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Micron-pore-sized metallic filter tube membranes for filtration of particulates and water purification ☆

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Abstract

Robust filtering techniques capable of efficiently removing particulates and biological agents from water or air suffer from plugging, poor rejuvenation, low permeance, and high backpressure. Operational characteristics of pressure-driven separations are in part controlled by the membrane pore size, charge of particulates, transmembrane pressure and the requirement for sufficient water flux to overcome fouling. With long term use filters decline in permeance due to filter-cake plugging of pores, fouling, or filter deterioration. Though metallic filter tube development at ORNL has focused almost exclusively on gas separations, a small study examined the applicability of these membranes for tangential filtering of aqueous suspensions of bacterial-sized particles. A mixture of fluorescent polystyrene microspheres ranging in size from 0.5 to 6 μm in diameter simulated microorganisms in filtration studies. Compared to a commercial filter, the ORNL 0.6 μm filter averaged approximately 10-fold greater filtration efficiency of the small particles, several-fold greater permeance after considerable use and it returned to approximately 85% of the initial flow upon backflushing versus 30% for the commercial filter. After filtering several liters of the particle-containing suspension, the ORNL composite filter still exhibited greater than 50% of its initial permeance while the commercial filter had decreased to less than 20%. When considering a greater filtration efficiency, greater permeance per unit mass, greater percentage of rejuvenation upon backflushing (up to 3-fold), and likely greater performance with extended use, the ORNL 0.6 μm filters can potentially outperform the commercial filter by factors of 100–1000 fold.
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1. Introduction

To meet the increasing need for potable waters, the use of ultrafiltration in large-scale processes has been increasing in recent decades. Robust filtering techniques capable of efficiently removing particulates and biological agents from water represent a critical need to overcome impending water shortages (Reisch, 2007). Pressure driven tangential flow membrane processes, where the feed stream is split into retentate and permeate fractions, are gaining widescale use (Van der Bruggen et al., 2003). Operational characteristics of pressure driven separations are in part controlled by the membrane pore size, charge of the retained particles, the pressure exerted on the membrane and requirement of sufficient water flux at low pressure to overcome fouling (Van der Bruggen et al., 2003; Goosen et al., 2005). With extended use membranes exhibit a decline in permeate flux as a result of filter-cake accumulation on the filtration surface, pore plugging, fouling, or membrane deterioration (Kilduff et al., 2005; Zularisam et al., 2006). Much has been written on the declining efficiency of membranes as well as methods to control fouling (Hilal et al., 2005) or better engineer membranes. For example, Kilduff et al. (2005) observed that surface roughness exacerbated fouling by

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reducing the advancing/receding contact angle hysteresis. They also demonstrated improvements in performance with increased wettability of membrane surfaces, and use of membrane surfaces that minimized attractive interactions with natural organic matter in water. Indeed, natural organic matter suspended in water was determined to be a major cause of membrane fouling by Zularisam et al. (2006). They found organic matter fouling substantially reduced permeate flux at lower pH, high ionic strength, and at high divalent cation concentrations, particularly calcium. They called for further research on advanced membrane materials resistant to chemical and mechanical attack with prolonged membrane performance.

Oak Ridge National Laboratory (ORNL) has considerable experience in the fabrication and use of metallic filter tubes that appear to have considerable potential for water purification applications in small to municipal public sector operations or by industry. Production of ORNL's inorganic membranes was initiated in the 1940's. Information on chemical and mechanical resistant filter tubes, including those produced in Oak Ridge, can be obtained from reviews and books (Bischoff et al., 2004; Judkins and Bischoff, 2004a,b; Mansur et al., 2004).

The purpose of this project was to utilize metallic filter tubes developed for other applications and demonstrate their potential feasibility to be used for water purification. In addition to evaluating the ORNL filter tubes, a comparison of the performance of these filters with comparable commercial filter tubes revealed advantages ORNL products may have over the state-of-the-art. Our results indicate that ORNL metallic filter tubes appear well suited for water filtration as they exhibit high permeance at low backpressures, are robust, and can be rejuvenated upon combustion at 500 °C for 8 h, and appear to have cheaper material costs than traditional depth filters.

2. Materials

2.1. ORNL metallic filter tubes

Using the Oak Ridge metallic filter tube technology miles of filter tubes have been manufactured with individual tubes used for decades (Judkins and Bischoff, 2004a,b). Though the vast majority of the metallic filter development has been for the purpose of gas separation (Bischoff et al., 2004; Judkins and Bischoff, 2004a,b; Mansur et al., 2004), versions with larger pores (~1 μm) have recently been recognized for potential utility to filter microorganisms and particulates from water. Common features of the ORNL-type of metallic filter tubes (Fig. 1) include:

- Membranes are extremely rugged, and last tens of years.
- Manufacturing costs are low.
- They tolerate transmembrane pressures >400 kPa (~60 psi) with low backpressures (<10 kPa).
- Filtration is performed at surface (i.e. not a depth filter) exhibiting high recovery when cleansed.
- They can operate as an end-member filter or in tangential-flow to minimize fouling.
- They are capable of removing/concentrating bacteria and viruses from fluids or gases.
- Specific pore sizes for water filtration may accurately range from 0.05 to >3.0 μm.
- The versatile fabrication process has led to 17 commercial products (Pall Corporation) including corrosion resistant 300–400 series stainless steel (SS) and Hastelloy X.

ORNL metallic filter tubes accomplish particulate filtration at their surface versus more traditional depth filters (Fig. 1). Surface filtration is accomplished by designing the membrane surface to have pore sizes that inhibit particles being entrapped within the pores. More traditional depth filters exhibit larger pores with the filtration occurring within the depth of the filter medium. The design of ORNL's filters maximizes permeance, capture efficiency and recovery efficiency by minimizing the effective thickness of the filter. Thus, potentially, most microbial cells or particulates retained by the ORNL filter tubes can be subsequently dislodged during rejuvenation processes enabling high efficiency for an extended period of time. The scanning electron micrograph in the left side of Fig. 1 shows a several-micron-thick metal filter tube layer with individual pore sizes of less than 0.5 μm deposited on a metallic backbone with micron-sized pores. The backbone structure extends ~500 μm below the bottom of the SEM providing structural integrity for the filter tube. A picture of an individual filter tube is shown in Fig. 2. A cross-section of an ORNL metallic filter tube compared with a traditional

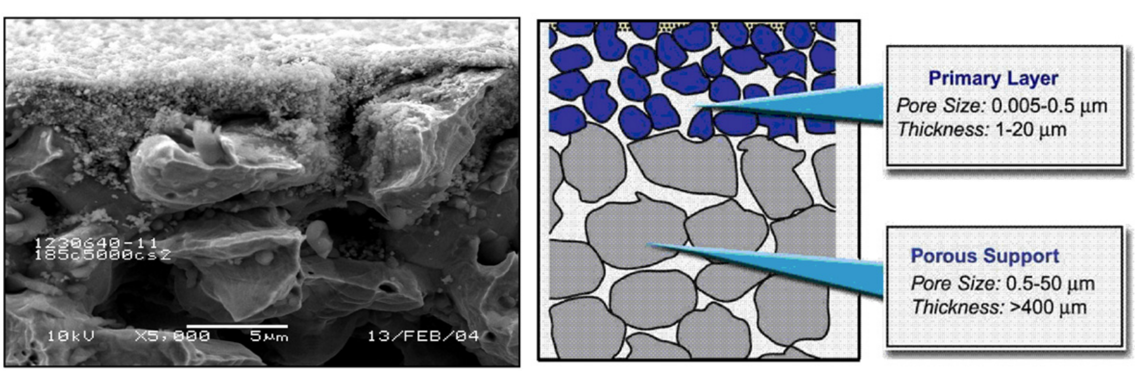

Fig. 1. a and b. ORNL metallic filter tube visuals. At left is a scanning electron micrograph of a cross section of a two-layer ORNL metallic filter tube membrane while the cartoon on the right shows the characteristics for a typical filter tube: the porous support of several micron-sized pores and a primary separation layer.