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Comparison of Nanoparticle Filtration Performance of NIOSH-approved and CE-Marked Particulate Filtering Facepiece Respirators

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The National Institute for Occupational Safety and Health (NIOSH) and European Norms (ENs) employ different test protocols for evaluation of air-purifying particulate respirators commonly referred to as filtering facepiece respirators (FFR). The relative performance of the NIOSH-approved and EN-certified 'Conformité Européen' (CE)-marked FFR is not well studied. NIOSH requires a minimum of 95 and 99.97% efficiencies for N95 and P100 FFR, respectively; meanwhile, the EN requires 94 and 99% efficiencies for FFRs, class P2 (FFP2) and class P3 (FFP3), respectively. To better understand the filtration performance of NIOSH- and CE-marked FFRs, initial penetration levels of N95, P100, FFP2 and FFP3 respirators were measured using a series of polydisperse and monodisperse aerosol test methods and compared. Initial penetration levels of polydisperse NaCl aerosols [mass median diameter (MMD) of 238 nm] were measured using a method similar to the NIOSH respirator certification test method. Monodisperse aerosol penetrations were measured using silver particles for 4–30 nm and NaCl particles for 20–400 nm ranges. Two models for each FFR type were selected and five samples from each model were tested against charge neutralized aerosol particles at 85 l min^{-1} flow rate. Penetrations from the 238 nm MMD polydisperse aerosol test were <1% for N95 and FFP2 models and <0.03% for P100 and FFP3 models. Monodisperse aerosol penetration levels showed that the most penetrating particle size (MPPS) was in the 30–60 nm range for all models of FFRs tested in the study. Percentage penetrations at the MPPS were <4.28, <2.22, <0.009 and <0.164 for the N95, FFP2, P100 and FFP3 respirator models, respectively. The MPPS obtained for all four FFR types suggested particle capturing by electrostatic mechanism. Liquid isopropanol treatment of FFRs shifted the MPPS to 200–300 nm and dramatically increased polydisperse as well as monodisperse aerosol penetrations of all four FFR types indicating that all the four FFR types share filtration characteristics of electret filters. Electrostatic charge removal from all four FFR types also increased penetration levels of 400–1000 nm range particles. Particle penetration data obtained in this study showed that the eight models of NIOSH-approved N95 and P100 and CE-marked FFP2 and FFP3 respirators used in this study provided expected levels of laboratory filtration performance against nanoparticles.

Keywords: filtration; monodisperse aerosol; NaCl particles; nanoparticle; particle penetration; respirator; silver particles

INTRODUCTION

The rapid growth of nanotechnology industries has introduced engineered nanomaterials into the workplace. Engineered nanomaterials show unique properties different from the bulk materials. Workers handling or manipulating nanomaterials can generate aerosolized nanoparticles (Schulte *et al.*, 2008) which may be inhaled, ingested or absorbed through skin. Among the different routes of nanoparticle entry, inhalation is considered to be the primary mechanism. Once inhaled, nanoparticles with increased solubility can reach parts of a biological system which

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are not readily accessible by larger particles. Nanoparticle inhalation has been shown to cause adverse effects on pulmonary and systemic functions (Pope *et al.*, 2002; Elder *et al.*, 2006). Many organizations recommend the use of personal respiratory protection devices when engineering controls and other control technologies do not reduce the occupational exposure to nanoparticles to acceptable levels. Because of concerns regarding respirator performance, in particular the filtration of nanoparticles, the National Institute for Occupational and Safety and Health (NIOSH, 2008), Nanotechnology Environmental and Health Implications working group (NEHI, 2008), International Council on Nanotechnology (ICON, 2008) and other organizations have called for increased emphasis on research to better understand the effectiveness of respirators.

Respiratory protection devices throughout the world are often regulated nationally. In the US, NIOSH certifies N, R and P series particulate filtering respirator types 95, 99 and 100 with minimum filtration efficiencies of 95, 99 and 99.97%, respectively. Several countries including Canada, Mexico and Chile recognize NIOSH certification of respirators, while in Europe, respirators marked with 'Conformité Européen' (CE) such as FFP1, FFP2 and FFP3 types meet minimum filtration efficiencies of 80, 94 and 99%, respectively. NIOSH and European Norm (EN) certifications of particulate respirators employ different test protocols for approval. NIOSH conducts respirator certification testing according to 42 CFR Part 84 (Federal Register, 1995) and the approved products are required to be labeled with 'NIOSH' in capital letters and with other information including part and lot number and company name. The European Community (EC) legislation specifies that EN standards must be followed for testing respirators (European Directive, 1996). A CE mark on the product indicates EC conformity. Both NIOSH and EN respirator certification programs are widely known in different parts of the world.

For certification of particulate respirators, NIOSH and EC notified bodies or test houses conduct filtration tests using different protocols. NIOSH regulations for N-series respirator testing require a polydisperse distribution of NaCl particles with a count median diameter (CMD) of 0.075 ± 0.020 μm and a geometric standard deviation (GSD) of <1.86 (NIOSH, 2005a). The mass median diameter (MMD) of the target distribution of test particles is 238 nm with a mass median aerodynamic diameter (MMAD) of 347 nm. For R- and P-designated respirators, a polydisperse distribution of dioctyl phthalate (DOP) particles with a CMD of 0.185 ± 0.020 μm and a GSD of <1.60 is used (NIOSH, 2005b). The MMD of DOP aerosol corresponds to 356 nm with a MMAD of 359 nm. The NIOSH certification test is conducted using charge neutralized polydisperse aerosol particles (NaCl and DOP) at 85 l $\min^{-1}$ flow rate using a TSI 8130 Automated Filter Tester, which employs a forward light scattering photometer to measure the flux of light scattering from particles. A reported limitation of the photometer used in the TSI 8130 is that it has a higher measurement efficiency for particles >100 nm size (Eninger *et al.*, 2008b). On the other hand, CE-marked particulate respirators are tested with non-neutralized polydisperse NaCl as well as paraffin oil particles at 95 l $\min^{-1}$ according to EN standards (BS EN 2000, 2002). For NaCl aerosol, the diameter of the particles varies from 40 to 1200 nm with a MMD of 600 nm. NaCl aerosol particles upstream and downstream of respirators are passed through a hydrogen flame and vaporized. The intensity of light emitted at 589 nm is measured, which is proportional to sodium concentration. For polydisperse oil aerosol production, paraffin oil is atomized at 100°C and diluted with filtered air. The particle size distribution is a log-normal distribution with a number median Stokes diameter of 400 nm and a GSD of 1.82. The aerosol concentration is measured before and after the test filter by a light scattering photometer.

Laboratory filtration performance of air-purifying particulate filtering respirators which include filtering facepiece respirators (FFRs) is well characterized for a wide size range of aerosol particles most commonly found in workplaces (Moyer and Bergman, 2000; Lee *et al.*, 2005; Balazy *et al.*, 2006; Rengasamy *et al.*, 2007; Eninger *et al.*, 2008a). Moyer and Bergman (2000) reported $<5\%$ initial percentage penetration levels of NaCl aerosols for three models of N95 FFRs. In one study, initial penetration levels of 50 nm monodisperse NaCl particles (most penetrating particle size, MPPS) $>5\%$ was reported for one of two N95 FFRs tested at 85 l $\min^{-1}$ (Balazy *et al.*, 2006). Further studies with additional N95 FFR models showed that penetration levels at the MPPS for some FFR models slightly exceeded NIOSH allowed 5% level, but the increase was not significantly different from 5% (Rengasamy *et al.*, 2007). Some studies also reported the filtration performance of other types of FFRs and filter media including R and P types (Martin and Moyer, 2000; Richardson *et al.*, 2006; Eninger *et al.*, 2008a; Rengasamy *et al.*, 2008b). NIOSH-approved P100 FFRs showed penetration levels within approved levels ($<0.03\%$) at 85 l $\min^{-1}$ flow rate. The MPPS for P100 FFRs was found to be in the 40–50 nm range (Richardson *et al.*, 2006; Rengasamy *et al.*, 2008b). A recent study reported $>1\%$ penetration for size-fractioned NaCl (20–500 nm) and viral aerosols (100 nm) for two models of N99 FFRs at 85 l $\min^{-1}$ flow rate (Eninger *et al.*, 2008a).

Very few studies reported the filtration performance of CE-marked FFR against nanoparticles (Wake *et al.*, 1992; Wilkes, 2002; Checchi *et al.*, 2005; Golanski *et al.*, 2008). One study assessed the respirator performance against radon daughter aerosols by measuring the filtration efficiency of filtering facepieces and filters approved by the British Standard Institution and Health and Safety Executive of UK with monodisperse NaCl aerosols (Wake *et al.*, 1992). The results showed that penetration levels of neutralized aerosols were higher than that of charged aerosols.

Recent studies reported the penetration of a wide size range of particles through respirators and filters (Huang *et al.*, 2007; Golanski *et al.*, 2008). Huang *et al.* (2007) measured the filtration performance of respirators against nanoparticles by determining the penetration levels of 4.5 nm to 10 μm NaCl aerosols through one CE-marked FFP1 model and one NIOSH-approved N95 FFR model. The results showed that particles below 10 nm were effectively captured by the FFP1and N95 FFR models studied. Another study reported the penetration levels of graphite nanoparticles ranging from 5 to 100 nm for FFP3 and other filter media (Golanski *et al.*, 2008). FFP3 filter showed maximum penetration levels of ~0.1% at the MPPS (30–40 nm) with varying penetration levels for high-efficiency particulate air (HEPA) and other filter media at a face velocity of 5.3 cm s^{-1}.

NIOSH and EN certification of particulate respirators employ different test protocols and a comparative performance of these FFR is not available for a wide range of particle sizes, in particular those particles <100 nm (i.e. nanoparticles). This study compared the filtration performance of two models each of NIOSH-approved N95 and P100 and CE-marked FFP2 and FFP3 FFRs using a polydisperse aerosol test (PAT) method similar to the method used in NIOSH certification and two monodisperse aerosol test methods. The relative filtration performances of the various respirators are discussed and data are presented on their filtration mechanisms.

MATERIALS AND METHODS

Filtering facepieces

Two models each of NIOSH-approved N95 and P100 and CE-marked FFP2 and FFP3 FFRs were purchased commercially. For comparison of filtration performance, class N95 and class FFP2 respirators as well as class P100 and class FFP3 respirators were selected. It could be argued that comparison of the filtration performances of NIOSH-approved class N99 respirator with the FFP3 respirator would be better because these two types are both certified to meet <1% particle penetration levels. However, a class P100 FFR was selected in this study to compare with FFP3 because it allows us to compare the results from this study with our previous work (Rengasamy *et al.*, 2008b). In addition, class P100 respirators are far more commonly used than class N99 respirators in the US. The manufacturers were randomly selected from the NIOSH- and CE-marked lists. A single respirator model was selected from each manufacturer, excepting FFP3. Two different models of FFP3 were selected from one manufacturer because of procurement difficulties.

Polydisperse NaCl aerosol penetration test (PAT)

Initial penetration levels of polydisperse NaCl aerosol were measured using a TSI 8130 Automated Filter Tester (TSI 8130) as described previously (Rengasamy *et al.*, 2007). Penetration levels were measured for 1 min of loading, instead of carrying out the entire NIOSH 42 CFR Part 84 test procedure (NIOSH, 2005a). Initial penetration levels were measured in order to be consistent with aerosol testing for various size monodisperse particles described below. Percentage particle penetration was measured at 85 l min^{-1} flow rate with the mask mounted in a Plexiglas box (20 × 20 × 10 cm). Five samples from each model were tested for particle penetration measurements.

Monodisperse 4–30 nm silver particle penetration test (MAT-1)

Silver nanoparticles were generated by an evaporation and condensation method and tested for penetration as described previously (Rengasamy *et al.*, 2008b). Briefly, pure metallic silver (Alfa Aesar, 99.99%) in a ceramic boat was placed inside a ceramic tube kept in a furnace (Lindberg/BlueM model: TF55035A-1) and heated at 1050°C (Figure 1). Polydisperse silver nanoparticles produced were transported by HEPA-filtered nitrogen gas at 2 l min^{-1} flow rate into a scanning mobility particle sizer (SMPS; TSI model 3080) equipped with a nano-differential mobility analyzer (Nano-DMA, TSI model 3085). Six different size (centered at 4, 8, 12, 16, 20 and 30 nm) monodisperse silver particles were produced based on electrical mobility. The size of the monodisperse aerosol particles generated by the test system was verified (Rengasamy *et al.*, 2008b). The exiting monodisperse particles were mixed with HEPA-filtered room air and passed through a ^{85}Kr neutralizer (TSI 3012). The charge neutralized monodisperse particles were passed into the Plexiglas respirator test box. Upstream and downstream particle numbers at 85 l min^{-1} flow rate were counted alternately using an ultrafine condensation particle counter (UCPC; TSI 3025A). Leakage of nanoparticles into the test system was checked by operating the nano-DMA at 0 V and measuring the counts by the UCPC. The absence of any leakage was ensured by measuring zero counts for 20 min. An equilibration time of ~5 min was allowed between upstream

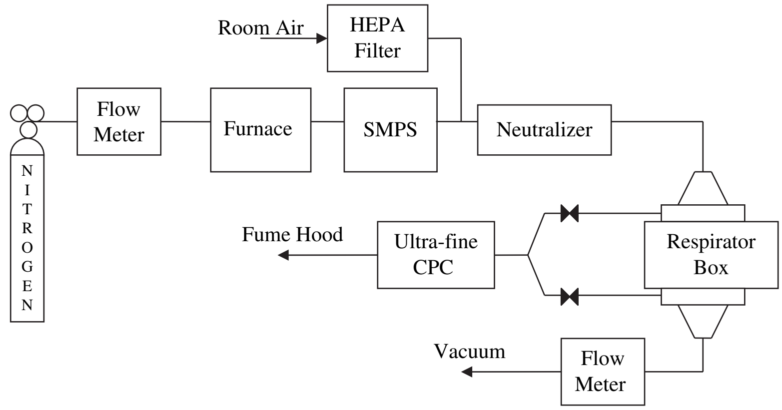

Fig. 1. Schematic diagram of the silver particle test system (Rengasamy *et al.*, 2008b. J. Occup. Env. Hyg. 5: 556–564, 2007).

and downstream sampling. Five samples from each model were tested for penetration of monodisperse silver particles.

For N95 and FFP2 respirator penetration studies, the furnace temperature was set at 1050°C to produce sufficient number of particles for measuring the penetration of the six different size monodisperse silver particles. For P100 and FFP3 respirators, furnace temperatures were kept at 950°C for 4 nm particles, 1050°C for 8 and 12 nm particles and at 1100°C for 16, 20 and 30 nm size particles to optimize the number of the test particles as described previously (Rengasamy *et al.*, 2008b).

Monodisperse 20–400 nm NaCl aerosol penetration test (MAT-2)

A different set of FFR samples from the same models that were employed for the PAT experiments were tested against monodisperse NaCl particles using a TSI 3160 Fractional Efficiency Tester (TSI 3160) equipped with a long DMA (TSI 3081) as described previously (Rengasamy *et al.*, 2007). Initial percentage penetration levels of 10 different size (centered at 20, 30, 40, 50, 60, 80, 100, 200, 300 and 400 nm) monodisperse particles were measured in one test run for each FFR at a flow rate of 85 l min^{-1}. Five samples from each model were tested for different size monodisperse particle penetrations.

Penetration of NaCl particles as a function of particle size from 30 to 1000 nm

To better understand the penetration of submicron size particle (<1000 nm), penetration was measured as a function of particle size from 30 to 1000 nm. NaCl aerosol was generated using a constant output atomizer (Model 3076, TSI) and the aerosol concentrations and size distributions (30–1000 nm range) were measured using a SMPS and a condensation particle counter (CPC) instead of using the TSI 3160 filter tester (Figure 2). Polydisperse NaCl particles were passed through a drier, a ^{85}Kr neutralizer and then into the Plexiglas box containing the test respirator. Particle number concentrations and size distributions upstream and downstream of the FFR were measured alternately using a SMPS in scanning mode. Percentage penetration was calculated from the ratio of the particle number concentration downstream to the concentration upstream.

Isopropanol treatment

Class N95 respirators typically capture particles by electrostatic and other mechanisms. It is unclear whether most P100, FFP2 and FFP3 respirators capture particles by mechanical or a combination of both mechanical and electrostatic mechanisms. The exact filtration mechanism of various respirator types is useful for filtration theory modeling and theoretical simulations (Balazy *et al.*, 2006) and for research to develop improved filters and air-purifying respirators. The physical interactions between particles and filter fibers can change dramatically when electrostatic charges on the fibers are introduced. To address this question for the models studied here, the FFR samples were subjected to isopropanol treatment, which is known to remove electrostatic charges on filter media and to increase particle penetration in laboratory tests (Chen *et al.*, 1993; Chen and Huang, 1998; Martin and Moyer, 2000; Kim *et al.*, 2007a). In the first set, five FFR samples were tested using the PAT method and then the FFRs were carefully removed from the test box and dipped into liquid isopropanol in a container for 1 min. FFR samples were removed from isopropanol solution, dried by evaporation overnight in a fume hood at room temperature and tested again using the PAT method with polydisperse NaCl aerosol particles. The second set consisting of five FFR samples was tested using the MAT-2 method using monodisperse NaCl aerosols (20–400 nm range) on the TSI 3160 and then treated with isopropanol and processed as described previously. The samples were tested again for particle penetration using the MAT-2 method. For the third

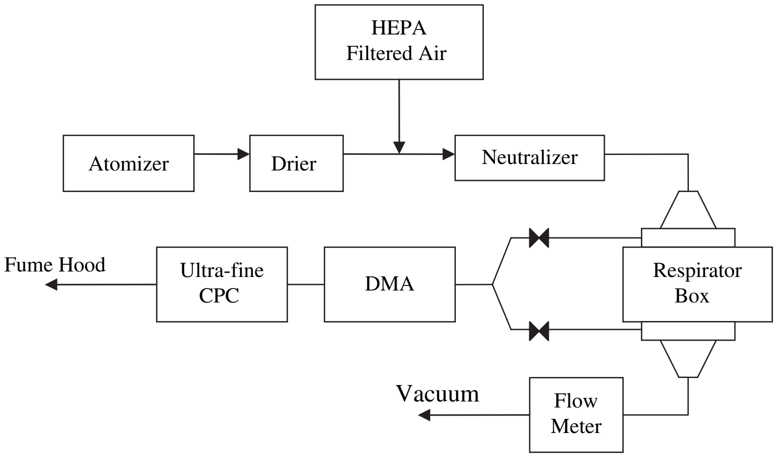

Fig. 2. Schematic diagram of test system for penetration of 30–1000 nm particles.

set, five FFR samples were tested for particle number concentrations and size distributions of NaCl particles from 30 to 1000 nm size using a SMPS in scanning mode and then removed from the test box, treated with isopropanol, processed as described previously and measured again the particle number concentrations and size distributions.

Data analysis

The data were analyzed using the SigmaStat computer program. Average, standard deviation and 95% confidence interval penetration levels were calculated for each model. Correlation coefficients between variable parameters were calculated using the Pearson's product-moment correlation method.

RESULTS

Table 1 shows the initial penetration levels of polydisperse NaCl aerosol and standard deviations for two models each of N95, FFP2, P100 and FFP3 FFR types at 85 l min^{-1} flow rate using the PAT method. Both N95 and FFP2 respirators showed average penetration levels of <1%. P100 and FFP3 respirators showed average penetration levels of <0.03%.

Percentage penetrations of six different size monodisperse silver particles in the 4–30 nm range were measured for the different FFR types using the MAT-1 method. Monodisperse particle penetration levels decreased with decreasing particle size for all N95, FFP2, P100 and FFP3 respirators tested at 85 l min^{-1} flow rate (Figure 3). Average penetration levels of the two N95 FFR models tested were similar to the two FFP2 models (top panel). Among the N95 and FFP2 respirator models tested, one FFP2 model showed no penetration for 4 nm particles. For P100 FFR models, the average penetration levels were one to two orders of magnitude less than the levels obtained for the two FFP3 respirator models (bottom panel).

Figure 4 shows average penetration levels of 10 different size monodisperse NaCl particles in the 20–400 nm range for N95 and FFP2 (top panels) and P100 and FFP3 respirators (bottom panels) at 85 l min^{-1} flow rate from the MAT-2 method. Average penetration levels increased from 20 to 30–60 nm and then decreased up to 400 nm particle size for all the respirator models tested. The MPPS for all the four FFR types was in the 30–60 nm range. Both N95 models showed penetration levels comparable to the FFP2 models for the different size particles in the 20–400 nm range (top panels). Penetration levels of both P100 models were approximately one order less than the FFP3 respirator models (bottom panels). Figure 5 shows the correlation of polydisperse (PAT) and monodisperse MPPS particle penetrations (MAT-2) for the NIOSH- and EN-certified FFRs. A significant correlation (r = 0.97; P = 0.00006) was obtained for each of two N95, FFP2, P100 and FFP3 respirator models.

Filter penetration was measured before and after isopropanol treatment of FFR to assess particle capturing by electrostatic mechanism. Penetration levels from the PAT method test were <1% for control N95 and FFP2 respirators (Figure 6, top panel). Isopropanol treatment increased the penetration levels by one to two orders of magnitude for both N95 and FFP2 respirator types. Figure 6 (bottom panel) shows polydisperse aerosol penetration levels of control and isopropanol-treated P100 and FFP3 respirators obtained using the PAT method. Average penetrations were <0.03% for the controls, which increased two to three orders of magnitude after isopropanol treatment.

Figure 7 shows the average penetration levels of monodisperse particles in the 20–400 nm range (MAT-2 method) for the four different FFR types before and after isopropanol treatment. The MPPS for the controls was ~30 to 60 nm, which shifted to

Table 1. Penetration levels from the PAT for the different FFR types

Respirator class	N95		FFP2		P100		FFP3	
Manufacturer	M1	M2	M1	M2	M1	M2	M1	M1
Mean penetration (%)	0.703	0.565	0.270	0.505	0.0034	0.0222	0.0098	0.0144
Standard deviation	0.200	0.525	0.096	0.275	0.002	0.036	0.004	0.011

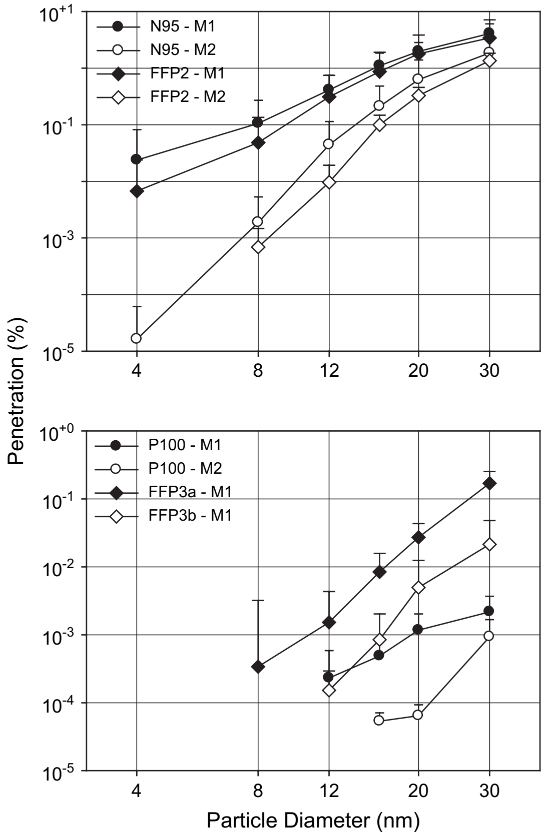

Fig. 3. Percentage penetrations of monodisperse silver particles (4–30 nm) through N95, FFP2, P100 and FFP3 FFR from two different manufacturers (M1 and M2) at 85 l min^{-1} flow rate (MAT-1 method). Error bar indicates the 95% confidence interval ($n = 5$).

the 200–300 nm range after isopropanol treatment. Isopropanol dramatically increased the penetration levels of different size monodisperse particles tested in the 20–400 nm range. The increase in penetration was greater for 200–300 nm particles compared to other size particles for all respirator models tested. The magnitude of increase in penetration was less than two orders for N95 and FFP2 respirators compared to more than four and more than two orders for P100 and FFP3 respirators, respectively. Figure 8 shows average penetration curves for NaCl aerosol particles as a function of particle size from 30 to 1000 nm range for control FFRs. Figure 8 (top panels) shows penetration levels of <3% for N95 and FFP2 respirators and <0.5% for P100 and FFP3 respirators for particles <100 nm. All respirator types showed negligible penetration levels for particles >400 nm. Figure 8 (bottom panels) shows the subsequent penetration levels for liquid isopropanol-treated FFRs. In general, the increase in penetration levels for 20–400 nm particles after isopropanol treatment agreed with the data obtained for individual monodisperse NaCl aerosols tested using the TSI 3160 filter tester (MAT-2 method). In addition, all the four respirator types showed a significant increase in penetration levels for 400–1000 nm particles after isopropanol treatment.

DISCUSSION

NIOSH and EN respirator programs employ different test protocols for certification of particulate FFR for respiratory protection. Penetration measurements and the test conditions used in this study are different from the penetration tests required by the NIOSH and EN certification protocols. The penetration results

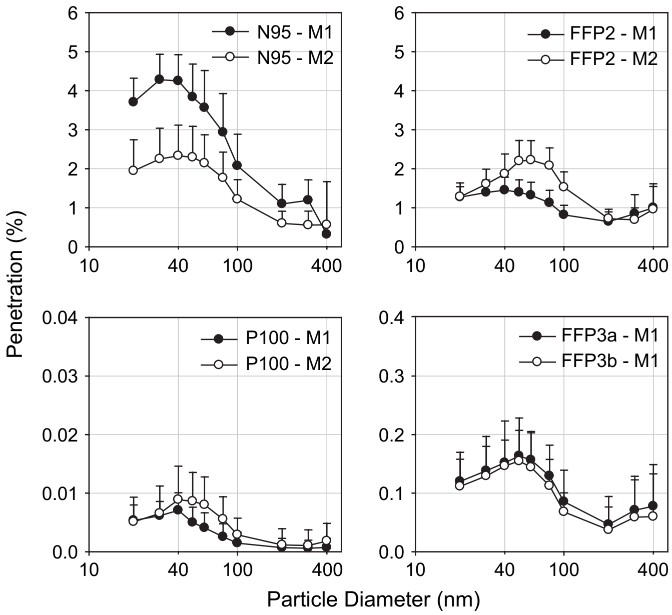

Fig. 4. Percentage penetrations of monodisperse NaCl (20–400 nm) particles through N95, FFP2, P100 and FFP3 FFR from two manufacturers (M1 and M2) at 85 l min^{-1} (MAT-2 method). Error bar represents the 95% confidence interval (n = 5).

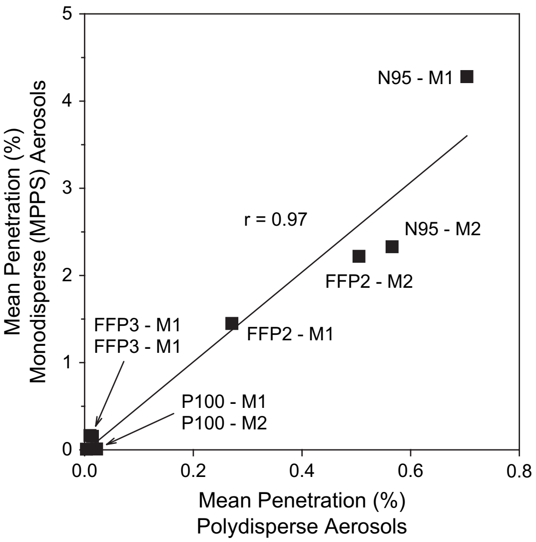

Fig. 5. Correlation of 238 nm MMD polydisperse (PAT method) and monodisperse (MAT-2 method) MPPS (30–60 nm) sodium chloride aerosol penetrations for N95, FFP2, P100 and FFP3 respirator models tested (r = 0.97, P = <0.00006) (n = 5).

obtained with the three test methods used in this study may not be predictive of the penetration results received using the respective certification test methods. For this reason, the results obtained in the study cannot be directly compared with the filtration performance of FFRs approved by the NIOSH and EN certification programs. Across all test methods employed, the penetration levels for N95 and P100 were within the NIOSH allowed <5 and <0.03% levels, respectively. Similarly, FFP2 and FFP3 respirators showed penetration levels <6 and <1%, respectively, as allowed by EN regulations. A comparison of the filtration performance from the PAT method showed that penetration levels were similar for N95 and FFP2 class respirators, as well as for P100 and FFP3 class respirators. Similar classifications of NIOSH and EN particulate respirators demonstrated similar penetration levels for polydisperse particles with a MMD of 238 nm. This observation is consistent with a previous report which compared the penetration levels of different breathing system filters using a TSI 8130 used in NIOSH certification tests and a Moore's Test Rig (CEN Bench Rig) (SPF Services, Christchurch, UK) (Wilkes, 2002) approved for CE marking. NIOSH respirator certification tests are conducted at 85 l min^{-1} with the TSI 8130 which uses charge neutralized polydisperse NaCl aerosols having a MMD of 238 nm. Particle penetration was measured using forward light scattering as described previously (Johnson and Smith, 1988). On the other hand, Moore's Test Rig uses non-neutralized NaCl particles of 40–1200 nm range with a MMD of 600 nm and the filter test is conducted at 95 l min^{-1}. NaCl particle penetration was measured using a neutral hydrogen flame photometer for different filter media (Wilkes, 2002). Their results showed no significant difference in the penetration values for the two methods. Although the NIOSH and EN FFR test methods employ polydisperse aerosol particles in the 22–259 nm (~95%) and 40–1200 nm ranges, respectively, the vast majority of particles that penetrate through the FFR are <300 nm size. Particle penetration results for N95 and FFP2 respirators are expected to be similar because of their expected

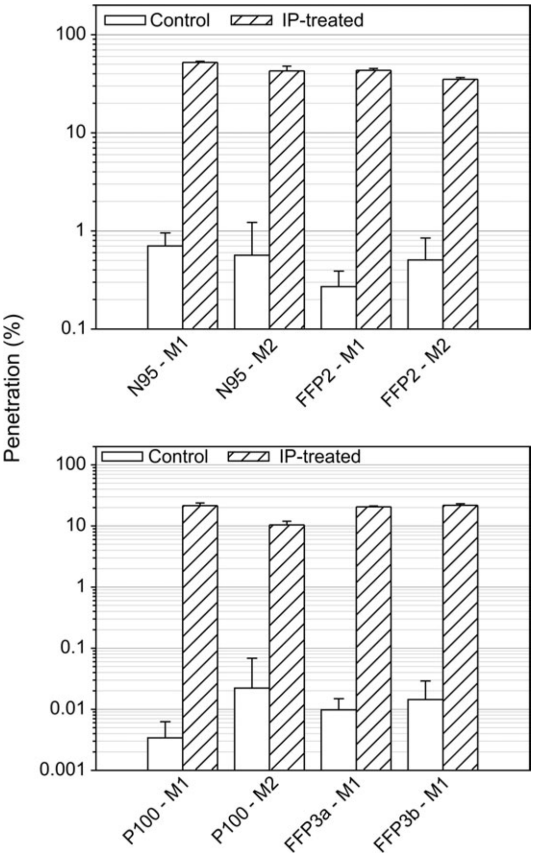

Fig. 6. Percentage penetration levels of 238 nm MMD polydisperse aerosols (PAT method) for different FFR types and manufacturers (M1 and M2) before (control) and after isopropanol treatment (IP-treated) at 85 l $\min^{-1}$ flow rate. Error bar indicates the 95% confidence interval ($n = 5$).

penetration levels <5 and <6%, respectively. Similarly, P100 and FFP3 class respirators allowed for <0.03 and <1.0% penetrations, respectively, are expected to show comparable penetration levels.

Monodisperse aerosol penetration results from this study showed that particle capture increased with decreasing size from 30 nm down to 4 nm for NIOSH-approved class N95 and P100 and CE-marked FFP2 and FFP3 FFR as expected by the single-fiber theory. The results are consistent with previous reports on the filtration performance of respirator filter media (Kim *et al.*, 2007a) and NIOSH-approved and CE-marked FFR (Huang *et al.*, 2007; Rengasamy *et al.*, 2008b). No measurable penetration levels for particles below 10 nm were obtained for one N95 and one FFP1 FFR models tested (Huang *et al.*, 2007). This is partly due to the particle generation method that produced fewer particles in the <10 nm range for penetration measurements. Recently, five N95 FFR models were tested against a relatively high concentration of monodisperse particles in the 4–30 nm range and showed measurable penetration levels for all different size monodisperse particles (Rengasamy *et al.*, 2008b). At the same time, no penetration was obtained for 4 nm particles for all the P100 and FFP3 models tested which is attributed to higher filtration efficiency compared to N95 FFR.

Similar penetration levels were obtained for N95 and FFP2 respirators using the PAT method. This may be partly due to the design of N95 and FFP2 respirators by manufacturers to meet 5 and 6% penetrations required by NIOSH and EN regulations, respectively. On the other hand, the penetration levels of some monodisperse aerosols for P100 FFR were one to two orders of magnitude less compared to FFP3 respirators while no significant difference in penetration was obtained for the PAT. This suggests that only a test method that is based on particle number instead of mass can reveal differences in penetration levels between the different FFR types. The PATs provide an overall penetration of different size particles based on mass of the particles as in the case of TSI 8130 as well as the EN approved equipment. The mass of particles <100 nm is a small fraction compared to the larger size particles and photometric test methods based on particle mass may not adequately measure light scattering of particles in this size range (Eninger *et al.*, 2008b).

Interestingly, the MPPS for all the four FFR types tested in this study was found to be in the 30–60 nm range at 85 l $\min^{-1}$ aerosol flow rate. This is consistent with previously reported MPPS values for N95 and P100 FFR (Balazy *et al.*, 2006; Richardson *et al.*, 2006; Rengasamy *et al.*, 2008b). The four types of FFRs studied also agree on the relative filtration performance measured using the monodisperse (MAT-2) and PAT methods. A consistent rank ordering and statistically significant linear correlation (Figure 5) of filtration performance of all four FFR types was obtained. Similar correlations between submicron polydisperse aerosol and monodisperse aerosol tests have been reported for N95 FFRs (Rengasamy *et al.*, 2007), dust masks (Rengasamy *et al.*, 2008a) and HEPA filter media (Lifshutz and Pierce, 1996; Pierce, 1998).

Using the MAT-2 method, percentage penetrations at the MPPS were <4.28, <2.22, <0.009 and <0.164 for the N95, FFP2, P100 and FFP3 respirator models, respectively. These data suggest the eight models of NIOSH-approved N95 and P100 and CE-marked FFP2 and FFP3 respirators used in this study provide expected levels of laboratory filtration performance against a wide range of particles, including those <100 nm (i.e. nanoparticles). A limitation of this study is that only two models from each respirator type were tested. Thus, the laboratory filtration performances seen in this study may not be representative of all commercially available respirators within the four types studied here. Indeed, studies both in our laboratory (Rengasamy *et al.*, 2007) and by other laboratories (Balazy *et al.*, 2006; Eninger

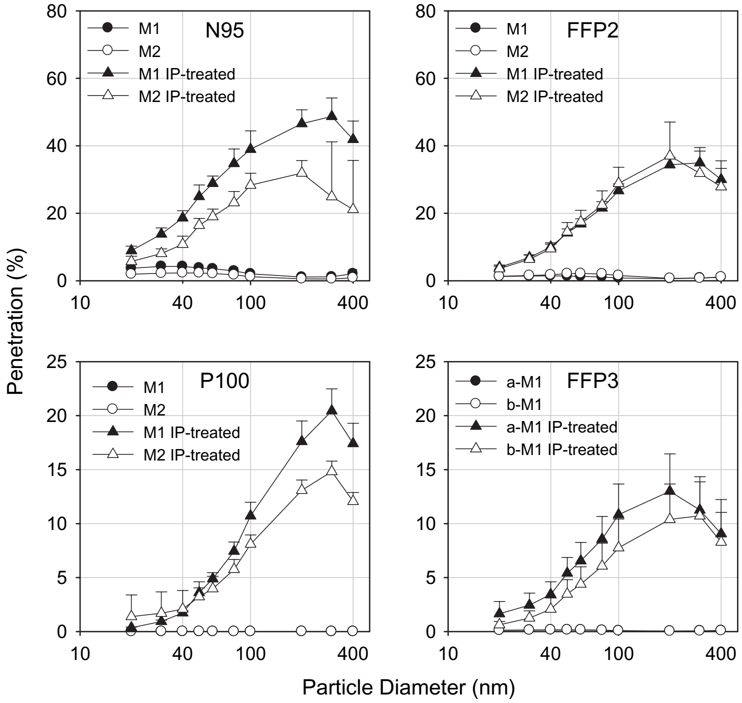

Fig. 7. Percentage penetrations obtained using the MAT-2 method of monodisperse NaCl (20–400 nm) particles for different FFR types and manufacturers (M1 and M2) before and after liquid isopropanol treatment (IP-treated) at 85 l min^{-1}. Error bar represents the 95% confidence interval ($n = 5$).

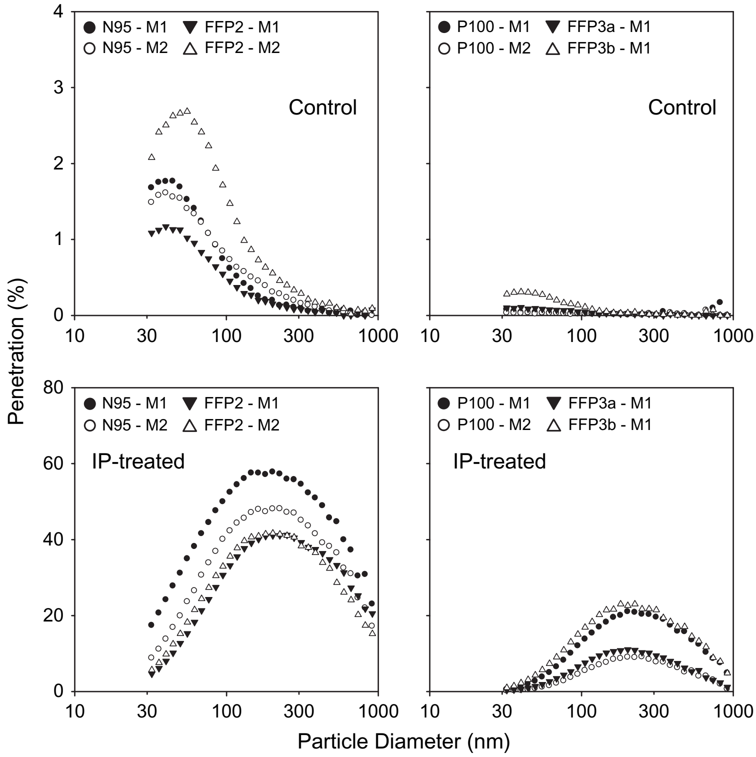

Fig. 8. Average penetration levels of NaCl particles as a function of particle size (30–1000 nm) particles through N95, FFP2, P100 and FFP3 respirators from two manufacturers (M1 and M2) before (control) and after isopropanol treatment (IP-treated) at 85 l min^{-1} ($n = 4$).

et al., 2008a) demonstrate that laboratory respirator filtration performance against nanoparticles in the MPPS range can vary widely, even within a specific respirator type.

The MPPS results obtained for the four different FFR types suggest that the NIOSH-approved as well as the CE-marked FFR models used in this study share filtration properties of electret filters. This was verified by exposing FFR to liquid isopropanol, which is known to remove the electric charge on filter media and to increase particle penetration in laboratory tests. Isopropanol treatment increased PAT penetration levels of N95 and FFP2 models by one to two orders of magnitude and P100 and FFP3 models by two to three orders of magnitude. Based on these results, one may speculate that the P100 and FFP3 FFR models used in this study have more electric charges on filter media fibers than that of the N95 and FFP2 models used in this study. The discrepancy can partly be explained by the filtration efficiency levels of FFR. The percentage penetration levels range from 0.270 to 0.703 for both N95 and FFP2 respirators and from 0.003 to 0.022 for both P100 and FFP3 respirators. Electret charge removal by isopropanol treatment can increase the percentage penetration levels up to only 100% even if the FFR is assumed to be fully electret. Based on the initial penetration levels for the control N95 and FFP2, electret charge removal can only increase penetration levels approximately two orders of magnitude (i.e. from 0.270–0.703 to 100%). At the same time, the penetration levels of P100 and FFP3 FFR can increase to three to four orders of magnitude (i.e. from 0.003–0.022 to 100%) after isopropanol treatment. Thus, isopropanol treatment of an electret filter with 10% penetration level can reach a maximum increase of 10-fold at the maximum. Indeed, two dust mask models with average penetration levels of 10–12% range showed 6- to 7-fold increase in penetration levels after isopropanol treatment (Rengasamy *et al.*, 2008a). Although, liquid isopropanol treatment is assumed to remove all the electret charge on the fiber, a small amount of residual electric charge might be expected to remain on the filter media (Chen *et al.*, 1993). The mechanism of removal of electric charge from filter fibers is not completely understood. Some studies suggested that isopropanol treatment did not remove electret effect from filter media, but caused swelling and dissolution of low-molecular weight polymers resulting in high penetration values (Myers and Arnold, 2003). On the contrary, a recent study employed electrostatic force microscopy and showed a significant removal of electric charges after isopropanol treatment (Kim *et al.*, 2007a). It is possible that isopropanol treatment may disrupt the bonding of non-woven fabric materials and release particles to produce increase in penetration levels. This was tested using HEPA-filtered air with no particles going through the isopropanol-treated respirators. The results showed no release of particles suggesting that the increase in particle penetration after isopropanol treatment may be due to removal of electric charges on filter medium.

Oil aerosols such as DOP decrease electret filter efficiency by mechanisms including neutralization of the charge on the fiber, masking the fiber charge by captured particles or disruption of the charge carrying fiber (Tennal *et al.*, 1991; Barrett and Rousseau, 1998). P-type NIOSH-approved FFRs are not degraded by oil aerosol particles, unlike the N-type electret FFRs. Based on this, NIOSH certification tests for P-type respirators use DOP liquid aerosol particles. This raises a question why a P-type respirator is resistant to oil particles and not an N-type, although both respirator types are electrostatic and susceptible to laboratory filter performance degradation via isopropanol treatment. This may be explained partly due to differences in the manufacturing process of the different types of FFRs (Barrett and Rousseau, 1998). The use of filter media with different chemical composition, different methods of introducing charge onto filter fibers and respirator design using hydrophilic and hydrophobic filter layers in some fashion may also contribute to this difference. Further studies are needed to better understand the mechanisms behind electret filter degradation of different types of respirators.

FFR upon exposure to liquid isopropanol showed a shift in the MPPS from 30 to 60 nm toward a larger size in the 250–300 nm. The results are consistent with the data obtained for filter media and FFR (Chen *et al.*, 1993; Chen and Huang 1998; Martin and Moyer, 2000). The increase in penetration levels of FFR after removal of electret charge by isopropanol treatment clearly shows that electrostatic mechanism plays a significant role in capturing particles of 250–300 nm size compared to particles outside the range as reported previously (Huang *et al.*, 2007). Particle penetration data obtained as a function of particle size after isopropanol treatment suggest that electrostatic forces also play a significant role in capturing particles at >400 nm size range.

The penetration levels measured using the three test methods for the eight models of FFRs were significantly less than the levels allowed by the NIOSH and EN certification test protocols. However, expected protection performance provided by these types of respirators is dependent upon both filtration performance and face seal leakage. Thus, worker protection levels are likely to be much less than the filtration levels seen in this study, which involved sealing the FFRs to the test system in the laboratory. Leakage is dependent upon several factors including proper respirator selection, fit and donning. Further research on leakage of nanoparticles is important to better understand the effectiveness of FFRs in workplaces where nanoparticles are present.

CONCLUSIONS

Initial particle penetration data obtained in this study showed that the eight models of NIOSH-approved N95 and P100 and CE-marked FFP2 and FFP3 respirators provided expected levels of laboratory filtration performance against nanoparticles. Penetration levels of different size monodisperse particles from 4 to 400 nm showed that the MPPS was in the 30–60 nm range for all four FFR types tested in the study. Monodisperse aerosol particles below the MPPS showed a decrease in penetration levels with decreasing particle size as expected by the single-fiber filtration theory. The NIOSH approved and CE-marked FFR models tested in the study were found to share filtration characteristics of electret filters as shown by the shift in the MPPS from 30–60 to 200–300 nm range after the electric charges were removed.

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