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Size-Resolved Penetration of Filtering Materials from CE-Marked Filtering Facepiece Respirators

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ABSTRACT

Throughout the years, the performance of the FFRs has been the topic of extensive studies, resulting mainly in the characterization of aerosol penetration through filters using aerosols that are thought to be similar to those encountered in workplaces. There are limited number of studies in the literature reporting CE-marked respirator performance, and there is a need to perform a penetration evaluation of CE-marked respirators through all 3 filtering classes: FFP1 (80%), FFP2 (94%), and FFP3 (99%). In this study, the percentage penetrations and the most penetrating particle size (MPPS) of 47 mm filters cut out from 13 different CE-marked respirator half masks (2 samples of each FFR) were evaluated size-selectively using nine sizes of charge-neutralized monodisperse aerosol, which ranged from 20 to 400 nm (CMD). Comparison of the penetrations at MPPS from all the examined filters showed that the percentage penetration ranged 3.2–16.3% (FFP1), 2.4–34.3% (FFP2), and 0.02–3.3% (FFP3). Experimental data also revealed that the penetration difference between 2 samples from the same respirator was in most cases up to 6.8%, and between 2 identical respirators up to 2.5%. The MPPS was found to be between 30 and 60 nm (CMD) in all measurements. By comparing the obtained results to the European Standard we conclude that the standard method underestimates particle penetration (especially for particles < 100 nm) due to the usage of non-neutralized, polydisperse test aerosol, detection methods burdened with measurement artifacts, and the assumption that the MPPS is at ≈ 600 nm (MMD) as the criterion for filtering facepieces to pass the penetration test.

Keywords: Size-resolved penetration; CE-marked respirator; Monodisperse ammonium sulfate; 47 mm filter sample; EN 149.

INTRODUCTION

Various industrial processes produce a wide range of aerosol particles of different composition and size. Thus, workers in these workplaces may be exposed to a broad range of aerosol particles possibly causing adverse health effects (Flanagan *et al.*, 2003; Muller *et al.*, 2005; Donaldson *et al.*, 2006; Poland *et al.*, 2008). Many organizations worldwide recommend protective respiratory devices as a preventative form of protecting workers from exposure to hazardous aerosol particles. Filtering facepiece respirators (FFR) are widely used against harmful aerosol particles and protect the user if worn properly. In Europe, the Personal Protective Equipment directive (89/686/EEC) requires that personal protective equipment (PPE) placed within the European market is certified by European Norm (EN) and marked with 'Conformité Européen' (CE), indicating European Community (EC) conformity.

All respirators must be approved and tested to the performance requirements of the corresponding European Standard (ES), which forms the following categories: filtering half masks, half masks and quarter masks, full face masks, powered air respirators, and supplied air respirators. A filtering half mask is a facepiece that consists entirely or substantially of filter material or comprises a facepiece in which the main filter(s) form an inseparable part of the device. Legislation of European Standards for filtering half masks is covered by EN 149:2001.

There are three categories of filtering facepieces (FFP) classified according to their maximum total inward leakage (TIL) and filter efficiency: FFP1 (80%), FFP2 (94%), and FFP3 (99%), which are designed to protect against solid, non-volatile water-based, and oil-based aerosol particles. Therefore, all respirators have to meet both the solid and liquid filter performance requirements. This norm was followed by the amended version EN 149:2001+A1:2009 (further referred to in this article as EN 149) in July 2009. This amendment introduced two usability classes of disposable respirators: single shift only (non-reusable, marked 'NR') and reusable (marked 'R'). Moreover, all

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reusable respirators must endure cleaning and disinfection as described by the manufacturer and pass the dolomite test for clogging (giving the user better and longer lasting breathing resistance), which is indicated by the printing of the letter ’D‘ on the mask (previously only required on FFP1 and FFP2 respirators) (BS EN, 2001).

Performance tests include filter penetration, extended exposure (loading), flammability, breathing resistance, TIL, and dolomite dust clogging (optional). EN 13274-7:2008 (Determination of particle filter penetration) is used for testing filter penetration, and dictates the use of a non-neutralized, polydisperse 1% solution of sodium chloride (NaCl) against solid particles and paraffin oil against oil based aerosol at flow rate of 95 L min^{-1}. The test aerosol is fed into the chamber where the examined filtering device is mounted in a leaktight way on an adaptor. The NaCl aerosol is generated by a Collison atomizer under pressure of 3.45 bar at flow rate of 13 L min^{-1}, with a particle size range of approximately 40–1200 nm and a mass median diameter (MMD) of ≈ 600 nm. The output is mixed with 82 L min^{-1} of dry dilution air (to give total of 95 L min^{-1}) in a chamber with volume of ≥ 1750 cm^3. The generated polydisperse NaCl upstream and downstream of the respirator is then vaporized through a hydrogen flame and the intensity of light emitted at 589 nm, proportional to sodium concentration, is subsequently measured by flame photometry and penetration based on mass concentration ratio is obtained.

For testing against liquid and oil aerosol particles, paraffin oil (paraffinum perliquidum CP 27 DAB 7) is atomized at 100°C under a pressure of 4 bar, diluted with 50 L min^{-1} of filtered air, the generated test aerosol is reduced to concentration of 20 ± 5 mg m^{-3}, by releasing a fraction of oil mist, and further dilution in the cyclone with 83 L min^{-1} of filtered air. Under these conditions the particle size distribution is lognormal with a median Stokes diameter of 400 nm and GSD of 1.82. The mass concentration of aerosol is measured upstream and downstream of the respirator by a 45° light scattering photometer (BS EN, 2008).

Throughout the years, the performance of the FFRs has been the topic of extensive studies, resulting mainly in the characterization of aerosol penetration through filters using aerosols that are thought to be similar to those encountered in workplaces (Myojo and Sugimoto, 1997; Lee *et al.*, 2005; Rengasamy *et al.*, 2008; Jung *et al.*, 2014, Rengasamy *et al.*, 2015; Vo *et al.*, 2015; Gao *et al.*, 2016), while other studies were aimed at determining the most penetrating particle size (MPPS) and to examine whether biological aerosols were collected in a similar manner to inert aerosols of the same particle size distribution (Eninger *et al.*, 2008; Lee *et al.*, 2008; Rengasamy *et al.*, 2010; Lore *et al.*, 2012; Zuo *et al.*, 2013; Rengasamy *et al.*, 2014). Nonetheless, these studies of respirator penetration were focused mainly on N95 facepieces (not oil-proof with filtering efficiency ≥ 95%) or other type of facepieces approved by the National Institute for Occupational Safety and Health (NIOSH), rather than on CE-marked respirators.

Even though EN and NIOSH certifications are widely recognized in many parts of the world, they employ different test protocols for the certification process. Nowadays, the N95 facepieces are certified according to regulations in NIOSH 42 CFR 84, which dictate that filters are tested with charge-neutralized NaCl at 85 L min^{-1}. For these tests the particle size of ≈ 300 nm in diameter thought to be the MPPS (NIOSH, 1997). However, before first significant changes were made and the NIOSH regulation 42 CFR 84 was revised in 1990s, Moyer (1986) suggested ’worst-case‘ type aerosol for testing of electret filter penetration, including using dried, charge neutralized test aerosol (NaCl and DOP) of different sizes and a CMD based measurement method. Further studies on filter efficiency as a function of particle size and flow rate proved the shortcomings of the former NIOSH regulation 30 CFR 11 (Moyer and Stevens, 1989a, b; Stevens and Moyer, 1989). Some studies urged the need to employ methods and conditions allowing the measurement of fractional penetration over a range of particle sizes under a ’worst-case‘ scenario and pointed out the importance of particle size and flow rate in particle capture by electret filter (Brosseau *et al.*, 1989; Chen *et al.*, 1990, 1992; Brosseau *et al.*, 1993). Other studies focused on surgical masks penetration or the use of an inert aerosol to predict the collection of a biological aerosol by respirators (Chen and Willeke, 1992; Weber *et al.*, 1993; Brosseau *et al.*, 1994; Chen *et al.*, 1994). All this research had a significant impact and contribution to the improvement of the filter penetration testing and certification process of NIOSH-approved respirators. These issues are somehow similar to those in EN 149 and yet, up to this day, EN 149 still lacks critical updates in methodology used for CE-marked facepieces certification, taken by NIOSH over 20 years ago. There are limited number of studies in the literature reporting CE-marked respirator performance (Huang *et al.*, 2007; Plebani *et al.*, 2010; Ciotti *et al.*, 2012; Plebani *et al.*, 2012; Penconek *et al.*, 2013). Although in vast majority of these studies a MPPS of 30–60 nm was found, and charge-neutralized test aerosol and detection method relying on CMD were used, these studies investigated the penetration through the entire half masks in an experimental chamber (or with human subjects) using their own test method (or with a commercially available filter tester) focusing on a specific problem (diesel exhaust particles, oily aerosol, bioaerosol, filtering material properties, particle collecting mechanism, etc.).

Therefore, there is a need to perform a comparison study using a size-resolved method for the penetration evaluation of CE-marked respirators through all 3 filtering classes. We compared the penetration performance of filters from 13 commercially available filtering facepieces from FFP1, FFP2, and FFP3 filtering classes by utilizing 9 sizes of charge-neutralized monodisperse ammonium sulfate. EN 149 sets the MPPS on ≈600 nm MMD (40–1200 nm) as a certification criterion for the respirator penetration tests, while this study is focused on the size range 20–400 nm (CMD). The goal of this research is to demonstrate the disadvantages and shortcomings of the currently valid European Norm and show by independent measurement method that for this particle size range the penetration is much higher than it would be by following the CE certification protocol.

MATERIALS AND METHODS

Filtering Facepieces

Thirteen CE-marked commercially available FFRs from five different brands were selected (3M, Refil, Moldex, Respair, and Segre) from all three protection classes: FFP1 (3 respirators), FFP2 (4 respirators), and FFP3 (6 respirators). Two replicates of each FFR were purchased and randomly marked from 1 to 13. Two representative 47 mm diameter samples were cut out from each FFR as received (i.e., without any conditioning) in order to determine the penetration performance of the material throughout the respirator. The samples were cut from all masks always from the same position, one from the right nose side and one from the left side. Some of the respirators had a plastic mesh keeping the structure and shape of the respirator, and this was removed prior to inserting the filter into the filter holder. Table 1 contains the list of filtering facepieces used in this study and their characteristics.

Experimental Setup

The filter tester used in this study was an automated system developed in the Laboratory of Aerosol Chemistry and Physics (LACP) of the Institute of Chemical Process Fundamentals, v.v.i. (ICPF) of The Czech Academy of Sciences. The program operating the experiments employed an algorithm developed in the LACP laboratory and ran in LabView (National Instruments Co., Austin, TX, USA), which controlled the whole test system, the data recording, and the data analysis. Fig. 1 presents the schematics of the experimental setup used to measure particle penetration through the filters. In order to reveal any system leakage, zero volts were applied to the differential mobility analyzer (DMA) as a zero count check prior to each measurement. Two blank tests (without a filter in the filter holder) were also performed; one occurred before the launch of the very first measurement and one was at the end, after the measurements of all filters were done.

In order to generate the challenge monodisperse aerosol particles, clean, dry, pressurized air was delivered to the particle generator and salt solution was dispersed by a nebulizer (AGK 2000, Palas GmbH, Karlsruhe, Germany) under pressure of 2.5 bar. A 1 g L^{-1} solution of ammonium sulfate ($(NH_4)_2SO_4$) was chosen as the challenge aerosol over sodium chloride, which is used for the certification tests of CE-marked facepiece half masks (according to EN 13274-7:2008). This was due to the dodecahedron shape of generated ammonium sulfate particles, which represent the sphere shape better than the cubic crystals of sodium chloride (being closer to the theoretical assumption for particle sizing in electrostatic classifier). A sample flow of 2 L min^{-1} of generated polydisperse ammonium sulfate passed through a liquid droplet separator to remove the larger drops and the polydisperse aerosol was then dried in a diffusion dryer (homemade, ICPF workshop, Prague, Czech Republic). The aerosol flow rate was checked prior to the start of each experiment with a Gilibrator-2 flow calibrator (Standard cell, Sensidyne LP, St. Petersburg, FL).

In the next step a Boltzmann charge equilibrium was imparted on the particles in the ^{85}Kr neutralizer (10 mCi, 370 MBq). The monodisperse aerosol particles of the required mobility diameter were subsequently selected in the electrostatic classifier of the DMA (homemade, Vienna type, ICPF workshop, Prague, Czech Republic) with active length of 22 cm. During all experiments the sheath airflow inside the DMA was maintained at ≈ 10 L min^{-1} and was recorded by a mass flow meter (Model 4040, TSI Inc., Minneapolis, MN, USA) along with pressure and temperature. The monodisperse particles then went through another ^{85}Kr neutralizer and the sample was diluted by 8 L min^{-1} of clean, dry air before entering the mixing volume. This was checked by another mass flow meter (Model 4040, TSI Inc., Minneapolis, MN, USA) prior to each measurement.

Afterwards, the generated monodisperse ammonium sulfate particles passed through the stainless steel filter holder (homemade, ICPF workshop, Prague, Czech Republic) with the tested 47 mm filter in it. Our filter holder consists of two 1-inch stainless steel tubes with customized tri-clamp ferrule

Table 1. List of filtering facepieces respirators and their characteristics.

FFR test number	FFR model	Protection class	Certifi-cation	Respirator characteristics		
				Filter Layer Material	Dispos-ability	Dolomite clogging
1	[a]Refil 511	FFP1	CE 1024	Polypropylene	Non-R	No
2	[b]Moldex 3505	FFP3	CE 0121	Polypropylene	Non-R	No
3	[c]3M 9312^{+}	FFP1	CE 0086	Polypropylene	Non-R	Yes
4	[d]RespAir C	FFP3	CE 0086	Polypropylene	Non-R	No
5	[a]Refil 831	FFP2	CE 1024	Polypropylene	Non-R	No
6	[c]3M 8835	FFP3	CE 0086	Polypropylene	Reusable	Yes
7	[b]Moldex 3405	FFP3	CE 0121	Polypropylene	Reusable	Yes
8	[e]Segre CN P3	FFP3	CE 0194	Polypropylene	Reusable	Yes
9	[b]Moldex 2405^{+}	FFP2	CE 0121	Polypropylene	Non-R	Yes
10	[c]3M 9310	FFP1	CE 0086	Polypropylene	Non-R	Yes
11	[c]3M 9322	FFP2	CE 0086	Polypropylene	Non-R	Yes
12	[a]Refil 731	FFP2	CE 1024	Polypropylene	Non-R	No
13	[e]Segre CN P3 V	FFP3	CE 0194	Polypropylene	Reusable	Yes

[a] Refil spol. s.r.o., Karlovy Vary, Czech Republic; [b] Moldex, Culver City, CA, USA; [c] 3M, Maplewood, MN, USA; [d] Respair, Aylesbury, Buckinghamshire, UK; [e] Segre AB, Örebro, Sweden.

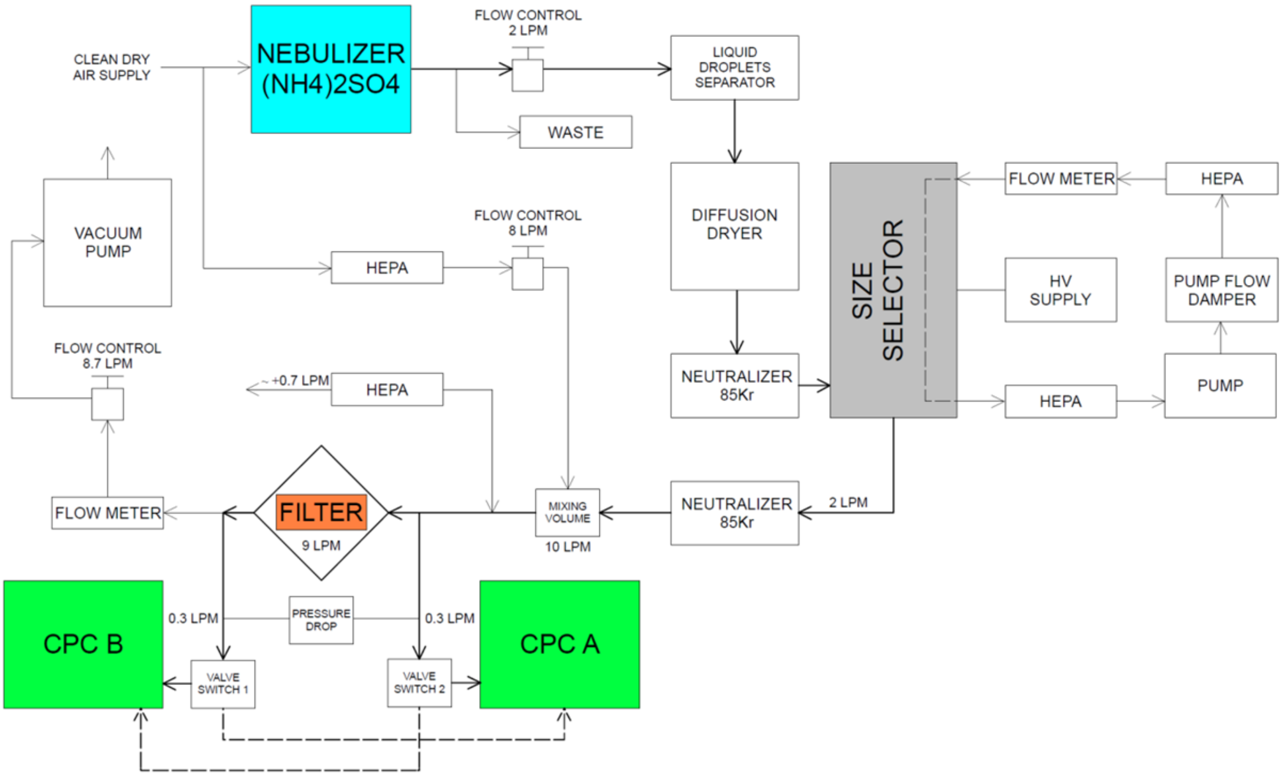

Fig. 1. Schematics of experimental setup used in this study to measure the particle penetration through the filters.

add-ons placed on the end. The filter is then placed between two customized ferrules. The two tubes are connected with a simple clamp, which can be removed in order to disconnect the tubes to insert/remove the filter. Instead of a classical gasket there are two customized PTFE rings ensuring perfect fit and seal of the filter in the filter holder. All filters were challenged under a flow of 9 L min^{-1}, which corresponded to a face velocity of 10.56 cm s^{-1} (with effective filter area 14.27 cm^2). In order to maintain the same face velocity for the cut sample and the whole respirator, the flow rate of 9 L min^{-1} through the sample was calculated from flow rate of 95 L min^{-1} (representing heavy work breathing conditions) with assumption of effective filter area estimated to 150 cm^2.

Particle number concentrations upstream and downstream of the filter were simultaneously measured by two ultrafine condensation particle counters (UCPC 3025A, TSI Inc., Minneapolis, MN, USA), which were sampling on low flow mode at 0.3 L min^{-1}. Additionally, our test system included two three-way electromagnetic switching valves that altered the position of the two CPCs after each particle size measurement in order to compensate for possible differences in the readings of the two CPCs. Thus, each size was measured twice both upstream and downstream of the filter with alternating measurement position of the two CPCs.

As a steady-state particle concentration during every scan is crucial for the correct calculation of the penetration of filtering materials and for the estimation of the MPPS, a regular concentration stability check was done after every change in the particle size and during every single measurement. In addition to the flow rate and the upstream and downstream concentrations, the pressure drop across the filter was also recorded by a pressure sensor (GMSD 25MR-K31, Greisinger Electronic GmbH, Regenstauf, Germany) connected to a pressure measuring device (GMH 3155, Greisinger Electronic GmbH, Regenstauf, Germany). Measuring the pressure drop across the filter also gave us an indication of the leak-free placement of the filter in the filter holder.

Data Analysis

The calculation and data analysis of penetration and standard deviation were performed by the program algorithm that controlled the entire test system. Penetration (*P*) can be expressed as:

$$\mathrm{P}(\%)=\frac{\mathrm{C}_2}{\mathrm{C}_1}\,100, \tag{1}$$

where *P* is the penetration defined as the ratio of the concentrations downstream (*2*) and upstream (*1*) of the filter. Taking into account the altering of the CPCs' position, the aerosol particle penetration through the filter and the standard deviation σ_P of the measured penetration were determined for each particle size as follows:

$$\mathrm{P}(\%)=\sqrt{\frac{c_{2B}\cdot c_{2A}}{c_{1B}\cdot c_{1A}}}\,100, \tag{2}$$

$$\sigma_{\mathrm{P}}(\%)=\frac{1}{2}\sqrt{\frac{c_{2B}\cdot c_{2A}}{c_{1B}\cdot c_{1A}}}\;?\sqrt{\frac{\sigma_{C1A}^2}{C_{1A}^2}+\frac{\sigma_{C2A}^2}{C_{2A}^2}+\frac{\sigma_{C1B}^2}{C_{1B}^2}+\frac{\sigma_{C2B}^2}{C_{2B}^2}}\,100, \tag{3}$$

where c_{1A}, c_{1B}, c_{2A}, and c_{2B} represent the concentrations upstream (*1*) and downstream of the filter (*2*) measured by the CPCs (*A* or *B*) and σ_{C1A}, σ_{C2A}, σ_{C1B}, and σ_{C2B} are the corresponding standard deviations to these measurements.

Eq. (3) was calculated by the error propagation principle from the Eq. (2), similarly to work of Zíkova *et al.* (2015). The previously mentioned variables in the Eq. (2) are the median values calculated from all the scans done while measuring a certain particle size for the specific variable (c_{1A}, c_{1B}, c_{2A}, c_{2B}). The length of each scan was one minute. Certain time delay was given to the system to stabilize after changing the size of the particles. This delay depends on number concentration measured by downstream CPC (starting from 5 seconds for concentrations > 1000 # cm^{-3} and going up to 5 minutes for concentrations <1 # cm^{-3}). After this time delay the system checks the stability prior to the measurement - taking the measurements for the whole selected measurement period and checking if the concentration changes more than 5% during the scan. If the change in concentration is larger than 5% than it continues checking until this criterion is met for both upstream and downstream CPC. Moreover, before the set of scans for given particle size the concentration on downstream CPC is measured and based on the measured concentration, the number of scans for this particular particle size is set (again starting from 2 scans per size for concentrations > 1000 # cm^{-3} and going up to 10 scans for concentrations <1 # cm^{-3}). This procedure should improve statistics in the case of lower concentrations of testing particles. The last stability check is done after measuring every single scan. The data from both CPC downstream and upstream the filter are checked in similar way like during the pre-measurement check and if the criterion is not met, the measurement continues until the total number of pre-set correct scans is reached. Furthermore, each scan could have theoretically been repeated an infinite number of times. The algorithm compares the stability of the concentration upstream and downstream of the filter at the end of each scan and evaluates it as sufficient or insufficient; in the latter case, the scan was repeated until a stability criterion was met. After the selected particle size was measured successfully in CPC position A-B the program altered the switchers into B-A position and this particle size was measured again the same way.

The nine particle sizes selected for the comparison of the filter penetrations ranged from 20 to 400 nm (20-35-50-70-100-140-200-280-400 nm) of count mobility diameter (CMD). This size range, as well as the particle sizes, remained the same for all the measurements. The selection of the particle size range for the size-resolved measurements, and the lower and upper size limits (20 and 400 nm), were given by the used aerosol generator. Particles with smaller and bigger diameters were generated with too low concentrations to overcome the CPCs' minimum counting limit downstream, and, therefore, they were under the detection limit of our filter testing system. The calculated MPPS were determined using the lognormal fit for each separate penetration curve. Measured MPPS can differ from the fitted one up to 10 nm, which is a considerable difference in some cases. Even though the resolution of size-resolved method allows to measure with better resolution, in order not to make the single measurement unnecessary long only limited amount of sizes was selected. Thus this information does not offer in detail where exactly the real MPPS is, the lognormal fitting can estimate the position of MPPS reliably. All presented results of MPPS, as well as penetrations, are the median values of individual filters representing the specific FFRs. Median values were chosen over mean values in all calculations mainly due to the fact that the mean values of the repeated measurements do not reflect the prevailing penetration values while median value is less burdened with the possible outliers in the results.

This test system has several advantages over commercial automated testers. As the system was developed in the LACP laboratory, there is full control of the key parameters, and all important parts of the system are exchangeable. Thus, the system is adaptable for various testing needs, gives detailed size-resolved penetration results, and allows for the determination of the real MPPS. Corrections for possible differences in measurement of the two detectors are not dependent on any initial calibration or estimation of a correction factor.

Evaluation of the System with Standard Filter Medium

We regularly used the system over the last few years to measure penetrations of filtration materials for several companies and manufacturers with excellent repeatability. So far the only published work proving the stability and advantages of our system and measurement method was published by Zíková *et al.* (2015) where the results had very good repeatability. In order to verify the performance of our filter testing system, we performed repeated tests using standard filter medium HB 5893 (Hollingsworth & Vose, East Walpole, MA, USA). This test was performed at the same conditions as all the tests of the commercial FFRs, flow rate through the filter 9 L min^{-1} (corresponding face velocity 10.56 cm s^{-1}) and the diameter of the cut sample of 47 mm. The test was performed using ammonium sulfate particles (1 g L^{-1} solution). The main reason of this setup is the limitation of our system, especially the face velocity of 2.5 cm s^{-1} (as described in HB 5893 technical data sheet), which would be on the edge of the measurement range of our system.

As can be seen in Fig. 2, the measured penetration during repeated measurements are close to each other suggesting very good homogeneity of the standard material and very good repeatability of our measurement system. The MPPS of HB 5893 is close to 100 nm and the corresponding maximum penetration in this case was 0.02–0.03%. The comparison of these standard filter medium results obtained by our system with the values given in the HB 5893 technical data sheet was not possible due to several important factors:

1. Different measurement procedures–the standard test measures overall penetration (one penetration value), one the other hand, our test gives detailed size-resolved penetration.
2. Different detection systems used–standard test uses photometers, whereas our method uses single count CPCs.

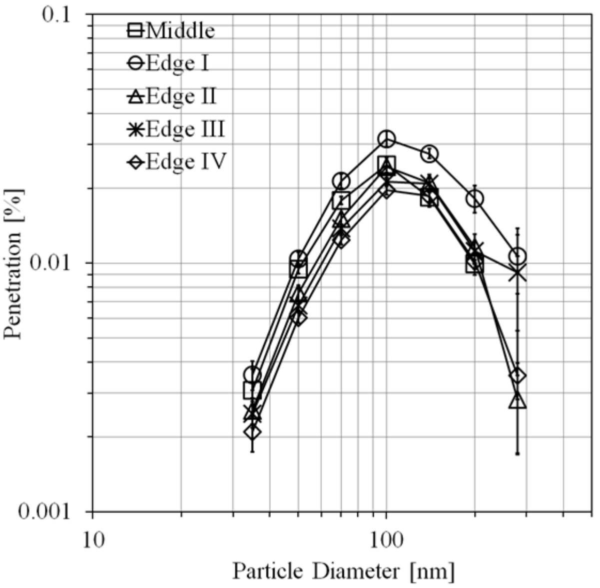

Fig. 2. Penetration of ammonium sulfate through standard filter material HB 5893. The error bars represent the standard deviation.

3. Different face velocities (due to the limitations of our system).

RESULTS

FFP Penetrations

Three facepiece respirators were challenged from the filtering class FFP1; Fig. 3(a) demonstrates the results from these measurements. Although all three respirators were within the FFP1 maximum penetration limit of 20% (at 600 nm MMD), the measurements showed a rather wide differences between filters from respirator 1, which had median penetration (CMD) at MPPS equal to 13.9%, and filters from FFRs 3 and 10, which had penetrations reaching 4.9 and 4.1%, respectively.

Differences in the FFRs penetration and performance were also observed in the FFP2 class (see Fig. 3(b)). The percentage penetration criterion of this filtering class is ≤ 6% (at 600 nm MMD). With exception of FFR 12, penetrations of all the filters from FFRs in this class ranged from 3.5 to 6.4% (CMD). Respirator 12 had, in all four filter measurements penetrations > 6%, with a median penetration of 17.7% (11.8–34.3%) at MPPS.

Considering that the FFP3 has the highest protection level, requiring a maximum penetration ≤ 1% (or a filtration efficiency 99% at 600 nm MMD), we chose 6 half masks to be tested. 4 out of 6 respirators showed median penetration higher than 1%, as seen in Fig. 3(c). It is also worth mentioning that none of the 13 filters among these four respirators met the ≤ 1% criteria. On the other hand, filters from respirators 4 and 6 provided results with median penetrations of 0.4% and 0.03% (CMD), respectively.

Comparison of Within-Respirator Penetration

As we examined two filter samples from each respirator, we also compared the within-respirator penetrations from these filters, and also from two identical respirators (see Figs. 4(a), 4(b) and 4(c)). In some cases (FFRs 2II, 4, 5, 6, 7II, 8, 8II, 9, 9II, 10, 11 and 13II) the penetration difference between two samples from the same respirator varied from 0.02–0.8%, which is less than 1%, and in other cases, the penetration difference was found to be over 1%, between 1.3–6.8% (FFRs 1, 1II, 3, 5II, 11II, 12 and 13). Results from respirator 12II from FFP2 reached exceptionally high penetrations (equal to 17.9 and 34.3%) and also had wide difference (16.4%) between the two examined samples.

Penetration also varied between two identical respirators and ranged from 0.02 to 2.5% (see Fig. 4(c)). The highest penetration difference, however, was found between respirators 12 and 12II, and was equal to 11%. Comparing the total penetrations from all the examined filters representing the individual FFRs show that the percentage penetration results from filtering classes FFP1, FFP2, and FFP3 overlap (3.2–16.3% for all filters from the FFP1, 2.4–34.3% for all filters from the FFP2, and 0.02–3.3% for all filters from the FFP3).

The MPPS was determined from the position of the maximum on the penetration curve by fitting lognormal peak shape and the hereby presented results are the median values for all filters representing the FFRs (see Table 2). The MPPS range for all the FFRs was found to be between 28–47 nm with exception of respirator 1 (FFP1), 2 (FFP2) and 9 (FFP3) ranging from 52 to 59 nm.

DISCUSSION

Conditions and test protocol for measurements in this study were different than those used for CE certification, thus the results obtained in this study may not predict the results that would be obtained by the CE certification test method. It is acknowledged that the method used in this study to evaluate the penetration performance of the filtering half masks is based on isolated measurement of 47 mm filtering material cut out of the FFRs unlike the EN 149 method, which tests the FFRs in a test chamber tightened on a speciman. Thus, the results presented in this article represent the percentage penetrations obtained by our measurement method against the FFP limits set by the EN 149. This method offers several advantages. First, penetration measurement of any specific part of the chosen respirator and adjustment of the diameter of the filter is possible according to the special needs of the measurement. Second, comparing the performance of several filters from the same facepiece would allow for the possibility of determining the homogeneity of the FFR filtering material if needed. As was previously mentioned, detailed size-resolved penetration results allow for the determination of the real MPPS based on CMD. Hence, this method provides more critical results in regards to leak control, non-charged particles, and finding the real MPPS.

One could argue that hereby presented results were obtained by a method that is not based on the European Norm. EN 149 uses a protocol where the FFRs are challenged with non-neutralized polydisperse aerosol nebulized from a

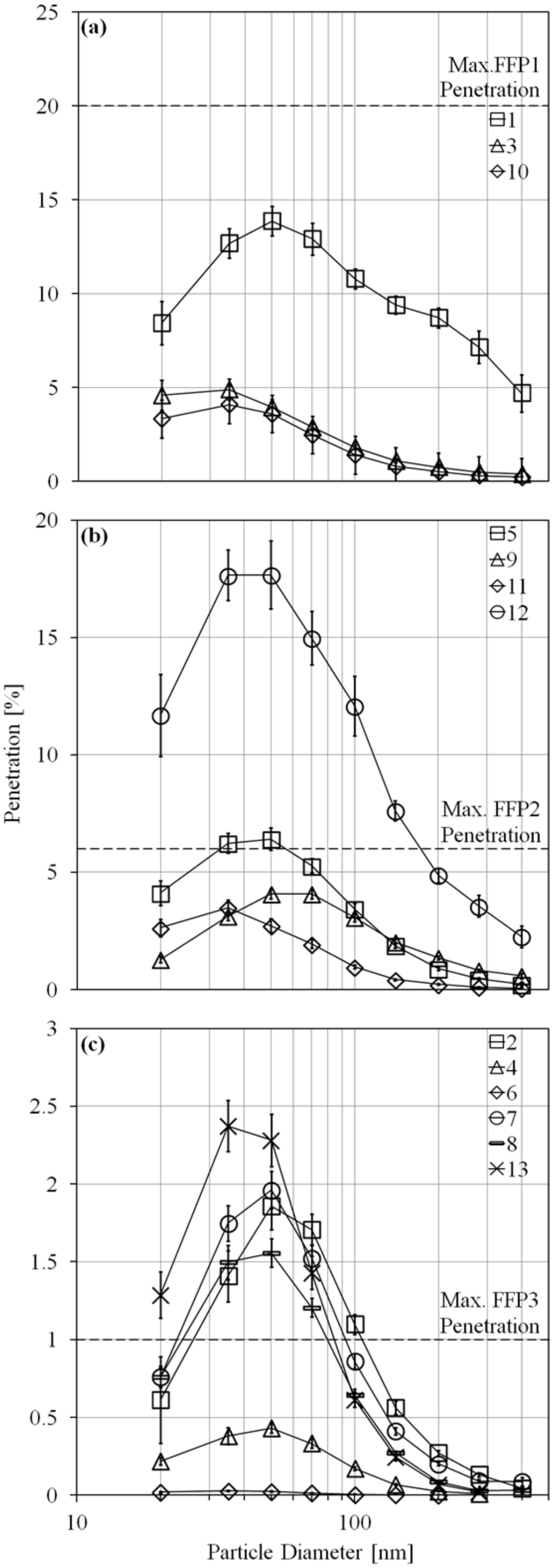

Fig. 3. (a) FFP1, (b) FFP2, and (c) FFP3 filtering classes median size-resolved penetrations. The error bars represent the standard deviation.

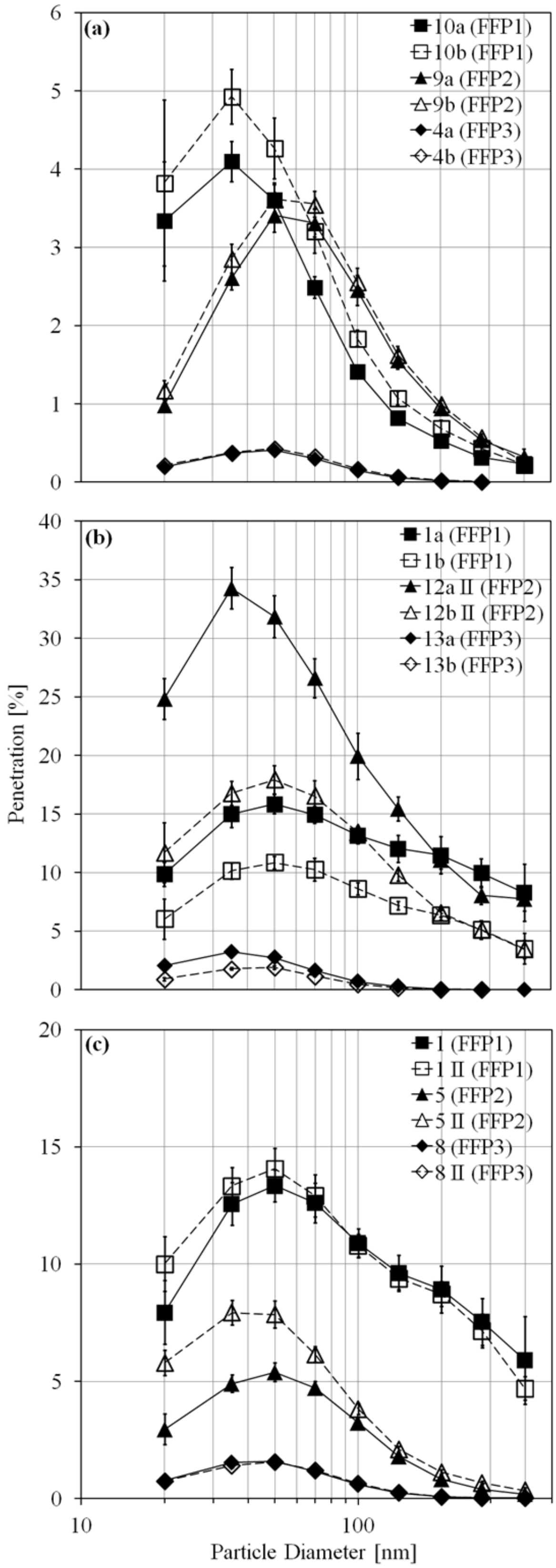

Fig. 4. Penetration difference: (a) < 1%, (b) > 1%, and (c) between 2 identical respirators. Letters 'a' and 'b' indicate two filters from the same FFR, and 'II' indicates filters from the second (identical) FFR. The error bars represent the standard deviation.

Table 2. Median values of MPPS, pressure drop, and the respirators performance.

FFR test number	Protection class	MPPS - lognormal fit [nm]	MPPS - measured [nm]	MPPS percentage penetration [%]	Pressure drop [kPa]
1	FFP1	58	50	13.9	0.07
2	FFP3	52	50	1.9	0.15
3	FFP1	28	35	4.9	0.07
4	FFP3	43	50	0.4	0.21
5	FFP2	43	50	6.4	0.11
6	FFP3	37	35	0.03	0.23
7	FFP3	47	50	2.0	0.15
8	FFP3	44	50	1.6	0.21
9	FFP2	59	50	4.1	0.18
10	FFP1	33	35	4.1	0.08
11	FFP2	33	35	3.5	*0.12
12	FFP2	40	50	17.7	*0.04
13	FFP3	39	35	2.4	*0.19

* pressure drop data available only for one of the measurements.

1% solution of polydisperse NaCl (40–1200 nm) with MMD on ≈ 600 nm. However, utilizing non-neutralized polydisperse NaCl for the certification of filtering facepieces does not represent the worst-case scenario (the maximum penetration) for testing of the electret filter and the use of polydisperse test aerosol does not provide with detailed size-resolved penetration, thus, it is not possible to find the real MPPS.

Neutralization of the test aerosol is of a great importance; when both the filter fibers and aerosol particles are charged, the Coulombic forces significantly enhance the capture of the particles and reduce the particles penetration. On the other hand, for a mechanical filter (i.e., with non-charged filter fibers) with low efficiency there are no significant differences between penetrations of charged and non-charged aerosol particles (Balazy *et al.*, 2006). For a charged filter fiber (electret filter) the MPPS of 300 nm (or > 300 nm) is insufficient as a limit for FFRs to pass the penetration test, and usage of a non-neutralized test aerosol may lead to distorted results of protection level. Based on the Single Fiber Filtration Theory, NIOSH accepts the size of 300 nm in diameter as the MPPS for particulate filters. Investigations have demonstrated that the MPPS is quite dependent on the operational conditions, and for non-charged filter fibers MPPS would be indeed ≈ 300 nm, but the particle penetration peak would reach as high as ≈ 80%. In order to provide adequate filter efficiencies, electret filters rely heavily on their electrostatic charge. Therefore, when compared to mechanical filters with non-charged filter fibers, the polarization force on the charged filter fiber with a fiber charge density of 13 nC m^{-1} would decrease the filter penetration from ≈ 80% to ≈ 5% and cause the shift of MPPS towards ≈ 50 nm (Martin and Moyer, 2000; Balazy *et al.*, 2006).

According to the Fuchs theory (Fuchs, 1963), in the size range where the MPPS occurred in all measurements (30–60 nm) less than 2% of particles have a double charge while over 20% of 60 nm particles are single charged (> 4.4%). Therefore, the penetration results are more sensitive at these particle sizes (≈ 15% of 400 nm particles have double charge and only about ≈ 20% have a single charge), which can cause uncertainty in the penetration measurement. Nevertheless, the mode of the aerosol size distribution produced by our generator, under the operational conditions and with given ammonium sulfate solution, is around 70 nm, which means that particles bigger than 70 nm will have every time much higher concentration then their doublets (doubly charged particles). Even though in the case of this study it rather caused an overestimation of the penetration for particles bigger than 100 nm, the position of MPPS was little affected by double charged particles and was still within the allowed uncertainty limit of the measurement.

EN 149 sets the criterion for the respirator penetration tests even lower; it is on ≈ 600 nm (MMD). Interestingly, for all the measured filters from FFRs in this work, the MPPS occurred in the range of 30–60 nm (CMD) at 9 L min^{-1} flow rate (with a face velocity equal to 10.56 cm s^{-1}, which is equivalent to 95 L min^{-1} aerosol flow at 150 cm^2 of effective filter area). The current findings are consistent with previous studies that report MPPS in the same range during CE-marked and NIOSH-approved half masks penetration measurements (Huang *et al.*, 2007; Rengasamy *et al.*, 2008; Rengasamy *et al.*, 2009). Under the assumption that the generator dispersing the NaCl has a lognormal size distribution with a GSD of 1.90 and taking into account the change from aerodynamic to mobility particle diameter of NaCl particles (ρ_p = 2.16 g cm^{-3}), the Hatch-Choate equations (Hinds, 1999) can be applied to calculate the CMD used in our method, which would be ≈ 80 nm (equivalent to ≈ 600 nm of MMD).

Although it may appear that ≈ 80 nm is much closer to the MPPS results found in this study, the results from these two methods are not comparable and, therefore, the previously mentioned calculation has an informative character only. The MPPS, however, is characterized as the size of a particle with the highest relative particle number concentration passing through a specific filtrating material and is also dependent on the detection method and operational conditions. The particle mass size distribution downstream of the filter shifts the MPPS towards particles with smaller diameter (< 400 nm) and lower mass, while the filtering material retains a larger part of bigger particles (≈ 400–1200 nm). Subsequently, this leads to underestimation of the particle penetration, as the flame photometry detection method is less sensitive to particles with smaller diameters (due to their lower mass contribution), which results in a misleading estimation of penetration. Thus, detection methods based on MMD should not be used for estimation of the penetration and MPPS of filtering facepieces.

Additionally, toxicity and health effects of nanoparticles do not necessarily depend on their mass. Aerosol of biological origins, such as viruses and bacteria, can be highly infectious if inhaled even at a very low dose and the related health effects may depend on the number of inhaled particles, not particle mass (McCullough and Brosseau, 1999; Donaldson *et al.*, 2006; Poland *et al.*, 2008; Jones and Brosseau, 2015). Furthermore, Huang *et al.* (1998) suggest that the aerosol penetration of the MPPS may locally be higher than the required penetration limit because of the fiber structure of the respirator. This may be caused by the non-homogeneity of the filter medium, which could be explained by manufacturing process, different chemical composition of the filter media, or different methods of introducing electric charge onto the filter fibers.

Based on the 89/686/EEC directive, during the process of CE certification in the country where the respirators are manufactured the notified body conducts the tests under EN 149 and EN 13274 protocols and is responsible for both type examination and checks on the final product (or monitoring of production). All respirators tested in this study were EN certified and therefore already passed the EN 149 penetration tests, assuming all notified bodies follow the same norm for examining and certifying the filtering facepieces. It is unclear why only one of the five manufacturers, which was examined and certified by the same notified body, had satisfactory maximum penetration levels according to our tests and as well to EN 149 as the tested half masks were already certified. Although the vast majority of the other respirators exceeded their FFP penetration limits according to our testing method, they successfully passed the EN 149 penetration tests.

CONCLUSIONS

Experimental data obtained by a size-resolved method (20–400 nm CMD) of measuring penetrations of 47 mm filters from CE-marked filtering facepieces revealed that the penetration difference between 2 samples from same respirator was in most cases up to 6.8%, and between 2 identical respirators up to 2.5%. Comparison of the total penetrations from all the examined filters showed percentage penetration ranges 3.2–16.3% (FFP1), 2.4–34.3% (FFP2), and 0.02–3.3% (FFP3). The MPPS determined from lognormal fitting of the penetration curves was found to be between 30 and 60 nm (CMD) in all measurements. As expected, in many cases the real penetration of these respirators was beyond of what is allowed by the European Standards, which is mainly due to usage of non–neutralized, polydisperse test aerosol, inadequate detection method relying on the particulate mass, and the assumption that the MPPS is on ≈600 nm (MMD) as the criterion for filtering facepieces to pass the penetration test. These conditions and test methods lead to underestimation of particle penetration, especially in the nanoparticle size range (< 100 nm). Based on this study, we conclude that the recently valid European Norm EN 149:2001+A1:2009 is neither sufficient nor efficient for estimation of particle penetration through filtering half masks and may need modification in relation to a more sophisticated testing method and conditions.

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DISCLAIMER

The authors declare no conflict of interest relating to the material presented in this Article. Mention of commercial product or trade name does not constitute endorsement by the Technical University of Crete nor the Academy of Sciences of the Czech Republic. The findings and conclusions of this work are solely those of the authors.

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