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In Situ Visualization of Particle Motions during Wipe Sampling of Explosives and Other Trace Particulate Materials

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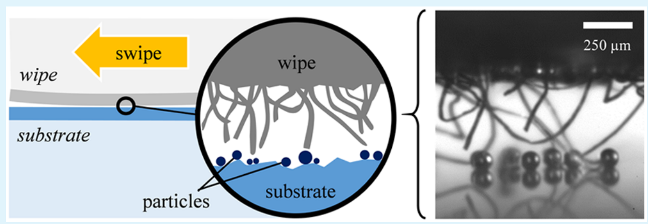
Supporting Information



ABSTRACT: Surface texture tailoring has the potential to increase the effectiveness of dry particle collection wipes, as a wipe's topographical features control the intimate surface contact made with particles on the substrate (critical for van der Waals-governed adhesion). However, texture-tailoring approaches have not yet been widely explored, in part because of a lack of understanding of the specific wipe topographies and wipe/particle interactions that maximize particle collection. Here we describe an in situ optical microscopy technique that enables direct observation of micrometer-scale particle−wipe interactions occurring at the wipe−substrate interface during contact sampling. The technique is demonstrated for nonwoven meta-aramid (Nomex) collection wipes with particles ranging from 1 to 90 μm in diameter and substrates of different topographies (glass and nylon coil zipper). Experiments with hemispherically coated Janus particles allow rolling motions to be distinguished from sliding motions, providing detailed information about how particles move prior to capture or release by the wipe. Particle−fiber and particle−particle interactions are seen to play important roles in particle capture, suggesting that conventional sphere-on-plane models are inadequate to describe adhesion behavior in these systems. Micrographs show how loose, flexible fibers in roughened textile wipes interrogate the valleys of uneven substrate topographies, allowing capture of particles that might otherwise be trapped within the substrate's grooves and depressions. The materials used in this work are specifically relevant to explosives detection, but the in situ visualization technique is transferable for the study of any application involving dry particle collection, such as toxic substance sampling and dust removal.

KEYWORDS: *dry particle adhesion, particle motion, particles at interfaces, trace detection, swipe sampling, particle collection, in situ optical microscopy, surface texture*

INTRODUCTION

Effective wipe- or swab-based processes for the removal of particulate matter from surfaces are important for applications such as trace contraband detection,[1−3] toxic substance sampling,[4−9] and dust removal and cleaning.[10−13] In many applications, ineffective particle collection can have serious repercussions, including national security risks (failure to detect explosives at security checkpoints) and public health emergencies (failure to detect chemical warfare agents or toxic metals). Despite these consequences, wipes used in trace detection typically collect only a small fraction of particles present on a given surface. Low particle collection efficiencies (PCE) may not present a problem if the concentration of target particles on persons or objects of interest tends to be high, as collection of at least a few particles on the wipe is likely. However, if the quantity of targeted particles on a given surface is small (such as trace residues deposited by fingerprints after handling hazardous materials), a low PCE may result in a false-negative detection result. Explosive materials, for example, sublimate relatively rapidly in air (on a time scale of a few minutes to a few days[14]), and consequently, the volume of material available for sampling may be quite small. The need for accurate detection of sparse residues has stimulated recent interest in the development of wipe materials and techniques that increase PCE.

In some sampling applications, such as heavy metal and biological sampling, it is standard practice to moisten wipes with water or another wetting solvent prior to wiping. This practice tends to increase PCE[5,15,16] but is not possible in all applications. In trace explosives and narcotics applications, for example, dry wipes are required to avoid chemical interference with ion mobility spectrometry (IMS) detectors.[17] Beryllium sampling is also performed with dry wipes in some situations to avoid damage to delicate surfaces.[18] Dry particle sampling presents a particular challenge for trace residue collection, as PCE values for dry wiping techniques are not only low but also quite variable, as illustrated by the data aggregation of Figure 1. Collection efficiencies of 1% or less are reported frequently, and large relative standard deviations (20% RSD or higher) are typical even in identical replicates of well-controlled studies.

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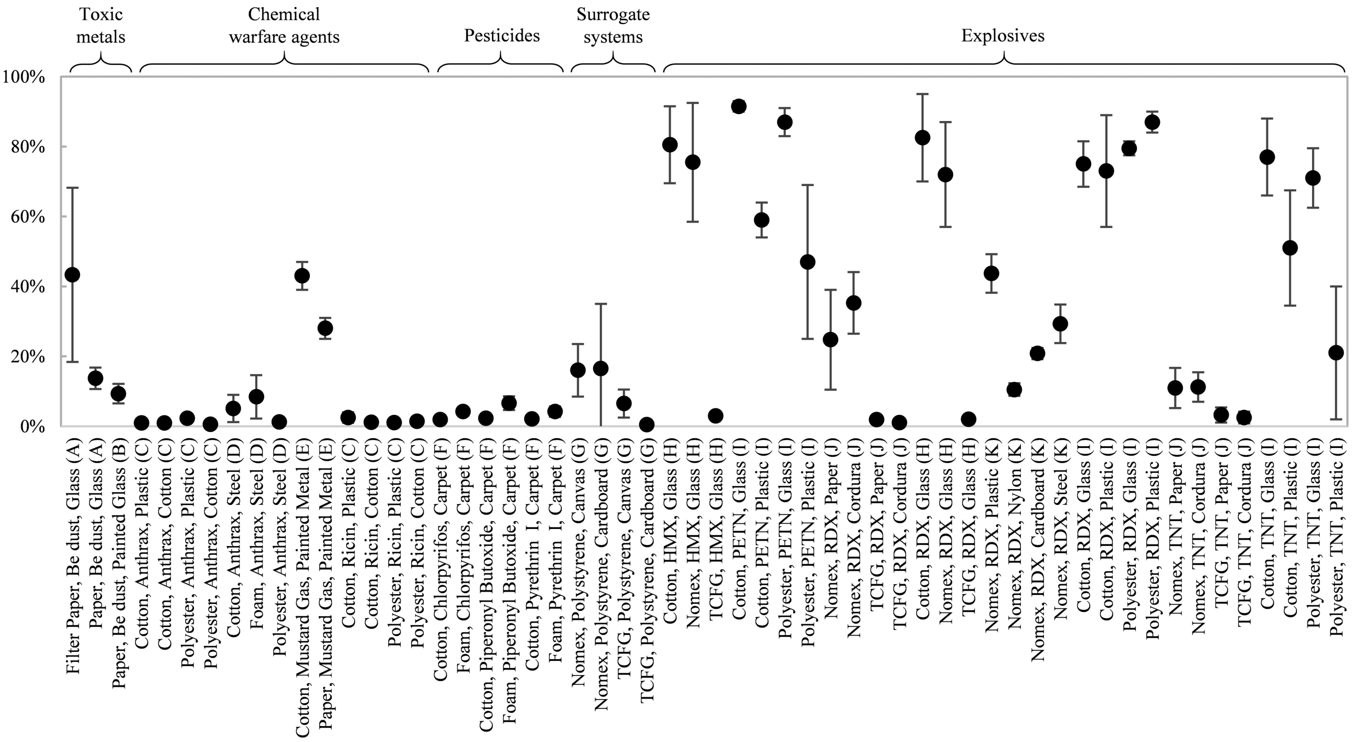

Figure 1. Dry particle collection efficiencies reported in the literature for dry wiping processes in various trace detection applications. Error bars represent one standard deviation of the mean. Acronyms and abbreviations: beryllium (Be); Teflon-coated fiberglass (TCFG); octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine explosive (HMX); pentaerythritol tetranitrate explosive (PETN); 1,3,5-trinitroperhydro-1,3,5-triazine explosive (RDX); and 2,4,6-trinitrotoluene explosive (TNT). Reference key: (A) Dufay 2006;[16] (B) Kerr 2004;[30] (C) Frawley 2008;[6] (D) Rose 2004;[5] (E) Opstad 1999;[31] (F) Fortune 1998;[32] (G) Staymates 2016;[33] (H) Fisher 2017;[2] (I) Song-im 2012;[15] (J) DeGreeff 2019;[34] (K) Robinson 2018.[3] Aggregated data set used to generate this figure is provided in the Supporting Information (Table S1).

This variability raises concerns about the reliability of common wiping materials and techniques and suggests that subtle (and often stochastic) differences in wiping technique, particle distribution, and interfacial configuration play important roles in particle adhesion and capture.

Multiple authors have used atomic force microscopy (AFM) to measure the adhesion of micrometer-scale explosive particles to a range of surfaces.[19−23,2] These studies have shown that adhesive forces generally decrease with surface roughness, consistent with van der Waals models that incorporate effects of surface texture.[24−28,20] The degree of interpenetration between peaks on one rough surface and valleys on the other also contributes to the magnitude of van der Waals forces.[20] Since van der Waals forces decay with separation distance (and are expected to govern particle adhesion only at separation distances of less than ∼15 nm[29]), variation in particle shape and orientation with respect to surface topography helps to explain the broad distributions in experimental measurements of adhesion forces.[20] Adhesive forces also depend upon the material properties of the two interacting media, though these effects tend to be small in comparison to textural factors, and have been observed mainly in studies using carefully prepared smooth surfaces with root-mean-squared roughness no more than a few hundred nanometers.[21,22]

In sampling applications, it is important to note that the goal is to maximize PCE for the entire collection of particles present on a surface, rather than the strength of interaction between the wipe and any individual particle. The relative strength of particle−wipe and particle−substrate adhesive forces is relevant only for those particles that are in close contact with both surfaces. In realistic scenarios involving rough surfaces and nonuniform particles, the collection wipe will be in contact with certain particles and not at all with others during swiping. In this case, statistical knowledge of particle locations and their accessibility to the wipe is important, since a particle that is not in contact with any part of the collection wipe will not be picked up, no matter how strong the hypothetical attractive force.

Chaffee-Cipich et al.[35] published an interesting study addressing this challenge in 2016. The authors developed a contact simulation approach to evaluate interfacial contact between rough, deformable collection wipes and typical test surfaces. They adopted the Greenwood and Williamson microcontact model to estimate contact area resulting from surface asperities and assumed Timoshenko clamped/clamped beam bending behavior to predict deformation of the wipe in areas spanning consecutive asperities. Simulation results were then analyzed to determine the number of void locations where a particle (10−60 μm in diameter) could remain undetected by the wipe due to lack of contact. Results showed that the likelihood of particle detectability increases with particle size (as smaller particles can be concealed in smaller voids) and decreases with surface roughness (as rougher surfaces provide deeper and/or more frequently occurring voids where particles can be concealed). Chaffee-Cipich and co-workers concluded by ranking wipe materials in order of effectiveness, as determined by the fraction of particles with which the wipe would be expected to make contact. The authors rated Nomex meta-aramid wipes as most effective in interrogating surfaces due to their smooth topography, and

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muslin cloth wipes as least effective due to their comparative roughness.

Considering that experimental AFM studies and van der Waals adhesion models both indicate that attractive forces between particles and surfaces are maximized in the case of smooth surfaces, one might expect that smooth wipes would yield the highest PCE values. Chaffee-Cipich's conclusion that smooth collection wipes are most effective at surface interrogation seems to reinforce that idea. However, the opposite outcome has been reported in multiple experimental studies: rougher wipes tend to exhibit significantly *higher* PCEs than smoother wipes.[2,34,33,36] Indeed, in several studies muslin cloth (judged as least effective by Chaffee-Cipich et al.) has been reported as the most effective wipe material in practice.[2,37] These seemingly inconsistent findings suggest that certain important wipe properties may not be captured in either the Chaffee-Cipich contact model or the van der Waals adhesion models. For example, the Chaffee-Cipich model assumes that material properties of the wipe are uniform through the thickness, including the small roughness features at the surface; their simulation results indicate that these features deform relatively little as the wipe is compressed. In reality, loose textile fibers and other features contributing to textile wipe roughness may be quite flexible and significantly more compliant than the wipe itself. Soft, pliable surface features that deform readily around asperities as the wipe passes over the test surface would likely *increase* the level of surface interrogation rather than decrease it. Loose fibers extending from the dense wipe surface provide a soft and porous layer with a high surface area and interstitial spaces available for particle binding and entrapment, possibly increasing surface contact, particle holding capacity, or both. Laster et al.[38] recently demonstrated a similar effect in polymer polypyrrole collection wipes, finding that the removal of particles from rough surfaces could be enhanced by patterning the wipe surface with an array of microscale pillars.

Given that flexible fibers and other soft surface features in a collection wipe appear to play a key role in particle collection, the goal of the present work was to build a technique for studying the mechanisms by which these features deform and interact with surfaces and particles during swipe sampling. In this paper, we describe the development and use of a controlled swiping system with silhouette microscopy that provides a direct, cross-sectional view of the wipe/substrate interface during real-time swipe sampling. This setup enables in situ visualization of contact between surfaces and motions of particles and textile wipe fibers leading up to capture. Information about wipe−particle−substrate interactions and their influence on particle capture afforded by this new technique is expected to inform strategic modifications of collection wipes that maximize particle collection efficiency, e.g., by tailoring wipe topography to increase the frequency of wipe/particle interactions that lead to particle pickup. In demonstrating the technique, we present a number of interesting observations and findings regarding particle behavior for wipe sampling of explosives-relevant material systems.

■ MATERIALS AND METHODS

Mechanical Swiping and Microscopy System. A custom mechanical swiping system, shown schematically in Figure 2, was built to provide precise control of vertical and horizontal wipe motions while simultaneously providing a direct cross-sectional view of the interface during swiping (Figure 3). Motion control was achieved with a pair of precision translation stages (Thorlabs, PT1). Each stage provides 1.00 in. (25.4 mm) of total travel through a manually operated micrometer drive. The stages were attached at right angles to one another, with the horizontal stage suspended in a gantry configuration using an L-shaped bracket (Thorlabs, PT102). A self-built adapter plate, secured between the bracket and the vertical stage, allows for a full range of motion of the horizontal stage. The entire translation stage assembly was affixed to a large angle bracket (Thorlabs, AP90RL) which can be secured directly to an optical table or alternatively to the stage of an inverted microscope. The inverted microscope setup provides a secondary (bottom-up) view of the interface during swiping when used with a transparent substrate.

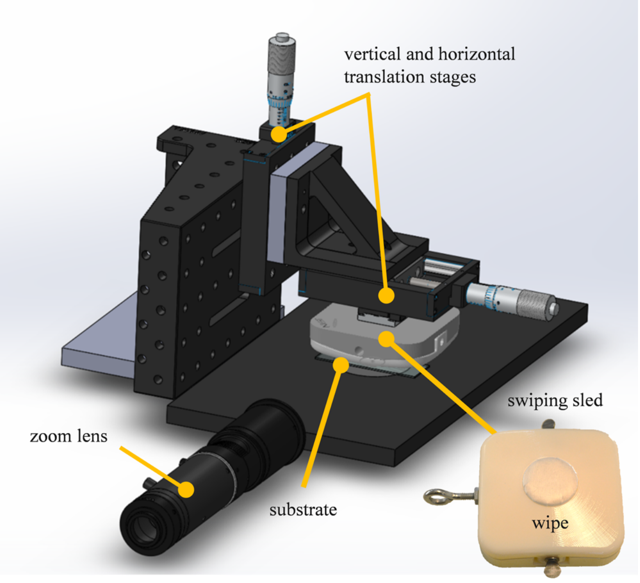

Figure 2. Mechanical swiping and side-view imaging system.

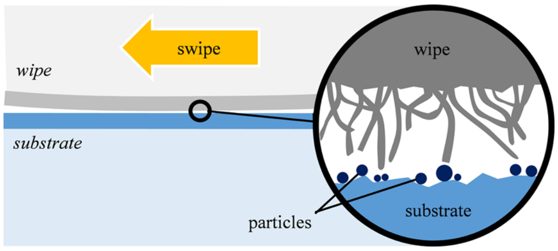

Figure 3. Schematic representation of a wipe−particle−substrate interface as visualized by the in situ microscopy system.

The swiping sled was provided by researchers at the National Institute of Standards and Technology (NIST) and is described in detail (including 3D printing files for reproduction) elsewhere.[1] This swiping sled is regarded as a test standard in the trace explosives sampling community and has been used in multiple research studies, including a recent Department of Homeland Security (DHS)-commissioned round-robin test.[39] The sled secures a collection wipe in a circular, slightly convex mount backed with felt. In our apparatus, the sled was attached to the horizontal translation stage using a dovetail clamp and locking screw (Thorlabs, X34D2-30 and XT34HP), allowing for motion control as well as easy removal of the sled for wipe mounting and setup.

Side-view microscopy was performed using a horizontally mounted zoom lens (Navitar Inc., 12X Zoom) with a 165 mm working distance. The zoom lens was equipped with a machine vision adapter (Navitar Inc., models 1-6233 and 1-6010) and Gigabit Ethernet (GigE) camera (Prosilica, GC750). Video capture was achieved in the

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LabVIEW (National Instruments) software environment using a timed collection to record images at a rate of 6 frames per second. The zoom lens has available magnification settings ranging from 0.58× to 12.0×, and the range of settings used in this work was 2.0× to 7.0×, depending on the size of particles and the detail needed. The system can be set up in any of three illumination configurations (front lit, back lit, or dual lit) depending on experimental needs. Front lighting was achieved using a compact white light-emitting diode (LED) flashlight (McMaster-Carr, 6545T5) mounted to a support post on the same side of the stage as the zoom lens. Back lighting was achieved using a blue LED light (First Ten Angstroms, B0001) aligned with the optical axis of the zoom lens on the opposite side of the object plane.

The back-lit configuration provides a "silhouette" image of the interface, which is convenient for image processing since silhouette images can be readily converted to binary for analysis. The front-lit configuration provides additional information about spatial relationships since shadowing and object order (foreground to background) is apparent. However, in this setup we encountered difficulties with sufficiently illuminating fibers in the field of interest without washing out foreground objects. Finally, the dual-lit configuration (involving both light sources) combined many of the benefits of both illumination setups, providing a good view of objects at the interface while also allowing for observation of shadowing and spatial detail. A comparison of the three illumination configurations is given in Figure 4, which shows 50 μm diameter Janus particles positioned at the interface between a Nomex wipe and a glass substrate. Only the dual-lit configuration provided a clear view of the orientation of the black/white half shell coating on the Janus particles. Meanwhile, the back-lit silhouette images provided the highest contrast. This configuration facilitates particle identification and tracking across the image plane, particularly for smaller particles and for use with automated image-processing routines. The back-lit and dual-lit illumination configurations were used exclusively in the remainder of this study, as the front-lit configuration did not appear to provide independent benefits in this work.

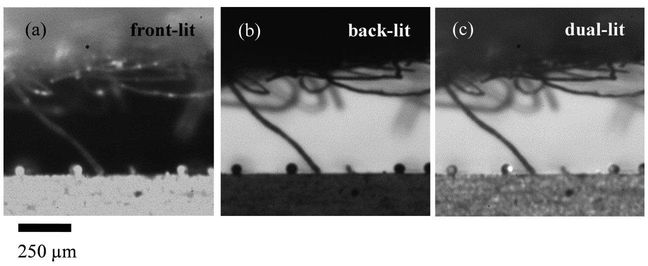

Figure 4. Comparison of three different illumination configurations: (a) front lit; (b) back lit; and (c) dual lit. Micrographs show a Nomex wipe interacting with 50 μm diameter Janus particles on a glass substrate.

Collection Wipes, Particles, and Substrates. Materials used in this work are relevant to trace explosives detection applications. All experiments were performed using DHS-approved Nomex collection wipes (DSA Detection, DSW8066). Nomex is a nonwoven meta-aramid textile, and virgin wipes have a texture similar to paper. Roughening of the wipe surface through repeated use liberates long, flexible fibers that can extend up to several millimeters from the wipe surface and are often visible to the naked eye. Wipes were either used as received or roughened by swiping repeatedly (100 times) across a Cordura (canvas-like) surface to simulate reuse, using the procedure described by DeGreeff et al.[34] The roughness average (R_A) for this material was found to be 9 ± 12 μm for the virgin wipes and 156 ± 29 μm for roughened wipes, including the contribution of the flexible surface fibers. A detailed analysis of surface roughening effects in this material is described elsewhere.[34] The wipe material used in this study is hydrophilic and fully wets on contact, consistent with findings of other authors for aramid fabrics,[40,41] indicating a high surface energy.

Particles used in experiments included spherical polystyrene latex (PSL) particles of diameters 1 and 90 μm (Polysciences, 16905-1 Polybead Sampler Kit III) and spherical polyethylene Janus particles with a manufacturer-specified diameter range of 45−53 μm (Cospheric, HCMS-BLK-WHT 45−53 μm), referred to hereafter as 50 μm diameter. The Janus particles comprised a black polyethylene core hemispherically coated on one-half-shell (30−50% areal coverage) with a white polymeric coating. This aided in discrimination between rolling and sliding particle motions. Polymer microspheres are common surrogates for explosive residues in studies of particle collection,[33,37,42,38] and the 1−90 μm size range used in this work was chosen for consistency with explosive particles found in typical fingerprint residues.[43] The surface free energies for the polystyrene and polyethylene materials are estimated at 43 and 33 mN/m, respectively.[44,45]

Substrates included precleaned glass microscope slides (Corning, 2948 microscope slides) and a nylon coil zipper (YKK Fastening, #3 zipper). These materials are representative of test surfaces encountered in trace screening settings, such as airport security checkpoints.

Optical micrographs of the 90 μm diameter PSL particles, 50 μm diameter Janus particles, nylon coil zipper, and Nomex wipe are given in Figure 5 to illustrate the morphology and texture of the materials used in this work. The 1 μm diameter PSL particles had a powder-like appearance under optical microscopy and are not pictured.

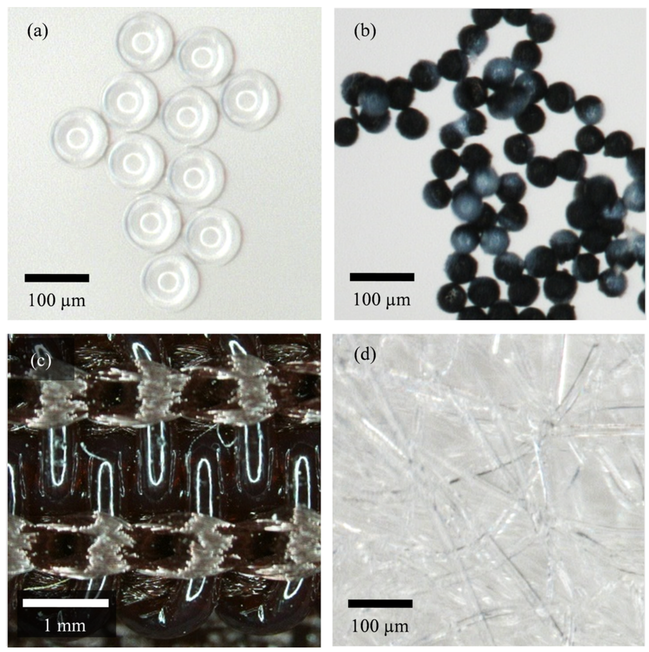

Figure 5. Micrographs of (a) 90 μm diameter PSL particles, (b) 50 μm diameter Janus particles, (c) a nylon coil zipper; and (d) a Nomex wipe.

RESULTS AND DISCUSSION

Role of Flexible Surface Fibers. In situ imaging results immediately supported the hypothesis that roughening of collection wipes can enhance surface contact. Comparisons of wipe/substrate interfaces for smooth and roughened Nomex wipes demonstrate that loose fibers can provide surface contact even when primary surfaces are significantly separated. As an example, Figure 6 shows the interface between a Nomex wipe and a nylon coil zipper separated by a distance of approximately 300 μm. In the case of the smooth virgin wipe (Figure 6a), no contact with the zipper substrate is made; conversely, the roughened wipe (Figure 6b) made contact with

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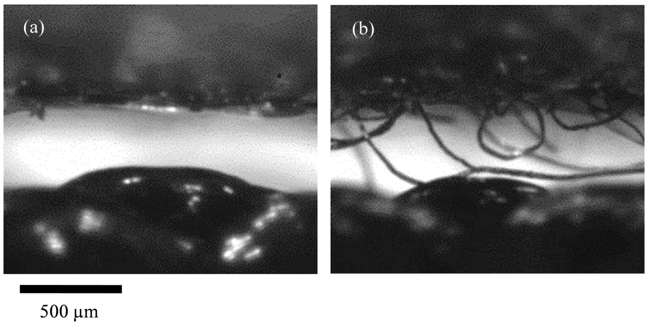

Figure 6. Comparison of contact between a Nomex wipe and a nylon zipper substrate (separated by ∼300 μm) for (a) an unused wipe and (b) a roughened wipe. While no contact was observed in the unused wipe case, loose fibers on the roughened wipe resulted in wipe/substrate contact in several locations, despite the separation between surfaces.

the substrate in several locations via its loose fibers. This is notable because in an interface between a wipe and textured surface we would not expect the wipe to be flush with the substrate across the entire sampling area during swiping. Separations would occur frequently across the contact surface as a result of nonuniformities in swiping pressure and substrate surface profile. Further, particles may preferentially accumulate in depressions on the substrate surface, where material is less likely to be shed through incidental contact and cleaning. This is illustrated by the example of Figure 7, which shows 90 μm diameter PSL particles embedded into topographical valleys in a nylon zipper. The compliant nature of loose textile fibers (not accounted for in roughness-based contact models) explains why loose surface fibers could penetrate into depressions without hindering the wipe's ability to make flush surface contact across the global surfaces.

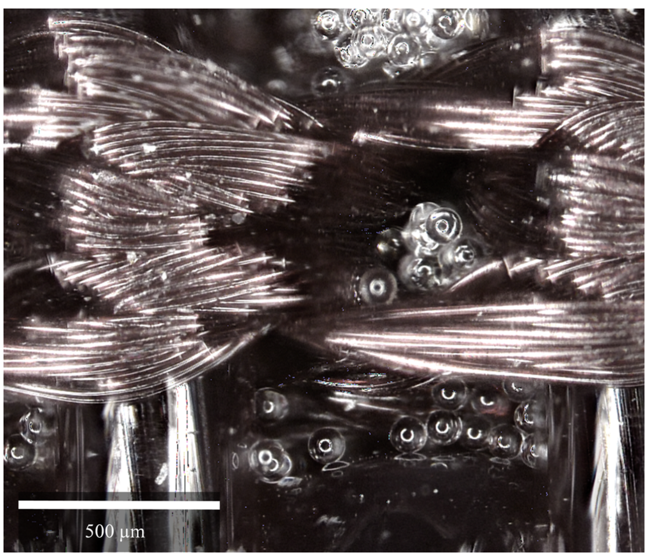

Figure 7. Nylon zipper substrate with numerous 90 μm diameter PSL particles embedded into depressions in its topology.

Particle Sliding, Rolling, Lifting, and Electrostatic Motions. By visualizing interfaces in situ during swipe sampling we are able to observe the motions of individual particles as they interact with wipe fibers. This is useful for understanding mechanisms of particle detachment and removal. One question of interest was whether swiping-induced particle motions tend to involve rolling, sliding, lifting, electrostatic effects, or a combination of these mechanisms. In particle removal studies involving hydrodynamic shear flows, rolling is generally agreed to be the predominant mechanism of particle detachment from smooth surfaces,[46−50] in which case a torque balance governs particle release. Conversely, in experiments involving perpendicular removal forces, the predominant removal mechanism is lifting,[51,52] and particle release is governed by the ratio of externally applied lifting forces to the adhesive forces. Sliding is less common than the other mechanisms because the force required for sliding onset is generally higher than that required for rotation[51,53] but can occur preferentially in certain situations, such as in systems involving flat-sided particles.[54,55]

No specific data could be found in the literature comparing release mechanisms in dry wipe-based particle removal. Verkouteren et al.[37] noted that while particle lifting is necessary for particle capture by the wipe, sliding and/or rolling could also play important roles, as initial particle detachment should significantly lower the force required for liftoff. It is not immediately obvious which mechanisms or combination of mechanisms should contribute most strongly, given that swiping motions involve the time-dependent application of multidirectional (normal and tangential) forces to the substrate and particles.

Janus particles were found to be particularly useful for studying the nature of particle motions and removal mechanisms in this work as the hemispherical coating enabled distinction between rolling and sliding. We found that both types of motion occurred in our samples, as illustrated by Figure 8, which shows a 50 μm diameter Janus particle undergoing rolling and sliding during manipulation by an individual Nomex fiber. A video showing additional examples of rolling and sliding motions is available as Supporting Information for this article. Rolling motions were observed more frequently, consistent with the theoretically lower onset force for this mechanism, and which may also be related to the exclusive use of spherical particles in this work. Meanwhile, sliding motions could be explained by adhesive van der Waals forces between wipe fibers and particles that overcome the forces favoring either rolling or reattachment to the substrate.

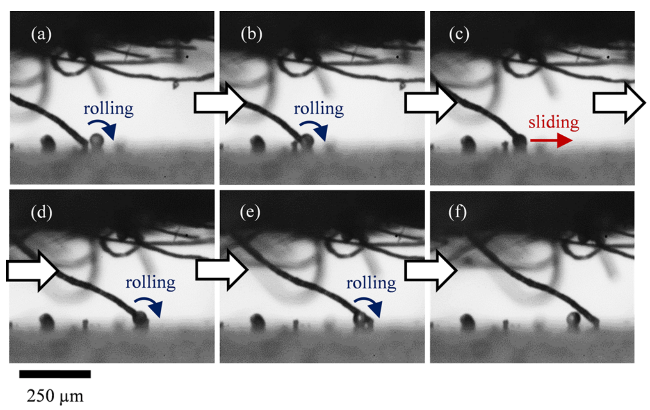

Figure 8. Sequence of dual-lit images, showing particle rolling motions occurring between frames a−b, b−c, d−e, and e−f, and sliding motions occurring between frames c−d, resulting from interactions between an individual Nomex wipe fiber and a 50 μm diameter polyethylene Janus particle on a glass substrate.

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The observation of sliding without ultimate particle removal from the surface may therefore suggest that the particle initially adhered to the wipe fiber but that the adhesive forces were disrupted prior to particle liftoff.

Electrostatic effects are known to contribute to micrometer-scale dry particle adhesion and transfer between surfaces.[56−58,37,53] Many commercial dust cloths (such as Swiffer dry wipes) leverage the triboelectric effect to maximize cleaning efficacy.[59−62] Laster et al. recently attempted to use the same effect to enhance trace particle sampling efficiency with some success.[38] In situ microscopy allows for direct observation of these electrostatic particle motions. An example is presented in Figure 9, which shows a pair of 50 μm diameter

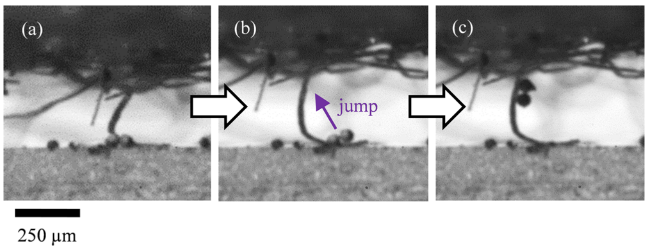

Figure 9. Sequence of dual-lit images, ordered sequentially as a−c, showing 50 μm diameter polyethylene Janus particles triboelectrically "jumping" across a 300 μm gap between a glass substrate and a Nomex wipe fiber.

Janus particles "jumping" across a 300 μm gap between a glass substrate and Nomex wipe. This motion was attributed to electrostatic effects as the particles traveled suddenly across the gap without significant movement of the wipe or substrate.

Particle Size and Substrate Considerations. Particle size (in the 1−90 μm size range explored in this study) was not observed to strongly affect particle physics or adhesion behavior. This finding is consistent with work of other authors, as the particles were either smaller or similar in size to topographical surface features on the wipes and substrates[26] and were small enough that van der Waals interactions would be expected to dominate over gravitational effects (see, e.g., Quesnel et al.[63]). Due to resolution limits inherent in optical systems, the in situ visualization system developed here is best suited for visualizing the motions of particles larger than 10 μm because at this size range particles can be distinguished from one another in the field of view, and rolling and sliding motions can also be distinguished. The technique was also used successfully with particles as small as 1 μm, albeit with reduced visibility of features. Figures 10 and 11 compare image series for Nomex swab fibers interacting with 90 and 1 μm

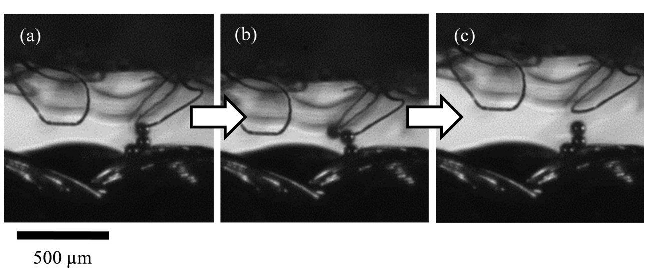

Figure 10. Sequence of dual-lit images, ordered sequentially as (a) through (c), showing a Nomex swab fiber interacting with three 90 μm diameter PSL particles on a nylon coil zipper substrate.

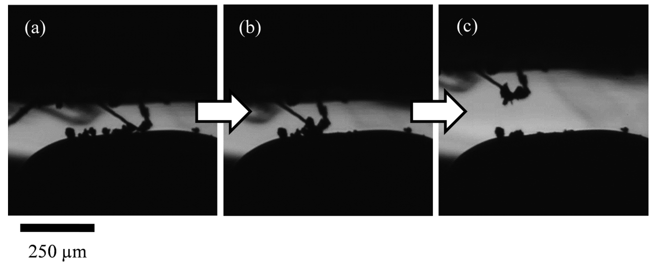

Figure 11. Sequence of back-lit images, ordered sequentially as (a) through (c), showing a Nomex swab fiber gathering and lifting numerous 1 μm diameter PSL particles from a nylon coil zipper substrate.

particles on a nylon coil zipper. In the case of the larger 90 μm microspheres (Figure 10), the particles are clearly distinct from one another, rolling/sliding motions can be clearly observed, and contact points between the particles and fibers, other particles, and the substrate can be identified. Conversely, the smaller 1 μm microspheres (Figure 11) could not be visualized individually, so it was not possible to examine contact points, but it was possible to observe group motions, providing information about overall behavior. At all scales, complex particle−particle and particle−fiber interactions were prevalent. This is significant because these interactions would not be well represented by the classical (or roughness-corrected) sphere-on-plane adhesion models[27,25,64,65] that are conventionally used to describe van der Waals driven particle adhesion at this scale. Results indicate that an accurate model would need to account both for multiparticle interactions and for nonplanar surfaces, such as loose fibers.

Figures 12 and 13 show Nomex wipe fibers interacting with 90 and 1 μm diameter PSL particles, respectively, on a smooth

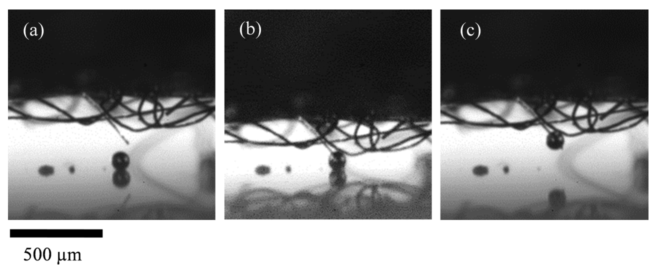

Figure 12. Sequence of dual-lit images, ordered sequentially as a−c, showing a Nomex swab fiber lifting a 90 μm diameter PSL particle from a glass substrate.

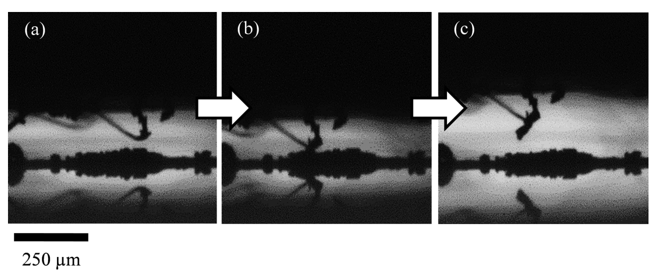

Figure 13. Sequence of back-lit images, ordered sequentially as a−c, showing a Nomex swab fiber lifting numerous 1 μm diameter PSL particles from a glass substrate.

glass surface. It was noted that imaging was somewhat more challenging in the case of the glass substrate, as it was

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sometimes difficult to identify the surface plane due to the material's transparency, and the reflectivity of the glass surface created mirror images across the interface. However, general behavior was found to be similar for both substrates, with particle−particle interactions again playing a significant role. Figure 13 demonstrates that particles need not all be in direct contact with the wipe to be removed from the substrate, as particles can also form chain-like structures with other particles and be lifted as a group.

■ CONCLUSIONS

This work employed a new optical interface visualization technique to observe the motions of micrometer-scale particles in situ during wipe-based surface sampling. Experiments were performed for several combinations of particle size (1, 50, and 90 μm) and substrate material (glass and nylon coil zipper) to demonstrate the capabilities of the technique and highlight interesting findings. Micrograph image series demonstrated the important effects that topographical surface features can play on particle behaviors. For example, loose textile fibers that protrude from the surface of a roughened collection wipe can bridge separation gaps between wipe and substrate to provide intimate contact with particles located in a depression or region without flush wipe/substrate contact. As a result, a roughened wipe may provide better contact with particles on a substrate as compared to a smooth wipe. This finding may at first glance appear to be in conflict with conventional contact models, which predict that surface contact should generally decrease with surface roughness. However, these models do not take into account the compliant nature of loose textile fibers in comparison with the wipe overall. The resulting depth-dependent properties allow soft wipe fibers to penetrate into substrate depressions locally without inhibiting the wipe's ability to make intimate surface contact at a global scale.

Novel experiments with hemispherically coated Janus particles enabled visual distinction between particle rolling and sliding behaviors. Both types of particle motion were observed in this study. Further work is planned to explore each type of motion in more detail, including analysis of a statistically significant population of particles to assess the prevalence of each type of motion as well as any configurational factors contributing to each occurrence. Preliminary work suggests that rolling motions are more common, consistent with a minimization of the theoretical force required to initiate the motion; however, thus far these observations are incidental and require quantification.

Particle−particle interactions were also found to be significant in particle capture phenomena. Chain-like particle interactions, wherein a wipe or wipe fiber would interact with certain particles that would in turn interact with others, resulted in group particle motions involving both wipe−particle and particle−particle adhesive interactions. This could result in particles being lifted from the surface in entangled groups where only a subset of particles is in direct contact with the wipe.

The prevalence of group motions of particles, particle−particle interactions, and the complex effects of topographical surface features observed in this study suggest that particle adhesion in the context of wipe-based sampling would not be well represented by existing sphere-on-plane adhesion models. Further work is needed to characterize wipe-based particle capture mechanics further, including the development of new models that account for particle−particle and particle−fiber interactions. The in situ visualization technique presented here enables particles of interest to be deliberately "activated" by bringing them in contact with a wipe, allowing for the study of dynamic interactions on the scale of individual wipe fibers and particles. Aggregated data can then be quantified via statistical analysis of representative populations of individual particles, including classification of incipient motions and other behaviors associated with particle adhesion and pickup. Beyond trace detection applications, the in situ visualization technique developed here (along with resulting mechanistic analyses and models) could be applied in a wide range of interface science applications where motions and adhesion behaviors of micrometer-scale particles are of interest.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.9b06761.

Aggregation of dry particle collection efficiency data from the literature (PDF)

Examples of Janus particles undergoing rolling and sliding motions (MP4)

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Notes

The authors declare no competing financial interest.

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