

Effect of phase of toluene on filtration performance of electret filter media against di-octyl-phthalate aerosols

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ABSTRACT

Performance degradation of electret filter media when exposed to toluene in a laboratory setting was investigated using a custom-made exposure apparatus. Three types of electret filter media were exposed to toluene in liquid and gas phases. Experimental data indicated that the penetration percentage of di-octyl-phthalate (DOP) aerosols through electret filter media was not influenced by exposure to toluene vapors for periods up to eight hours. The filtration performance for all three types of electret filter media decreased more than 30% when the samples were exposed to controlled amounts of liquid toluene. Pressure drop changes of the filter media prior and after exposure to liquid toluene were statistically negligible. Neither morphological changes nor degradation of the polymer fibers were observed after exposure of the filter media samples to liquid toluene. Thermodynamic simulations were performed to determine the molar flux of liquid and gas through the filter media specimens. Experimental results obtained via capillary gas chromatography were within 6% of the predicted toluene concentration values. The decrease in performance was attributed to changes in the density and spatial distribution of the electret charges on the surface of the polymer fibers.

Keywords: Electret filter media, toluene, liquid exposure

INTRODUCTION

The effects of permanent static electric fields on filtration performance have been well established in the scientific literature [Chen and Huang, 1998; Brown, 1992; Brown et al., 1998; Brown, 1981; Ando et al., 1990]. It is also well established that electret filter media (polymeric fibers containing trapped electrical charges) exhibit better performance for particles in the 0.5-3 μ m particle diameter range and lower pressure drop than mechanical filter media (uncharged fibers). Two theories have been advanced to explain the location and strength of the permanent electric field: a phenomenological theory, in which various polarization mechanisms play the primary role and charge injection is secondary, and the charge injection theory, where the electric field is caused by the external injection of charged particles, which may induce permanent electric dipoles [Yovcheva et al., 2004]. In either case, it is known that the charge effect is localized to the surface of the polymeric material, perhaps to the outermost two or three nanometers.

Previous laboratory studies [Biermann et al., 1982; Jasper et al., 2005] have shown that direct exposure to liquid organic solvents decrease filter media performance. However, some of these results have been under controversy as the experimental conditions were not properly controlled. It is the objective of this research work to determine the effect of exposure to toluene (a common organic solvent recognized as a neurotoxin by NIOSH), in gas and liquid phases, on the performance of electret filter media under controlled experimental conditions.

METHODS

Materials

Three different types of electret filter media as well as one type of mechanical filter media were used in this study. Technical specifications for these materials are shown in Table I. All filter media were tested as received from the manufacturers. The electret filter media was composed of meltblown polypropylene nonwoven mats charged by means of corona treatment. The mechanical filter media consisted of a nonwoven mat of glass fibers without electrostatic charge. The chosen materials are typically used as a component of commercially available particulate respirators rated by NIOSH as R, P and N.

ACS grade toluene and di-octyl-phthalate (DOP) (also known as diethylhexyl phthalate) was obtained from Sigma Aldrich (Milwaukee, WI) and used as received. Commercial grade nitrogen was used as the carrier gas, and it was provided by Machine and Welding Supply Company (Dunn, NC).

Analytical Instrumentation

A CertiTest 8130 (TSI Incorporated, St. Paul, MN) automated filter tester was used to determine the performance (percentage of penetration) and pressure drop across the filter media. In this work, DOP was used in the oil aerosol generator to produce particles with a count median diameter of 0.2 μm and a geometric standard deviation of less than 1.6. The filter media were tested as flat circular samples with a 15.2 cm diameter. All the tests were done according to the criteria established in 42 CFR 84 [NIOSH, 1995] for challenging respirator filter media at a flow rate of 42.5 lpm.

The electrostatic potential at a distance of 25 mm was measured using a SIMCO FMX-002 Electrostatic Fieldmeter (SIMCO, Hatfield, PA). The units of the field meter are given in kV. The instrument is calibrated with a measurement error of $\pm 10\%$.

A gas chromatograph (GC-17A Shimadzu Instruments, Columbia MD) equipped with a 30 m capillary column, 0.53mm ID (Stabilwax® crossbond® carbowax®-PEG) and a flame ionization detector was used to determine the concentration of toluene in the gas entering and leaving the exposure apparatus. The accuracy of the method was corroborated using calibrated mixtures certified by The Specialty Gas Group (Raleigh, NC).

Table I. Filter Media Specifications

Test Code	Basis Weight (g/m^2)	Thickness (mm)	Fiber	Charge
E1	112	0.99	Meltblown Polypropylene	Corona
E2	185	1.78	Meltblown Polypropylene	Corona
E5	80	0.76	Meltblown Polypropylene	Corona
Mechanical	78	0.40	Microglass	N/A

Filter Media Testing

Filter media testing was carried out in accordance with 42 CFR 84 Subpart K, which establishes performance criteria for all negative pressure particulate air-purifying respirators and pre-filters. The filter media was placed on the lower portion of the filter holder, exposing an area of 182.4 cm^2 to the test agent. The DOP aerosol traveled from the generator to the filter holder and down through the filter media. Two solid-state laser photometers measured the aerosol concentration upstream and downstream of the tested filter media. Filter penetration was determined from the ratio of the two aerosol concentration

measurements. Electronic pressure transducers located upstream and downstream of the test holder were used to determine pressure drop across the filter media. All the signals and values were time stamped and stored in a SQL database.

Exposure Apparatus

Nitrogen was bubbled through a three nozzle rounded vessel containing liquid toluene in order to generate a saturated vapor. The mole fraction of toluene in the saturated vapor stream was determined to be 0.037. Gas flow through the saturator was controlled at $1000 \text{ cm}^3 \text{ min}^{-1}$ using a flow-meter. The stream of saturated vapor was connected to the exposure chamber using Tygon tubing. By measuring the gas concentrations up and down stream of the filter sample and the flow rate, one can calculate from mass conservation the molecular fraction of gas in the exposed chamber and the rate of liquid toluene deposition onto the filter media. This value is temperature dependent, as the toluene in the gas phase precipitates onto the fiber when cooled below its dewpoint temperature.

The vapor exposure chamber was a custom design that was fabricated by Prism Research Glass, Inc. (Research Triangle Park, NC). The vessel was a round tube closed on one end with a spherical bottom (approximately 25 cm in diameter and 32 cm tall). A base was molded to the tube so that the chamber would be free standing. A shelf was pressed into the edges of the glass tube that would serve, in conjunction with a matching glass ring, as means to suspend the sample materials in the middle of the vessel. Two nozzles were attached to the vessel in opposite sides, one above the shelf and one below. The top closure was achieved by adding a flange and grinding the surface to match a ground-glass lid. The interface was sealed with inert grease. The exhaust of the exposure chamber was connected to a common ventilation system and was intermittently sampled with the gas chromatograph to verify the toluene concentration. The temperature of the exposure apparatus and the gas bubbler was controlled at 25 degrees Celsius. A schematic of the experimental setup is shown in Figure 1.

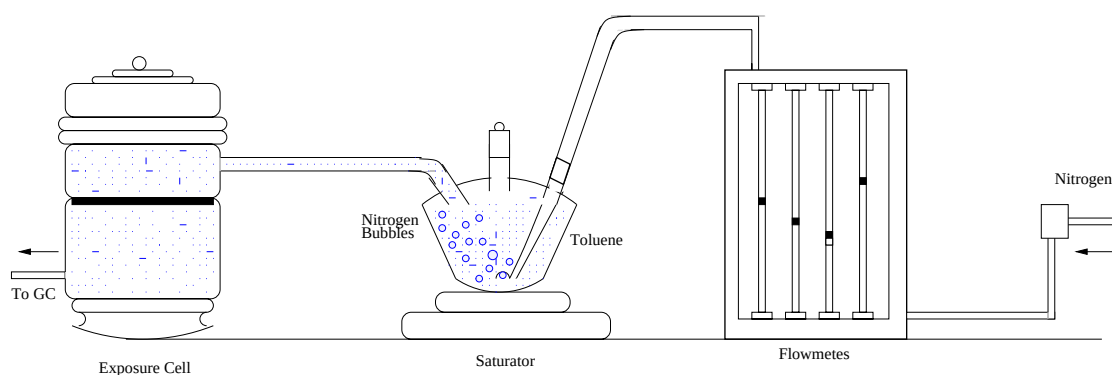


Figure 1. Experimental Setup for Chemical vapor exposure tests.

Exposure Experiments

The purpose of this work was to evaluate the performance of electret filter media against a worst case aerosol after being exposed to toluene in gas and liquid phases. In order to generate two-phase flow, dry-ice packs were placed at the entrance nozzles of the exposure chamber. Four sets of experimental conditions were envisioned to decouple the effect of exposure to the gas and liquid phases of toluene.

The first set of experimental conditions introduced the saturated gas into the exposure apparatus using the upper nozzle and exhausting through the lower one. This set of experimental conditions was labeled as *downward flow*. The second set of experimental conditions introduced the saturated gas into the exposure chamber through the lower nozzle and exhausted through the upper nozzle, and will be referred in this paper as *upward flow*. The third experimental condition resembles the downward flow experimental

condition with the difference that dry-ice packs were placed around the outside of the upper chamber in order to generate a two-phase flow. This condition is referred to as *ice-bath downward flow*. The fourth experimental condition introduced the saturated gas into the exposure chamber using the lower nozzle and exhausted through the upper nozzle like condition two, except that dry-ice packs were placed around the lower chamber. This condition is referred to as *ice-bath upward flow*.

In all cases, the filter media was first exposed for a predetermined amount of time to toluene (for example 1, 2, 4 or 8 hours), and then tested for filtration performance with the TSI 8130. After testing with DOP, samples were not further exposed to toluene.

During experimental conditions three and four (ice-bath downward and ice-bath upward flow), liquid toluene was observed at the bottom of the exposure chamber confirming the presence of a two-phase flow. It was evident that only during experimental condition three (ice-bath downward flow) was the filter media directly exposed to liquid toluene. In order to determine the flux of toluene liquid and gas through the exposure chamber, thermodynamic simulations were performed.

In order to accurately determine the toluene concentration after the bubble-saturator as well as the flux of liquid and gas phases inside the exposure chamber, thermodynamic simulations were performed using NRTL (Non-Random Two Liquids), UNIFAC (Universal Functional Activity Coefficient) and NRTL-RW (Non-Random Two-Liquids-Redlich-Kwong) activity coefficient models. These models have been extensively used in the past to model the activity of organic vapors in inert gases.

The NRTL (Non-Random Two Liquid) model uses three binary interaction parameters (BIPs) for each binary pair in a multi-component mixture. UNIFAC is based on chemical group contributions, precluding the need for molecule/molecule interaction parameters. NRTL-RW uses the NRTL algorithm adding the RW equation of state to predict phase equilibrium parameters as well as non-ideal mixture properties for each phase.

Experimental values for toluene concentration were determined using the gas chromatograph. The predicted values were around $\pm 6\%$ of the experimental values, indicating good quantitative agreement between the prediction of the models and the actual experimental value.

Table II shows the molar flux and molar concentration for toluene at the saturator and the exposure chamber. As the temperature inside the chamber decreased due to the presence of dry-ice around the top or bottom of the exposure chamber, a two phase environment was experimentally observed. All three thermodynamic models used in this work predicted a multiphase environment in agreement with experimental observations. The variation among the predicted values by the three models used was less than 3%.

Table II. Predicted Molar Flux and Concentration of Toluene

Model	Molar fraction of toluene from the saturator	Flux of toluene gas to the exposure chamber [mols/sec]	Flux of toluene liquid in the exposure chamber [mols/sec]	Molar fraction of toluene gas in the exposure chamber
NRTL	0.0375	0.00523	0.00689	0.0165
UNIFAC	0.0381	0.00530	0.00701	0.0168
NRTL-RK	0.0381	0.00532	0.00701	0.0168
Experimental	0.0401			0.0171

RESULTS

A total of 80 experiments were carried out using DOP aerosol as a test agent. Four repetitions were performed at each one of the four experimental conditions for the filter media selected for this study. In addition, four repeats for each filter media (not exposed to toluene) were carried out in order to establish the performance and resistance baselines.

Baseline Tests

Figure 2 illustrates the baseline performance and pressure drop for filter media not exposed previously to toluene. As expected, electret filter media E1, E2, and E5 exhibited lower pressure drop than the mechanical filter media.

While E1, E2 and the mechanical filter media samples exhibited negligible changes in penetration as a function of time, E5 exhibited a monotonic increase in penetration as a function of time. This behavior was expected, as E5 is not designed to filter oil aerosols.

Exposure Tests

Figures 3-5 show the performance of filter media as a function of exposure time under the four experimental conditions stated previously. For all three electret filter media specimens, E1, E2, and E5, a marked decrease in performance (increase in penetration percentage) was noted for experiments performed under experimental condition three (ice-bath downward flow). Negligible changes in performance were noted for experiments carried out under experimental conditions one, two and four. No change in performance was detected for the mechanical (uncharged) filter for all four experimental conditions.

Experimental condition three, ice-bath downward, is the only one in this study in which liquid toluene is in direct contact with the filter media. Liquid toluene was observed at the bottom of the exposure chamber during these experiments as the presence of two phase flow was also predicted by the thermodynamic simulations. During experimental condition four, liquid toluene was observed at the bottom of the exposure chamber, but due to the direction of the flow, the liquid was not in direct contact with the filter media.

Since an increase in aerosol penetration was only noted when the filter media was directly exposed to toluene in the liquid phase, the decrease in performance could be attributed to a reduction in the filter media's static electric field caused by the creation of a thin layer of liquid toluene over the surface of the charged polymer fibers. The exact spatial location and distribution of the net charge is an open question in the literature. An upper limit on the surface charge density can be shown to be less than 8 atoms per million that reside on the surface of the fiber [Cross, 1987]. When toluene plasticizes the surface of the polypropylene fiber, electron mobility increases and charge transfer can occur. This change is confirmed by the significant decrease in the bulk electrostatic field measurement as shown in Table III. Furthermore, no significant changes in the electrostatic field measurements were noted when the specimens were exposed to toluene vapors as indicated in Table III. This behavior may be explained by the large affinity between toluene and polypropylene as expressed by their solubility parameters (18.30

and $16.81 \text{ MPa}^{\frac{1}{2}}$) [Michaels et al., 1969]. The closeness of the solubility parameters is consistent with a rapid sorption of the toluene vapor into the polypropylene matrix preventing the formation of a conformal liquid layer and hence not affecting the electrostatic charge on the specimens as indicated in Table III. It is evident from Figures 3-5 that toluene in the gas phase did not alter the performance of electret filter media for exposure times up to eight hours. This behavior may be explained by the fact that the sorption and diffusion process for toluene into polypropylene occurs at different time scales with diffusion being the rate-limiting step.

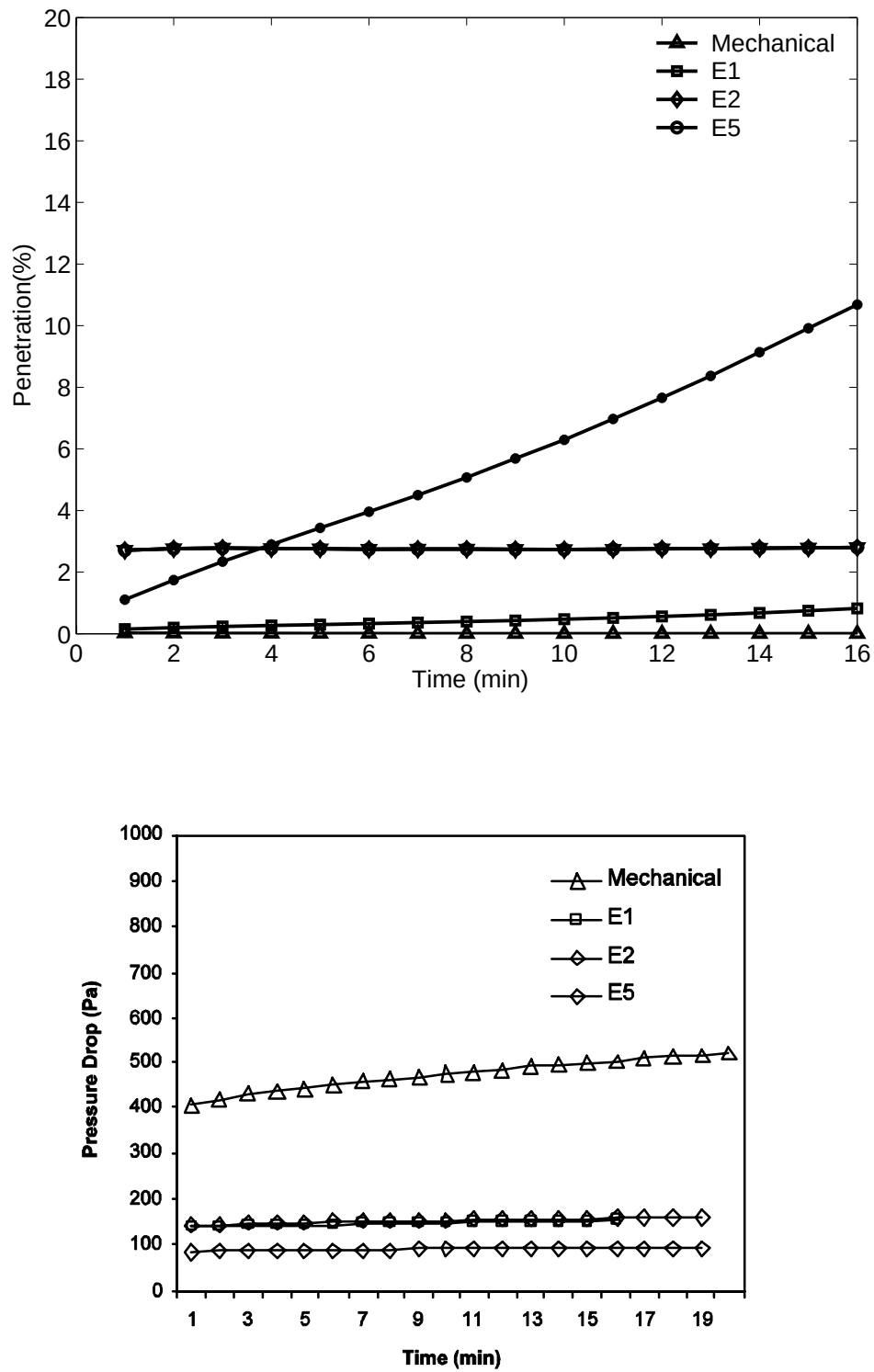


Figure 2. Baseline performance data.

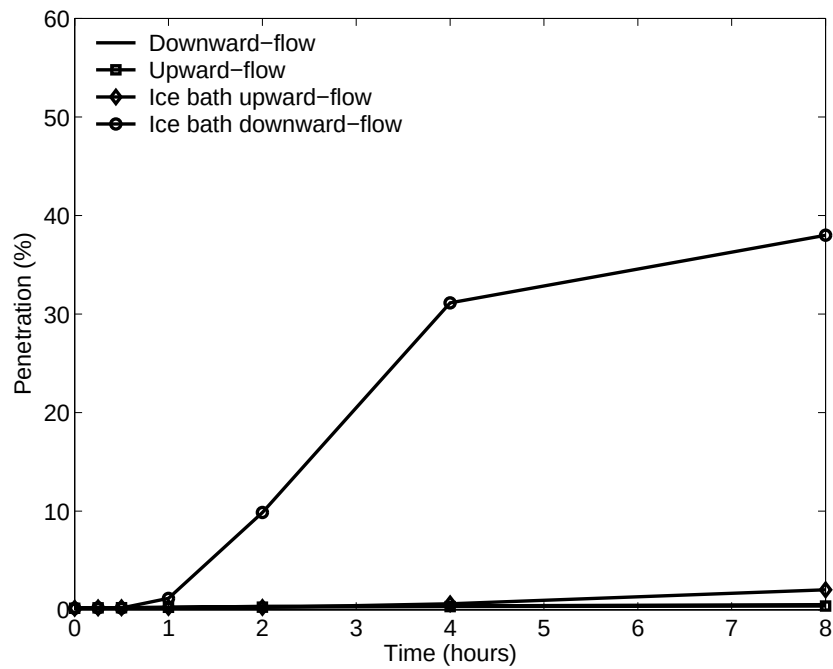


Figure 3. Experimental results of E1 exposure to toluene.

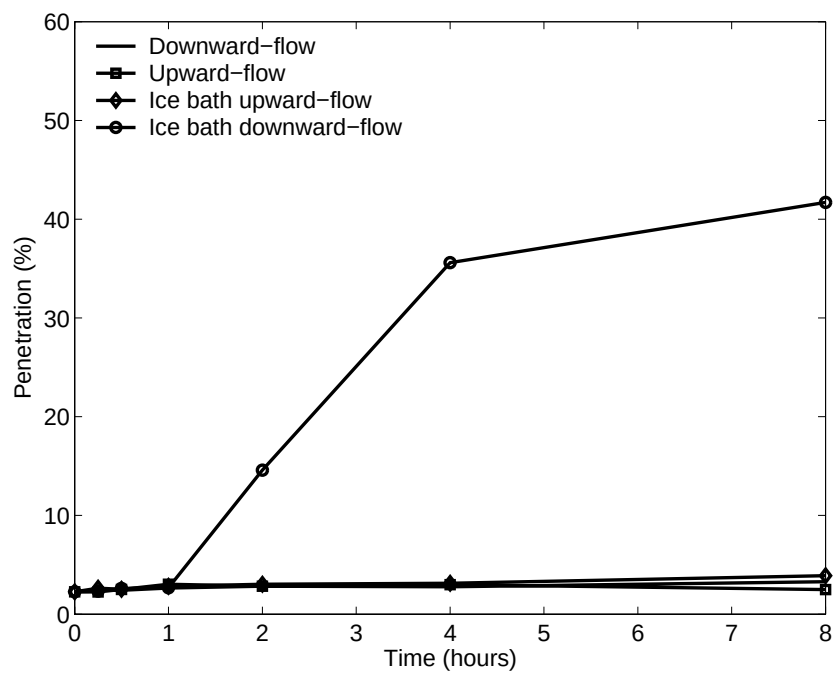


Figure 4. Experimental results of E2 exposure to toluene.

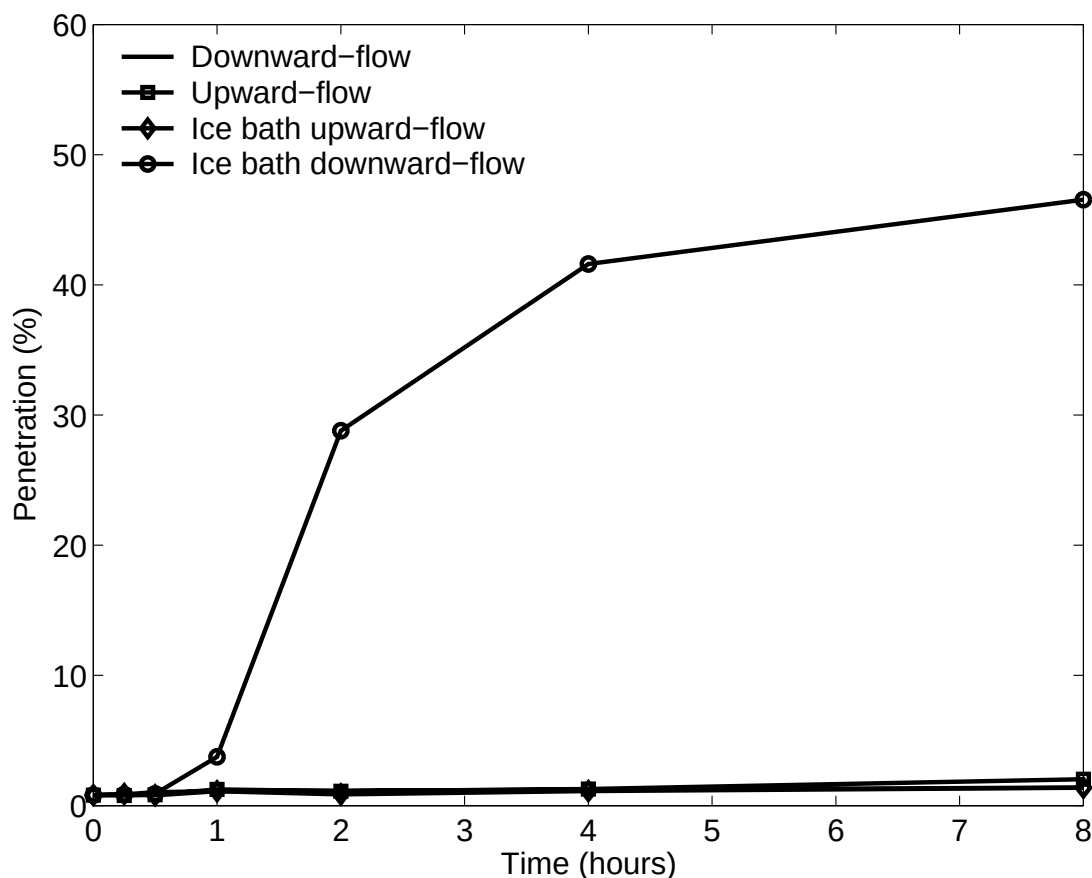


Figure 5. Experimental results of E5 exposure to toluene.

Table III shows the average of 5 measurements of the static electric field taken 25 mm above the surface of the filter media before and after exposure to toluene in the liquid and vapor forms. The filter media consists of a middle meltblown layer between two spunbound layers. Measurements were taken on the middle meltblown layer. For the case of E1 and E2, the polarity was opposite when measured from the top and bottom. The core of E5 consists of 2 meltblown layers that were combined such that the top and bottom surfaces both have negative potential.

Table III. Electrostatic Field Measurements

Sample	Unexposed specimen (kV)	Filter media exposed to toluene specimen (kV)	Filter media exposed to toluene liquid (kV)
E1	2.80	2.52	0.39
E2	1.15	1.00	0.23
E5	-2.18	-1.86	-0.26

CONCLUSIONS

A set of experiments were conducted to determine the effect of exposure to toluene on the performance of electret filter media. It was established that the penetration percentage of DOP aerosol through the electret filter media was not affected by exposure to toluene vapors for up to eight hours. Direct contact of electret filter media with toluene in the liquid phase caused a decrease in filtration performance up to 40% when tested under worst case conditions. The decrease in performance was attributed to changes in the static electric field caused by the presence of liquid toluene on the surface of the fiber. The performance of the mechanical filter media was unaffected by the exposure to toluene in either the liquid or gas phases.

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