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# Feasibility test of cellulose filter for collection of sulfuric acid mists

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ABSTRACT

Exploring affordable and reliable filters to sample sulfuric acid mists is of interest because the polytetrafluoroethylene filters recommended by NIOSH Method 7908 are relatively costly. In this study, the feasibility of hardened cellulose filters such as Whatman grade 540, 541, and 542 filters were investigated for sampling sulfuric acid mists. Thirty-minute collection efficiency tests showed low collection efficiency for Whatman grade 540 and 541 filters while Whatman grade 542 filters exhibited high collection efficiency because of its smaller pore size. Eight-hour collection efficiency tests for the concentration of ca. 5000 μg/m$^3$ showed Whatman grade 542 filters retained 95% collection efficiency. The collection efficiency over eight hours neither increased nor decreased because the mists might have been absorbed or have formed a thin layer on the fiber and the filter structural integrity did not change. The higher collection efficiency was at the expense of a higher pressure drop; however, the pressure drop was still under the back pressure limit for most personal pumps. The integrity test showed that under the typical concentration in the occupational environment the hardened cellulose filters can maintain integrity for more than seven days. In summary, Whatman grade 542 filter is a suitable alternative for measuring sulfuric acid mists.

## 1. Introduction

In the steel, fertilizer and petroleum industries, about 200 million tons of sulfuric acid ($H_2SO_4$) are used each year worldwide [1]. $H_2SO_4$ mists incidentally released into the air during the manufacturing processes can irritate skin and eyes and damage the respiratory system when concentrations are excessive. Such damage has been linked to laryngeal cancer, and accordingly the National Toxicology Program has identified strong inorganic acid mists containing $H_2SO_4$ as "human carcinogens" [2]. To evaluate workers' exposure to $H_2SO_4$ mists, NIOSH Method 7908 specifies 37-mm diameter quartz fiber filters or 0.45 μm pore size polytetrafluoroethylene (PTFE) filters to sample sulfuric acid mists [3]. This method overcomes the chemical interference issue of filter materials in the previous and now abandoned NIOSH Method 7903 [4].

PTFE filters are chemically inert and have low chemical interference; hence, they are good candidates for accurately sampling sulfuric acid mist. However, for large-scale or long-term sampling programs, the PTFE filters can be expensive. The current (2017) price of one piece of 37 mm PTFE filter ranges from 2.4 to 3.4 US dollars. The cost of quartz fiber filters ranges from 1.6 to 2.4 US dollars. In comparison, cellulose filters provide the most economic choice because each cellulose filter only costs 0.1–0.5 US dollars. Consequently, from the perspective of sampler developers, PTFE would be the last material of choice when no cheaper and equally effective alternative is available. Alternative filter materials such as the cellulose ester membrane filter might be used to sample $H_2SO_4$ mists as indicated in NIOSH Method 6004. However, Murdoch et al. [5] observed decomposition of MCE filters under high concentration (0.3–7 mg/m$^3$) of acidic mists. Such an observation suggests durability should be considered for new material selection.

Cellulose filters were selected as an alternative in this study because of their affordable cost and good tensile strength. Mader et al. [6] collected $H_2SO_4$ mists on Whatman grade 4 cellulose filters and obtained 91–97% recovery by pH titration method. However, $H_2SO_4$ may esterify cellulose or deteriorate the filters by attacking the bond in the cellulose [7]. In this study, hardened cellulose filters (e.g. Whatman grade 540, 541, and 542) were proposed as a possible solution to this

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problem because they are specially treated with acid, which makes them mechanically stronger and more resistant to acids. Alwine et al. [8] illustrated their good mechanical strength and resistance to acidic 1-[(m-nitrobenzyloxy)methyl]pyridinium chloride, and Moore [9] successfully impregnated them with phosphoric acid for ambient ammonia gas collection. Hence, the first objective of this study was to assess the collection efficiency and pressure drop of different grades of hardened cellulose filters. Secondly, their integrity and chemical recovery were tested with 1% $H_2SO_4$ solution to evaluate the durability for the purpose of storage.

## 2. Methods

### 2.1. Experimental setup for collection efficiency and pressure drop testing

Sulfuric acid aerosol was used to evaluate the performance of hardened cellulose filters. Pure sulfuric acid ($H_2SO_4$, MW = 98.079 g/mole) was not directly applied as atomization liquid because it is corrosive and could damage the atomization system. According to the National Toxicology Program (NTP), strong inorganic acid mists containing sulfuric acid were identified as a "known human carcinogen" [2]. In other words, the challenging aerosols are mists containing sulfuric acid as a mixture rather than pure sulfuric acid mists. In this study, a 1% w/w sulfuric acid solution (pH approximates to 1), which was diluted from 98% sulfuric acid (ACS grade, Fisher Scientific) with DI water, was adopted for atomization.

Micron aerosols rather than nano-particles were of interest in this study. In phosphate fertilizer manufacturing facilities, sulfuric acid in micron size was dominantly present because of its hygroscopic property [10,11]. The Collison nebulizer has been applied to generate sulfuric acid mists for exposure studies [12,13]. Accordingly, as shown in Fig. 1, a one-jet Collison nebulizer was used to inject polydisperse liquid sulfuric acid mists into a chamber (41 cm in diameter and 71 cm in length) maintained at 30% RH (monitored by an Omega HX94C RH transmitter) and 22 °C (measured by an Omega thermocouple). Mist size would shrink and increase concentration due to water evaporation at 30% RH. Three sets of cassettes were connected to the chamber, and the coefficient of variation among the cassettes was < 6%, proving they were mixing well. All 37 mm round hardened cellulose discs were prepared from larger cellulose discs of Whatman grade 540, 541, and 542 using a laser cutter (Zing 25 W, Epilog). Each upstream cassette contained a hardened cellulose filter that continuously collected the sulfuric acid aerosol, while the downstream hardened cellulose filter was changed during a pre-set time. The sampling flow rate was 2 Lpm. The first set of experiments of 30 min were conducted to determine the type of filters among Whatman grades 540, 541, and 542 that would be suitable for the following 8-h experiment. The downstream filter was followed by a thick cellulose pad in the second filter pack to ensure 100% collection efficiency. Note Whatman grade 41 was also tested as reference to compare 540, 541, and 542. The pressure drop of the upstream filters pumped by a Universal PCXR8 personal pump (SKC Inc. Eighty Four, PA) was measured with a manometer (Magnehelic, Dwyer Inst., Michigan City, IN).

In this study, the collection efficiency by mass rather than by count was applied, because the standard of exposure limit for sulfuric acid is based on mass concentration [2]. Also, real-time monitor was not applied because acidic mists may damage the instrument. The following determination of sulfate mass was modified from NIOSH Method 7908. After sampling, the hardened cellulose filters were soaked in DI water for 24 h to extract sulfuric acid. The extracted solution was then evaluated with an Ion Chromatography System (ICS-1500, Thermo Fisher Scientific Inc., Waltham, MA) equipped with an AS9-HC column. After the analysis, all aerosol mists deposited on the upstream filter ($M_{upstream\ filter}$) and the downstream filters ($M_{downstream\ filters}$) were calculated to determine the collection efficiency ($C$). The 30-min collection efficiency for various filters is defined by

$$\eta_{30\ \min} = 1-\frac{M_{downstream\ filters}}{(M_{upstream\ filter}+M_{downstream\ filters})} \quad (1)$$

For the 8-h collection efficiency, the collection efficiency at $i_{th}$ hour is defined by

$$\eta_{i_{th}\ hour} = 1-\frac{M_{downstream\ filters,i}}{(M_{upstream\ filter}+\sum_{i=1}^{8} M_{downstream\ filters,i})/8} \quad (2)$$

### 2.2. Experimental setup for durability test

The filter integrity and the recovery of $H_2SO_4$ solution were conducted using 50 µL $H_2SO_4$ solution with concentrations of 1552 ppm (76 µg as $SO_4^{2-}$), 5670 ppm (278 µg as $SO_4^{2-}$), 17,467 ppm (856 µg as $SO_4^{2-}$), and 151,062 ppm (7399 µg as $SO_4^{2-}$) at room temperature (24 °C and 50% RH). Each solution was pipetted onto Whatman grade 542 filters. These filters were manually folded and visually examined for physical damage every day. Recovery tests were conducted to simulate repeated mists wetting on the cellulose filters. After spiking a known amount of $H_2SO_4$ solution on the filter, the filter was analyzed by the Ion Chromatography System after 7 days to determine the recovery rate.

### 2.3. Quality assurance and quality control

#### 2.3.1. Collection efficiency test

The fresh mist aerosolized from the Collison nebulizer with DI water was measured by an APS for its size distribution. The Mass Median Aerodynamic Diameter (MMAD) and Geometric Standard Deviation (GSD) were 6.4 µm and 1.6, respectively. Because RH in the chamber

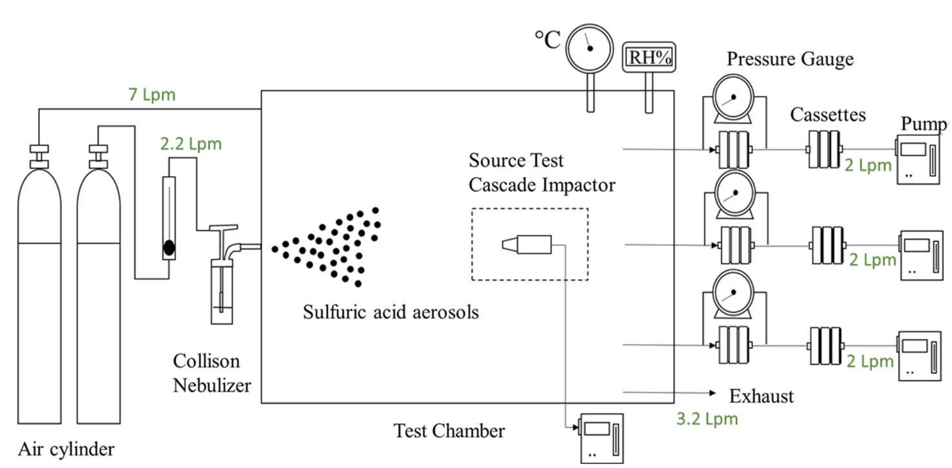


Fig. 1. Experimental setup for measuring the collection efficiency and pressure drop of the hardened cellulose filter.