

# Detachment of tobacco-smoke-material carriers from surfaces by turbulent air flow

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Received 9th May 2008, Accepted 3rd October 2008

First published as an Advance Article on the web 6th November 2008

DOI: 10.1039/b807918h

The influence of tobacco-smoke-material (TSM) on the detachment of microparticles from surfaces by turbulent air flow was investigated experimentally. Both clean and dusty glass surfaces were subjected to TSM either before or after the deposition of 64  $\mu\text{m}$  to 76  $\mu\text{m}$ -diameter stainless steel microparticles onto the surfaces. The TSM was generated by mechanically puffing research-grade cigarettes inside a smoking box that contained the surfaces. Microparticle detachment characteristics were studied in a wind tunnel using video microphotography. Measured nicotine concentration was used to determine the amount of TSM deposited on a surface.

The 5% and 50% threshold velocities for detachment were used to quantify the effect of TSM on microparticle detachment. These velocities were compared with those obtained using a clean surface with no TSM exposure. The effect of TSM exposure on microparticle detachment depended significantly on whether exposure occurred before or after microparticle deposition. TSM exposure before microparticle deposition had little effect. TSM exposure after deposition delayed detachment to much higher velocities. The presence of dust on the surface with TSM also delayed detachment and increased the variability in the detachment velocities as compared to the case of a clean surface with no TSM exposure.

## 1 Introduction

Particles of different shapes and sizes are ubiquitous in the environment and can be entrained into heating, ventilation and air conditioning (HVAC) systems. These particles can act as tobacco-smoke carriers (TSCs) when tobacco smoke material (TSM - the vapor, liquid and solid material present in tobacco smoke) is deposited on them. Surface-resident TSCs can be detached by air flow, re-entrained into circulating air inside HVAC systems, and eventually respired into the lungs. In contrast to the usual direct and secondary pathways of TSM to humans, this represents an additional, indirect pathway.

The authors previously have studied the detachment of microparticles from clean surfaces by air flow.<sup>1,7-9</sup> The present work aims to expand this understanding to particles that interact with tobacco smoke and become entrained into HVAC systems. In these situations, TSM can be continuously generated and deposited onto surfaces. These surfaces can contain previously deposited particles (for example, dust). TSM deposition also can occur on particles that enter into or are re-entrained within a tobacco-smoke environment, which includes its ventilation system. This continuous process of deposition and entrainment

causes new TSCs to be deposited on surfaces already exposed to TSM and also subjects surface-resident TSCs to new TSM.

The situation under consideration involves several related areas that include the mechanics of particle detachment from surfaces, tobacco smoke materials, and particle transit through HVAC systems. Reviews on particle detachment and entrainment mechanisms have been presented by Ziskind,<sup>17</sup> Ziskind *et al.*,<sup>18</sup> Nicholson<sup>10</sup> and Sehmel.<sup>13</sup> Reviews on the contents, physical, chemical properties, health effects and collection of tobacco smoke have been made by Husgafvel-Pursiainen,<sup>6</sup> DeMarini,<sup>4</sup> Witschi *et al.*<sup>16</sup> and Dube and Green.<sup>5</sup> Studies on particle movement in HVAC systems include, for example, those of Sippola and Nazaroff,<sup>15</sup> and Siegel and Nazaroff.<sup>14</sup>

This work is an initial study to identify the overall effect that TSM exposure has on the detachment of microparticles from clean and dusty surfaces by air. In these experiments, only *one* TSM-exposure concentration was studied. More detailed studies involving variable TSM concentrations are definitely helpful but are beyond the scope of the present work.

In the following, the results of experiments conducted to quantify three limiting situations of the aforementioned process for both clean and dusty surfaces are presented. The first situation, a base case, considers the detachment of particles from a clean surface without TSM exposure. This situation was studied previously by the authors in detail and is repeated here for comparative purposes. The second situation represents a limiting case when TSCs that are deposited on surfaces containing TSM. For the present experiments, the TSCs were deposited within minutes of TSM exposure. The third situation represents a limiting case when surface-resident TSCs are exposed to TSM.

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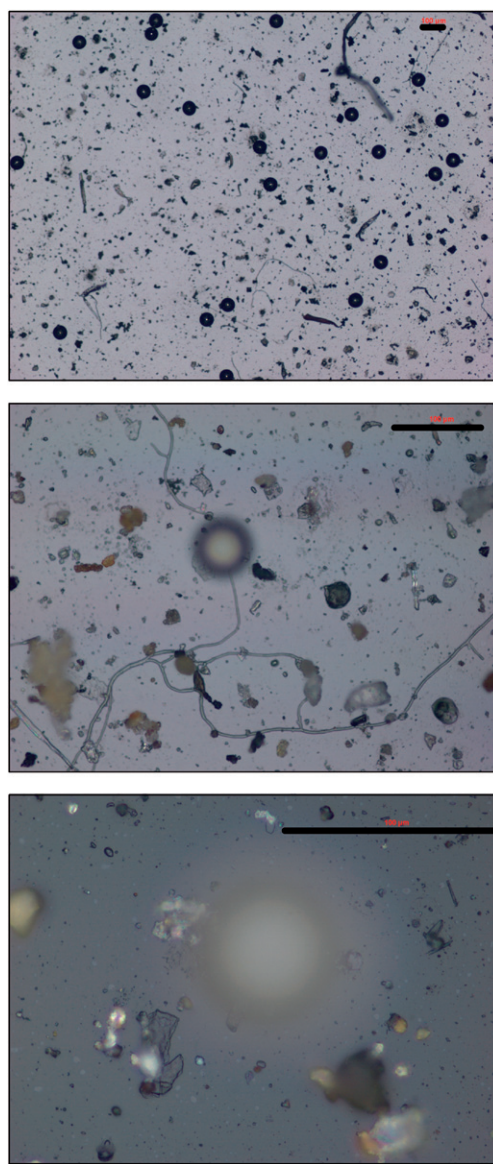
For the present experiments, existing TSCs are exposed to TSM generated just minutes before the detachment experiment.

## 2 Experimental approach

The experimental approach consisted of two phases. In the first phase, described in subsection 2.1, a glass plate was placed inside a custom-made cigarette-smoking box and exposed to TSM. The nicotine component of the TSM was selected as measure that could be used to determine the amount of TSM present on a plate. Nicotine was chosen because the extraction and analysis methodologies used for it have been clearly established. A gas chromatograph/mass spectrometer (GC/MS) system was used to quantify the amount of nicotine deposited on the plate. In the second phase, described in subsection 2.2, a plate was exposed to a slowly-accelerating air flow in a wind tunnel and video microphotography was used to view and quantify the amount of particle detachment.

Stainless steel microspheres (Duke Scientific; diameter: 64  $\mu\text{m}$  to 76  $\mu\text{m}$ ; density: 8000  $\text{kg}/\text{m}^3$ ; Poisson's ratio: 0.28; Young's modulus: 215 GPa) were used as TSC simulants. These were chosen because their detachment characteristics have been studied before in detail by the authors, they are available commercially in near mono-disperse diameters, and they provide significant visual contrast on the glass surface, which permits accurate counting. Further, their size is comparable to the TSCs that act as carriers of smaller, inhalable particles within HVAC systems. The microparticles were deposited (either before or after TSM exposure) as a sparse monolayer (less than  $\sim 50$  microparticles/ $\text{cm}^2$ ) onto a glass plate (Amersham Pharmacia; 10 cm  $\times$  10.5 cm  $\times$  1.27 mm). Previous work<sup>7,8</sup> has shown that the particle layer may be considered uniform for detachment studies as long as the number of particles per unit area in different randomly selected areas within the field of view are on the same order of magnitude. This was confirmed in the present study by manually counting the number of particles in random squares of equal area in the images acquired before initiating flow. Deposition was accomplished *via* gravitational settling from a height of approximately 10 cm immediately before placing the plate into the smoke-exposing box. The microparticles and plates were chemically inert with respect to all of the TSM components.

'Clean' glass plates were prepared using the technique described by Phares *et al.*<sup>11</sup> One plate's surface was scanned using an atomic force microscope, revealing an average standard deviation in the asperity heights of approximately 17  $\text{\AA}$ . The histogram of the standard deviation of the plate's asperity heights is presented in Ibrahim *et al.*<sup>9</sup> This surface roughness reduces the force required to detach the particle from the surface (the pull-off force) to approximately 1% of its smooth-surface value, according to the theory of Cheng *et al.*<sup>1</sup> Any larger values of the standard deviation of the asperity heights, such as those for Formica<sup>®</sup> or galvanized-steel surfaces, will cause slightly less than a 1% further reduction in the pull-off force. Thus, the glass surface provides a reasonable representation of real surfaces for detachment studies, while providing suitable contrast with the microparticles for both clean and dusty situations. A dusty plate was characterized by microscopic images taken with an optical microscope (Olympus microscope BX51M) at different magnifications, as shown in Fig. 1.



**Fig. 1** Optical microscope digital images at different magnifications of a dusty glass plate used in these experiments. The solid line is 100  $\mu\text{m}$  in each image.

### 2.1 TSM exposure methodology

A cigarette smoking apparatus (Fluid Metering, Inc; stroke rate controller model V200 and pump model QV1-PM6014) was used to puff research cigarettes (K2R4F from the Kentucky Tobacco Research and Development Center). Each K2R4F cigarette contained 11.70 mg of total particulate matter, 9.70 mg tar, 0.85 mg nicotine, 1.12 mg water, 13.00 mg CO and 0.22 mg  $\text{NO}_x$ . The cigarettes were smoked from an initial length of 84 mm to a final butt length of 35 mm. The outlet of the apparatus was connected to a custom-made smoke exposure box (60 cm  $\times$  29 cm  $\times$  29 cm) that contained the glass plates. The plates were placed on the floor of the box in a horizontal position. Two cigarettes were smoked for seven minutes and the smoke was left in the box for 15 minutes. These exposure duration and volume conditions were sufficient to achieve a nicotine concentration of

0.04 mg per mL of solvent, which was above the detection limit of the GC/MS system used (0.01 mg of nicotine per mL of solvent). The box then was opened and the plates removed. The TSM deposited on one glass plate was extracted with an alkaline solvent as described by the CORESTA recommended method No. 9.<sup>12</sup> The alkaline solvent used for samples and standards was made of 500 mL of methanol, 0.5 g of sodium hydroxide and 0.25 g of n-heptadecane. Another plate subjected to the same exposure history was placed in the wind tunnel for microparticle detachment experiments.

GC/MS analysis was performed using a GC system (Hewlett Packard 6890 series) equipped with a flame ionization detector and an HP-5MS column (30 mm length  $\times$  0.25 mm internal diameter  $\times$  0.25  $\mu$ m film thickness). Mass spectra were recorded on a JEOL GCmate in the EI<sup>+</sup> scan mode.

Nicotine (99% GC purity), n-heptadecane (an internal standard with 99% purity), and MeOH (LC MS grade with 99.9% purity) were used as received. Other solvents were distilled prior to use. Methanol containing 1 g/L of NaOH and 0.5 g/L of n-heptadecane served as an internal standard. A series of nicotine standards were prepared for calibrations by dissolving nicotine in the solvent. The concentrations of the nicotine standards covered the range expected to be found in the cigarette samples. The standards were stored in refrigerator until use.

The internal standard and the nicotine peaks were separated quantitatively by applying the following temperature program: The starting column temperature was 50 °C; the first temperature ramp of 30 °C/min was applied up to 160 °C; the second temperature ramp of 15 °C/min was applied up to 250 °C for a total run of 9 min with a delay time of 2 min. The injection temperature was 250 °C, the detector temperature was 285 °C, and the carrier gas was helium with an injection volume of 1 L. The total analysis time was about 9 min.

For the smoking conditions described above, the nicotine concentration was found to be approximately 0.04 mg/mL of the solvent. This corresponded to approximately 0.06 mg/plate, or 0.6  $\mu$ g/cm<sup>2</sup> of the plate. Therefore, approximately 3.5% of the nicotine and associated TSM that were present in the two cigarettes was deposited on the plate.

## 2.2 Particle detachment characterization

A schematic of the wind tunnel with its instrumentation is shown in Fig. 2. Its details are described in Ibrahim *et al.*<sup>9</sup> Several

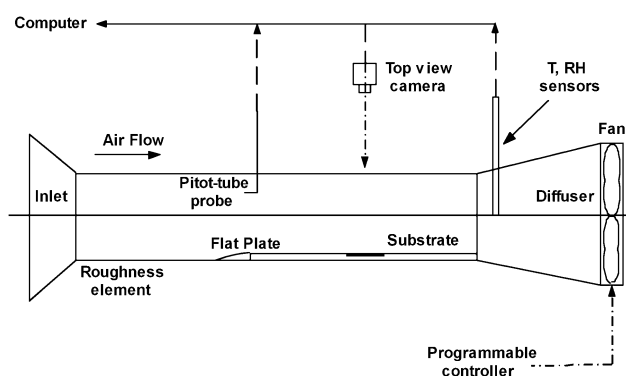


Fig. 2 A schematic of the experimental facility.

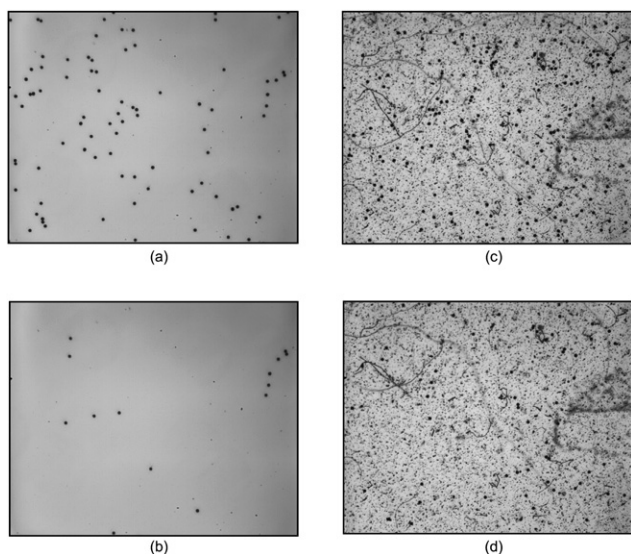
conditions were unique to this particular study. The tunnel's programmable controller was set to attain a *final* free-stream velocity of either 6 m/s or 18 m/s for all experiments. The microparticle-laden test plate was placed 1.4 m downstream from the wind tunnel inlet. This corresponded to Reynolds numbers of 400 000 and 1 200 000, based upon the two free-stream velocity values and the 1.4 m distance. A distributed roughness element was located at the beginning of the tunnel's inlet section to assure a fully developed turbulent boundary layer at the location of particle detachment. Subsidiary measurements of oil-film interferometry of the test-plate's surface and Preston-tube boundary-layer pressure measurements confirmed the developed extent of the turbulent boundary layer. The air temperature and relative humidity were approximately 22 °C  $\pm$  1 °C and 19%  $\pm$  2%, respectively.

For these experiments, the relative humidity of the air was not controlled directly. The results of Ibrahim *et al.*,<sup>8</sup> Corn<sup>2</sup> and Corn and Stein<sup>3</sup> have shown that low values of relative humidity produce almost no change in particle pull-off forces up to a relative humidity of approximately 30%. Above that humidity, the pull-off force increases. This has been well documented by Ibrahim *et al.*<sup>8</sup> Therefore, the results obtained in this work are directly applicable for relative humidity up to approximately 30%, and can be extrapolated using the results of Ibrahim *et al.*<sup>8</sup> to higher relative humidity.

The extent of microparticle detachment was characterized through the detachment fraction versus the free-stream velocity. This fraction is defined as  $n^*(t) = 1 - [n(t)/n(t = 0)]$ , where  $n(t)$  is the number of non-detached microparticles on the surface at time  $t$ . One indicator of detachment progress is the threshold velocity for detachment,  $U_{th, 50\%}$ , which is defined as the free-stream velocity at which  $n^*(t)$  equals 0.5. Another indicator, introduced in this work, is  $U_{th, 5\%}$ , which is defined as the free-stream velocity at which  $n^*(t)$  equals 0.05. The selection of the 5% threshold value is somewhat arbitrary. Another, relatively low value could have been used. One obvious use of  $U_{th, 5\%}$  is to characterize cases where the  $U_{th, 50\%}$  is beyond the maximum free-stream velocity of the experimental facility. The physical basis of each detachment velocity is discussed in section 3.

The motion of the microparticles was recorded using a Basler A501K progressive-scan CMOS monochromatic digital camera equipped with magnifying lenses (Micro-Nikkor 105 mm lens and 4X Nikon teleconverter) to achieve enough optical magnification to resolve individual microparticle motion. The camera had a 1280 pixel  $\times$  1024 pixel resolution and was operated at a rate of 5 frames/s and a shutter speed approximately equal to the inverse of the frame rate.

The camera output was connected to a personal computer for image analysis. Simultaneously, the voltage output of a pitot-static tube/pressure transducer system was digitized and recorded on a computer. Thus, the free-stream velocity and time were known for each image. The number of microparticles removed from the plate by the air flow (and not by collisions from other microparticles, which were very few) was counted manually from the time-sequenced images of a 5.2 mm (parallel to the flow direction) by 7.8 mm (normal to the flow direction) area in the center of the plate. This area was the largest area possible over which individual particles still could be resolved. The initial number of surface-resident microparticles on the 5.2 mm by



**Fig. 3** Images of surface-resident microparticles for clean (a and b) and dusty (c and d) surfaces upon which the microparticles were deposited after TSM exposure. Images a and c are at  $t = 0$  s and  $U_{\infty} = 0$  m/s, having 82 and 137 microparticles, respectively. Images b and d are at  $t = 129$  s and  $U_{\infty} = 18$  m/s, having 15 and 43 microparticles, respectively. The flow direction is from bottom to top in each image.

7.8 mm surface area ranged from 77 to 202 for the 10 runs. Previous experiments on microparticle detachment [Ibrahim<sup>7</sup>] established that an area initially having approximately 30 to 40 microparticles was sufficient to give the same threshold velocity to within  $\pm 6\%$  of those having a larger number of microparticles.

Four example images are shown in Fig. 3 of surface-resident microparticles for clean (left two images) and dusty (right two images) surfaces upon which the microparticles were deposited after TSM exposure. The top two images are the surfaces at  $t = 0$  s and  $U_{\infty} = 0$  m/s, having 82 and 137 initial microparticles, respectively. The bottom two are at  $t = 129$  s and  $U_{\infty} = 18$  m/s, having 15 and 43 remaining microparticles, respectively.

### 3 Results and discussion

The conditions of the experiments and their threshold velocity results are summarized in Table 1. The results of a series of five experiments are presented in Fig. 4, in which microparticles were deposited on clean glass substrates. Run No. 1 (open square)

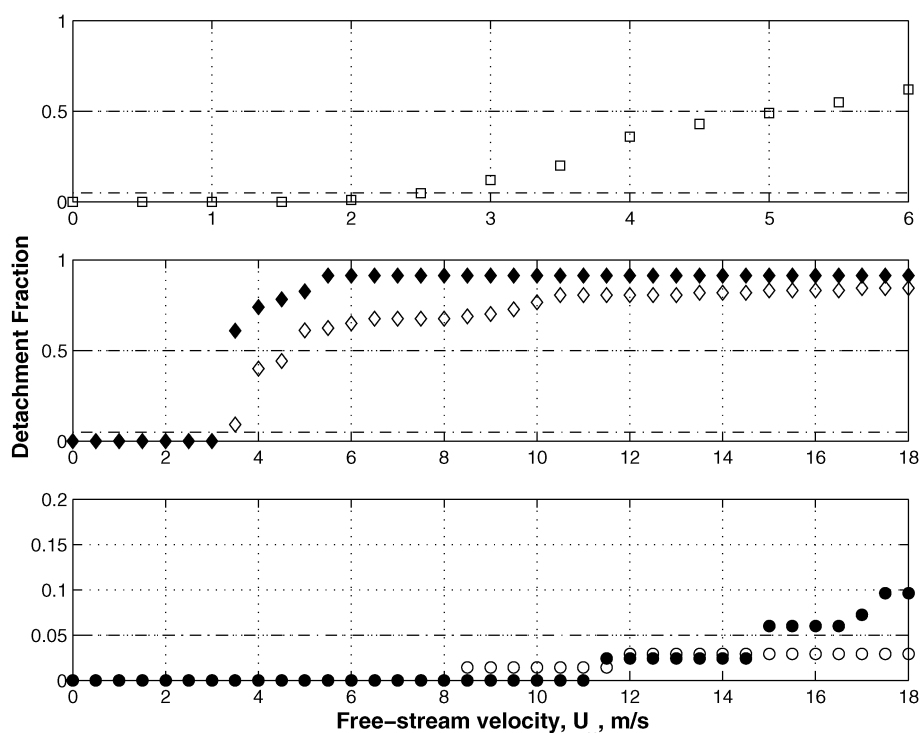
represents the progress of the microparticle detachment without any TSM exposure. Run Nos. 2a and 2b (solid diamond and open diamond) denote the progress of microparticle detachment with TSM applied to the surface *before* microparticle deposition. Run Nos. 2a and 2b each were repeated once under the same conditions. Run Nos. 3a and 3b (solid circle and open circle) reveal the progress of microparticle detachment with TSM applied *after* microparticle deposition. Run Nos. 3a and 3b each were repeated once under the same conditions.

The  $U_{th, 50\%}$  for the base case (Run No. 1) was 5.0 m/s. For the two cases (Run Nos. 2a and 2b),  $U_{th, 50\%}$  changed on the average by approximately 1.2 m/s. The uncertainty in  $U_{th, 50\%}$  (estimated at 95% confidence) was approximately 25%. Most of this uncertainty arises from the inherent sensitivity of particle detachment to the 'local' effects, such as, for example, the roughness of the individual particle/surface contact.<sup>9</sup> These effects manifest themselves in the variability observed between experiments that are repeated under the same operating conditions, such as for Run Nos. 2a and 2b. The extent of this variation is comparable to that found in previous, carefully controlled, repeated-trial, TSM-free experiments.<sup>9</sup> Such similar variations suggest that the detachment characteristics of the TSCs deposited on surfaces *after* TSM exposure are relatively similar to their counterparts that were not subjected to any TSM exposure. This implies that existing data base on microparticle detachment from clean surfaces could be used as an approximate indicator of TSC detachment from similar surfaces when TSC deposition has occurred before TSM exposure. This also suggests that the amount of TSM used in these experiments, as assessed solely by changes in  $U_{th, 50\%}$ , did not affect the primary factors that govern the detachment process. These factors and the extent of their effects on detachment were presented by Ibrahim *et al.*<sup>8</sup>

A much larger change in  $U_{th, 50\%}$  occurred when TSM was applied *after* microparticle deposition. The  $U_{th, 50\%}$  in both cases (Run Nos. 3a and 3b) was beyond the maximum free-stream velocity of the wind tunnel, which was 18 m/s. This indicates a significant increase in the force required to pull a microparticle off of the surface. Most likely, the condensation and deposition of TSM onto the microparticle and its interface with the surface effectively increases the surface area of contact between the microparticle and the surface. A similar effect was observed by the authors (see Fig. 5,<sup>8</sup>) when microparticles remained on a surface for long periods of time at high relative humidity before detachment. In those experiments, the  $U_{th, 50\%}$  increased to beyond the maximum free-stream velocity of the same wind tunnel. However, it should be recognized that other

**Table 1** Summary of experimental conditions and threshold velocity results

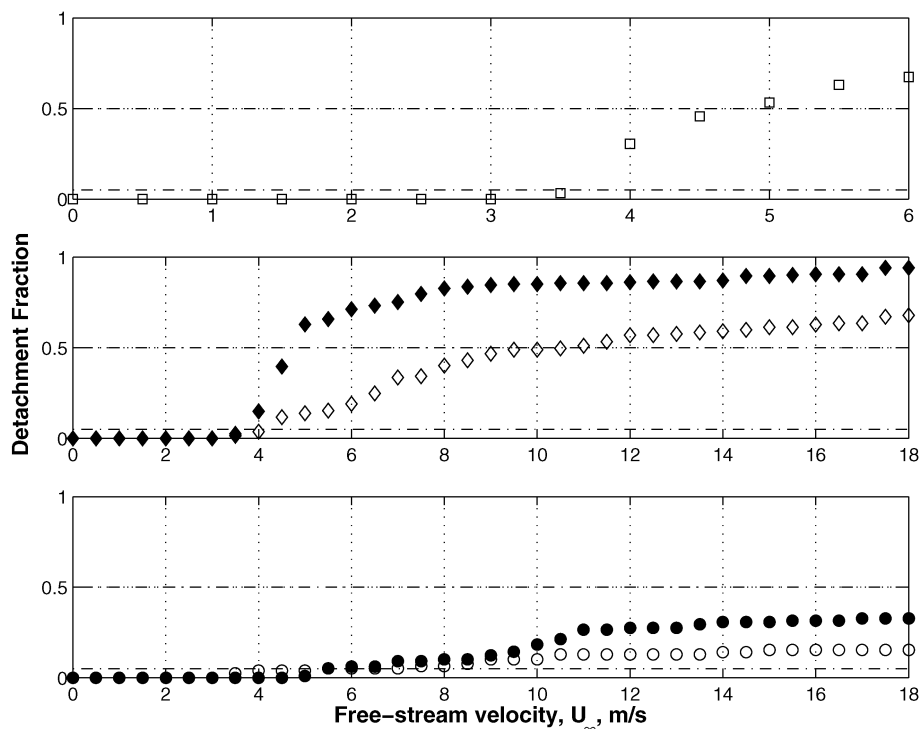
Run No.	Figure No. & Symbol	TSM exposure	Plate	$U_{th, 50\%}$ , m/s	$U_{th, 5\%}$ , m/s
1	4, ■	none	clean	5.0	2.5
2a	4, ◆	before particle deposition	clean	3.2	3.1
2b	4, ◇	before particle deposition	clean	4.4	3.5
3a	4, ●	after particle deposition	clean	> 18	15
3b	4, ○	after particle deposition	clean	≫ 18	> 18
4	5, ■	none	dusty	4.7	3.6
5a	5, ◆	before particle deposition	dusty	4.7	3.7
5b	5, ◇	before particle deposition	dusty	10	4.1
6a	5, ●	after particle deposition	dusty	> 18	5.5
6b	5, ○	after particle deposition	dusty	≫ 18	6.5



**Fig. 4** The progress of microparticle detachment from a **clean** glass plate. The square, diamond and circles are for TSM exposure made without, before and after microparticle deposition, respectively. The full and empty symbols denote two experiments conducted under the same conditions.

circumstances, such as repetitive vibrations that can occur in HVAC ducts, may break this strong adhesion bond and cause detachment at lower free-stream velocities.

The results of another series of five experiments are shown in Fig. 5. In this series, similar microparticles and surfaces were used, except that the plates were exposed passively to common



**Fig. 5** The progress of microparticle detachment from a **dusty** glass plate. The square, diamond and circles are for TSM exposure made without, before and after microparticle deposition, respectively. The full and empty symbols denote two experiments conducted under the same conditions.

indoor dust for a period of two months before the experiments. To maximize the consistency of this exposure, the plates were placed next to each other for the same period of time. In general, the same qualitative trends were observed here as in the first series. However, there were some noticeable differences.  $U_{th, 50\%}$  was the same for the TSM-free case (Run No. 4) and one TSM exposure case (Run No. 5a). However,  $U_{th, 50\%}$  for the repeated case (Run No. 5b) was more than twice the base-case value. This relatively large variability in  $U_{th, 50\%}$  (approximately 5.3 m/s) between similar cases is most likely the cause of dust on the surface.

Comparing the results in Figs. 4 and 5 helps to further elucidate the effect of dust with TSM on the plate. For the TSM-free case,  $U_{th, 50\%}$  differed by approximately 0.3 m/s (Run No. 1 compared to Run No. 4). With dust and TSM both present on the surface, a relatively large increase in  $U_{th, 50\%}$  occurs (from an average of approximately 3.8 m/s for Run Nos. 2a and 2b compared to an average of approximately 7.4 m/s for Run Nos. 5a and 5b). The ubiquitous presence of dust on the surface, as shown in Figs. 1 and 3 (images c and d), will reduce the average *local* flow velocity that a microparticle experiences as compared to its isolated counterpart. This results in reduced aerodynamic removing forces and moments and, hence, a higher velocity for detachment. However, it should be noted that when the number of microparticles per unit surface area (or dust of comparable size) increases beyond a certain limit, more collisions between the microparticles can occur and  $U_{th, 50\%}$  would decrease.<sup>7</sup>

The non-uniform spatial distribution of dust with TSM on the surface leads to relatively large variations in the strength of microparticle/surface contacts. This produces variations in the aerodynamic and resisting forces/moments of the microparticles. Typically in previous studies,  $U_{th, 50\%}$  has been used to characterize the detachment process. This velocity essentially describes the balance between the *average* aerodynamic forces/moments and the *average* resisting forces/moments. The present work suggests the utility of an additional threshold velocity, namely  $U_{th, 5\%}$ . This velocity indicates when microparticles start to detach, and is a measure of the balance between the *strongest* aerodynamic forces/moments and the *weakest* resisting forces/moments.  $U_{th, 5\%}$  is very useful in situations when there is a large spatial scatter in the forces and moments and when collisions between microparticles is a dominant detachment mechanism. The  $U_{th, 5\%}$  values for both series of experiments is also listed in Table 1.

Further inspection of the values in Table 1 shows that experiments made under similar conditions with TSM exposure before particle deposition have relatively small variability in the values of  $U_{th, 5\%}$  as compared to a larger variability in the values of  $U_{th, 50\%}$ . Run Nos. 2a and 2b, for example, show a 6% average variation in  $U_{th, 5\%}$  and a 16% average variation in  $U_{th, 50\%}$ . Likewise, Run Nos. 5a and 5b show a 5% average variation in  $U_{th, 5\%}$  and a 36% average variation in  $U_{th, 50\%}$ . Similar comparisons between experiments made under similar conditions with TSM exposure after particle deposition can not be made because most  $U_{th, 50\%}$  values exceeded the tunnel's maximum velocity. The small variability in  $U_{th, 5\%}$  when dust is present on the surface probably results from the fact that the strongest removing mechanisms (caused by the air flow) and the weakest resisting forces/moments are relatively similar for both runs. In contrast, the larger variability in  $U_{th, 50\%}$  probably is due to the

large spatial scatter in the removing and resisting forces/moments.

## 4 Summary and conclusions

Experiments were conducted on microparticle detachment caused by turbulent air flow from clean and dusty surfaces where TSM was applied to the surfaces either before or after microparticle deposition. The amount of TSM applied was characterized by its nicotine content, which was approximately 0.6  $\mu\text{g}/\text{cm}^2$  per plate or 3.5% of the nicotine present in two cigarettes. The  $U_{th, 5\%}$  and  $U_{th, 50\%}$  threshold velocities were used to quantify detachment progress.  $U_{th, 5\%}$  was a more sensitive indicator of detachment. The variability in  $U_{th, 5\%}$  was always less than that in  $U_{th, 50\%}$ .

When TSM exposure was made *after* microparticle deposition, there was a considerable increase in both  $U_{th, 5\%}$  and  $U_{th, 50\%}$  with respect to their TSM-free and TSM-exposure-*before*-deposition cases. The extent of TSM exposure *before* microparticle deposition suggests that its effects are minimal and within typical uncertainties for the threshold velocities observed in TSM-free cases. This implied that the existing data base on microparticle detachment from clean and dusty surfaces could be used as an approximate indicator of TSC-detachment situations when TSM exposure has occurred *before* TSC deposition. The presence of dust on the surface with TSM also delayed detachment and increased the variability in the detachment velocities as compared to the TSM-free case.

## Acknowledgements

The authors wish to acknowledge Professor Marya Lieberman, Department of Chemistry and Biochemistry, University of Notre Dame, for many fruitful discussions about quantifying the amount of deposited TSM, and the Kentucky Tobacco Research and Development Center for donating the K2R4F reference cigarettes. Research described in this article was supported by Philip Morris USA Inc. and Philip Morris International.

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