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Membrane distillation pilot plant trials with pharmaceutical residues and energy demand analysis

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HIGHLIGHTS

- Pharmaceutical residues removal efficiency of membrane distillation was tested.
- Integration approaches of district heating with membrane distillation at pilot plant scale has been assessed.
- Membrane distillation system's specific energy demand was determined.
- Techno-economy of large-scale MD systems were analyzed.

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ABSTRACT

In this study, an air gap membrane distillation (AGMD) system at pilot scale is applied for purification of effluent from a municipal wastewater treatment plant. A district heating network (DHN) is considered as a heat source for the membrane distillation system. Removal performance of pharmaceutical residuals, specific heat demand, and economic assessments were analyzed on the membrane distillation plant. Almost all targeted pharmaceutical compounds were removed to a very high degree, often below the method detection limit. The heat requirement for the MD process could be sufficiently supplied by the low-temperature district heating return line. Specific heat demands for the AGMD ranges from 692 to 875 kWh/m^3 without heat recovery and as low as 105 kWh/m^3 when heat recovery is possible. Different approaches to integrating the MD within the DHN system were analyzed; the advantages and shortcomings of each are discussed with emphasis on the MD system's capacity requirement and annual heat demand. The thermoeconomic analyses from this study presented the potential for energy optimization regarding heat recovery and module design improvement of the current MD equipment.

1. Introduction

Membrane distillation (MD) is a separation process driven by an imposed vapor pressure gradient across a hydrophobic microporous membrane. This technology is relatively new and still developing with a possibility of overcoming some limitations of the already established and popular membrane-based technologies. MD can produce ultrapure water in a single step and treats high feed concentrations. MD process takes place at temperatures below 100 °C and ambient pressure [1]. MD can also be powered by low-grade heat such as waste heat supply which is advantageous for large scale applications in industries with waste heat. Hence, recently much attention is given to MD processes driven by waste heat [2] and solar [3]. However, the specific energy demand is still higher as compared to the conventional membrane processes when internal heat recovery is not considered.

There are four major types of MD modules: Direct Contact Membrane Distillation (DCMD) in which the vapor condenses into a cooling stream on the cooling side of the membrane, Air Gap Membrane Distillation (AGMD) where the vapor passes through the membrane into an air-gap and condenses on the other end of the gap, Vacuum Membrane Distillation (VMD) where a vacuum is created instead of the air gap to increase flux across the membrane, and Sweeping Gas Membrane Distillation (SGMD) in which the vapor is swept away after passing through the membrane and condenses in a separate condenser.

Some applications of membrane distillation that have been investigated are: production of drinking water by desalination [3], concentration of acids [4], treatment of high salinity brines [5], and production of ultrapure water, e.g. in the semiconductor industry [6]. Gethard et al. [7] investigated an MD module

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enhanced by carbon nanotubes for purifying pharmaceutical wastewater and producing pure water for use at the same time. Food processing such as in dairy plants [8], juice concentration [9] and ethanol-water separation [10] are also some of the other applications for which MD has been tested and showed interesting results.

The capability of MD process to produce ultrapure water makes it a potential candidate for removal of very low concentration contaminants like pharmaceutical residues from domestic and industrial wastewater since these are otherwise difficult to remove by traditional wastewater treatment plants (WWTPs). A study carried out on effluents from four WWTPs in Sweden [11] showed that 85 out of the 101 pharmaceuticals considered in the survey were detected in at least one of the effluents of the WWTPs with diclofenac having the highest concentration at 3.9 μg/L. Some levels of pharmaceuticals were also detected in surface water, biota and in drinking water samples [11]. The presence of pharmaceutical residues in WWTP effluents can be an indication that there is a significant need for improved technologies to treat liquid effluents and process streams before releasing them to water bodies, where they may cause environmental problem including affecting fish behavior [12].

Many types of treatment technologies have been studied and used in municipality wastewater treatment systems. The performance of a biological reactor using activated sludge was evaluated for removal of ibuprofen, naproxen, and sulfamethoxazole in a Spanish WWTP [13]. Percentage removals of 70% for ibuprofen, 40–55% for naproxen and around 67% for sulfamethoxazole were obtained [13]. Clara et al. [14] studied the removal efficiency of a membrane bioreactor (MBR) and made comparisons with activated sludge plants in Austria. The results from their study showed that high removal percentages (97–99%) could be attained for ibuprofen by both MBR and conventional WWTPs. The MBR process was found to remove a maximum of 65% of sulfamethoxazole and up to 32% of it by the conventional WWTP. The studied methods did not show the removal of carbamazepine [14]. In another study [15], ultrafiltration (UF) and nanofiltration (NF) were tested for retaining pharmaceuticals. Even though average retention by NF for some pharmaceuticals like naproxen was as low as less than 10%, it was found to be higher percentage of retention (30–90%) than by UF which had typical retention percentages of less than 30%. In another study [16], nanofiltration showed retention of 90% for hydrochlorothiazide and 100% for diclofenac, even though higher retentions were obtained from reverse osmosis. Researchers from China [17] experimented with degradation of pharmaceuticals by photolysis and oxidation in the presence of hydrogen peroxide which resulted in the destruction of pharmaceuticals in the range of 25–95%, but here the decomposition reaction produced smaller organic compounds whose effect should be considered. Treatment by activated carbon and ozone were studied as a polishing step after MBR treatment to remove remaining pharmaceutical residues [18] Membrane distillation was also tested as a real-time concentration tool for analysis of pharmaceuticals by Gethard & Mitra [19]. Enrichment factor and solvent reduction for ibuprofen, for example, were 5.6 and 48% respectively. To date, there are not complete studies that include the energy demand and economy of a specific technology in addition to the removal performance for pharmaceuticals.

In this paper, membrane distillation powered by low-temperature district heating (DH) is explored for removal of pharmaceutical residues from treated domestic wastewater. The main objectives of this study are to evaluate MD's performance for removing pharmaceutical residues and to design and make a comparison between different MD-DH integration approaches regarding heat demands, capacity requirements, and economies.

2. Experimental set-up and methods

2.1. The AGMD pilot plant

The MD pilot plant was built in a joint project between KTH Royal Institute of Technology and IVL Swedish Environmental Institute. An illustration of the pilot plant is shown in Fig. 1 and a photo of the plant in Fig. 2. The plant consists of ten AGMD modules arranged in five cascades, each containing two modules connected in series (denoted a and b in Fig. 1). The MD modules are assembled and supplied by Xzero AB and installed at Hammarby Sjöstadsverk, Stockholm, Sweden. The active membrane area for each module is 2.3 m^2 (with a total membrane area of 2.8 m^2) and they are constructed from a stack of ten cassettes each containing two membranes. The membrane material is polytetrafluoroethylene (PTFE) with polypropylene (PP) support. The membrane characteristics are 0.2 mm thickness, 0.2 μm average pore size, and 80% porosity. The size of one module is 63 cm wide and 73 cm high and a stack thickness of 17.5 cm. During the experiments, data was logged for parameters connected to the control system. Type-T thermocouples installed close to the MD module's inlet and outlet streams were used to measure temperatures, which were recorded by the control system. System control and data (temperature, pressure, and flow rates of feed and cooling water) were registered on a personal computer with Citect Runtime SCADA software installed. A YOKOGAWA DC402G dual cell conductivity analyzer (ranges of detection 1 μS/cm–25 mS/cm and accuracy of ±0.5%) was introduced to check the conductivity of the product water. A Pt100 Ω temperature sensor with an accuracy of ±0.4 °C was used for the measurement of the permeate temperature. Rotameters (±5% accuracy) were used to measure feed and cooling water flow rates.

2.2. MD-district heating integration cases

District heating (DH) is considered here to power the MD process and three different approaches to integrating it with AGMD were investigated. The three cases were chosen based on the temperature levels in the supply and return lines of the DH. These three cases are presented in the following subsections. In all the cases, flow rates of feed and cooling are fixed at 1200 L/h, and the same modules and similar feeds are considered.

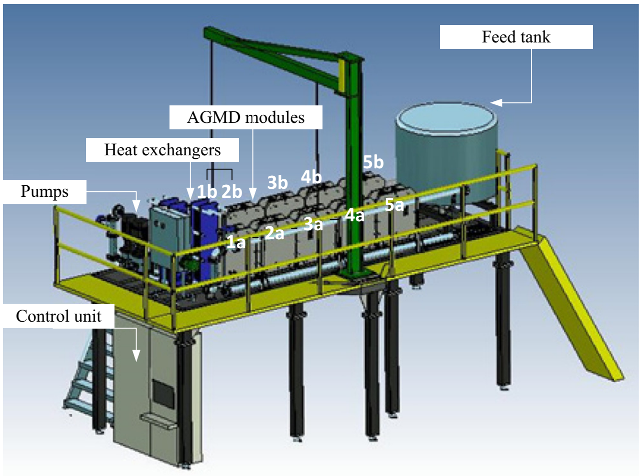

Fig. 1. Illustration of the membrane distillation setup at Hammarby Sjöstadsverk.