

Total Inward Leakage of Nanoparticles Through Filtering Facepiece Respirators

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Nanoparticle (<100 nm size) exposure in workplaces is a major concern because of the potential impact on human health. National Institute for Occupational Safety and Health (NIOSH)-approved particulate respirators are recommended for protection against nanoparticles based on their filtration efficiency at sealed conditions. Concerns have been raised on the lack of information for face seal leakage of nanoparticles, compromising respiratory protection in workplaces. To address this issue, filter penetration and total inward leakage (TIL) through artificial leaks were measured for NIOSH-approved N95 and P100 and European certified Conformer Europe-en-marked FFP2 and FFP3 filtering facepiece respirator models sealed to a breathing manikin kept inside a closed chamber. Monodisperse sucrose aerosols (8–80 nm size) generated by electrospray or polydisperse NaCl aerosols (20–1000 nm size) produced by atomization were passed into the chamber. Filter penetration and TIL were measured at 20, 30, and 40 l min⁻¹ breathing flow rates. The most penetrating particle size (MPPS) was ~50 nm and filter penetrations for 50 and 100 nm size particles were markedly higher than the penetrations for 8 and 400 nm size particles. Filter penetrations increased with increasing flow rates. With artificially introduced leaks, the TIL values for all size particles increased with increasing leak sizes. With relatively smaller size leaks, the TIL measured for 50 nm size particles was ~2-fold higher than the values for 8 and 400 nm size particles indicating that the TIL for the most penetrating particles was higher than for smaller and larger size particles. The data indicate that higher concentration of nanoparticles could occur inside the breathing zone of respirators in workplaces where nanoparticles in the MPPS range are present, when leakage is minimal compared to filter penetration. The TIL/penetration ratios obtained for 400 nm size particles were larger than the ratios obtained for 50 nm size particles at the three different flow rates and leak sizes indicating that face seal leakage, not filter penetration, contributing to the TIL for larger size particles. Further studies on face seal leakage of nanoparticles for respirator users in workplaces are needed to better understand the respiratory protection against nanoparticle exposure.

Keywords: aerosols; face seal leakage; filter penetration; manikin; nanoparticles, N95 respirator

INTRODUCTION

Nanoparticle is defined as a nano-object with all three external dimensions in the size range from ~1 to 100 nm (ISO/TS, 2009). Nanoparticles are generated by various natural as well as industrial pro-

cesses (Biswas and Wu, 2005). Nanoparticles have unique properties, which differ from the bulk form of the same materials. The recent increase in the production of engineered nanoparticles in workplaces has raised concern on their effects on human health and the environment. Exposure to different levels of nanoparticles has been shown to produce adverse health effects (Schulte *et al.*, 2008). With the increase in production of nanoparticles, high-level

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exposure in some workplaces can be expected (Bello *et al.*, 2009; Johnson *et al.*, 2010). Human exposure to particulates is primarily reduced by implementing engineering and administrative controls. When these measures are insufficient or not installed, respiratory protection is required to reduce the inhalation of harmful particles.

In the USA, >3 million workers are required to wear respirators in 282 000 establishments (BLS/NIOSH, 2003). Workplaces that have an OSHA mandated respiratory program in place are required to use NIOSH-approved respirators (OSHA, 2008). Many workplaces select disposable filtering face-piece respirators (FFRs) for their availability and low cost. Several studies reported that NIOSH-approved FFRs provided expected levels of filtration efficiency against polydisperse and monodisperse aerosols >20 nm size (Qian *et al.*, 1998; Willeke and Qian, 1998; Martin and Moyer, 2000; Rengasamy *et al.*, 2007). Many studies have reported that the most penetrating particle size (MPPS) for many NIOSH-approved and European certified Conformer Europe (CE)-marked FFRs were in the 30 to 60 nm size range (Martin and Moyer, 2000; Balazy *et al.*, 2006; Huang *et al.*, 2007; Rengasamy *et al.*, 2008, 2009). Recent studies also showed that nanoparticles <20 nm were also efficiently captured by NIOSH-approved FFRs as predicted by the single fiber theory (Huang *et al.*, 2007; Rengasamy *et al.*, 2008, 2009).

Respiratory protection, however, is dependent not only on the filtration efficiency of the respirator but also on the face seal leakage. Filter penetration and face seal leakage of different size particles through respirators have been reviewed (Shaffer and Rengasamy, 2009; Rengasamy *et al.*, 2010). Several studies addressed this issue by studying filter penetration and face seal leakage for different types of respirators using a manikin head (Cooper *et al.*, 1983a,b; Tuomi, 1985; Hinds and Bellin, 1987; Hinds and Kraske, 1987). In one study, the filtration efficiency was >95% under sealed conditions for particles >5 µm diameter and face seal leakage resulted in higher levels of penetration for two half-mask respirators and two surgical masks (Tuomi, 1985). Subsequent studies measured filter penetration and leakage for wide size ranges of particles under different flow rates employing different types of respirators (Hinds and Kraske, 1987; Chen *et al.*, 1990). Effects of artificial leaks on penetration were studied with half-mask and single-use respirators mounted on a manikin in a chamber exposed to 0.10 to 11.34 µm diameter oleic acid particles (Hinds and Kraske, 1987). Particle penetration through the filter media and artificially induced

leaks was measured at different flow rates between 2 and 150 l min⁻¹. The results showed that aerosol penetration through the filter was strongly dependent on particle size and flow rate, while particle leakage was dependent strongly on particle size and less strongly on pressure drop. In a similar study on a human subject using particles in the 0.07 to 4.4 µm size range, particle leakage through three circular holes (0.57, 1.07, and 1.68 mm diameter and 4 mm long) in the body of a half-mask respirator was measured (Holton *et al.*, 1987). The authors showed a decrease in particle leakage with increasing particle diameter from 1.0 to 4.4 µm as well as with decreasing particle size from 0.20 to 0.07 µm, which were attributed to inertial and diffusional losses, respectively. However, comprehensive information on face seal leakage for smaller diameter size particles (<100 nm) is lacking.

Government agencies and other organizations have raised concern on nanoparticle exposure in workplaces and recommended further studies in the area of nanoparticle penetration through filter media as well as through face seal leaks (Aitken *et al.*, 2004; European Commission Joint Research Centre (ECJRC), 2009; NIOSH, 2009). The high mobility of nanoparticles (<100 nm) suggests that they may more readily penetrate leaks in the respirator face seal compared to larger size (>100 nm) particles. In this study, the total inward leakage (TIL) representing filter penetration and face seal leakage with artificial leaks for different size particles in the 8–400 nm range were measured for FFRs sealed to a manikin under breathing conditions. The TIL values for 50 nm size particles at different leak sizes and flow rates were compared with those values obtained for smaller and larger size particles.

MATERIALS AND METHODS

Filtering facepiece respirators

NIOSH-approved N95 and P100 as well as European certified CE-marked FFP2 and FFP3 FFRs were employed for the TIL measurement. One model for each of N95, P100, FFP2, and FFP3 filtering facepieces, employed in previous studies from our laboratory, was selected and three samples from each model were tested to measure filter penetration and TIL. All four model FFRs contained electrostatic filter media and showed a MPPS in the 30 to 60 nm size range (Rengasamy *et al.*, 2009).

Manikin set-up

Nanoparticle leakage was measured using a manikin set-up as shown in Fig. 1. Briefly, a FFR was

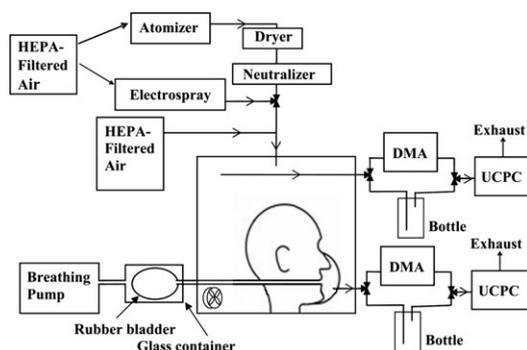


Fig. 1. Diagram of experimental set-up for measuring nanoparticle penetration and TIL through FFRs under simulated breathing conditions.

equipped with a sample probe and two additional probes for introducing artificial leaks. The sample probe similar to that used for fit factor measurement for respirators with a PortaCount® (TSI, Inc., Minnesota, MN, USA) was inserted into the middle of the FFR along the centerline to eliminate effects of asymmetry. Two additional probes were placed on both sides of the FFR at the same height as the sample probe. These probes were placed ~ 1.5 cm from the sealing area of the FFR with the manikin face on each side. The probes were filled with non-hardening putty and the FFR was then sealed to the manikin face using a silicone sealant. No attempt was made to measure any micro leaks at the manikin face and FFR interface. The manikin was placed in a sealed acrylic chamber ($30 \times 30 \times 30$ cm³). Increasing leak sizes were obtained by carefully drilling hypodermic needles (20, 18, 16, and 13 gauge needles with outer diameters of 0.90, 1.27, 1.65, and 2.41 mm, respectively) through the putty. For each test, two similar size holes were introduced simultaneously. A metal tube (2.5 cm diameter) from the back of the mouth opening was connected to an inflatable rubber bladder inside an isolated airtight glass container connected to a breathing pump. This prevented any particles generated by the pump from entering the breathing zone of the FFR on the manikin. The breathing pump was a cam-driven piston pump with a displacement of 1.66 l of air per breath.

Aerosol generation and measurement

Polydisperse NaCl aerosol particles were generated using 2% NaCl solution with an atomizer (TSI 3076), dried, and passed through a charge neutralizer (TSI 3077A). The aerosol was then diluted with high efficiency particulate air (HEPA)-filtered air and passed into the manikin chamber at a flow rate of

100 l min⁻¹ through a hole (1.3 cm diameter) on the top. The aerosol was mixed with a small fan placed at the bottom of the chamber. Excess aerosol from the chamber exited through a hole (1.3 cm diameter) at the back of the box. The resulting aerosol was sufficient to give monodisperse aerosol particles in the 20 to 400 nm size range at ~ 10 000 particles cm⁻³ concentration. In some experiments, charge neutralized monodisperse sucrose aerosol particles in the 8–80 nm range were generated with an electro-spray (TSI 3480), mixed with HEPA-filtered air, and passed into the chamber (data shown only for 8 to 20 nm size range). Varying concentrations of sucrose solutions (0.01–0.5%) were employed to produce different size monodisperse sucrose aerosol particles at ~ 10 000 particles cm⁻³ concentration. In each experiment, the size of sucrose particles generated by the electro-spray was verified using a nano-differential mobility analyzer (Nano-DMA; TSI 3085).

Samples from inside and outside of the FFR were withdrawn for analysis simultaneously with two particle counting systems. To measure particle concentrations, samples were drawn from inside of the respirator through the sample probe on the FFR and outside the FFR from behind the manikin head to avoid sampling the exhalation air directly from the FFR. For measuring NaCl aerosol concentration, the sample was passed through a scanning mobility particle sizer (SMPS; TSI 3080) system with a long DMA (TSI 3081) to select the desired size particles in the 20 to 400 nm size range. This step was bypassed when monodisperse sucrose aerosols were employed in the tests. The sample was then passed into a 1.0-l glass bottle to reduce pressure fluctuations and then to an ultrafine condensation particle counter (UCPC; TSI 3776) for particle counting.

In other experiments, two SMPS (TSI 3080) systems were used in scanning mode to obtain the size-

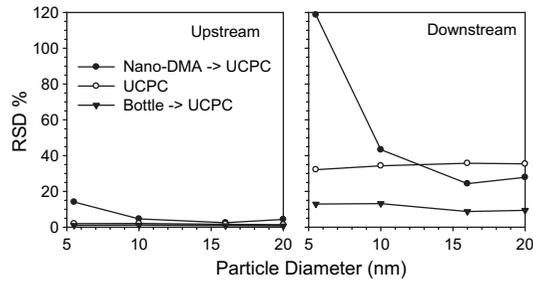


Fig. 2. Comparison of different methods of dampening pressure fluctuations due to the manikin breathing. Aerosol sample was passed into a UCPC directly or through a nano-DMA or a 1.0-l glass bottle.

dependent penetration curves. Both SMPS systems were operated with a sheath flow of 5 l min^{-1} to allow a scan of 20–400 nm, using a long DMA. To minimize the periodic variation due to breathing, the average value of three scans, each taken over a 3-min period was used for each data curve. Penetration was calculated as the ratio of the particle concentration inside of the respirator (C_{in}) to the outside concentration (C_{out}) for each size bin. For a given aerosol concentration inside the test chamber, the two SMPS systems showed a difference in the particle size distribution. To make a correction for the difference, the two SMPS systems were employed to measure the aerosol size distribution inside the test chamber simultaneously. Based on the aerosol size distributions measured by the two SMPS systems, a correction factor was obtained for each size bin and applied to the data collected in the experiments. For sucrose aerosol concentration measurements, only the UCPC with a glass bottles was employed. The counting efficiency of the two UCPCs was compared and found to vary $<5\%$ and no correction was made for the difference.

The UCPCs employed in the study were designed to measure aerosol concentrations in constant pressure environments. Aerosol sampling inside the breathing zone of the filtering facepiece sealed to a manikin generates relatively large changes in pressure causing uncertainties in the UCPC data. This did not cause any significant problem with UCPC counting of NaCl aerosols because the polydisperse aerosol was passed through a DMA and then to an UCPC. On the other hand, smaller size ($<20 \text{ nm}$) monodisperse sucrose aerosol counting by the UCPC showed wide variations in the data obtained in the experiments. Figure 2 shows the relative standard deviation (RSD) for the upstream and downstream concentration data obtained for particles ($<20 \text{ nm}$) for a typical N95 FFR sealed to a manikin breathing at 40 l min^{-1} flow rate. Aerosol sample passed di-

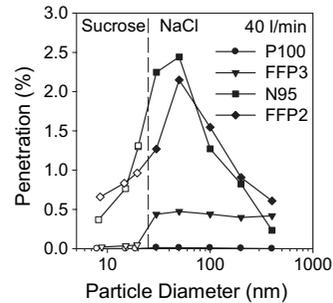


Fig. 3. Typical filter penetration levels for the four FFR models at sealed conditions at 40 l min^{-1} breathing flow rate. Sucrose (open symbols) and NaCl (filled symbols) particles.

rectly to the UCPC or through the nano-DMA and UCPC combination showed relatively high RSD values, specifically in the downstream sample. Measurement with the UCPC under manikin breathing conditions showed higher RSD because of pressure changes and fewer numbers of particles inside the respirator. The variations in the data obtained with the nano-DMA and UCPC combination can be attributed to the loss of particles. A significant decrease in RSD was obtained by passing the aerosol sample through a 1.0-l airtight glass bottle and then to the UCPC. For the data obtained in Figs. 3, 4 and 5, monodisperse sucrose aerosols were passed through an airtight glass bottle (1.0 l) and then to an UCPC to measure particle concentrations.

Filter penetration and TIL measurement

Particle concentrations outside and inside of an FFR sealed to the manikin head were measured by two UCPCs simultaneously. C_{in} at sealed conditions accounts for particle penetration through the filter media only. With the introduction of artificial leaks, C_{in} represents particle penetration through the filter

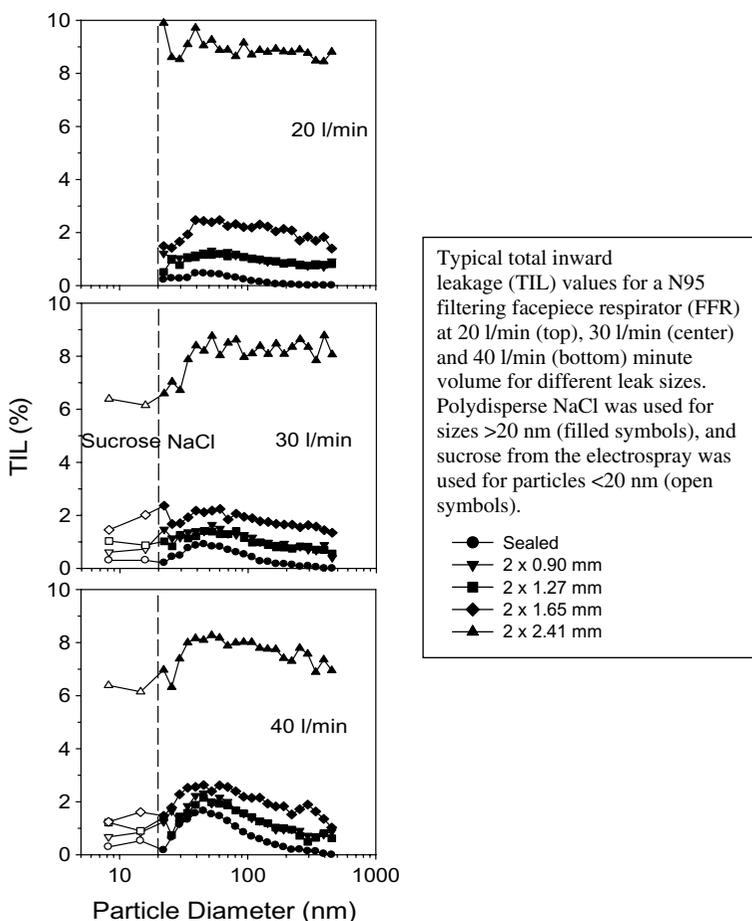


Fig. 4. Typical TIL values for a N95 FFR at 20 l min⁻¹ (top), 30 l min⁻¹ (center), and 40 l min⁻¹ (bottom) volume for different leak sizes. Polydisperse NaCl was used for sizes >20 nm (filled symbols) and sucrose from the electro spray was used for particles <20 nm (open symbols).

media as well as artificial leaks. Filter penetration as well as TIL were calculated as the ratio of C_{in} of the FFR to C_{out} . Measurements were made with a breathing volume of 1.66 l at three different breathing flow rates 12, 18, and 24 breaths min⁻¹, equivalent to minute volumes of ~20, 30, and 40 l min⁻¹, respectively.

Data analysis

The data were analyzed using the SigmaStat® (Jandel Corporation) computer program. Average penetration values and 95% confidence intervals were calculated for each model. A two-way analysis of variance (ANOVA) was conducted to evaluate the significance of penetration results for different size particles at the three flow rates. The TIL and TIL/penetration data for particle size, flow rate, and leak size, were analyzed using a three-way ANOVA. All

pairwise multiple comparisons were performed using the Tukey's test.

RESULTS

Figure 3 shows filter penetration values for N95, P100, FFP2, and FFP3 model respirators against 8 to 400 nm particles at sealed conditions at 40 l min⁻¹ flow rate. All four model FFRs showed a MPPS in the 50 nm size range. Particle penetration was several times higher for the particles in the MPPS than for 8 and 400 nm size ranges. P100 FFR does not show the MPPS at 40–50 nm range in the figure because of the low penetration value (<0.03%).

Percentage penetrations for a typical N95 FFR at sealed conditions and TIL values with different artificial leak sizes for particles in the 8–400 nm at 20, 30, and 40 l min⁻¹ breathing flow rates are shown

in Fig. 4. For N95 FFRs, the MPPS was in the 50 nm size range at sealed conditions and penetrations increased with increasing breathing flow rates. Percentage filter penetration level for 50 nm size particles increased from 0.57 to 1.6 with increasing flow rates from 20 to 40 l min⁻¹ (Table 1). TIL for all size particles increased with increasing leak sizes and flow rates. The MPPS was in the 50 nm size range at smaller leak sizes. TIL for different size particles increased significantly and showed similar values at the largest leak size tested in the study.

The TIL value for N95 FFRs with artificial leaks increased with increasing leak sizes for the three flow rates tested in the study (Table 1). The TIL data obtained for the first leak size (2 × 0.90 mm) and second leak size (2 × 1.27 mm) were similar. For this reason, the data obtained for these two leak sizes were presented as the data for Leak size 1 (2 × 0.90 mm) in Table 1 and for the statistical analysis. The TIL values for particles in the 50 and 400 nm size ranges were 1.53 and 0.77%, respectively, at 30 l min⁻¹ breathing flow with the smallest leak size introduced. At the same flow rate, the TIL values for 50 and 400 nm size particles were 6.97 and 7.16%, respectively, with Leak size 3 (2 × 2.14 mm). In general, the TIL for 50 nm size particles was approximately two times higher than the value for 400 nm size particles at smaller leak sizes at different breathing flow rates. No difference in the TIL for different size particles was obtained at Leak size 3 tested at any flow rate. The TIL/penetration ratios for 50, 100, and 400 nm size particles at different flow rates and leak sizes were calculated.

A dramatic increase in TIL/penetration ratios was obtained with increasing particle sizes at different leak sizes and flow rates. At 30 l min⁻¹ breathing flow, the TIL/penetration ratios for 50 and 400 nm size particles were 2.2 and 34.7, respectively, for Leak size 1. At Leak size 3, the ratios increased to 10.0 and 432 for the same size particles.

A two-way ANOVA was used to analyze the filter penetration data and a three-way ANOVA for the TIL values and the TIL/penetration ratio data and the *P*-values are presented in Table 2. Penetration data showed statistically significant interaction between particle size and flow rate groups. All pairwise multiple comparison analysis showed that the mean penetration values for 50, 100, and 400 nm size particles were significantly (*P* < 0.05) different between particle sizes and between flow rates. For 50 and 100 nm size particles, the mean penetration values were significantly (*P* < 0.05) different between the three different flow rates. However, the mean penetration values for 400 nm size particles at the three flow rates were not significantly different from each other. Within each flow rate, the mean penetration values for 50, 100, and 400 nm size particles were significantly different from each other.

A three-way ANOVA for the TIL values showed no statistically significant interaction between the particle size and flow rate, particle size and leak size, and flow rate and leak size groups. All pairwise multiple comparison analysis showed that the mean TIL values for 50, 100, and 400 nm size particles at Leak Size 3 were significantly (*P* < 0.05) higher than the mean TIL values obtained at Leak Size 1 and Leak Size 2

Table 1. Comparison of filter penetration, TIL, and TIL/penetration ratio as a function of particle size at three different flow rates and leak sizes

Leak size (mm)	Flow rate (l min ⁻¹)	Penetration or TIL (%)			TIL/penetration ratio		
		Particle size (nm)			50	100	400
		50	100	400			
Sealed	20	0.57 ± 0.12	0.24 ± 0.09	0.05 ± 0.08	N/A	N/A	N/A
	30	0.94 ± 0.13	0.46 ± 0.05	0.05 ± 0.06	N/A	N/A	N/A
	40	1.6 ± 0.06	0.66 ± 0.09	0.09 ± 0.09	N/A	N/A	N/A
2 × 0.90 (Leak Size 1)	20	1.14 ± 0.17	0.94 ± 0.15	0.72 ± 0.02	2.1 ± 0.8	4.3 ± 1.8	59 ± 54.5
	30	1.53 ± 0.12	1.02 ± 0.12	0.77 ± 0.11	2.2 ± 0.3	3.4 ± 1.1	34.7 ± 26
	40	1.85 ± 0.13	1.21 ± 0.15	0.71 ± 0.06	2.3 ± 1	3.5 ± 1.6	25.8 ± 23.9
2 × 1.65 (Leak Size 2)	20	2.01 ± 0.46	1.84 ± 0.4	1.71 ± 0.23	3.7 ± 1.6	8.7 ± 4.4	152 ± 143
	30	2.01 ± 0.27	1.78 ± 0.19	1.58 ± 0.20	2.9 ± 0.5	6 ± 2.7	76.9 ± 68.8
	40	2.33 ± 0.16	1.94 ± 0.26	1.43 ± 0.24	2.9 ± 1.2	5.9 ± 3.5	61.2 ± 68.6
2 × 2.41 (Leak Size 3)	20	7.62 ± 4.36	7.68 ± 4.56	7.76 ± 4.73	14.3 ± 9.1	38.4 ± 26.6	791 ± 694
	30	6.97 ± 3.89	7.18 ± 4.19	7.16 ± 4.20	10.0 ± 6	26.2 ± 23.2	432 ± 412
	40	7.07 ± 3.97	6.88 ± 4.2	6.69 ± 4.54	8.7 ± 7	22.7 ± 23.8	365 ± 461

N/A, not applicable.

Table 2. ANOVA for filter penetration, TIL, and TIL/penetration ratio data

Variable	ANOVA results, <i>P</i> -value		
	Filter penetration	TIL	TIL/Penetration
Particle size	<0.001*	0.806	<0.001*
Flow rate	<0.001*	0.966	0.393
Particle size × flow rate	<0.001*	0.997	0.513
Leak size	N/A	<0.001*	0.001*
Particle size × flow rate	N/A	0.997	0.513
Particle size × leak size	N/A	0.994	<0.001*
Flow rate × leak size	N/A	0.971	0.785
Particle size × flow rate × leak size	N/A	1.000	0.931

N/A, not applicable.

*Statistically significant ($P < 0.001$).

(2×1.65 mm). No significant difference in the mean TIL values between Leak Sizes 1 and 2 was obtained.

A three-way ANOVA for the TIL/penetration ratio data showed a significant interaction between particle size and leak size groups. No significant interaction between particle size and flow rate or flow rate and leak size groups was obtained. All pairwise multiple comparison analysis showed that the mean TIL/penetration ratios for 400 nm particles were significantly ($P < 0.05$) higher than the mean TIL/penetration ratios for 50 nm as well as 100 nm particles. No significant difference was obtained between 50 and 100 nm size mean TIL/penetration ratios. The mean TIL/penetration ratio at Leak Size 3 was significantly ($P < 0.05$) higher than the ratios obtained at size Leaks 1 and 2. No significant difference between Leak Sizes 1 and 2 was obtained. Within 400 nm size particle mean TIL/penetration ratios, the values were significantly ($P < 0.05$) higher at Leak Size 3 than Leak Sizes 1 and 2. No significant difference in the mean ratios between Leak Size 1 and 2 was obtained. Within Leak Size 3, the mean TIL/penetration ratios for 400 nm size particles was significantly ($P < 0.05$) higher than the values for 50 as well as 100 nm size particles. No significant difference in the ratios for 50 and 100 nm size was obtained.

The average TIL values was between 5.4 and 14.3% for all four respirator models at the largest leak size (Leak Size 3) introduced (Fig. 5). For N95 respirators, no marked difference in the penetration levels for 8 to 400 nm size particles was obtained. Some variations in penetration levels for different size particles were obtained for the other three models tested in the study. For example, TIL values for FFP2 respirator were higher for 50 nm size particles than the values for other size particles.

In some experiments, penetrations at sealed condition and TIL at the largest leak size for monodisperse

sucrose particles in the 30–80 nm range were measured and compared with those values obtained for similar size NaCl aerosols at a 40 l min^{-1} flow rate (data not shown). Similar size sucrose and NaCl aerosols showed comparable filter penetrations as well as TIL values.

DISCUSSION

N95 FFRs challenged with different size particles in the 8–400 nm range at sealed conditions showed that penetration through filter media peaked at the 40–50 nm range and increased with increasing breathing flow rates. Filter penetration markedly decreased for smaller and larger size particles. Filter penetrations were significantly different among the different particle sizes, flow rates, and ‘particle sizes × flow rates’ as shown in Table 2. In the case of the TIL, 50 nm size particles had ~2 times higher values than the values obtained for 400 nm size particles at the three different flow rates at smaller leak sizes. The higher TIL value for nanoparticles obtained in the experiments was not statistically significant from the values for 400 nm size particles. The higher TIL values for 50 nm size particles with the smaller leak sizes may partly be due to the increased penetration through filter media and through leaks for 50 nm size particles compared to the smaller and larger size particles. Effective filtration of smaller and larger size particles by manikin breathing can reduce the TIL values for these particles and thereby show a higher TIL value for the MPPS. The TIL values for 50, 100, and 400 nm size particles increased with increasing leak sizes showing a significant increase at the largest leak size tested. This can be explained by the significant filter penetrations and smaller increases in TIL from the aerosols that enter

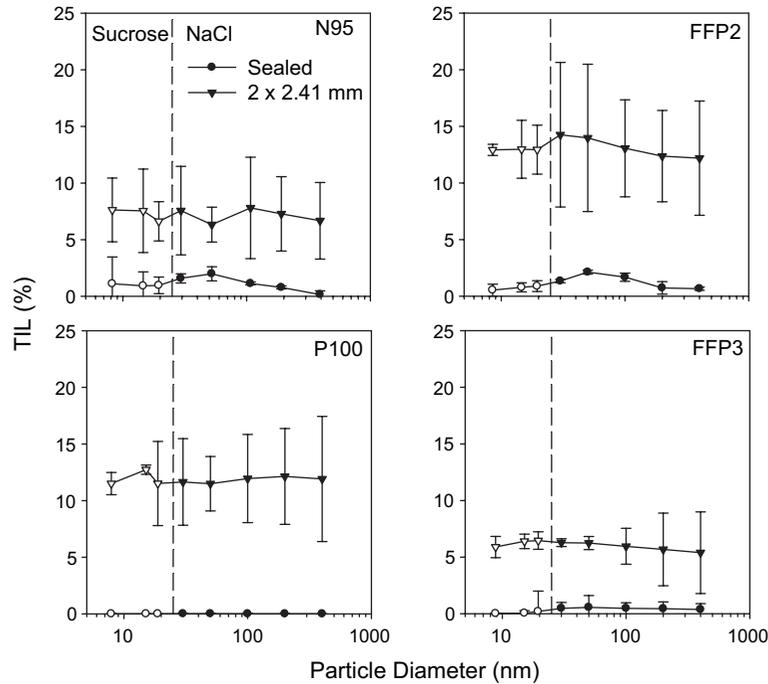


Fig. 5. Comparison of TIL values as a function of aerosol particle size for the four FFR models against sucrose (open symbols, 8–20 nm size) and NaCl (filled symbols, 20–400 nm size) particles at sealed and at 2×2.41 mm leak sizes at 40 l min^{-1} breathing flow rate.

through the smaller leak sizes. At Leak size 3, the greater influx of different size particles contributes to the significant increase in TIL values.

Similar to N95 FFR, P100, FFP2, and FFP3 respirators showed a MPPS in the 50 nm range at sealed conditions. These data are consistent with the MPPS obtained for different respirator models containing electrostatic filter media tested with a manikin or other test systems (Martin and Moyer, 2000; Balazy *et al.*, 2006; Rengasamy *et al.*, 2007; Eshbaugh *et al.*, 2009). The TIL value for 20 to 100 nm size particles at the largest leak size was slightly higher or similar to 100 to 400 nm size particles for the four models. FFP2 model FFRs showed higher TIL values for 50 nm size particles than for 400 nm size particles. The difference in TIL values among the respirator models obtained for nanoparticles (<100 nm size) and 100 to 400 nm size particles may be due to the variability in the manufacturing processes.

Penetration through filter media as well as through artificial leaks contribute to the TIL values obtained in the study. Filter penetration at sealed conditions decreased with increasing particle size markedly for the three different flow rates (Table 1). At the same time, the TIL/penetration ratio for 400 nm size

particles was several times higher than the ratio for 50 nm particles at all three flow rates and leak sizes. This can be partly explained by the lower filter penetrations for the larger size particles than the MPPS. Moreover, larger size particles are known to enter the FFR mainly through face seal leaks (Grinshpun *et al.*, 2009). These two factors contribute to the higher TIL/penetration ratios for larger size particles. These data are consistent with the results obtained in a recent study on the relative contribution of face seal leakage and penetration through filter media for subjects wearing N95 FFRs (Grinshpun *et al.*, 2009). Face seal leakage and filter penetration of different size particles in the 40 to 1000 nm range were measured employing test subjects and a manikin, respectively, and face seal leakage-to-filter (FLTF) penetration ratios were calculated. The FLTF ratio increased with increasing particle size indicating greater contribution of face seal leakage for larger size particles than nanoparticles (<100 nm size). The TIL/penetration ratio for 50, 100, and 400 nm size particles obtained in our study showed a decrease with increasing flow rate at all leak sizes. This decrease in TIL/penetration ratio may partly be due to the increased filter penetration at high flow rates.

Results from this study showed that the TIL value for nanoparticles is dependent on leak sizes. As seen in Table 2, leak size was the only variable that was found to be statistically significant in the three-way ANOVA for TIL. The TIL values for particles <100 nm size showed a peak at the MPPS at smaller leak sizes. TIL value was independent of particle size for the largest leak size tested in the study indicating that TIL crossed a threshold level. This agrees with a previous study, which measured particle leakage by reducing particle penetration through filter media employing relatively less penetrating 10 nm size monodisperse NaCl particles (Liu *et al.*, 1993). Particle leakage was measured at controlled leak holes at steady flow rates using dust/mist and high efficiency dust/mist/fume/radionuclide respirators. The TIL value was shown to be directly related to filter efficiency, leak size, and flow rate. The TIL became independent of the total flow through the respirator when the leak size exceeded a threshold value.

Results from the study showed that TIL values for particles <30 nm sizes were less than the TIL values obtained for the MPPS at all leak sizes. In contrast, a recent study reported higher leakage levels for 4 to 10 nm size particles than for 30 nm size particles indicating that respiratory protection for the smaller size nanoparticles may be a concern (Mouret *et al.*, 2009). The authors employed non-woven fiberglass filter media perforated with 400 to 2000 μm calibrated needles and showed increased penetration levels for particles <30 nm. However, the penetration levels for 4 to 10 nm size particles were below the penetration levels for 30 nm size particles even at the largest leak size (2000 μm size hole) tested in that study. This indicates that leakage of 4 to 10 nm size particles into the breathing zone may not be significantly higher than for 30 nm size particles.

Contrary to the data obtained in this study, an increase in leakage was obtained for a gaseous agent smaller than the diameter size of nanoparticles (Myers *et al.*, 1991). The authors measured apparent fit factors for respirators at different leak sizes for acetone vapor at different flow rates and compared with the levels for polystyrene latex (PSL) aerosol particles (0.36–2.5 μm diameter). The results showed that the apparent fit factors measured with acetone vapor were less than the values obtained for PSL particles. Based on these results, the authors suggested that fit test utilizing a vapor or gaseous agent may be a more critical test in terms of penetration than fit tests using an aerosol. Another study indicated that the lower apparent fit factors measured with acetone vapor may be due to the vapor

permeation through the filter media to increase the concentration inside the respirator (Gardner *et al.*, 2004). Further studies showed that the simulated fit factors measured with a gaseous challenge agent sulfur hexafluoride (SF_6) was $\sim 25\%$ higher than those measured for PSL aerosol challenge with no significant difference between the two test agents (Gardner *et al.*, 2004). However, the simulated fit factor values measured using isoamyl acetate (IAA) vapor showed a significant decrease at sealed conditions and an increase at larger leak sizes than the values obtained with PSL particles. Their results showed that IAA permeated through the respirator and increased the concentration inside the respirator to produce a lower simulated fit factor value at sealed conditions. On the other hand, the increase in simulated fit factors at large size leaks was attributed to the selective adsorption of IAA to the interior surface of the respirator and/or the test system.

The static leaks employed in our study using a manikin are different from the variable leak sizes created during workplace maneuvers of a respirator user (Krishnan *et al.*, 1994b; Myers *et al.*, 1995; Janssen and Weber, 2005). This hypothesis was tested by measuring the fit factors sequentially for three different exercises by an aerosol fit test method and a dichotomous-flow fit test method (Krishnan *et al.*, 1994a,b). Quantitative fit factors were measured for a test subject by both the aerosol and the dichotomous-flow methods and compared with the values obtained for a manikin. The variation in fit factors obtained in the human study was much higher than the values obtained with the manikin study suggesting that the higher variation in the human study was due to variations in face seal leaks. Similarly, leak size and shape changes with time were reported for respirator wearers in workplaces (Myers *et al.*, 1995).

The TIL data obtained in the study suggest potential concerns for respiratory protection in nanotechnology and other workplaces, where the MPPS is in the nanoparticle size range, warranting additional research. In this study, the TIL value for 50 nm size particles was higher than for particles in the 400 nm range for small leak sizes. Workers may be exposed to nanoparticles during nanoparticle generation, handling, and cleaning processes as reported (Bello *et al.*, 2009; Johnson *et al.*, 2010). This indicates that an increased TIL for respirators with MPPS < 100 nm remains a concern in workplace settings generating high levels of nanoparticles. Among the different size nanoparticles, the MPPS with relatively higher penetration levels can

contribute more to the TIL value. As noted elsewhere (Rengasamy *et al.*, 2007; Shaffer and Rengasamy, 2009), employers should factor in particle size during the respirator selection process. This could be accomplished by choosing a different type of respirator offering higher levels of protection when used in a complete respiratory protection program or by choosing a particulate respirator with a higher filtration rating. However, the data reported here shows the importance of getting a good respirator fit as leak size was the predominant factor affecting TIL.

LIMITATIONS

Some limitations of the study include the location of the fixed size artificial leaks created for measuring TIL. Several studies reported that face seal leaks occur mostly in the nose and chin area of the respirator wearer (Crutchfield and Park, 1997; Holton *et al.*, 1987; Oestenstad *et al.*, 1990). In our study, leaks were created at the cheek area of the FFR 1.5 cm from the sealing surface to make sure that the hole was well open during the experiment. The static holes employed in the study do not represent the dynamic leaks created by the different maneuvers of the respirator wearer in a workplace (Janssen and Weber, 2005; Krishnan *et al.*, 1994b; Myers *et al.*, 1995). Similarly, the leak sizes introduced in the manikin study are expected to be different from the leak sizes created by the respirator wearer's maneuvers. The aerosol particles employed in the study were dry and charge neutralized to produce higher filter penetrations, which represents a conservative scenario. Further studies on the TIL value for respirator wearers in workplaces are needed to assess the respiratory protection against nanoparticle exposure.

CONCLUSIONS

Filter penetrations for N95 and other electrostatic FFR models sealed to a manikin showed that the MPPS was ~50 nm size. Filter penetration for 50 nm size particles was several times higher than the penetrations 8 and 400 nm size particles at different flow rates. A consistent increase in penetration was obtained for the different size particles with increasing flow rates. With the introduction of artificial leaks, TIL values for all size particles increased with increasing leak sizes. At relatively smaller size leaks, the TIL value for 50 nm particles was ~2-fold higher than the value obtained for 8 and 400 nm size particles at different flow rates indicating that

TIL for the most penetrating particles was higher than the smaller and larger size particles. The TIL values for different size particles were significantly higher at the largest leak size than the values at the smaller size leaks. The data indicate higher concentrations of nanoparticles (<100 nm) inside the breathing zone of electrostatic respirators can be expected in some workplaces. The TIL/penetration ratios obtained for 400 nm size particles were larger than the ratios obtained for 50 nm size at the three different flow rates and leak sizes indicating that leakage, not filter penetration, was contributing to the TIL for larger size particles. Further studies on face seal leakage of nanoparticles in workplaces are needed to better understand the respiratory protection against nanoparticle exposure.

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