Hao et al., 2009). The removal of acetone, acetonitrile and ethanol from water by pervaporation was investigated by Khayet. The organic selectivity was found to be in the order of acetone > acetonitrile > ethanol (Khayet et al., 2008). GAO studied the separation of 1-butanol/water mixtures (5%) by PV using the PIM-1/PVDF thin-film composite membrane and found that the total flux can reach $9 \text{ kg m}^{-2} \text{ h}^{-1}$ with separation factors up to 18.5 (Gao et al., 2017). A mass transfer mathematical model has been developed, and the transfer rate of organics across the TFC membrane was measured in pervaporation using feed solutions with different NaCl concentrations (Cocchini et al., 2002a, 2002b).

The pervaporative removal of VOCs such as acetone (Zhang et al., 2016), chloroform (Urutiaga et al., 1999), toluene (Chovau et al., 2010), styrene ethyl benzene (Yahaya, 2008), xylene (Jian et al., 1996), hazardous organic solvents (MTBE, EtAc and BuOH) (Kujawa et al., 2015), and methanol (Yi and Wan, 2017) from solvent/water mixtures was also investigated. However, the recovery of acetonitrile from high-salt organic wastewater has not been studied. In this work, the pervaporation process for treating the volatile organic compound (VOC) acetonitrile in chemical wastewater at different feed concentrations, temperatures and salinities was evaluated. The wastewater contained acetonitrile with a concentration of 3000 ~ 40000 mg/L in a high salinity of approximately 20% sodium chloride. The pervaporative removal of acetonitrile from wastewater and acetonitrile concentrates is conducted in this paper. The condensed acetonitrile can be treated for reuse.

2. Materials and methods

2.1. Materials

The hydrophobic ceramic membranes with a functional PDMS layer used in the experiments were provided by Nanjing JiuSi Corporation (Nanjing, China). Acetonitrile (HPLC grade, purity 99.9%) and sodium chloride (reagent grade, purity 99.8%) were

supplied by Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China).

2.2. Wastewater quality

Wastewater containing the acetonitrile generated from C6-fluoroketone production was collected from SinoChem Environmental Protection Chemicals Corporation in TaiCang, China. Since C6-fluoroketone is a new product in China, there are no data about the wastewater from the literature. The Government required the project's environmental protection facilities and the product's production projects to be constructed and put into production at the same time. Therefore, we carried out the research on the treatment of this wastewater when the product was in the pilot stage. The characterization of the wastewater was performed to determine the composition and evaluate the potential for pervaporation separation. The effluents were analysed for COD, BOD, total nitrogen (TN), and total dissolved solids (TDS) according to standard methods. The sample concentrations of acetonitrile were determined by gas chromatography (HP6890). The concentrations of sodium ions and chloride ions were analysed by ion chromatography (Thermo Scientific Dionex ICS 5000). The typical wastewater characteristics are listed in Table 1:

2.3. Pervaporation tests

Pervaporation tests were performed with a lab-scale system schematized in Fig. 1. It consisted of a feed tank, a peristaltic pump, a ceramic membrane module, a cold trap and a vacuum pump. The ceramic PDMS membrane module (JiuSiCorp., Nanjing, Jiangsu, China) has a membrane area of 303 cm². The liquid solution was recirculated through the membrane module at a speed of 0.5 L/min, and the temperature of the solution was kept constant at 40 °C by using a water bath. The aim of recirculation is to maintain a constant concentration and temperature of the feed solution in the pervaporation cell. During the experiments, a permeate pressure of 4–15 mbar (2XZ-2 Huanyan Tianlong Vacuum Inc., China) was applied to the system by a vacuum pump. The cold trap was immersed in a liquid nitrogen bath to collect the acetonitrile-rich permeate for analysis.

Wastewaters containing low concentrations of acetonitrile were treated by pervaporation. For each set of experiments, the operating parameter was varied one at a time and covered the following ranges: feed concentration of 0.1 ~ 4 wt% acetonitrile and operating temperature of 18 ~ 55 °C. The collected permeate sample was weighed and analysed every hour.

2.4. Swelling experiment

The gravimetric method was used to determine the swelling degree of the PDMS/ceramic composite membrane. The membrane dry weight was first determined using an electrical balance (model AR522CN, OHAUS, Shanghai, China). The dry membrane was immersed in a container filled with acetonitrile wastewater at room temperature of 20 °C. After a fixed time interval, the free liquid on the surface of the swollen membrane was wiped out carefully by using tissue paper before weighting. The degree of swelling of the membrane, *S*, was determined by

$$S = \frac{W_s - W_d}{W_s} \quad (1)$$

where *w_s* and *w_d* represent the weight of the dry and swollen membranes, respectively.

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