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Polymer aerogels for efficient removal of airborne nanoparticles

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ABSTRACT

This study evaluates the potential of polymer aerogel monoliths in removing airborne nanoparticles. Macroporous monolithic aerogels of δ-form syndiotactic polystyrene (sPS) are synthesized for this purpose via thermoreversible gelation of a solution of sPS in a good solvent followed by supercritical drying. The air permeability and airborne nanoparticle removal efficiency are determined as function of the bulk density of aerogels. The data reveal a power-law dependence of particle removal efficiency and air permeability on bulk density. The data also reveal that efficiency greater than 99.95% can be achieved if the bulk density is kept at 0.042 g/cm^3 or higher. These materials show air permeability of the order of 10^{-10} m^2. The gradient density aerogels produced via sequential injections of sPS solutions show improvements in filtration efficiency attributed to additional skin layers. This idea is exploited in designing gradient density aerogel filters that offer higher efficiency and higher air permeability at a much lower bulk density than the single density monoliths of the same thickness.

1. Introduction

It is widely known that exposures to airborne particles can cause severe health hazards to human being including nausea, breathing difficulties, bronchitis, birth defects, serious developmental delays, weakened immune systems, and even cancers [1,2]. The significance of the hazards is a function of the size of the airborne particles. For example, smaller particles can easily penetrate into the human body and reach the pulmonary alveoli [3]. Fine particles of size 0.1–1 μm and ultrafine particles of size <0.1 μm are more detrimental to human health than the coarse grain particles of size >1 μm. The health hazards are compounded by the presence of pathogens of size 20–2000 nm residing in these particles [4].

Removal of small particles and pathogens by filtration can significantly improve the quality of air and reduce the health hazards. The use of high efficiency particulate absorption (HEPA) filters fabricated from fiber mats are widely used for the purpose. The filtration efficiencies of HEPA filters are high at 99.95% for removing around 0.3 μm particles as per EN 1822-1:2009 classification at comparably low pressure drops. However, published literature on evaluation of HEPA filters for removal of airborne nanoparticles of sizes 100 nm and smaller are scarce.

A wide range of industrial applications use porous media for filtration due to their large surface area and pore volume [5]. In this context, monolithic aerogels fabricated from inorganic or organic sources present several attributes as filter media. The aerogel monoliths have low solid contents, high porosity with various pore sizes, and large surface areas [6–26]. For example, silica aerogels derived from alkoxy silanes offer large surface areas, up to 1000 m^2/g, significant mesopore fractions, and porosities over 90% [6–18]. The δ-form syndiotactic polystyrene (sPS) aerogels also offer macropores and porosities up to 97%; the nanocavities of crystalline strands of sPS selectively absorb small organic molecules [19–26]. Despite offering several attractive characteristics, research on filtration of nanometric airborne particles by monolithic aerogels is scarce. A handful of studies used aerogel granules or microspheres in packed beds [27–30]. Some literature reports are available on the use of aerogels impregnated into HEPA filters for airborne particle filtration [31], colonization of bioluminescent organisms inside a monolithic silica aerogel to detect viral particles [32], and applications of heterogeneous catalyst for removal of pollutants, such as removal of cyanides or combustion gases by monolithic silica aerogel composites containing metal oxides [33–35]. Literature on direct investigations of filtration of airborne nanoparticles by monolithic aerogels is scarce.

This study investigated the utility of monolithic δ-form sPS aerogels for nanometric airborne particle filtration. The values of permeability and filtration efficiency are two key measured properties needed to quantify the performance of the aerogels for use as filtration media. In this context, monolithic sPS aerogels of single density and gradient density were examined.

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2. Materials and methods

sPS aerogel monoliths were fabricated using a two-step process involving thermoreversible gelation and supercritical drying [19–26]. The thermoreversible gelation of sPS occurred via spinodal decomposition of the polymer–solvent binary system. sPS pellets with 98% syndiotacticity and molecular weight of 300,000 g/mol was provided by Scientific Polymer Products Inc. (New York, NY). The sPS pellets were converted into powders for easy dissolution in a solvent. Reagent grade tetrahydrofuran (THF) purchased from Fisher Scientific was used as the solvent. The solutions of sPS in THF was turned into δ-form sPS gel via natural cooling [23–26]. The concentration of sPS in the solution was varied to obtain different values of porosity. sPS solutions with concentration 0.010, 0.015, 0.020, 0.025, 0.030, 0.040, 0.050, 0.060, 0.070, and 0.080 g/mL were prepared by dissolving sPS powder in THF in sealed glass containers kept in oil bath at 120 °C. The solution was poured in a disk shaped mold of 30 mm diameter and allowed to gel as the solution gradually cooled to room temperature. The thickness of the disk shaped gels was maintained at around 3.5–4.0 mm. The gels were aged in the mold for a day and washed and solvent exchanged with 200-proof ethanol for at least 3 days. sPS aerogels were obtained by supercritical drying in carbon dioxide. For this purpose, ethanol in sPS gel was exchanged with liquid carbon dioxide before the supercritical drying step.

Monolithic gradient density aerogels were produced by a sequential injection method, whereby sPS solutions of different solid content were sequentially injected into the mold before or after gelation of the previously injected solution. For example, one gradient density aerogel was obtained by first injecting a solution of 0.05 g/mL sPS into the mold followed by injection of 0.03 and 0.01 g/mL solutions. The time interval of each injection was varied based on the gelation time of each solution. The gelation time of the solution containing 0.05 g/mL sPS was about 4.5 min. Thus, the 0.03 g/mL solution was injected before the 4.5 min mark or shortly afterward. In this study, two types of gradient density aerogels were produced respectively by injecting solutions before and after gelation of the previously injected solutions.

The value of bulk density, ρ_b was obtained from the weight-to-volume ratio of the specimens. The permeability of air was obtained from Darcy's law, as in Eq. (1):

$$Q = \frac{kA}{\mu}\frac{\Delta P}{L} \tag{1}$$

In Eq. (1), Q is the measured volumetric flow rate of air, k is permeability, A is cross-sectional area of the face normal to the fluid flow, μ is the viscosity of air, ΔP is the measured pressure drop, and L is the thickness of the aerogel specimen. An air permeability tester (Frazier Precision Instrument, Hagerstown, MD) was used for this purpose. The disk shaped aerogel specimens were placed at the center of a perforated bottom plate with a central hole which allowed air flow (Fig. 1). The hole sizes were 25, 22, and 5 mm in diameter. Darcy's law can be modified with the shape factor, G for holes much smaller than the diameter of the aerogel specimen [36]. A sample holder covered the specimen and prevented lateral flow of air. The gaps between the specimen, the bottom plate, and the sample holder were eliminated using vacuum grease. Finally, the specimen with the assembly was placed inside the Frazier tester to measure the permeability (Fig. 2). As seen schematically in Fig. 2, a vacuum pump drew air through the aerogel specimen.

The power of the vacuum pump was controlled to obtain several values of pressure drop, ΔP and volumetric flow rate, Q. Fig. 3 presents a set of representative data on ΔP vs. Q used in the evaluation of air permeability, k.

Filtration efficiency (E) measures the fraction of the incident particles that a filter can capture after passing through the filter once, given in Eq. (2):

$$E\ [\%] = \frac{N_B - N_A}{N_B} \times 100 \tag{2}$$

In Eq. (2), N_B and N_A are respectively the number of particles before and after passing through the filter. Penetration, P is obtained from the value of E, where E is a fraction, as follows:

$$P = (1 - E) \tag{3}$$

In this work, a TSI-8130 filter tester (TSI Inc., Shoreview, MN) that generated in situ polydispersed sodium chloride nanoparticles was used. The diameter of generated particles ranged from 25 to 150 nm with an average diameter of 75 nm [37]. A wide-mesh metal net attached at the bottom of a specimen holder (Fig. 4) supported the aerogel specimen. The periphery of the aerogel specimen was sealed by a rubber O-ring. The face velocity (V_f) of air flow was varied between 25, 40, and 50 cm/s. For each specimen, the particles were allowed to go through the specimen 10 times and the value of filtration efficiency corresponding to each incident was measured and an average value was determined. The maximum efficiency that can be measured using TSI-8130 filter tester is 99.999%.

The internal structures of the sPS aerogel specimen were examined by scanning electron microscope (JEOL JSM5310) to visually identify how the trapped nanometric NaCl particles were retained. For this purpose, the aerogel specimens were recovered after filtration and frozen in liquid nitrogen. The frozen specimens were cut into pieces and a representative piece was mounted on an aluminum stub using carbon tape. The samples were sputter coated by silver (ISI-5400 Sputter Coater, Polaron).

The absorption capacity of the particles by the filter media is an important attribute as it relates to the life-time of a filter. In this

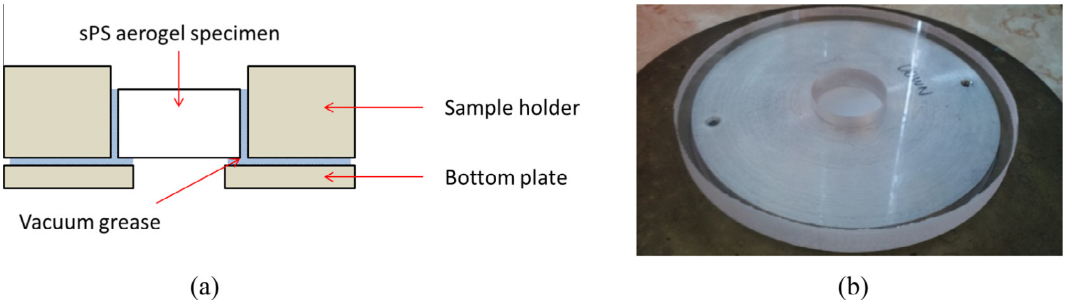

Fig. 1. (a) Schematic and (b) experimental image of the assembly of sPS aerogel specimen, sample holder, and bottom retaining plate for permeability measurement. Vacuum grease is used to seal any gaps.