Journal of Membrane Science 512 (2016) 13–20

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Dewatering of source-separated human urine for nitrogen recovery by membrane distillation

Lat Lat Tun [a,b], Dawoon Jeong [a,c], Seongpil Jeong [a], Kyungjin Cho [a], Seockheon Lee [a,*,1], Hyokwan Bae [a,*]

[a] Center for Water Resource Cycle Research, Korea Institute of Science and Technology, 39-1 Hawolgok-Dong, Seongbuk-Gu, Seoul 136-791, Republic of Korea
[b] Yangon Technological University, East Gyogone, Insein Township, Yangon, Myanmar
[c] Chemical and Biomolecular Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-Gu, Seoul 120-749, Republic of Korea

ARTICLE INFO

Article history:
Received 20 November 2015
Received in revised form
3 March 2016
Accepted 2 April 2016
Available online 6 April 2016

Keywords:
Human urine
Ammonia enrichment
Membrane distillation
Acidification

ABSTRACT

The nitrogen content of a synthetic ammonia wastewater was concentrated using direct contact membrane distillation (DCMD). The ratio of transferred ammonia to water (i.e., specific ammonia transfer: SAT) was controlled by operational conditions. With 20 °C on the permeate side, and a high temperature of 70 °C on the feed side, the process exhibited low SAT values for PTFE/PP (PTF045LD0A), PTFE/PP (TF-450), and PVDF (HVHP-14250) membranes. This was because the increase in water flux (>24 L/m^2 h) was greater than that of ammonia transfer. A positive relationship between SAT and free ammonia concentration was identified under different total ammoniacal nitrogen concentration and pH. The acidification pretreatment to pH 5 led to further reduction in the SAT value (as low as 6.91×10^{-5} g-N/g-H_2O). As a practical application, the dewatering process of source-separated human urine by DCMD required an additional filtration step to prevent fouling, but the filtration had an insignificant effect on the SAT. For the acidified and filtered source-separated human urine, total ammoniacal nitrogen was successfully concentrated with a low SAT value ($<2.06\times10^{-3}$ g-N/g-H_2O).

1. Introduction

The emission of nitrogen and other nutrients from sewerage systems currently causes severe eutrophication problems in public water bodies [1]. Ecological sanitation mitigates nutrient release in the water environment by treating nutrients in a safe manner in order to contribute to sustainable social and natural developments [2]. Human urine is the largest nutrient contributor to municipal wastewater [3]. The public health has been protected by hygienically stable disposal of human urine. A urine-diverting dry toilet for human urine has been implemented as an ecological sanitation technology. The human urine from the urine-separating toilet, i.e., source-separated human urine, allows it to be treated and applied on the farmland as fertilizer [4].

Source-separated human urine contains 3.5% organics (urea, creatinine, uric acid, etc.) and 1.5% inorganic salts (sodium, potassium, chloride, magnesium, calcium, ammonium, sulfates, phosphates, etc.), but it is highly diluted with approximately 95% water [5]. The TAN concentration of source-separated human urine varies depending on population, age, physical activity, feeding habits, and water consumption [6]. The TAN concentrations reported for source-separated human urine from households, schools, and workplaces ranged from 1.80 to 2.61 g-N/L [3,7,8]. The valuable nutrient compounds exist in a ratio of 11:1:2 for nitrogen, phosphorous, and potassium [6]. This chemical composition makes source-separated human urine a preferable fertilizer capable of replacing 20–25% of commercial fertilizers, instead of releasing urine into domestic wastewater [9]. However, direct application of source-separated human urine as fertilizer, would be inconvenient, unpleasant, and unhygienic [6]. Source-separated human urine also causes increased pH by urease activity, and consequent production of ammonia in soil environments [7]. To avoid inappropriate application, there is demand for a concentrated, microbial-free fertilizing product in crystalline form, such as NH_4NO_3. The major nitrogen fraction (NH_4^+) in source-separated human urine is produced by urease activity during storage, but biological nitrification is required to obtain the equal molar concentrations of NH_4^+ and NO_3^-. These NH_4NO_3 crystals can be produced from nitrified source-separated human urine by reduction of water content, evaporation, and crystallization [10].

Several processes (e.g., evaporation, freeze-thaw, and reverse osmosis) have been considered in finding an effective method to reduce the water content of human urine [8]. For example,

* Corresponding authors.
E-mail addresses: seocklee@kist.re.kr (S. Lee), lotus@kist.re.kr (H. Bae).
[1] He also equally contributed to this work.

http://dx.doi.org/10.1016/j.memsci.2016.04.004
0376-7388/

significant water reduction was achieved by evaporation (> 96%) and the freeze-thaw process (~75%) [8,11]. However, these two processes required unacceptably intensive energy. In addition, the dissolved ammonia contained in source-separated human urine can easily be lost (evaporated) to the atmosphere during thermal evaporation [8,11]. In contrast, the dewatering process using reverse osmosis membranes is a relatively cost-effective way to reduce the water content and remove micro-pollutants from the produced water [10]. However, membrane processes are often hampered by fouling problems that lower membrane porosity, which results in decreased efficiency and increased operational cost. Therefore, a clear need exists for development of a novel, cost effective technique that also solves the fouling problem.

The membrane distillation (MD) process is a process driven by a vapor pressure gradient caused by temperature difference [12]. The MD process could be an effective dewatering process to concentrate the human urine because it requires less energy than evaporation and has a lower tendency for membrane fouling, compared with the reverse osmosis process. Moreover, the operational cost of the MD process can be lowered using waste heat generated by industrial plants or heat from solar thermal sources. In the MD system, the vapor molecules transfer through a microporous hydrophobic membrane, and non-volatile matter can be completely rejected, theoretically [13]. As a result, the MD process is able to condense the ammonia and nitrate contents on the feed side by dewatering. However, high ammonia concentration and the alkaline condition of source-separated human urine lead to high volatile free ammonia (FA) content and consequent significant ammonia transfer to the permeate through the hydrophobic pores of the MD membrane. For this reason, the MD application is limited to membrane-based ammonia stripping (condensing NH_4^+ on the permeate side) to recover ammonia from highly concentrated ammonia wastewater such as source-separated human urine or swine manure [14–17]. Therefore, the optimized conditions to concentrate ammonia on the feed side are not available in our best of knowledge. The main aim of this study was to select appropriate membrane material, and to find optimal conditions for the best dewatering performance and the least ammonia transfer to the permeate side in the MD system. The parameters (type of membrane, pH, temperature, and total ammoniacal nitrogen (TAN) concentration) were optimized using synthetic nitrogenous wastewater. Then, a setup providing optimal conditions was applied to processing source-separated human urine. In addition, the effects of acidification and pre-filtration on flux and ammonia transfer were evaluated using source-separated human urine.

2. Materials and methods

2.1. Materials

Flat sheet commercial membranes with mean pore size of 0.45 μm were applied in the DCMD system [PTFE/PP (PTF045LD0A, Pall, USA), PTFE/PP (TF-450, Pall, USA) and PVDF (HVHP-14250, Millipore, USA)]. To investigate FA transfer through the DCMD membrane, synthetic wastewater samples were prepared in different concentrations of ammonium chloride (Sigma-Aldrich, Germany) in deionized water. Source-separated human urine was collected from the Water Quality and Treatment Laboratory, School of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST). The pH of synthetic wastewater (ammonium chloride solution) and source-separated human urine were adjusted using NaOH (10 mol/L) and HCl (11.3 mol/L). For the pre-filtration of source-separated human urine, a membrane filter paper (1.2 μm, GF/C, Whatman, UK) was used.

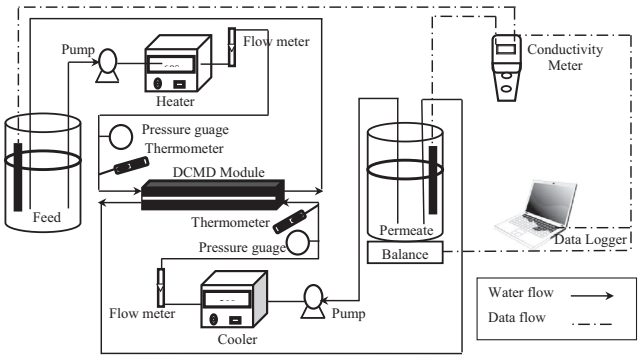

Fig. 1. Schematic diagram of experimental set-up for DCMD system.

2.2. Experimental conditions

In this study, direct contact MD (DCMD) was applied to concentrate human urine because DCMD is the simplest structure capable of producing reasonable high flux, among the various MD configurations [12]. Schematic representation of the DCMD system is shown in Fig. 1. Feed and permeate streams were designed to flow counter-currently with gear pumps, and circulated to their reservoir tanks. The temperature of feed and permeate solutions was controlled by thermostatic water baths (WCR-P22, Daihan Scientific, Korea). The temperature sensors and pressure gauges were placed in front of the feed and permeate inlets of the membrane module. The flow rates were set to 2 L/min (i.e., a cross flow velocity of 31.75 cm/s) in both hot and cold streams, into symmetrical, rectangular channels 3-mm deep. The flat sheet hydrophobic membrane was located in a horizontal module, which was connected to the feed and permeate tanks. The effective membrane area was 0.003 m^2 (8.6 cm long and 3.5 cm wide).

The DCMD dewatering, using PTFE/PP (PTF045LD0A), PTFE/PP (TF-450) and PVDF (HVHP-14250) membranes, was conducted at feed temperatures of 40, 50, 60, and 70 °C and at a fixed permeate temperature of 20 °C. The best membrane type was selected in terms of low FA transfer and high water flux. For the selected membrane, different pH conditions (5, 6, 7, 8, and 9) and TAN concentrations (0.465, 0.986, 1.945, 2.972, 3.972, and 4.940 g-N/L) were applied. Then, source-separated human urine processing was tested under the two conditions of initial pH 8.8 and acidified condition pH 6. Filtration pretreatment of acidified source-separated human urine was conducted using membrane filter paper of 1.2 μm pore size before the MD process. Water flux and ammonia enrichment on the feed side were evaluated for the acidified and filtered source-separated human urine. The concentrations of TAN, total nitrogen (TN), total phosphorus (TP), and total organic carbon (TOC) of all source-separated human urine samples are summarized in Table 1. The temperature on the feed and permeate sides were maintained at 60 and 20 °C, respectively, if not noted otherwise.

2.3. Analysis

The samples were taken from both sides every 30 min for 2 h to measure the TAN concentration on the feed and permeate sides. The concentration of TAN was analyzed using a Kjeltec TAN analyzer (Auto 2300 system, FOSS, Denmark). The ammonium and FA concentrations were calculated based on the TAN concentration, pH and temperature, as follows:

$$NH_3 + H^+ \leftrightarrow NH_4^+ \quad (1)$$