



Development of a Novel Porous Membrane Denuder for SO₂ Measurement

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ABSTRACT

Conventional denuders made of glass or metal are too heavy and bulky to be used in personal sampling systems. In this study, a portable Porous Membrane Denuder (PMD) was developed for personal sampling. The PMD utilizes the porosity of the membrane material and a configuration of multiple parallel flow channels to reduce the size and weight of the device, while increasing the gas collection efficiency. Four types of PMDs (PMD I, II, III and IV) with increasing numbers of channels and smaller channel openings were constructed and tested. Using 10% sodium carbonate coating and a feed concentration of 1 ppm, PMD Ia's collection efficiency for sulfur dioxide over 8 hours was higher than 99.9%. For a feed concentration of 10 ppm, the 5-hr time-weighted-average collection efficiency for sulfur dioxide was 73.1%, 82.8%, 90.9%, 97.2% for PMD Ib, II, III and IV, respectively, compared with 96.6% of the Glass Honeycomb Denuder (GHD), which has similar structure to PMD IV. However, the weight of PMD IV is only one-tenth of that of the GHD. It is clear that with a similar physical structure, PMDs have similar capacities to traditional Glass Honeycomb Denuders, yet are much lighter and less expensive. This study demonstrates the great potential of this new type of denuder for many applications in the field of environmental and industrial hygiene monitoring. Particle loss fractions of the four types of PMDs in the size range of 1 to 10 μm were also measured and were 2.9%, 5.2%, 5.7% and 7.3%, respectively.

Keywords: Porous membrane; Denuder; Sulfur dioxide; Diffusion; Collection efficiency.

INTRODUCTION

Diffusion denuders are well established devices to remove gases from ambient air (Dasch *et al.*, 1989; Huang *et al.*, 2004; Tsai *et al.*, 2004; Acker *et al.*, 2005; Hayami 2005; Pathak and Chan, 2005). The inner surface of the flow path is coated with a material that absorbs a particular gas or vapor. When air passes through the denuder, gas molecules diffuse to the surface of the flow path and get absorbed on the coated surface. Because of the gas removal capability, denuders have been used to measure the concentrations of gases in sampled air. They have also been used to remove gases or vapors from an aerosol stream to prevent their interference in aerosol measurement that can cause positive or negative sampling artifacts (Hinds, 1999; Keck and Wittmaack, 2006).

The original design of the denuder system is a straight tube (Ferm, 1979; Durham *et al.*, 1986) which is 1 m long to allow for an operating flow rate of 10 L/min. Denuders

with various shapes and dimensions have been developed thereafter, e.g., annular denuder (Possanzini *et al.*, 1983; Sekiguchi *et al.*, 2009), coiled denuder (Pui *et al.*, 1990), honeycomb denuder (Koutrakis *et al.*, 1993; Sioutas *et al.*, 1996), parallel plate denuder (Eatough *et al.*, 1993) and so on. Glass and stainless steel are commonly used for the fabrication of denuders. The honeycomb denuder made of glass (GHD) has been commercialized and integrated in a denuder-filter system, which is called "Chembomb Speciation Sampling Cartridge" (Model 3500, Thermo Electron Co., Inc.). However, this commercial denuder-filter system is quite bulky, heavy and relatively expensive, rendering it undesirable for personal sampling. In recent years, new types of denuders have been proposed. Continuously wetted denuders (Simon and Dasgupta, 1993; Dasgupta *et al.*, 1997; and Ku *et al.*, 2010) use a continuously renewed liquid as an absorber and the effluent can be used to determine gas concentrations online. However, these denuders are connected to an Ion Chromatography (IC) system or other sizeable infrastructure which makes them infeasible for personal sampling. Proposed by Fitz and Motallebi (2000), the fabric denuder is another type of new denuder. Fabric filter paper is coated and loaded in a filter stack, and collected pollutants are extracted in the same way as common filters.

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The fabric denuder has a relatively small loading capacity and suffers from significant particle loss (Fitz and Motallebi, 2000). Tsai *et al.* (2001) developed a personal porous-metal denuder specifically for adsorbing acidic gases and ammonia gas in an occupational environment. Field test results showed that the porous-metal denuder has very high collection efficiency; however, particle loss in the porous metal disc is also high because particles are forced to penetrate the curved channel of the porous metal discs.

The main objective of the present study was to develop a novel denuder with the advantage of compactness, easy operation, high capacity and low cost. This new denuder is aimed to be integrated into a personal sampling system and applied in occupational environments such as phosphate fertilizer manufacturing facilities where the acidic gas concentration may be much higher than the ambient level. A soft and porous membrane was chosen as the material for making this new type of denuder – the Porous Membrane Denuder (PMD). The principle of the PMD is to utilize the porosity of a filter membrane to increase the surface area. A configuration of parallel flow channels was used to compact the size, decrease the weight and increase gas collection efficiency.

PMD DEVELOPMENT

In constructing the denuder prototype, material strength and low cost are two important factors to be considered. Cellulose filter (Whatman Grade 40), which has a suitable rigidity and reasonable price, was selected to build the multi-channel grid-shape denuder. Cellulose filter papers were cut by a laser plotter (Epilog Zing 35W) into a grid of interconnecting parallel planes. Each interconnecting panel has tabs and inserts that help ensure straightness and uniformity of the grid structure. Within each grid cell is an insert made from the same material, and various densities of fold forms to accommodate different channel numbers and openings. The height and outside dimension of the assembly of cut pieces and inserts can be tailored to a predetermined size and shape. However, as the number of grid divisions or insert folds increases, the difficulty of assembling the grid cells and inserts also increases.

Four different types of PMDs, named as PMD I, II, III and IV respectively, have been built with the cellulose medium. The different types correspond to different numbers of channels and different dimensions of channel openings. The schematics of PMD I, II, and III are shown in Fig. 1. PMD IV has the same structure as PMD III, only with more zigzag inserts. The prototypes of these 4 types of PMDs are shown in Fig. 2, and their properties are listed in Table 1.

THEORY

The collection efficiency for a hollow tube that acts as a perfect sink for a certain gas was given by Gormley and Kennedy (1949):

$$\eta = 1 - 0.8191\exp(-11.489\mu) - 0.0975\exp(-70.1\mu) - 0.0325\exp(-179\mu) \quad (1)$$

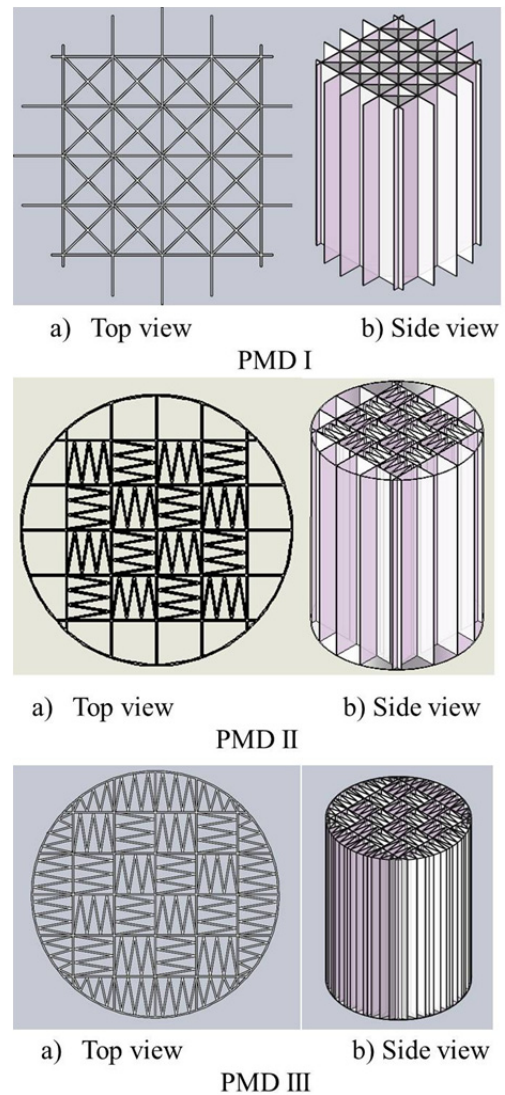


Fig. 1. Schematics of the PMDs.

$$\mu = \frac{nDL}{Q} \quad (2)$$

where η is collection efficiency of the denuder; μ is the dimensionless deposition parameter; D is diffusion coefficient of the penetrating gas or particles; Q is volume flow rate through the entire denuder; and L is the length of the denuder.

For particles, a simplified Gormley-Kennedy Equation was also given by Hinds (1999):

$$\eta = 5.50\mu^{2/3} - 3.77\mu \quad \text{for } \mu < 0.009 \quad (3)$$

$$\eta = 1 - 0.8191\exp(-11.5\mu) - 0.0975\exp(-70.1\mu) \quad \text{for } \mu \geq 0.009 \quad (4)$$

EXPERIMENTAL

Materials

The four types of PMDs were evaluated for their performance. To compare the PMDs' capacity with

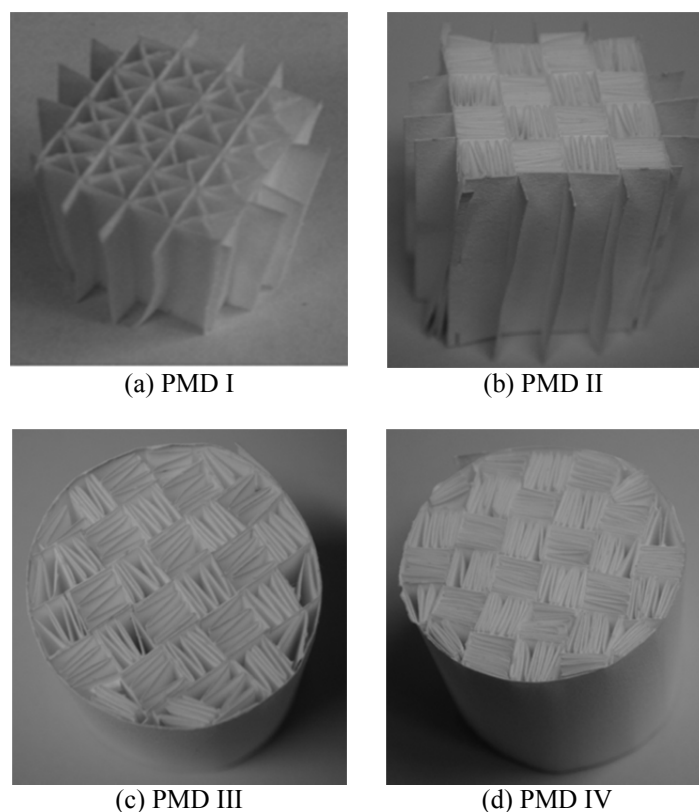


Fig. 2. Photos of prototype PMDs.

Table 1. Properties of 4 different types of PMDs.

Denuder	Diameter (mm)	Height (mm)	Number of Channels	Smallest Channel Opening Area (mm ²)	Largest Channel Opening Area (mm ²)	Weight (g)
PMD Ia	47	35	80	17.26	39.83	6–9
PMD Ib		50	80	17.26	39.83	
PMD II		50	112	10.51	39.83	
PMD III		50	192		8.58	
PMD IV		50	280		5.90	

commercially available denuders, experiments were also carried out with the glass honeycomb denuder (GHD). The GHD is 47 mm in diameter and 38 mm long. It has an internal surface area of 508 cm² which is made possible by 212 hexagonal flow channels that are 2 mm on each side. The GHD has a weight of 106 g.

Coating

Both the PMDs and GHD were coated with sodium carbonate (Na₂CO₃)/glycerin to absorb SO₂ gas. The use of this coating material is well documented in previous research (Perrino, De Santis and Febo, 1990; Perrino and Gherardi, 1999; Wittmaack, 2006; Lin *et al.*, 2010). However, the coating procedure was different in this study due to the special characteristic of the porous membrane denuder. The strength of the porous membrane was much less than that of glass or metal, and the upper operating temperature limitation of cellulose filter is generally in the range of 75°C to 80°C, which means the drying temperature of the

denuder should not be higher than 75°C. Sorbent of known mass was added into deionized (DI) water to make the coating solution. In coating denuders, the first step was to fully immerse each clean denuder into the solution. Then, the container was sealed with aluminum foil and placed into a sonication bath for 30-min. Thereafter, the denuders were taken out of the container using clean forceps and placed on a clean glass tray for drying in a pre-heated furnace with a temperature of 50–60°C for 3 to 5 hours.

Two concentrations, 5% (m/v) Na₂CO₃/glycerin and 10% (m/v) Na₂CO₃/glycerin in DI water were used in the experiments. In Experiment Sets 1 and 2, two PMD Ia were coated with 5% and 10% solution, respectively, to compare the effect of coating concentration on the capacity of the denuder. In Experiment Sets 3 to 6, all types of PMDs and the GHD were coated with 10% solution. Duplicated experiments were carried out for Experiment Sets 1 to 4 and triplicated experiments were carried out for Experiment sets 5 and 6.

Sampling and Analysis

Denuder Collection Efficiency

The experimental system for capacity testing is shown in Fig. 3(a). In Experiment Set 1, target gas, SO₂ supplied as 10 ppm from a cylinder, was mixed with compressed air in a ratio of 1:9 to obtain a concentration of 1 ppm to test PMD Ia's performance in the ambient environment. On the other hand, in testing PMDs' performance in occupational environment, the feed SO₂ concentration used in Experiment Sets 2 to 6 was 10 ppm, which is twice the Permissible Exposure Level (PEL) of SO₂ set by U.S. Occupational Safety and Health Administration (OSHA). The flowrate of the gas stream, 2 L/min was controlled by a mass flow controller (OMEGA, Model FMA 5520). An SO₂ monitor (International Sensor Technology, Inc., Model IQ-350) which can monitor the SO₂ concentration in real-time was connected to the upstream and downstream of the sampler to measure SO₂ gas concentrations. Two impingers in series were connected downstream of the sampler using 9 mM Na₂CO₃ solution to absorb gas that penetrated the denuder. The solution in the two impingers was changed every 30 minutes. Most of the exhaust gas was collected by the first impinger, while the second impinger was used to check whether the first one remained fully functional. After sampling, hydrogen peroxide (H₂O₂) was added to the sample solution to oxidize sulfite to form sulfate. The time weighted average (TWA) concentration of sulfate ions was determined by an IC system (Model ICS-1500, DIONEX Inc.).

The impinger method was used instead of the SO₂ monitor when the feed SO₂ concentration was low (e.g., 1 ppm), because the detection limit of the SO₂ monitor is 0.1 ppm, which does not have the resolution needed for low SO₂ concentration. For real-time gas monitoring using the SO₂ monitor, the collection efficiency (*Eff*) of SO₂ can be obtained by measuring the feed concentration (*C_u*) upstream and the exit concentration (*C_d*) downstream of the holders. *Eff* at any given time can be calculated by the following equation:

$$Eff = \frac{C_u - C_d}{C_u} = 1 - \frac{C_d}{C_u} \quad (5)$$

If the impinger method was used for measuring downstream gas concentration, the exit gas concentration in Eq. (5) was the summation of the concentrations in impinger 1 and impinger 2.

The consumption of Na₂CO₃ was calculated according to the amount of SO₂ adsorbed. First, the time weighted average (TWA) collection efficiency (*Eff_{TWA}*) of PMDs was determined using the real-time monitor measurements. Then, the total amount of SO₂ collected by the denuder can be determined accordingly. Since the exhaustion of Na₂CO₃ follows the stoichiometric reaction between Na₂CO₃ and SO₂,



the amount of consumed Na₂CO₃ can then be estimated accordingly.

In this study, the performance of the denuder was only tested in laboratory. Since inlet SO₂ concentration is unknown in field, two denuders in series can be used with the second denuder used to check the collection efficiency of the first denuder.

Particle Loss

Fig. 3(b) shows the experimental setup for measuring particle loss of the PMDs. A vibrating orifice aerosol generator (VOAG, Model 3450, TSI Inc.) was used to generate monodisperse particles. The droplet size depends on orifice size, solution feed rate, operating frequency and solution concentration. The aerosol solution was composed of oleic acid as the non-volatile solute and isopropyl alcohol as the solvent. The particles thus generated were dispersed with compressed air and went through a Kr-85 neutralizer column to be dried and neutralized. The airflow, 50 L/min,

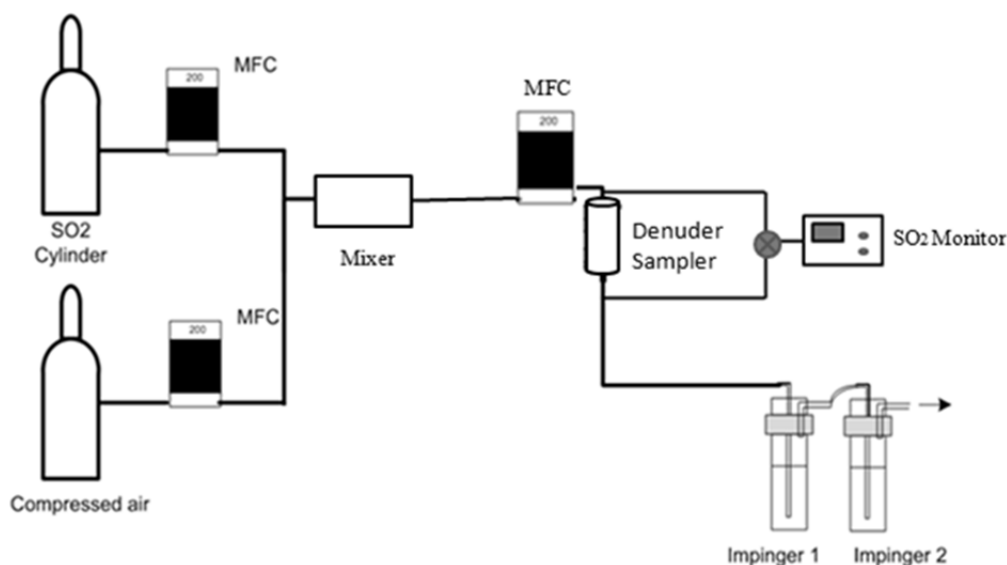


Fig. 3(a). Experimental setup for testing aerosol collection efficiency and particle loss.

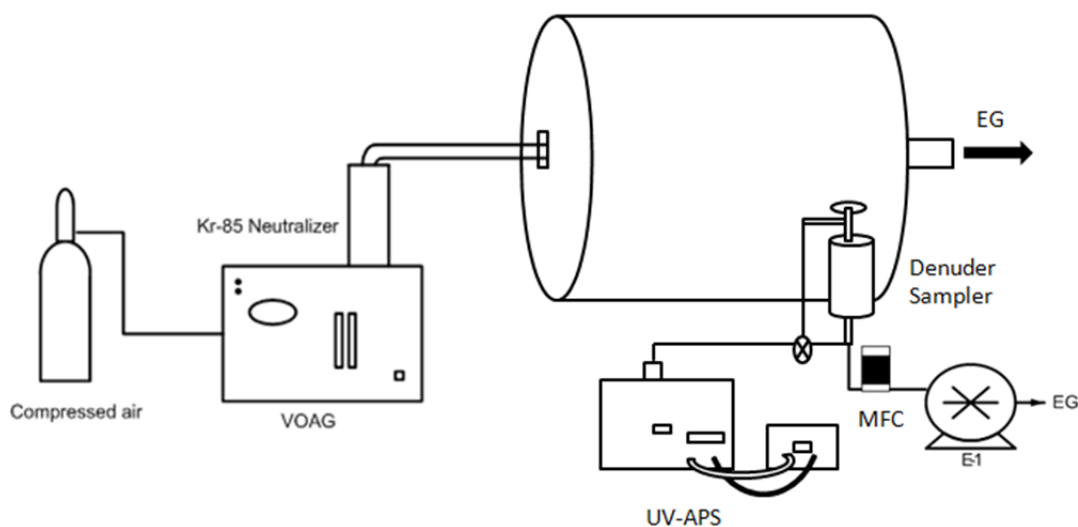


Fig. 3(b). Experimental setup for measuring particle collection efficiency of the impactor.

then went through the hose and entered one end of the chamber and exited the other end of the chamber. The aerosol chamber was made of stainless steel, and the cylinder was approximately 40.64 cm in diameter and 71.12 cm height. The corresponding air velocity through the chamber was about 6 mm/s, i.e. calm air conditions were applied in the test chamber (Trakumas and Salter, 2009). The denuder sampler was vertically installed in the chamber and connected with a mass flow controller and a pump outside. Sampling flowrate of the sampler was controlled to be 2 L/min by the mass flow controller. An ultraviolet aerodynamic particle sizer (UV-APS, TSI Model 3312) was connected to upstream and downstream of the sampler to measure the particle size distribution. To examine particle loss in the tubing and joints of the system, an empty sampler was also tested as a control, the result of which was used as a baseline for the PMDs.

RESULTS AND DISCUSSION

Denuder Collection Efficiency

The collection efficiency of a denuder can be predicted by Eq. (1) with a known diffusion coefficient of the target gas. Fig. 4 displays predicted collection efficiency versus denuder length of different denuder types of SO_2 gas. It can be seen that when the denuder length is greater than 1.5 cm, the collection efficiency of all types of denuder for SO_2 is close to 100%. However, because the reagent on the channel wall will be consumed during sampling, the capacity of the denuder will decrease as sampling time increases, and the assumption that the tube surface serves as a perfect sink for Eq. (1) becomes invalid. Other factors, such as incoming gas concentration and the amount of reagent coated on the denuder are also important factors that affect the denuder's capacity.

For PMD I, as shown in Fig. 5, when a feed SO_2 concentration of 1 ppm was used, analysis of all the samples obtained from impingers 1 and 2 downstream shows that the 30-minute average exhaust gas concentration was below 0.01 ppm during the entire 8 hours. The low exhaust

concentration means that the denuder has collection efficiency higher than 99.9% for the entire eight hours. According to U.S. Environmental Protection Agency (USEPA), the average ambient concentration of SO_2 was lower than 0.01 ppm from 1990 to 2009; Chen *et al.* (2011) reported the same ambient level of SO_2 in Taiwan. Our results demonstrate that the PMD has a capacity that is high enough to be used for ambient SO_2 sampling.

Under a feed concentration of 10 ppm, the collection efficiency of PMDs decreases quickly with time. The decreasing rate is opposite to the number of channels. As shown in Fig. 5, increasing the length of the denuder (Set 3 vs. Set 2) increases the efficiency, as it increases the amount of reagent and allows a longer residence time for gas molecules to diffuse to the wall and react with the reagent. Increasing the coating concentration (Set 2 vs. Set 1) also helps increase the overall collection efficiency. The reason is due to the finite capacity of the sorbent. In general, the more sorbents coated on the surface of the denuder (e.g., thicker coating), the more gas molecules can be trapped by the denuder. Meanwhile, the higher the incoming gas concentration, the quicker the sorbents become exhausted. Sorbents at the top of the coating react with target gas to form a different substance which covers the top layer of the coating. In this study, sulfur dioxide reacts with sodium carbonate to form sulfite which slowly oxidizes further to sulfate. This top layer becomes a barrier for the target gas to reach unreacted sorbents inside. Therefore, even if some reagents have not been consumed yet, the denuder's collection efficiency still decreases with time. Accordingly, a proper coating concentration should be selected according to designated capacity of the denuder.

Currently, there is no theoretical model that can be applied directly to the PMDs to predict the relationship between gas adsorption capacity and coating thickness. Surface condition of the porous membrane plays an important role in the process, but such information is not readily available. Research to develop such a model that can accurately describe the diffusion process and sorbent consumption is warranted.

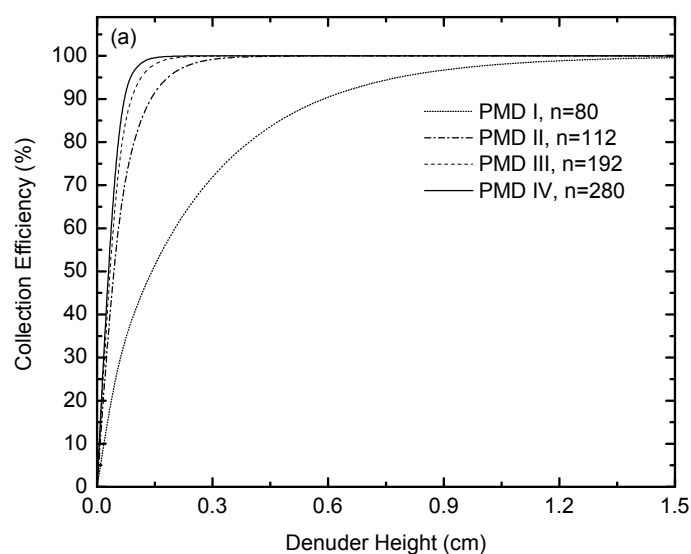


Fig. 4. Collection efficiency of different types of denuders (predicted gas: SO_2) vs. denuder height.

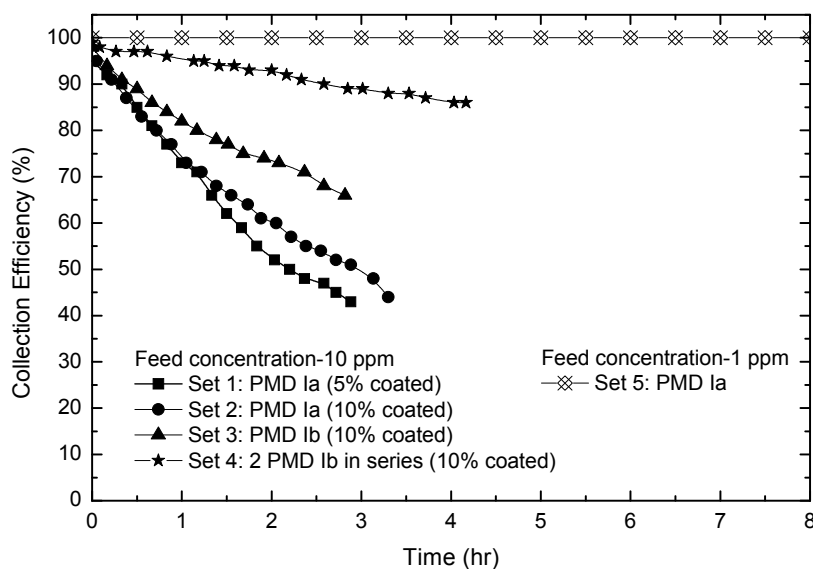


Fig. 5. SO_2 removal efficiency of PMD I as a function of time.

The 3-hr time weighted average (TWA) collection efficiency of Set 1 to Set 4 was about 68.2%, 73.4%, 81.3% and 94.6%, respectively, as shown in Table 2. The corresponding 3-hr consumption of Na_2CO_3 according to Eq. (6) was in the range of 10.4 to 14.4 mg for these 4 sets of experiments. The consumption was about 0.2% of the total coating, indicating the capacity had not been effectively utilized. It indicates that if a longer residence time is allowed for SO_2 gas to travel through the denuder and react with Na_2CO_3 , a higher efficiency and longer breakthrough time can be expected. Either increasing denuder length or decreasing gas flowrate can both achieve this goal, e.g., two denuders arranged in series as in Set 4.

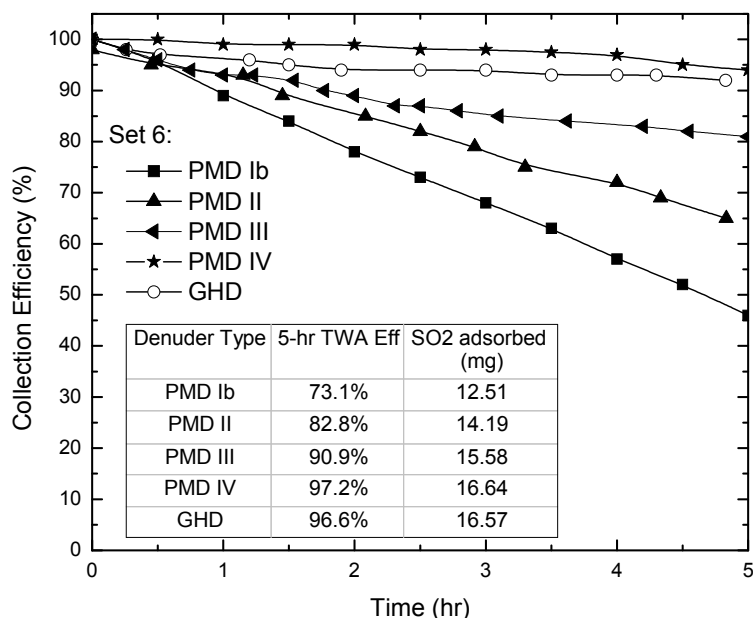
The result in comparing the collection efficiencies of the four different types of PMDs and the GHD are shown in Fig. 6. It can be observed that as the number of channels increases and channel cross-sectional area decreases, the

capacity of the denuder increases. Uniformity can also affect PMDs' performance. For PMD I and II, the grid density at the edge was lower than that at the center, causing overall lower performance. Therefore, PMD III and IV were designed in a more uniform grid for the entire cross-section to increase collection efficiency.

5-hour TWA collection efficiencies of PMD Ib, II, III, and IV were 73.1%, 82.8%, 90.9% and 97.2%, respectively, while that of the GHD was 96.6%. Compared with the GHD, PMD IV has a slightly higher collection efficiency. However, the weight of the GHD is about 10 times greater than that of the PMD IV. In addition, since the PMDs are made of filter paper, they are relatively cheap and can be disposable. Therefore, the PMDs have the potential to replace traditional denuders made of glass or metals for applications where light weight and low cost are important features.

Table 2. Experimental results of Sets 1-4 of PMD I.

No.	3-hr TWA Collection Efficiency	Amount of Absorbed SO ₂ (mg)	Amount of Consumed Na ₂ CO ₃ (mg)
Set 1	68.2	6.3	10.4
Set 2	73.4	6.7	11.2
Set 3	81.3	7.5	12.4
Set 4	94.6	8.7	14.4

**Fig. 6.** SO₂ removal efficiency of GHD and PMDs as a function of time.

Particle Loss

The mass based particle loss percentage of each type of PMDs is shown in Fig. 7. The particle size range selected for testing was from 1 to 10 μm . Particles larger than 10 μm usually should be removed by an upstream impactor or cyclone to avoid large impaction loss on the denuder. For particles smaller than 1 μm , although the diffusion effect will be enhanced, the total mass usually accounts for a very small percentage of all the particle loss. As shown in Fig. 7, as particle size increased, particle loss on all four types of PMDs increased. For larger particle size that was generated by the VOAG, particle number concentration was lower than that of smaller particle size. Accordingly, relative concentration fluctuation was larger compared with that of smaller particles. Due to impaction, large oleic acid particles were broken into smaller particles of various sizes. Some of them were collected on the denuder while the rest of them penetrated, leading to a varied downstream particle size distribution. These two factors resulted in an increased overall variance of particle loss as particle size increased. For the same particle size, particle loss increases as channel opening area decreases. Since PMD II and PMD III had the same channel opening in the center field, their particle loss tendencies were similar. The average particle losses (1–10 μm) of PMD I to PMD IV were 2.9%, 5.2%, 5.7% and 7.3%, respectively.

Particle loss in the denuder system usually occurs in four ways: diffusion across the denuder, impaction at the tube inlet,

gravitational sedimentation and interception. Sedimentation can be easily avoided by setting the denuder in the vertical position while interception usually causes low particle loss ($< 1 \mu\text{m}$) (Hinds, 1999). The loss due to diffusion is usually quite small, since gases usually have diffusion coefficients much higher than those of particles. The loss due to diffusion can also be calculated using Eqs. (1) to (3). With a gas flowrate of 2 L/min, diffusional collection efficiency/loss on the PMDs of SO₂ gas and different sized particles is shown in Table 3. As shown, the loss is smaller than 0.28% for any particle between 1–10 μm . Impaction is the most important mechanism for large particles. The increasing particle loss tendency with increasing particle size in the experiments also proves that particle loss on the denuder is mainly due to inlet impaction, which is consistent with published research (Possanzini *et al.*, 1983; Ferm, 1986; Koutrakis *et al.*, 1988).

Particle loss due to impaction can be reduced by optimizing the inlet geometry and fluid dynamic conditions. Alternatively, an elutriator, impactor or cyclone inlet with a mass median aerodynamic diameter (MMAD) of 2.5 mm cut size is commonly added in front of a denuder system to overcome problems of impaction loss of large particles on the denuder (Koutrakis *et al.*, 1988; Ianniello *et al.*, 2007).

CONCLUSIONS

A novel porous membrane denuder has been developed.

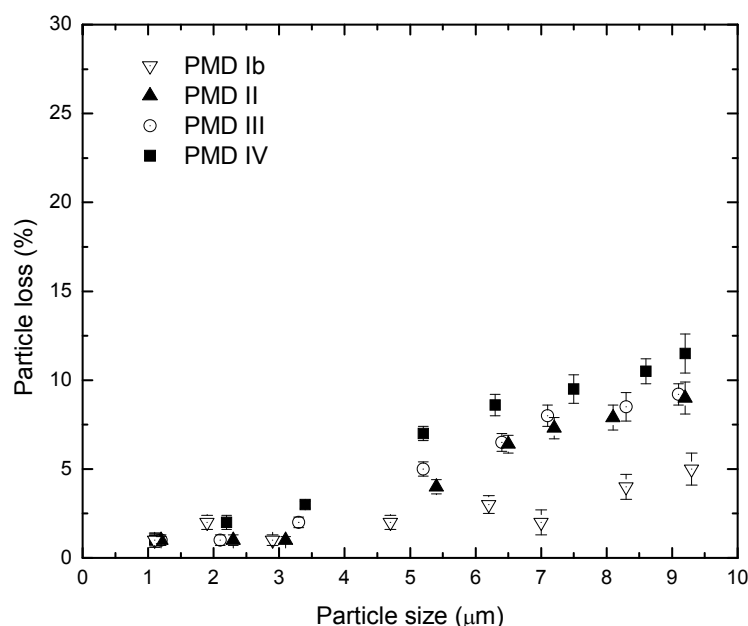


Fig. 7. Particle loss of PMDs in the size range of 1 to 10 μm .

Table 3. Theoretical diffusional collection efficiency of gases and particles by the PMDs.

Gases/Particles	Diffusion Coefficient (cm^2/s , 20°C, 1 atm)	Collection Efficiency (%)			
		PMD I	PMD II	PMD III	PMD IV
SO_2	1.370×10^{-1}	100	100	100	100
0.001 μm	5.228×10^{-2}	99.94	100	100	100
0.01 μm	5.329×10^{-4}	17.68	20.91	28.03	34.66
0.1 μm	6.865×10^{-6}	1.01	1.26	1.79	2.29
1 μm	2.817×10^{-7}	0.12	0.15	0.22	0.28
10 μm	2.419×10^{-8}	0.02	0.03	0.04	0.06

The characteristics of high capacity, light weight and disposability enable PMDs to be a great tool for personal sampling of gases in the ambient environment and occupational settings. Tested with a flow rate of 2 L/min and a feed SO_2 concentration of 1 ppm, PMD Ia has a collection efficiency higher than 99.9% for 8 hours, which demonstrates that the PMD has high enough capacity to be used in ambient SO_2 sampling. Further experiments of PMD I showed that coated reagent amount and denuder length are both very important factors. In addition, by increasing the number of channels and decreasing the opening area of channels, PMDs' capacity increases accordingly. With a feed concentration of 10 ppm, PMD IV, which has the highest number of channels and smallest channel opening areas, achieves a 5-hour time weighted average collection efficiency of 97.2%; slightly higher than 96.6% of the glass honeycomb denuder of similar matrix. However, the PMD weighs less than 9 g, which is only one-tenth of the glass honeycomb denuder.

Particle losses of four types of PMDs with the same length and diameter are 2.9%, 5.2%, 5.7% and 7.3%, respectively for 1–10 μm particles. Most particle losses in this size range are caused by impaction at the inlet of the denuder. An impactor or cyclone arranged before the denuder can remove the majority of large particles and reduce particle loss in the denuders.

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