Forensic Science International 267 (2016) 16–24

Contents lists available at ScienceDirect

Forensic Science International

journal homepage: www.elsevier.com/locate/forsciint

Nanomechanical mapping of latent fingermarks: A preliminary investigation into the changes in surface interactions and topography over time

Buddhika N. Dorakumbura[a,b], Thomas Becker[a,b,*], Simon W. Lewis[a,b,*]

[a] Nanochemistry Research Institute, Curtin University, GPO Box U1987, Perth, Western Australia 6845, Australia
[b] Department of Chemistry, Curtin University, GPO Box U1987, Perth, Western Australia 6845, Australia

ARTICLE INFO

Article history:
Received 31 March 2016
Received in revised form 4 July 2016
Accepted 26 July 2016
Available online 3 August 2016

Keywords:
Fundamental knowledge
Ageing
Adhesion
Atomic force microscopy

ABSTRACT

Crime scene investigations often rely on successful development of latent fingermarks for personal identification. In this context, exploring fundamental properties of latent fingermarks is vital for developing robust and more effective detection techniques. Here in a novel approach, PeakForce quantitative nanomechanical mapping (PF QNM) atomic force microscopy (AFM) has been used to study the variations in surface adhesion and topography of latent fingermark droplets over time. It was found that variation in adhesion was exhibited even across the surface of a single fingermark droplet, suggesting that individual droplets are heterogeneous in chemical composition on the nanoscale. The technique was successfully employed in observing the topographical variation of eccrine droplets, which has not been achieved using other optical microscopy techniques. In addition, the adhesion of fingermark droplets changed significantly as they aged. Propagation of a thin film of material from the fingermark ridges across the furrows, starting immediately after deposition, was captured in real-time, demonstrating the dynamic nature of the deposit. These results will aid in providing a more complete fundamental understanding of latent fingermark residue, allowing the more rational development of new detection techniques, especially those involving nanostructured materials.

1. Introduction

Fingermarks were first used as exchange evidence for criminal investigations in 1892 [1,2], and they remain extremely important in allowing forensic investigators to establish links between people, objects and locations [3]. To be successfully used for identification purposes, latent fingermarks must first be detected and made visible. Techniques for detecting latent fingermarks exploit the physical or chemical properties of the fingermark deposit in order to distinguish it from the surface upon which it lies [3,4]. Wet chemical techniques allow visualisation of the fingermark by means of a chemical reaction between a specific group of compounds present in the deposit and the development reagent. Physical techniques, such as dusting powders, instead rely on the overall properties of the deposit, for example the presence of moisture, greasy substances and electrostatic charge distribution [5].

The chemical composition of latent fingermarks is undeniably vital for any type of detection technique [2,6]. Latent fingermark deposits collected for research purposes are broadly categorised into three different types depending on their composition; natural, eccrine (obtained without touching any surfaces to have primarily sweat) and sebum rich deposits (deliberately charged with sebum from the face or scalp) [7]. It is considered to be a complex mixture of aqueous and lipid constituents with several exogenous contaminants such as food and cosmetic residues [8]. The sources for endogenous constituents are the eccrine and sebaceous glands in the dermis of the skin. The main constituents of eccrine skin secretions are water, amino acids and proteins, while sebaceous secretions are mainly comprised of glycerides, fatty acids, wax esters and squalene [6,8]. There has been sustained interest in the chemical composition of latent fingermarks, including variations across a wide array of influential factors in order to develop more effective visualisation techniques [9–14].

Abbreviations: 3D, three dimensional; AFM, atomic force microscopy; PF QNM, PeakForce quantitative nanomechanical mapping; IFRG, International Fingerprint Research Group; VMD, vacuum metal deposition.

* Corresponding authors at: Nanochemistry Research Institute, Curtin University, GPO Box U1987, Perth, Western Australia 6845, Australia.
E-mail addresses: t.becker@curtin.edu.au (T. Becker), s.lewis@curtin.edu.au (S.W. Lewis).

http://dx.doi.org/10.1016/j.forsciint.2016.07.024
0379-0738/

In contrast to the volume of studies that have been directed towards the chemical properties, there is a paucity of studies into the physical characteristics of latent fingermark residue with only a few studies from over four decades ago by Thomas [5,15,16] and a limited number of recent studies [17–20]. As proposed by Thomas, any physical measurements on fingermarks should be interpreted in relation to the distribution of the materials within the deposit [5]. The physical appearance of a fingermark deposit is highly influenced by its composition and the substrate on which it is deposited [21,22]. Microscopic examination of latent fingermarks by Thomas found that sweat rich (eccrine) deposits generally take the form of isolated circular droplets of material, whereas sebum rich deposits form ridges defined by continuous pools or by large, irregular shaped islands of material [5]. Recently, Moret et al. reported that in sebum rich deposits on glass, the droplet size was heterogeneous and the concentration of secretions was highest between the centre and the borders of the ridges [19]. While they could visualise the ridge outlines, no information on droplet size, shape and their ageing over time was provided for natural and eccrine deposits. It has also been reported that the contact angles for eccrine droplets on glass are larger in comparison to sebum rich droplets [23]. This observation to some extent is contradictory, but without information on the distribution of materials within the droplets, further explanations are impossible at this stage. The effect of the substrate on latent fingermark deposition has been reviewed by Bobev, who discussed the effects of temperature and structure of the surface upon adhesive forces that cause transfer of material on to surfaces [21]. Investigations carried out by Azoury et al. indicate that the surface energy of the substrate is a major factor responsible for the amount of skin secretions transferred on to substrates [22].

Powdering is one of the oldest techniques for latent fingermark detection and is routinely performed on fingermarks deposited on non-porous surfaces such as glass. The mechanics of powder adhesion to fingermarks were discussed by Thomas, noting the fact that powder formulations and their methods of application have been developed in an improvised manner, without a firm understanding of the mechanism of adhesion [5]. It is thought that the powder particles adhere to water and greasy substances via a pressure deficit mechanism [24]. When the powder particle is wetted on the lower side by the fingermark droplet, it creates a curvature of the meniscus, leading to a pressure difference inside the droplet, thereby causing the particle to adhere. Thus, the presence of water and grease in the deposit promotes the mechanical adherence of particles by (i) wetting the particle surface, (ii) forming capillary forces with the particles [25]. In a study into exploring adhesion of powder particles, the force required to detach aluminium powder particles (particle radius of 0.1–100 μm) from a deposit was reported to be in the range of 10^{-2} to 10^{-8} N [5]. The electrostatic attractions between the fingermark deposit and the dust powder particles are thought to play a minor role in powder adhesion [5].

In recent years, there has been an increased interest towards the development of novel detection techniques that involve nanoparticles and their conjugated forms [26–29]. Some of these techniques utilise solution baths containing nanoparticles as the mode of application and others apply nanoparticles as powders. These techniques are claimed to offer potentially superior outcomes across a wide range of substrates and on aged deposits [26–29]. However, there is as yet incomplete understanding about the mechanisms of these techniques [27,30]. For example, in the single metal deposition technique, the mechanism of deposition of colloidal gold nanoparticles onto the fingermark deposit is presumed to be via electrostatic attractions, although as yet this has not been confirmed [26,27]. In this context, exploring nanoscale features of the fingermark deposit such as surface interactions and spatial distribution of chemical compounds that contribute to the charge distribution of the deposit will be invaluable.

Ageing of the deposit, alteration in both the physical and chemical properties over time, is another confounding factor for latent fingermark detection [8]. For example, it is challenging to develop aged fingermarks, especially those exposed to tropical climates, using dusting powder and cyanoacrylate fuming [24,31]. Thus in recent years, more research has been directed toward studying trends in the ageing of fingermarks, but as indicated above these studies have mostly focussed on chemical rather than physical changes [9,12,13,32–35]. As comprehensively discussed by Moret et al., further investigations into fundamental physical properties of the fingermark deposit itself are vital to generate better understanding of the mechanisms of detection techniques which will in turn assist developing novel detection techniques or improving the existing ones [19].

To date, investigations into the physical properties of latent fingermarks have relied mainly on various optical microscopy techniques. What is missing in this array of techniques is the capability of resolving micro- or nanoscale features of the fingermark deposit without altering its properties. Although AFM has been applied in forensic research [36–39], it has had minimal impact on fingermark research [40,41]. Investigations into the effects of substrate surface topography on latent fingermark development [42] and imaging of latent fingermarks by localised corrosion on brass surfaces [43] by AFM are reported, but to the best of our knowledge, AFM has not been applied to the direct study of latent fingermark residue itself. We report here the potential of AFM to probe the physical properties of latent fingermarks such as surface interactions and topography and their variations over time. PeakForce quantitative nanomechanical mapping (PF QNM) is a novel imaging mode proprietary to Bruker atomic force microscopes, which allows simultaneous acquisition of high-resolution topography images and co-localised measurements of nanomechanical sample properties such as adhesion, stiffness, elastic modulus and deformation. The AFM probe is driven at a frequency well below the resonant frequency of the cantilever and periodically taps the sample surface. The feedback loop maintains a constant interaction force between probe and sample, which is measured via the cantilever deflection. The mechanical properties of the sample are derived in real-time, directly from the force curves obtained at every interaction between the tip and the sample. When the AFM probe taps the sample, its velocity is minimal due to sinusoidal motion of the piezo scanner, which makes this imaging mode ideal for soft samples [44–46]. This technique has been successfully applied to investigate interactions between the AFM tip and surface nanobubbles [47], to study stiffness and evolution of interfacial micropancakes [48], to observe assembly and disassembly process of hydrogels in situ [49] and to probe topography and microscopic contact angles of hydrocarbon oil droplets on a polystyrene surface [50].

2. Experimental

2.1. Collection of latent fingermarks

The sample collection protocol was based on the guidelines recommended by the International Fingerprint Research Group (IFRG), and the purpose of obtaining three types of deposits was only to study the physical properties of different deposits [7]. Eccrine, natural and sebum rich latent fingermark deposits from three donors (25–48 years of age) were obtained on plain microscope glass slides (Mikro-Glass, Australia), which were pre-cleaned with ethanol and ultra-pure water followed by drying under nitrogen. Immediately prior to sample collection, both hands were thoroughly washed with soap and warm water for