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Visualization of Latent Fingerprints Using Dextran-Based Micropowders Obtained from Anthocyanin Solution

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Abstract: Fingerprints are usually classified as patent, plastic, and/or latent. However, detection and visualization of latent fingerprints, as relevant traces found at the crime scene are very important for reliable identification of suspects by law enforcement personnel. Chemical and physical methods have now been used for years for that purpose, but they showed many deficiencies, where the most important one was related to their toxicity. Therefore, researchers are in constant search for producing complementary, less harmful methods, and some novel approaches are based on (bio)polymeric materials exploiting their specific properties. In this paper, dextran-based micropowders obtained from anthocyanin solution by the simple precipitating method were synthesized and characterized. Dextran is widely used in medicinal and pharmaceutical applications, but, up to our knowledge, no study has been reported using this biopolymer as a powder component for the developing agent for latent fingermarks visualization. Besides being biodegradable and biocompatible, dextran shows non-toxic properties and thus prevents detrimental effect on humans often present when commercial chemical and physical methods are being routinely employed, while at the same time reduces the overall cost of the obtained powder system. Four different formulations of dextran-based micropowders were prepared with the aim of evaluating their performances in visualizing latent fingerprints. FT-IR analyses confirmed interactions between components of the systems. Optical microscopy showed that prepared powdered samples were small and uniform in size, while at the same time confirmed their easy binding to the sweat and lipid residues present in the latent trace. The results demonstrated that these novel dextran-based powders have the potential to supplement routinely employed physical systems in detecting and visualizing latent fingerprints.

Keywords: Latent Fingerprints, (Bio)polymers, Dextran, *Brassica oleracea var. capitata f. rubra*, Anthocyanins, Forensic Science

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Antosiyanin Solüsyonundan Elde Edilen Dekstran Tabanlı Mikro Tozlar Kullanılarak Gizli Parmak İzlerinin Görselleştirilmesi

Nemanja Vučković, Stefan Dimitrijević, Nikola Milašinović

Öz: Parmak izleri genellikle patent, plastik ve / veya gizli olarak sınıflandırılır. Ancak, olay yerinde bulunan ilgili izler olduğu için gizli parmak izlerinin tespiti ve görselleştirilmesi, şüphelilerin kolluk kuvvetleri tarafından güvenilir bir şekilde tanımlanması için çok önemlidir. Bu amaçla kimyasal ve fiziksel yöntemler yıllardır kullanılmaktadır, ancak bu yöntemlerin özellikle toksik olmaları gibi çeşitli eksiklikleri vardır. Bu nedenle, araştırmacılar tamamlayıcı ve daha az zararlı yöntemler geliştirmek için sürekli araştırma yapmaktadır ve bu çalışmalar içerisinde bazı yeni yaklaşımlar, kendine has özellikleri olan (biyo) polimerik malzemelere dayanmaktadır. Bu makalede, basit bir çökeltme yöntemi ile antosiyanin çözeltisinden elde edilen dekstran bazlı mikro tozlar sentezlenmiş ve karakterize edilmiştir. Dekstran, tıbbi ve farmasötik uygulamalarda yaygın olarak kullanılmaktadır, ancak bilgimize göre, bu biyopolimeri, gizli parmak izlerinin görüntülenmesi için geliştirici ajan için bir toz bileşeni olarak kullanan hiçbir çalışma rapor edilmemiştir. Biyolojik olarak bozunabilir ve biyouyumlu olmanın yanı sıra, dekstran toksik olmayan özellikler gösterir ve bu nedenle ticari kimyasal ve fiziksel yöntemler rutin olarak kullanıldığında insanlar üzerinde zararlı etkileri önlerken, aynı zamanda elde edilen toz sisteminin toplam maliyetini azaltır. Gizli parmak izlerini görselleştirmedeki performanslarını değerlendirmek amacıyla dört farklı dekstran bazlı mikro toz formülasyonu hazırlanmıştır. FT-IR analizleri, sistem bileşenleri arasındaki etkileşimleri doğrulamıştır. Optik mikroskopi, hazırlanan toz numunelerin küçük ve muntazam boyutlarda olduğunu gösterirken, aynı zamanda gizli parmak izinde bulunan ter ve lipit kalıntılarına kolay bağlanabildiklerini doğruladı. Sonuçlar, bu yeni dekstran bazlı tozların, gizli parmak izlerini tespit etmek ve görselleştirmek için rutin olarak kullanılan fiziksel sistemleri tamamlama potansiyeline sahip olduğunu göstermiştir.

Anahtar kelimeler: Gizli parmak izleri, (Biyo) polimerler, dekstran, *Brassica oleracea var. capitata f. rubra*, Anthosiyaninler, Adli Bilimler

1. Introduction

Fingerprints represent unique features of individuals and, thus, they are one of the most valuable forensic evidence that could be recovered from the crime scene (Bumbrah, Sharma, & Jasuja, 2016). They often remain as random prints on surfaces of different objects, when sweat and sebum residues are deposited onto the surface. In the late 1800s, Ivan Vučetić gave basis in the identification of criminal offenders using fingerprints, by taking prints from all ten fingers and giving special marks for each basic form of a fingerprint (along with Henry-Galton classification system). That method, later named dactyloscopy, was a forerunner of a modern automated fingerprint identification system (AFIS) (Mozayani & Noziglia, 2006).

Based on the fingerprint structure, basic forms were classified as loops, arches, and whorls. Besides those evident features, fingermarks contain some tiny, specific, and distinguishing characteristics called minutiae points, which were necessary for reliable identification of persons (Mitrović, 1998). The papillary line traces are transferred by the sweat (eccrine) glands, which secrete sweat and other components through the sweat pores, leaving a trace characteristic for each person. Other compounds, such are blood, oil, ink, dye, etc. can often be found and transferred from finger surface to the substrate along with the fingerprint (Champod, Lennard, Margot, & Stoilovic, 2004). Therefore, three distinct types of fingerprint could be found in forensic practice: patent, plastic, and latent. Patent (traces deposited together with other compounds) and plastic (three-dimensional impressions) fingermarks are clearly visible to the human eye and can be easily examined to identify persons involved with a crime. On the other hand, latent fingermarks are imperceptible and consist of secretions of the eccrine, apocrine and sebaceous glands, where sweat contains, approximately, water (98%), minerals (0.5%), organic compounds (0.5%) and the 1.0% of other residuals (Färber, Seul, Weisser, & Bohnert, 2010). In order to analyze such traces, they must be visualized first, which is why different optical, physical, and chemical methods have to be employed. After applying these methods and developing traces, they can be processed in the same manner as visible ones (Färber, Seul, Weisser, & Bohnert, 2010; Trapecar & Balazic, 2007).

Chemical methods are based on chemical reactions between chemical means and fingerprint residues, followed by the formation of steady complexes (Datta, Lee, Ramotowski, & Gaensslen, 2001; Milašinović & Koturević, 2016). Ninhydrin and silver nitrate are routinely employed on porous surfaces, cyanoacrylate fuming is used on non-porous surfaces, while iodine fuming is applied on different porous, semi-porous and non-porous surfaces. However, these methods have disadvantages commonly related to their toxicity and potential formation of complexes (thus disabling further examination of fingermarks). On the other hand, physical methods include physical interactions or binding of certain pow-

ders or dyes to some specific residues from the (latent) prints. Commonly used powder formulations are regular, metallic, luminescent and thermoplastic, while their choice depends on the surface and its characteristics: illumination, texture, color, porosity, etc. (Bumbrah, Sharma, & Jasuja, 2016; Datta, Lee, Ramotowski, & Gaensslen, 2001; Milašinović & Koturević, 2016).

By marking toxicity and detrimental effect on human health as the biggest shortcomings of current approaches, scientists are aiming at developing some novel systems (or even methods) that will overcome the aforementioned problem and, additionally, satisfy cost-benefit demands. Therefore, researchers are resorting to various (bio)polymers, which utilization is still insufficiently known to the scientific public, especially in developing latent fingerprint traces (Milašinović, 2016). In recent years, researchers have used polymeric materials to map sweat pores on fingerprints (Lee, et al., 2014). The concept of sweat pore mapping is already well known, but present methods have failed to provide satisfactory results. One of the newest approaches, described in the research of Joosub Lee and colleagues, confirmed the adequate sensitivity in detecting individual pores of a human palm, with complete visualization of fingerprints. An article published in *Nature Communications* indicated that a polymer showing immediate fluorescence and color change in response to a small amount of water may be useful for detecting fingerprints (Lee, et al., 2014). Additionally, researchers are attempting to immobilize active ingredients (usually fluorescent dyes) into different polymeric materials to achieve stable and improved performances of the resulting systems (Araya-Hermosilla, Muñoz, Orellana, Yáñez, & Olea, 2014). Several studies have been conducted to evaluate the application of CdS nanoparticles encapsulated/immobilized into a chitosan matrix, showing their potential in developing of latent prints on various porous and semi-porous surfaces, regardless of their color (Dilag, Kobus, & Ellis, 2009; Milašinović, 2016; Wanga, Yang, Wanga, Shi, & Liu, 2009). However, all these systems are still insufficiently tested.

This paper deals with dextran-based micropowders, obtained by the precipitating method, using potassium periodate (KIO_4) as an initiator, *N*, *N*'-methylene bis(acrylamide) (MBA) as a crosslinking agent and methanol as a precipitation solvent. Dextran is a complex, branched and hydrophilic polysaccharide composed of anhydroglucose rings, obtained from bacteria (particularly from *Lactobacillus*, *Leuconostoc* and *Streptococcus* species), widely used in medicine and pharmacy, as a component of drug-delivering nanoparticle systems, material that reduces blood viscosity and prevents the formation of blood clots, etc. (Wang, Dijkstra, & Karperien, 2016; Wasiak, et al., 2016). However, up to our knowledge, there are currently no studies that address this biopolymer in latent fingerprints detection and visualization. Dextran was used due to its low price and biodegradable, biocompatible, and non-toxic properties, as well for its water solubility and easy filtration process (Wang, Dijkstra, & Karperien, 2016). KIO_4 is an oxidizing agent and it was used to obtain aldehyde functionalities of dextran chains,

to improve interactions with fingerprints trace residues (Maia, Carvalho, Coelho, Simões, & Gil, 2011). In this paper, four different microparticle formulations based on dextran biopolymer were prepared, and their potential in developing latent fingermarks was tested, showing that the prepared micro-formulations could complement currently used powdered systems.

2. Materials And Methods

2.1. Materials

Dextran powder was purchased from Sigma-Aldrich (USA), KIO_4 from Merck (Germany), MBA from Acros Organics (USA), and methanol from Centrohem (Serbia). Distilled water was used for all buffer solutions preparation. Acetate buffers of various pHs were prepared by dissolving sodium acetate and acetic acid in distilled water, to obtain buffer solution of desired pH value. Buffer solutions were used to extract anthocyanins from *Brassica oleracea* (var. *capitata*, f. *rubra*) and afterward the obtained solution was used to dissolve dextran powder, the initiator and the crosslinking agent. Besides *B. Oleracea* (var. *capitata*, f. *rubra*), all materials were used without further treatment and purification.

2.2. Preparation of Dextran-based Micropowders

Acetate buffer solution (pH ~ 3.52) (dissolving medium) was prepared by modifying experimental procedures described by Chandrasekhar, et al. (Chandrasekhar, Madhusudhan, & Raghavarao, 2012). This medium was used to extract anthocyanins from *B. Oleracea* (var. *capitata*, f. *rubra*). Briefly, 50 g of *B. Oleracea* (var. *capitata*, f. *rubra*), ground with blender *Bosch* (180W power, Germany), were added to 200 mL of acetate buffer solution, then mixed using a magnetic stirrer (~600 rpm) and heated (~30 min) until boiling of the solution was achieved. After cooling, the obtained solution containing anthocyanins was filtered using a metal mesh and kept in a refrigerator at 4 °C until further use. The obtained anthocyanin solution was used to obtain the different color of desired micropowders, as well for better enhancement through complexing with fingerprint sweat and lipid residues. Furthermore, four different formulations of dextran-based micropowders were prepared to determine their capability in visualizing latent fingermarks. Briefly, 2.0000 g of dextran powder was dissolved in 200 mL of a prepared solution containing anthocyanins. Afterward, the obtained solution was divided into 4 equal parts (volume of each 50 mL). The first solution was left as blank; in the second solution initiator in ratio 10:1 (dextran: KIO_4) was added; in the third solution crosslinking agent MBA (8 w/w.% by mass of biopolymer) was added; in the fourth solution, both KIO_4 and MBA were added taking the same ratios as was already stated. The samples were stirred at low speed and room temperature using

a magnetic stirrer. After homogenization, methanol in a 1:3 v/v ratio (solution sample: methanol) was added, to precipitate polymer from the solutions. When the precipitate was formed, samples were filtered using a filter paper, first dried at room temperature for ~ 24h, and then transferred at 37 °C for an additional few hours. Afterward, the obtained dry samples were ground with pestle and mortar to fine powders and kept in a desiccator until further application.

3. Characterization of the Prepared Powder Samples

3.1. FT-IR Analyses

The solid samples of synthesized micropowders were recorded in a dry state, using the *Bomem MB* 100 FT-IR spectrophotometer. Samples in the amount of 1.5 mg were mixed and ground with 75 mg of potassium bromide and then compressed into pellets at a pressure of 11 t for about a minute, using the *Graseby Specac* model: 15.011. The spectra were obtained in the wavenumber range between 4000 to 400 cm^{-1}, at 25 °C with 4 cm^{-1} resolution.

3.2. Optical Microscopy

The powdered samples were recorded with optical microscope *Leica FS C Comparison Macroscope*, equipped with The Leica IM Matrox Meteor II Driver Software Module. Samples were tested in a dry state, with and without backlight. Before imaging under the microscope, latent fingerprints left on microscope glass slides were developed using prepared powder samples.

3.3. Development of latent fingerprints

In order to determine the capability and performances of prepared powder samples, three different donors deposited their sebaceous and dry fingerprints onto a copy paper (porous), rubber (semi-porous), and glass (non-porous) surface. Following the guidelines proposed by the International Fingerprint Research Group (IFRG) ”natural“ (not specifically modified), sebaceous (oily) and dry fingerprints were deposited on mentioned surfaces using a technical scale, to simulate real manipulating procedures and determine the pressure on surfaces (force applied to accommodate 100-150 g, per fingerprint), and the prints were then left under laboratory (humid) conditions for a short period. That period allowed the traces to dry and reduce the residues, by the time the latent fingerprints were developed with synthesized powder samples and two control samples, using BVDA Squirrel hairbrush (BVDA, The Netherlands).

Optical microscopy was used to approximately determine the size and uniformity of prepared micropowders, as well as to evaluate their performances in visualizing latent fingermarks on the glass surface (showing the best results). The-

refore, sebaceous and dry fingerprints randomly deposited onto the glass microscopic slides (properly labeled) using a technical scale (according to the already described procedure), were left for a few minutes and then four types of prepared powders formulations and pure dextran powder (control sample) were used for their visualization. After the short period, fingerprints were halved with the thick slide barrier, and two different powders were applied on the same fingerprint (i.e. synthesized powders were applied to the left and pure dextran powder was applied to the right barrier side), using BVDA Squirrel hairbrush. Such type of visualization enabled a direct comparison between applied powder samples to assess their size range, uniformity, and efficiency in developing latent fingerprints.

Additionally, the prints were left under dry (desiccator) and humid (laboratory) conditions (relative humidity around 60%) and afterwards, fingerprints were developed in different time intervals – after 24 hours, 7 days and 1 month. The exception is the first series with 60 fingerprints (explained above), developed immediately after deposition used as a pretest to determine the direction of subsequent experiments. That resulted in total of 204 fingerprints, with 72 fingerprints per storage condition (and 60 additional fingerprints used as a pretest), where 180 were enhanced from a glass, 12 from a rubber and 12 from a paper surface.

IFRG recommends the implementation of 4 phases to evaluate new detection and visualization techniques and systems. By conducting the experimental procedures as stated above, Phase 1 (*Pilot Studies*) was completed, to determine the functionality of prepared powders (International Fingerprint Research Group (IFRG), 2014). The results are presented in detail in the Supplementary Material.

4. Results and Discussion

The synthesized powders were labeled as S(Dex/KIO_4/MBA), where "S" refers to the sample, "Dex" to dextran, "KIO_4" to the initiator and "MBA" to the crosslinking agent content used to prepare the desired micropowders. Obtained micropowders were tested on paper, rubber, and glass surface. Powder samples were applied on deposited fingerprints over 20 times on each of the surfaces, to determine the possibility and reproducibility of their application, i.e. to achieve desired visualization results. The best results were obtained with the sebaceous fingermarks deposited onto the glass surface. On the other hand, when applied onto the white paper surface with latent prints, it was not possible to achieve a satisfying contrast between the fingermark and the background, due to the white powder color. Additionally, the development of fingerprints on rubber surface was poor, since that surface contains many bulges and indentations, thus disabling the binding of the prepared micropowders to the fingerprint residues. Therefore, prepared micropowders did not provide satisfactory results on paper and rubber surfaces and additional research to improve this system should be conducted.

Figure 1 shows the sebaceous and dry fingermarks of one donor, developed with four prepared powdered samples and two control samples (BVDA Magnetic silver and pure dextran powder). The prints were then photographed under visible light with a 12 MP camera (aperture $f/2.2$, pixel size 1.22 μm) using a black background surface to achieve better contrast. When observing dry fingermarks, it was evident that their visualization was poor and satisfactory results were obtained with none of the applied powders. It is already well known that dry prints pose a great challenge in the forensic investigation of fingerprints (Lennard, 2007). On the other hand, development of sebaceous fingermarks was far better, since all powder samples have developed full fingerprint images, while at the same time visualizing the basic pattern type (double loop), the papillary lines (without disruption in their flow) and some minutiae points, as well.

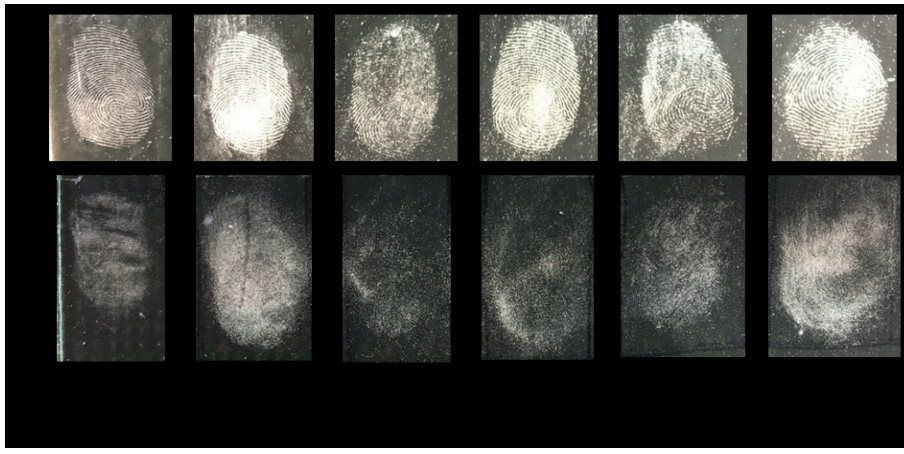

Figure 1: Sebaceous (1) and dry (2) fingerprints developed on the glass surface, using the following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under visible light using black background surface for better contrast.

When compared with control samples (Figure 1, 1-a) and 1-b)), the best results are obtained with sample S(1/1/0) (Figure 1, 1-d)), while other tested samples showed relatively satisfying results as well, which may be associated with a diameter size of powders' particles. As confirmed by optical microscopy, smaller particles better adhere and bind to sweat and lipid fingerprint residues, which was noticeable with synthesized powder formulations. Contrary, pure dextran powder somewhat "overfilled" a fingerprint due to larger and non-uniform distribution of particles' size (Gürbüz, Özmen Monkul, İpeksaç, Gürtekin Seden, & Erol, 2015).

Figure 2 represents the same fingerprints as those given in Figure 1, but recorded in dark, under yellow (lamp) light with red background surface to achieve better contrast. Different background colors enabled better contrast and visualization

of tiny friction ridges of sebaceous fingerprints (Figure 2, 1-c), d), e), f)), which were not clearly perceptible on prints recorded with black background (Figure 1).

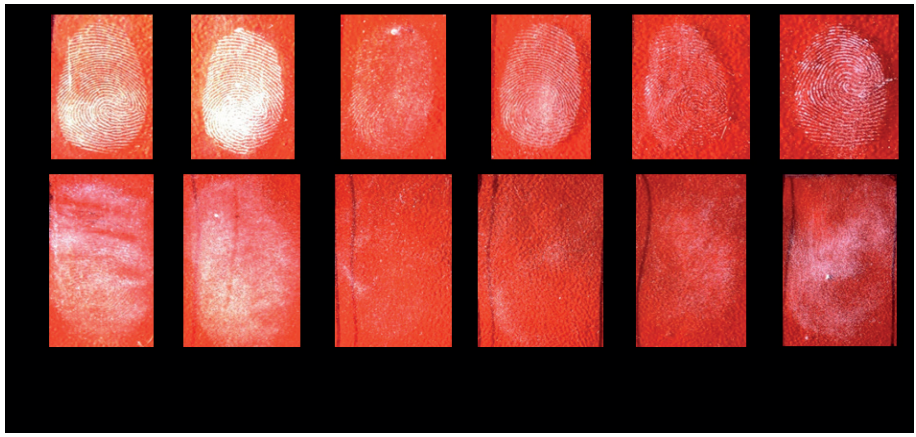

Figure 2: Sebaceous (1) and dry (2) fingerprints developed on the glass surface, using the following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under yellow (lamp) light with red background surface for better contrast.

4.1. FT-IR Analyses

FT-IR analyses were performed to evaluate interactions between components of prepared systems. Figure 3 shows the spectra of pure dextran and prepared powdered samples S(1/0/0), S(1/1/0), S(1/0/1), and S(1/1/1). All spectra in Figure 3 contain some characteristic bands: 3385 cm^{-1} due to O–H stretching and 2360 cm^{-1} due to the stretching of C–H (Carp, et al., 2010; Mehta, Rucha, Bhatt, & Upadhyay, 2006; Mitić, Cakić, & Nikolić, 2010). The band at 1154 cm^{-1} can be assigned to stretching vibrations of the C–O–C bond and glycosides bridge, while the band at 1017 cm^{-1} can be associated with stretching of C–O–H (Chiu, Hsiue, & Chen, 2004; Mehta, Rucha, Bhatt, & Upadhyay, 2006; Mitić, Cakić, & Nikolić, 2010). The weak band at 1110 cm^{-1} can be ascribed to the vibration of the C–O bond at the C4 position of the glucopyranose units (Mitić, Cakić, & Nikolić, 2010). Peaks at 905, 841, and 758 cm^{-1} can be assigned to α-glucopyranose ring deformation modes (Cakić, Nikolić, Ilić, & Stanković, 2005; Carp, et al., 2010). Additionally, weak shoulder peak at 1077 cm^{-1} may be due to complex vibrations involving the stretching of the C6–O6 bond with the participation of deformational vibrations of the C4–C5 bond (Guerrero, Kerry, & de la Caba, 2014; Nikolić, Cakić, Mitić, & Ilić, 2008). However, according to Mitić, et al. (Mitić, Cakić, & Nikolić, 2010), peaks at 1041 and 1017 cm^{-1} present in all spectra are related to the crystalline and amorphous phases, respectively, and can be responsible for more and less ordered

structures of dextran chains. Finally, the band at 3830 cm^{-1}, slightly more intense in spectra 4 and 5 (Figure 3) when compared with others, can be related to the N–H stretching frequency of the acrylamide (in MBA) (Fan, Zhang, & Feng, 2005).

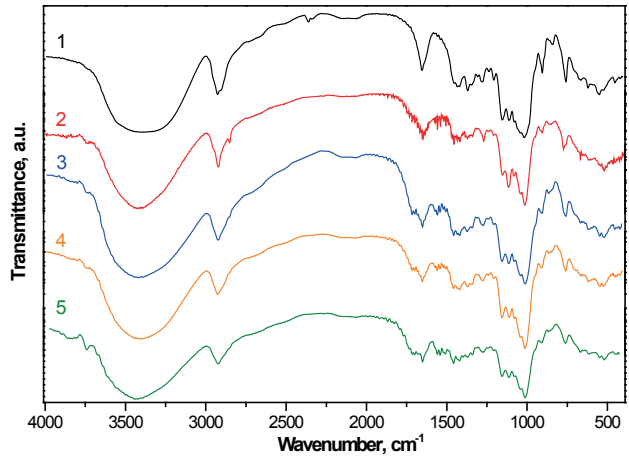

Figure 3: FT-IR spectra: 1) pure dextran; 2) S(1/0/0); 3) S(1/1/0); 4) S(1/0/1) and 5) S(1/1/1).

4.2. Optical microscopy

Since the best results were obtained with traces deposited onto glass (non-porous) surface, the same surface was used for further examinations. Sebaceous and dry (data are shown in Supplementary Material document) fingerprints randomly deposited onto the glass microscopic slides (properly labeled) using a technical scale (according to the described procedure), were left for a few minutes and then 4 prepared powders formulations and pure dextran powder (control sample) were used for their visualization. After the short period, fingerprints were halved with the thick slide barrier, and 2 different powders were applied on the same fingerprint – synthesized powders were applied to the left and pure dextran powder was applied to the right barrier side, using BVDA Squirrel hairbrush. Afterward, the samples were recorded under the optical microscope (magnification ×15), using dark-field (Figure 4, a1), b1), c1) and d1)) and bright-field (Figure 4, a2), b2), c2) and d2)) contrast techniques. Three different donors have left fingerprints and their visualization and scanning were repeated several times, with only the mean results shown in Manuscript for practical reasons (results are presented in detail in Supplementary Material).

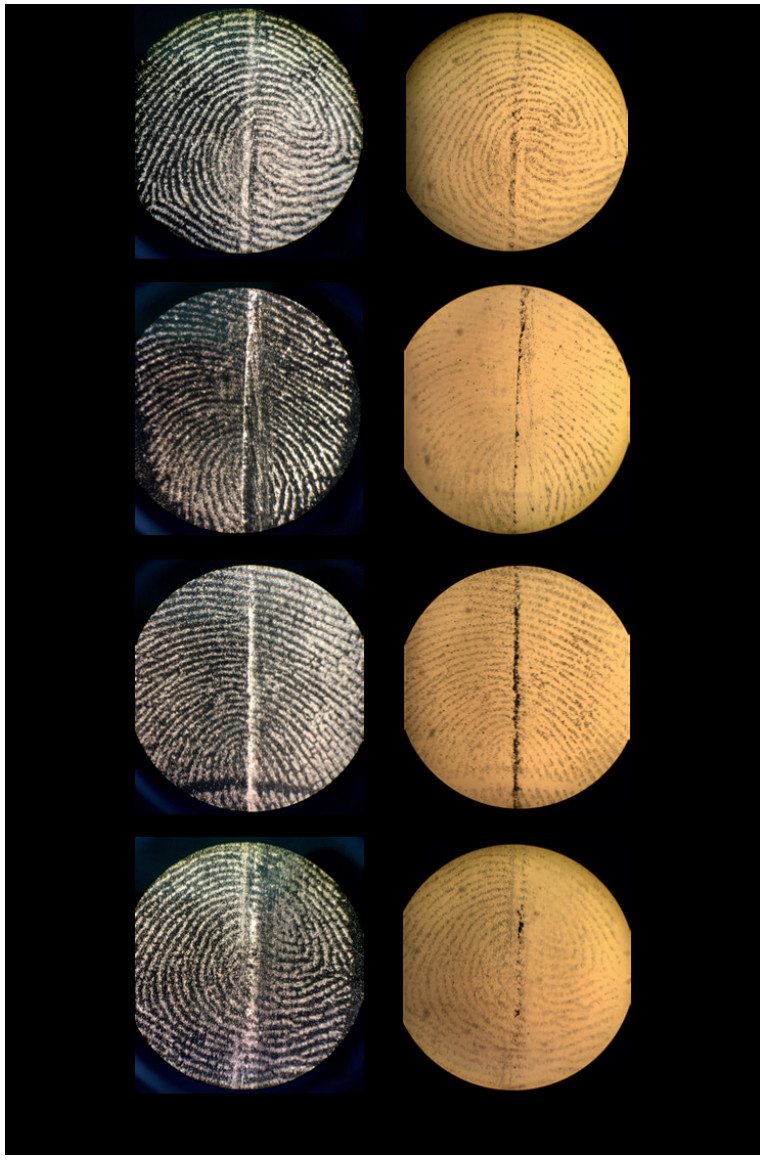

Figure 4: Sebaceous fingerprints deposited onto glass microscopic slides, left for a few minutes and developed using powdered samples: a) S(1/0/0); b) S(1/1/0); c) S(1/0/1) and d) S(1/1/1) (left side of the images) and pure dextran powder as a control sample (right side of the images), recorded with an optical microscope (magnification ×15), using: dark-field (a1-d1) and bright-field (a2-d2) contrast techniques.

When compared to the pure dextran powder (control sample), all prepared powder samples showed better results in terms of developing latent fingermarks, which was reflected in visualizing the papillary lines with their continuous flow and making perceptible some minutiae, as well. As we hypothesized previously, this may be associated with a smaller diameter size of prepared powders' particles, when compared with pure dextran powder (data not shown), which enabled their better adhesion and binding to fingerprint sweat and lipid residues (Gürbüz, Özmen Monkul, İpeksaç, Gürtekin Seden, & Erol, 2015). Additionally, when applied with a brush, prepared powdered samples bonded with fingerprint residues and did not remain in the interpapillary space when compared with the control sample. On the other hand, pure dextran powder has also developed fingerprints with persuasive results, but with noticeable "overfilling" of traces. Sample S(1/0/0) (Figure 4, a)) showed as good results as S(1/1/1) (Figure 4, d)) and even better results than samples S(1/1/0) and S(1/0/1) (Figure 4, b) and c)), which was very promising, since that sample contains only dissolved dextran powder precipitated with methanol, without initiator and/or crosslinker (KIO_4 and MBA show toxic, detrimental and irritating effect, while MBA is also potentially carcinogenic, as already explained and confirmed by Lent et al. (Lent, Crouse, & Eck, 2017; George, et al., 1998)). Therefore, a very satisfying visualization of sebaceous fingerprints was achieved using glass surface as substrates, and with the relatively cheap and less harmful dextran-based powdered system.

On the other hand, it was evident that dry fingerprints could not be developed with applied powders, due to the lack of lipid and sweat residues deposition onto different substrates. Those prints still must be continuously and thoroughly investigated to overcome one of the main problems in the forensic examination of fingerprints (Lennard, 2007).

Conclusions

In this paper, four different dextran-based micropowder formulations were obtained by a simple precipitating method and were characterized to determine their potential application in the development of latent fingerprints. Dextran was used due to its availability and low price, water solubility, and non-toxic properties. The initiator and the crosslinking agent were used for obtaining aldehyde functionalities of dextran chains and their crosslinking, respectively, to enhance interactions with fingerprint residues. However, KIO_4 and MBA showed a toxic and detrimental effect, which is unfavorable for the desired bio-based powder system. Based on the accomplished results, sample S(1/0/0) showed the best properties, with small and uniform particles, good binding to the fingerprint residues, and their apparent visualization, and the system is less harmful and satisfies the cost-benefit requirements. Additionally, all prepared powder samples showed

better results in terms of developing latent fingermarks when compared to the pure dextran powder (control sample). However, obtained results did not meet the expectations regarding color appearance, since the color of applied micropowders was not appropriate to visualize fingerprints on the (white) paper surface and the hypothesized complexing of anthocyanins has not contributed to the enhancement of fingerprints on the rubber surface. On the other hand, as many commercial dusting formulations, these micropowders are easily handled and applicable, requiring no prior knowledge and the method itself is non-destructive, avoiding irreversible loss of traces. Finally, additional research should include other (bio) polymers or addition of bio-based dyes and indicators, to expand the application of these systems on other surfaces and under different conditions, with the aim at supplementing some of the routinely employed physical methods in visualizing and enhancing latent fingerprints.

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Supplementary Material

In order to determine the functionality and performances of prepared powder systems in developing latent fingerprints, the guidelines proposed by the International Fingerprint Research Group (IFRG) were followed. Experimental procedures were set to conduct Phase 1 (*Pilot Studies*) of recommended guidelines, which briefly implies:

- 3-5 donors;
- 1-3 clean, low interference substrates;
- Donors briefed on how to deposit fingermarks, with assistance provided as required;
- Natural marks preferred, with groomed marks avoided where possible;
- Fingermarks should normally be allowed to "age" for a minimum of 24 hours prior to development and the actual age prior to treatment should be recorded and reported;
- The study should include at least a preliminary performance comparison against relevant routine detection methods (for example, the treatment of split marks can be used to provide an initial indication of relative performance);
- Qualitative or holistic scales should be employed for assessing the quality of fingermark development; and
- Reports must clearly indicate limitations and conclusions must be conservative (International Fingerprint Research Group (IFRG). (2014). Guidelines for the Assessment of Fingermark Detection Techniques. Retrieved from https://ips-labs.unil.ch/ifrg/wp-content/uploads/2014/06/IFRG-Research-Guidelines-v1-Jan-2014.pdf).

Therefore, three different donors deposited their sebaceous and dry fingerprints onto porous (paper), semi-porous (rubber) and non-porous (glass) surface using technical scale (force accommodated to 100-150g, per fingerprint), in order to simulate manipulation with objects under real conditions, and fingerprints were left under humid (laboratory) conditions for a few minutes. In order to estimate performances in visualizing latent fingerprints, besides four prepared powder formulations, two control samples (BVDA Magnetic silver and pure dextran powder) were employed with BVDA Squirrel hair brush. Afterwards, all fingerprints were photographed with a 12 MP camera (aperture $f/2.2$, pixel size 1.22 μm). The initial results are shown in Figures S1, S2, S3 and S4, which represent sebaceous and dry fingermarks developed on paper (Figure S1), rubber (Figure S2) and glass (Figures S3 and S4) surface, using

aforementioned powder samples. Figures S3 and S4 show the same fingerprints, but photographed under different conditions in order to obtain adequate contrast.

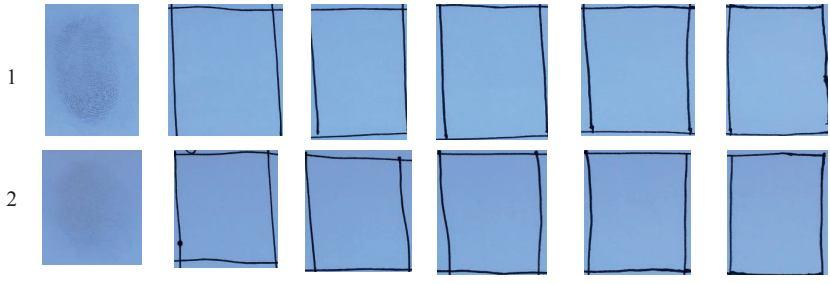


Figure S1. Sebaceous (1) and dry (2) fingerprints developed on paper surface, using following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under visible light.

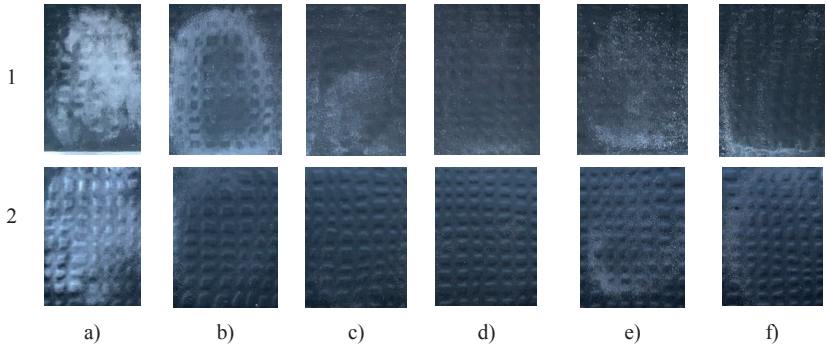
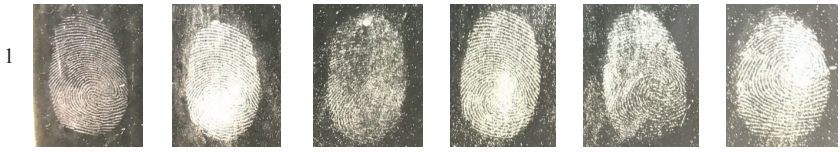


Figure S2. Sebaceous (1) and dry (2) fingerprints developed on rubber surface, using following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under visible light.



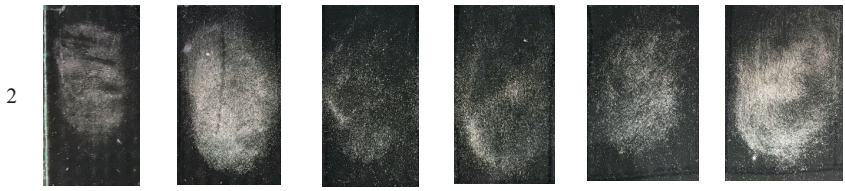


Figure S3. Sebaceous (1) and dry (2) fingerprints developed on glass surface, using following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under visible light using black background surface for better contrast.

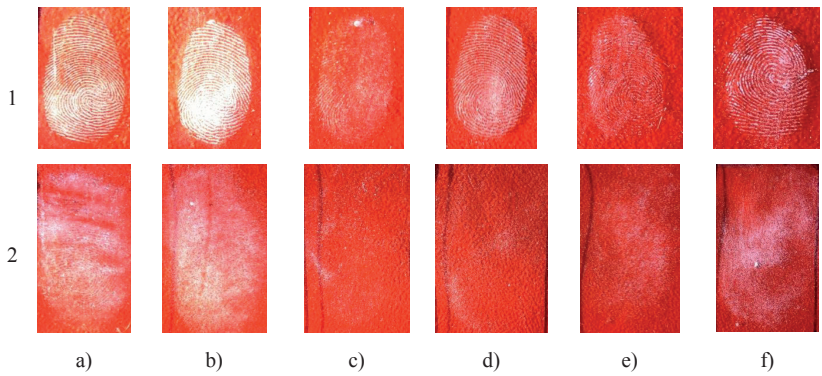


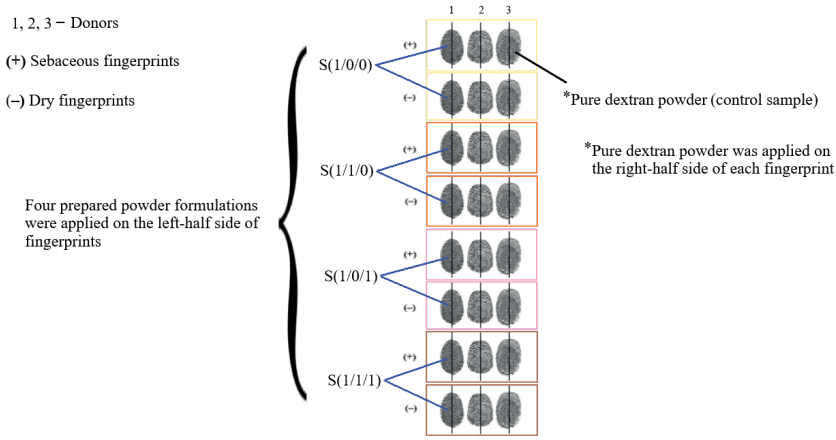
Figure S4. Sebaceous (1) and dry (2) fingerprints developed on glass surface, using following powder samples: a) BVDA Magnetic silver powder; b) Pure dextran powder; c) S(1/0/0); d) S(1/1/0); e) S(1/0/1) and f) S(1/1/1); and recorded under yellow (lamp) light with red background surface for better contrast.

Based on the obtained results, it is evident that visualization of dry fingerprints was poor and satisfactory results were not obtained with none of the applied powders. It is already well known that dry prints pose a great challenge in forensic investigation of fingerprints[1]. Sebaceous fingerprints have been successfully developed on glass surface with all applied powder samples, but the best results are accomplished with sample S(1/1/0). On the other hand, when applying

[1] Lennard, C. (2007). Fingerprint detection: current capabilities. *Australian Journal of Forensic Sciences, 39*(2), 55-71.

prepared powders onto the white paper surface with latent prints, it was not possible to achieve satisfying contrast between the fingermark and the background, due to the white powder color. Additionally, development of fingerprints on rubber surface was poor, since that surface contains bulges and indentations, thus disables the binding of the prepared micropowders to the fingerprint residues. Therefore, since the best results are obtained on glass surface, the same surface was used for further examinations.

Subsequently, in different time intervals, fingerprints deposited onto glass surface were split in halves with a thin barrier and the following powders were applied with BVDA Squirrel hair brush, as represented in the scheme bellow:



Afterwards, fingerprints were recorded with Leica FS C Comparison Macroscope, equipped with The Leica IM1000 Matrox Meteor II Driver Software Module, using magnification ×15 and two contrast techniques (backlighting).

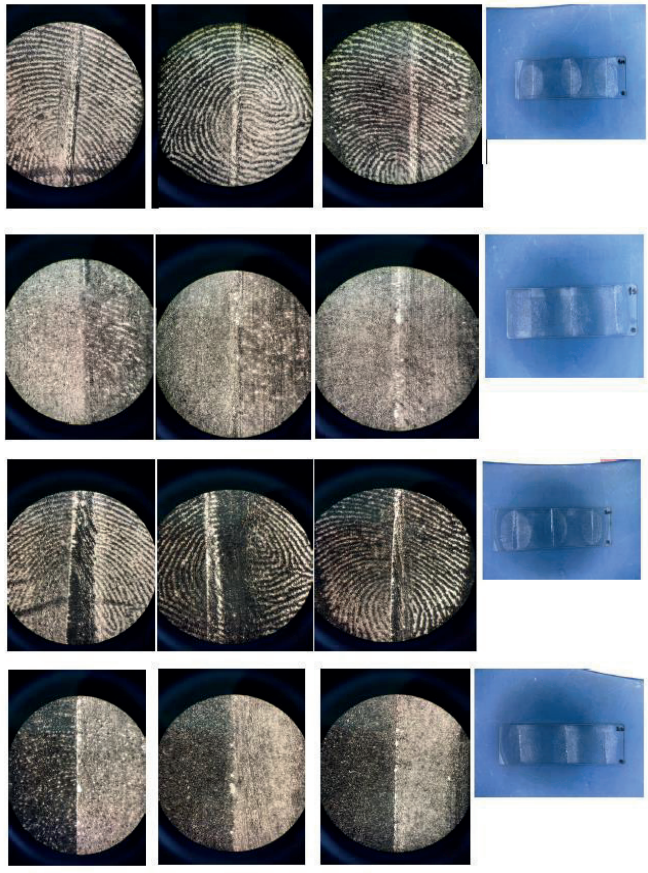
Immediate development of latent fingerprints – June 6, 2019

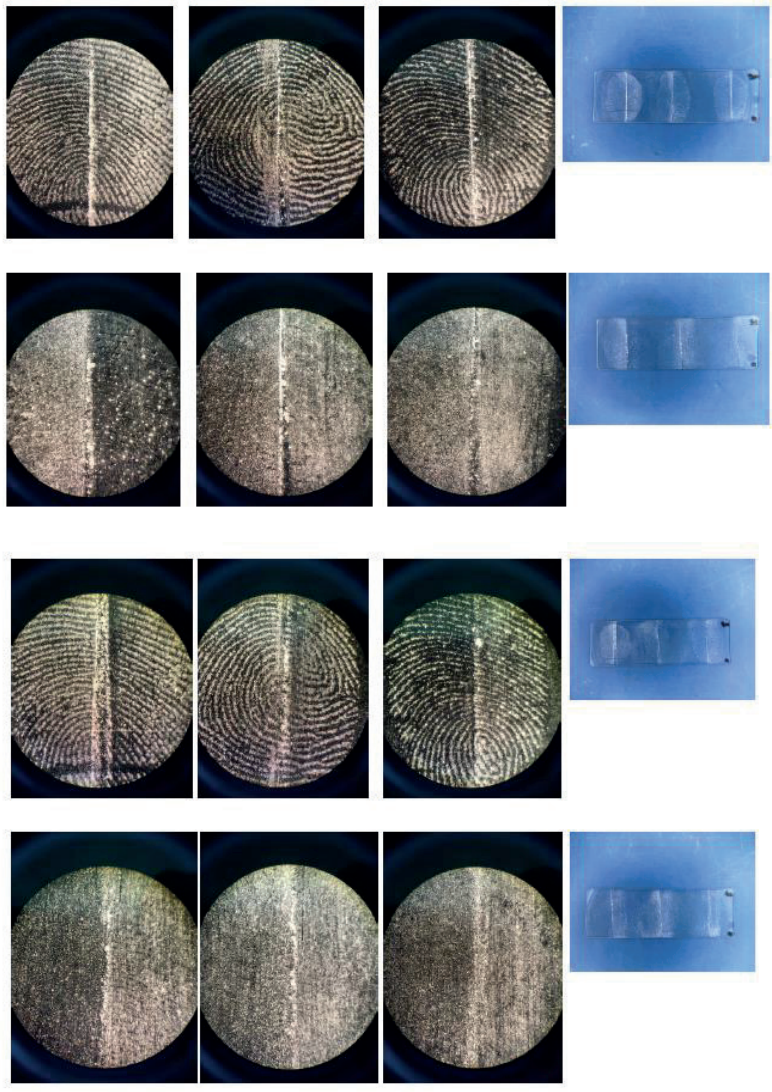
Sebaceous and dry fingerprints of three different donors developed immediately after deposition, with samples S(1/0/0), S(1/1/0), S(1/0/1), S(1/1/1) and pure dextran powder (as a control sample). Figure S5 represents fingerprints developed on glass (non-porous) surface, recorded using dark-field (Figure S5, a)) and bright-field (Figure S5, b)) contrast techniques. In the far right column all developed fingerprints photographed using a magnifying glass (magnification ×5) with a 12 MP camera (aperture f/2.2, pixel size 1.22 μm) were shown.

Fingerprints developed on glass (non-porous) surface

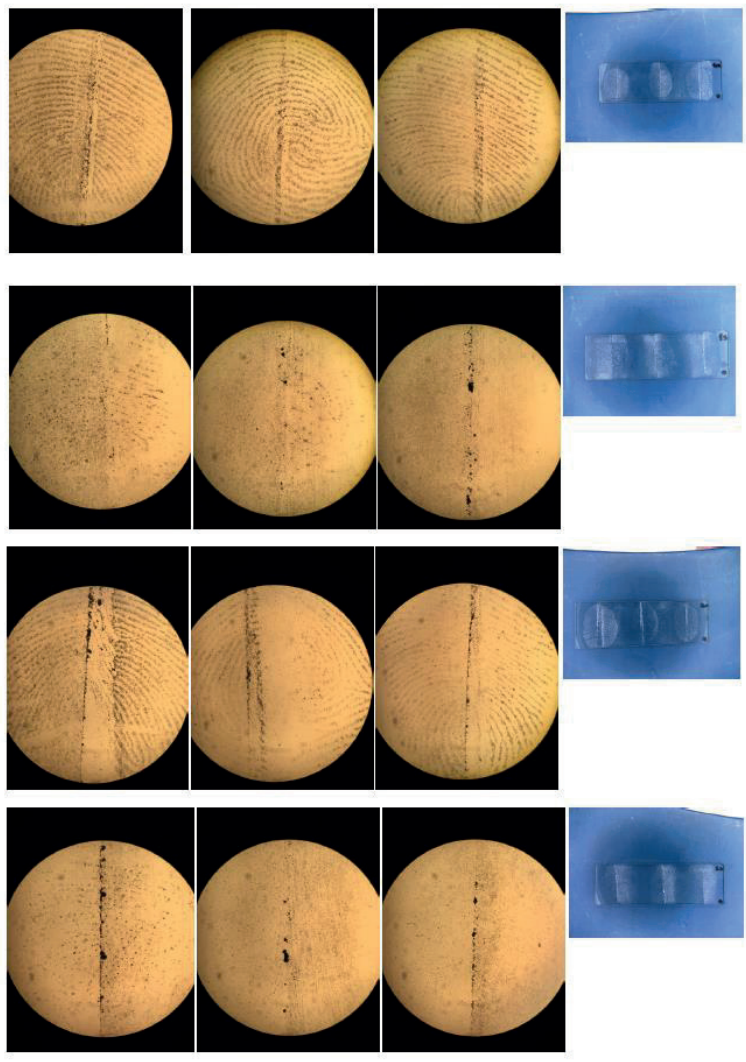
Fingerprints were recorded with the optical microscope, using:

a) Dark-field contrast technique





b) Bright-field contrast technique



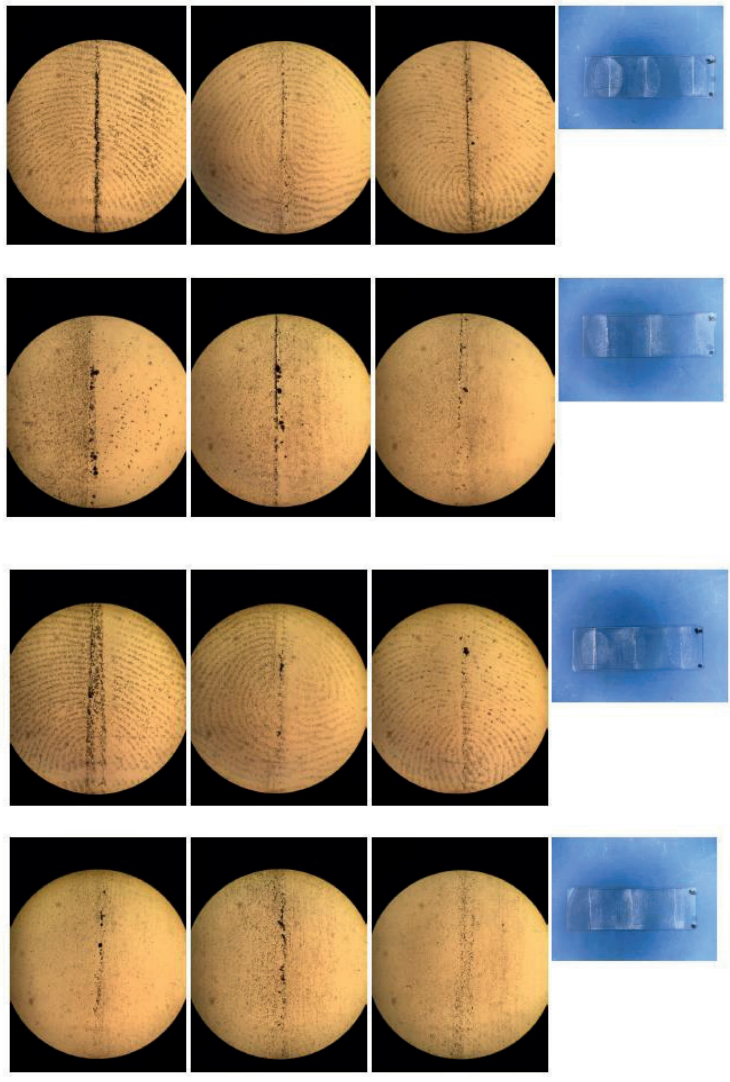


Figure S5. Sebaceous and dry fingerprints developed on glass (non-porous) surface with aforementioned powdered samples, recorded using: a) dark-field and b) bright-field contrast technique.

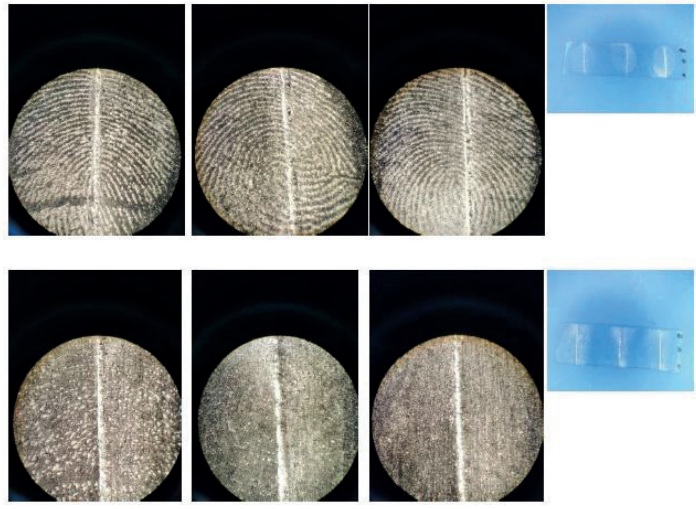
Development of latent fingerprints 24 hours after deposition – June 7, 2019

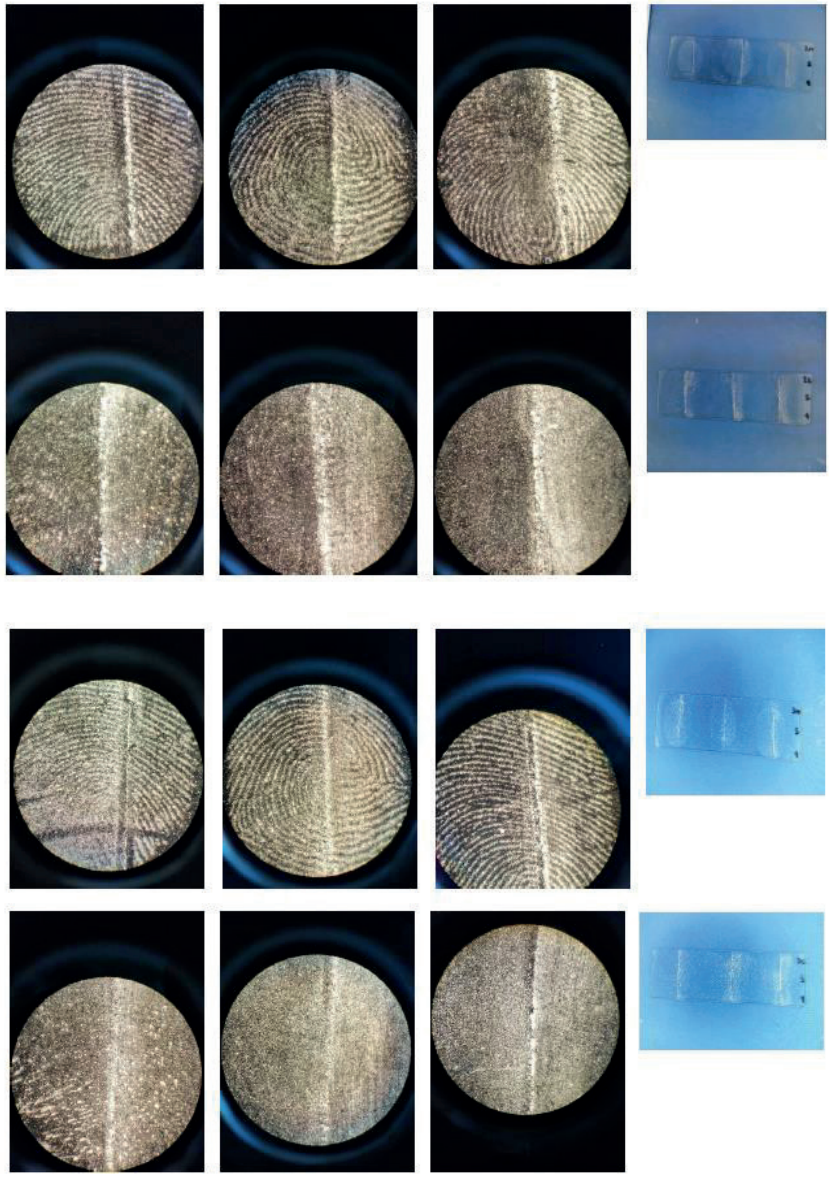
Sebaceous and dry fingerprints of three different donors, kept under dry (desiccator) and humid (laboratory) conditions, developed 24 hours after deposition, with samples S(1/0/0), S(1/1/0), S(1/0/1), S(1/1/1) and pure dextran powder (as a control sample). Figures S6 and S7 represent prints on glass (non-porous) surface kept under dry and humid conditions, respectively, recorded using dark-field (Figures S6 and S7, a)) and bright-field (Figures S6 and S7, b)) contrast techniques. In the far right column all developed fingerprints photographed using a magnifying glass (magnification ×5) with a 12 MP camera (aperture *f*/2.2, pixel size 1.22 μm) were shown.

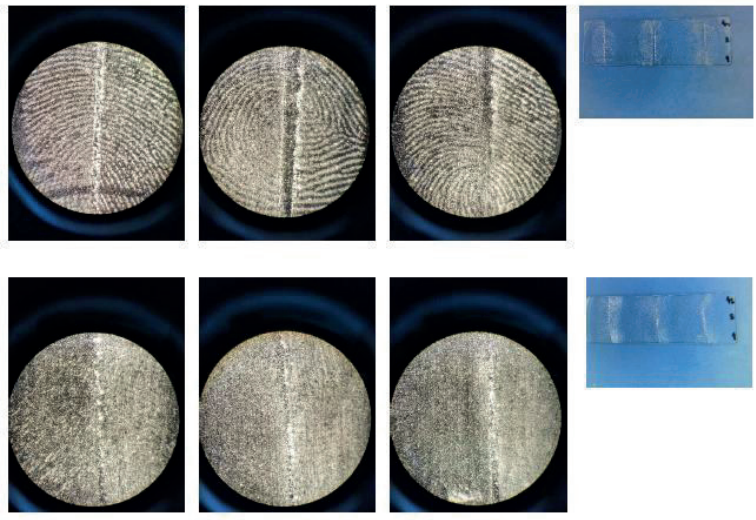
Fingerprints developed on glass (non-porous) surface (dry (desiccator) conditions)

Fingerprints were recorded with the optical microscope, using:

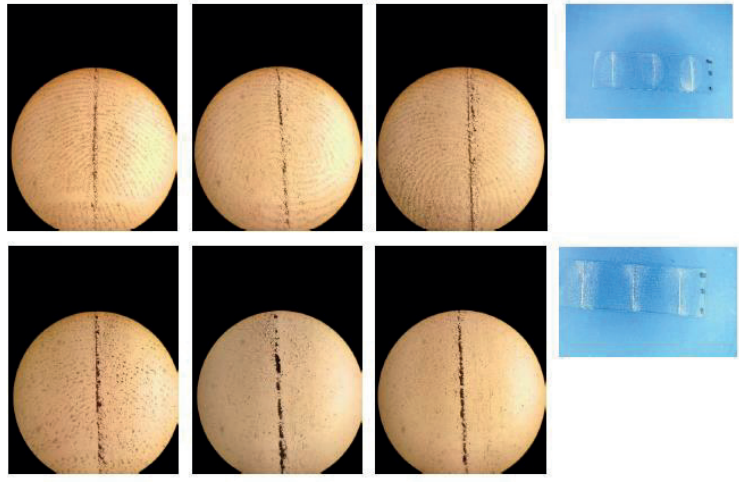
a) Dark-field contrast technique

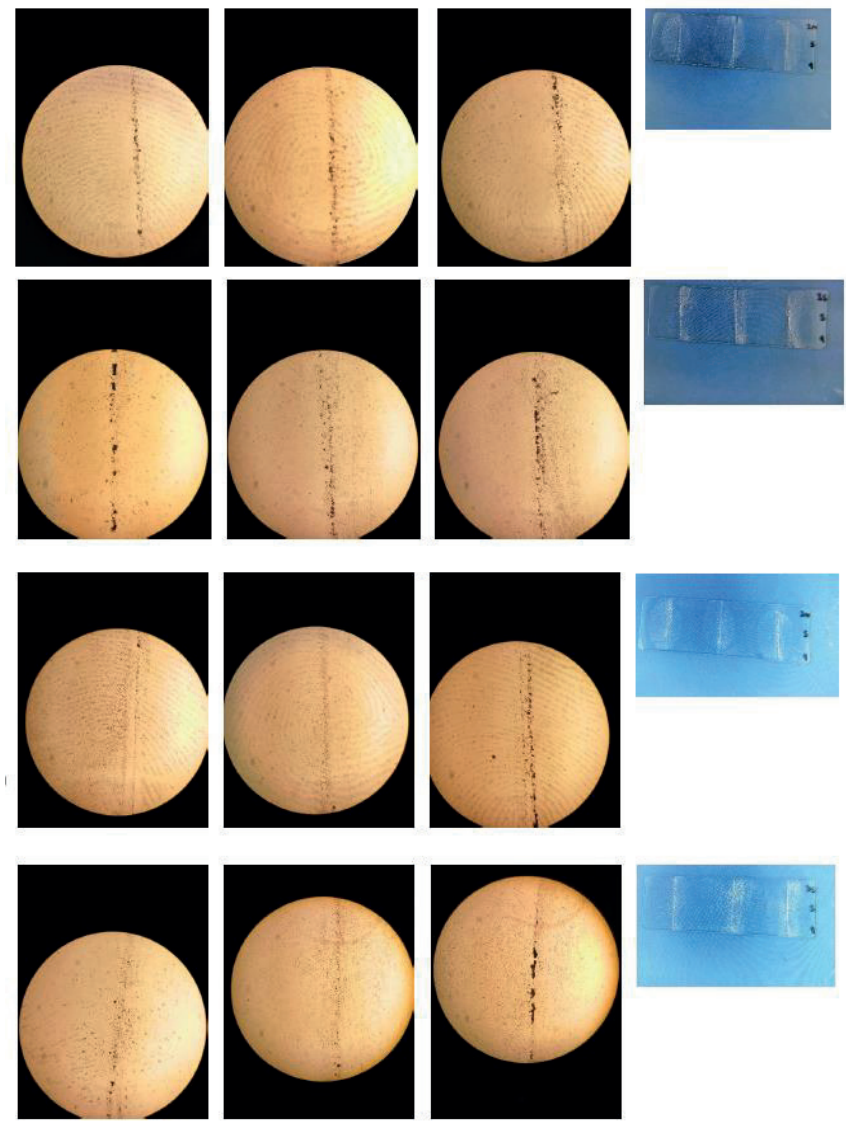






b) Bright-field contrast technique





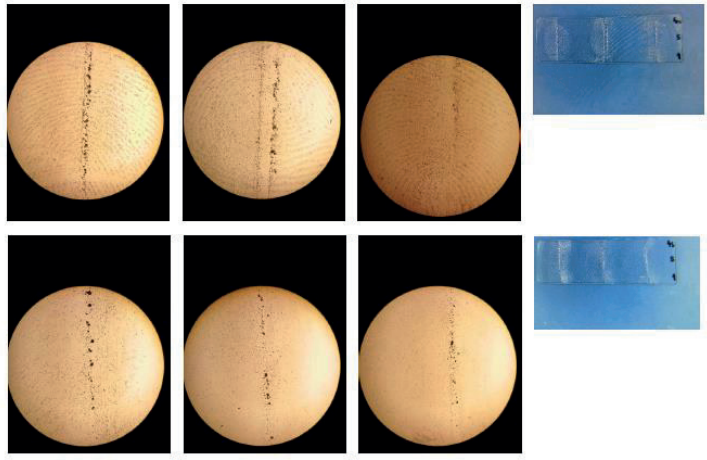
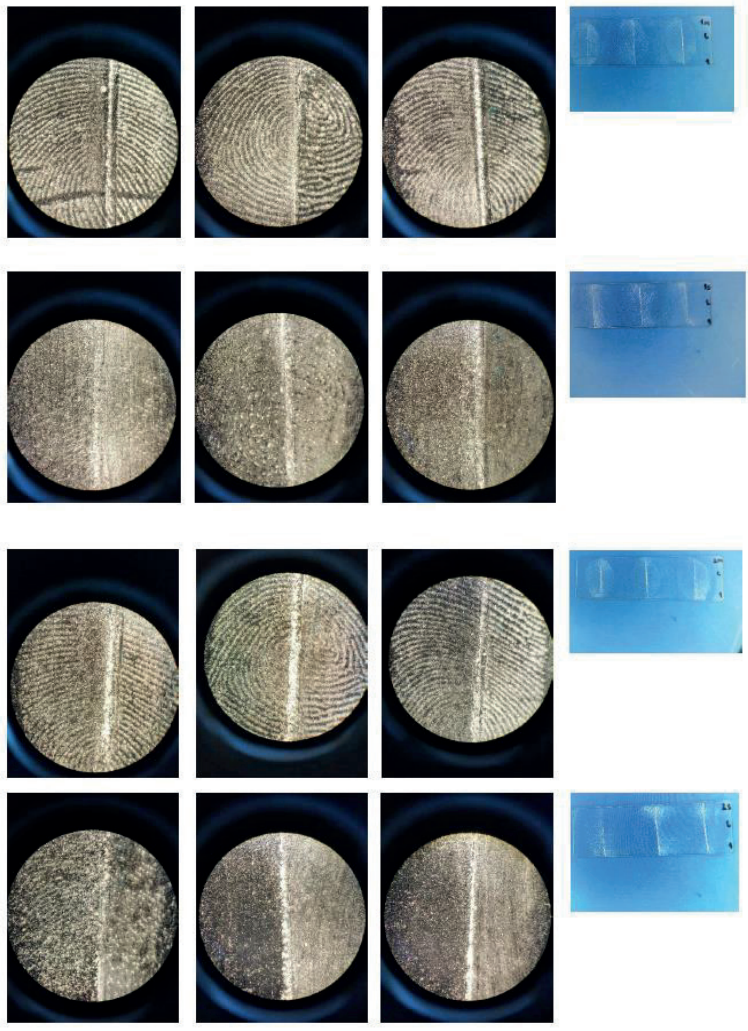


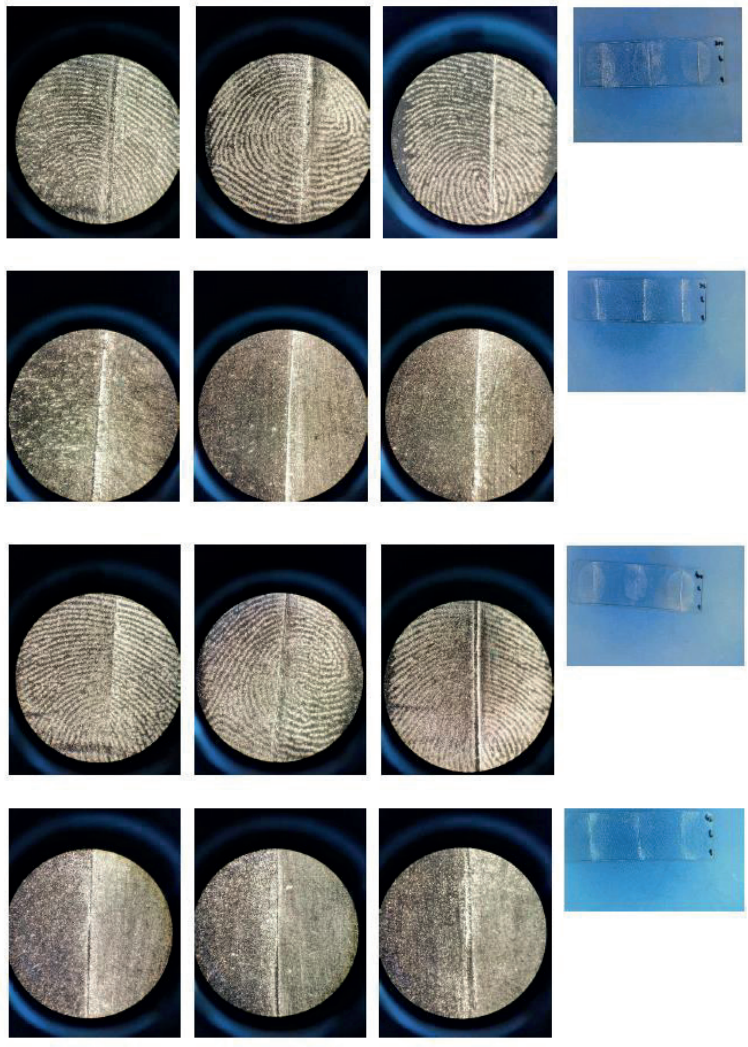
Figure S6. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under dry conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

Fingerprints developed on glass (non-porous) surface (humid (laboratory) conditions)

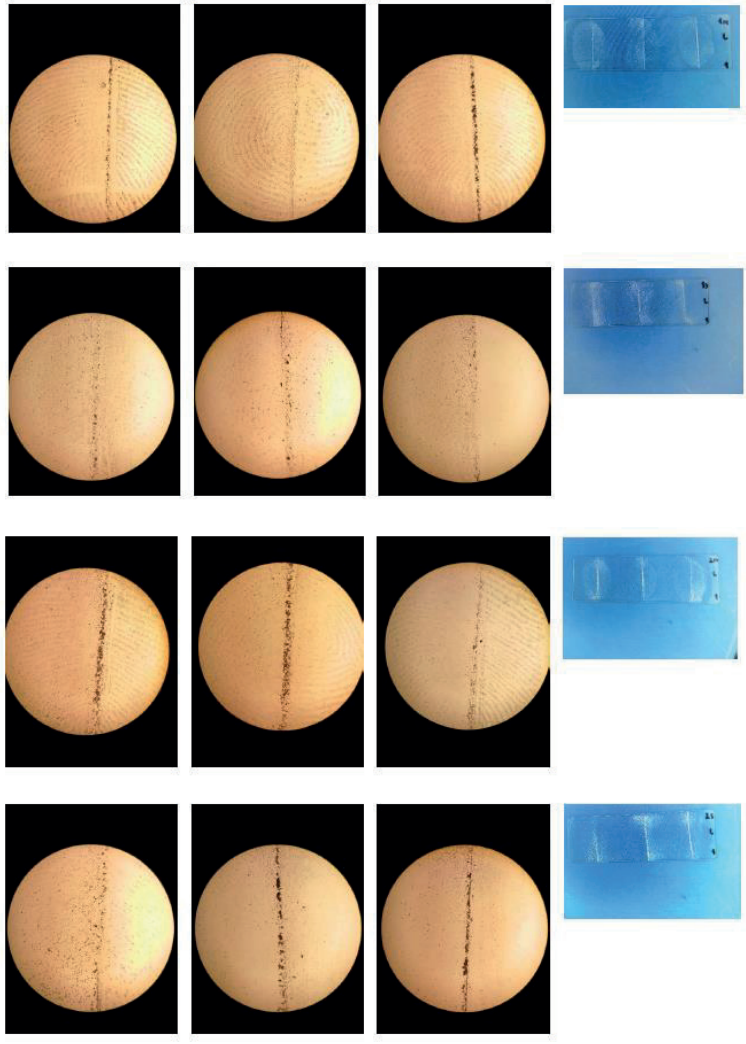
Fingerprints were recorded with the optical microscope, using:

a) Dark-field contrast technique





b) Bright-field contrast technique



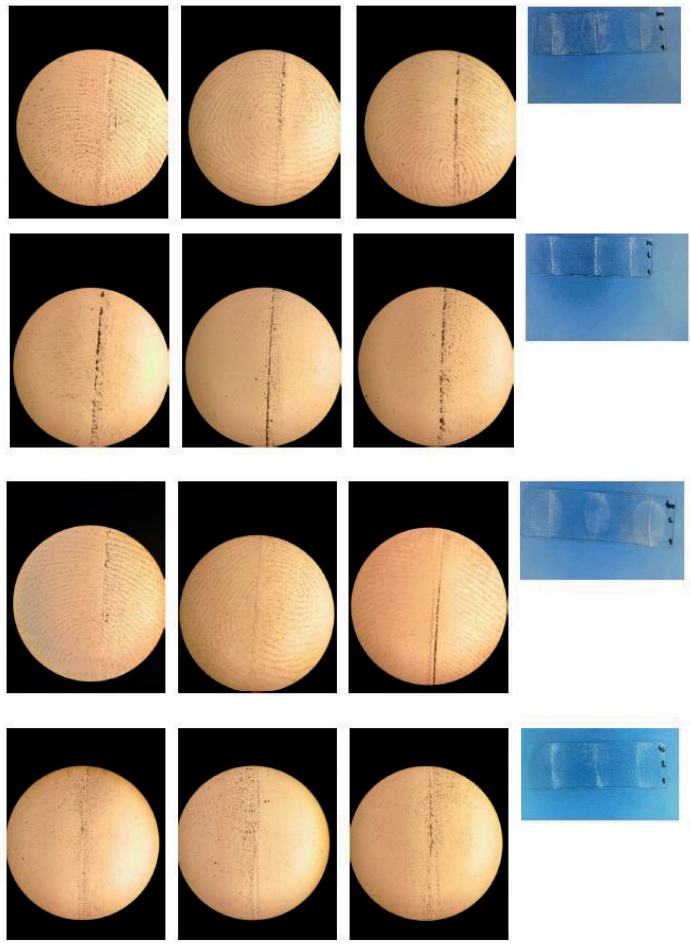


Figure S7. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under humid conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

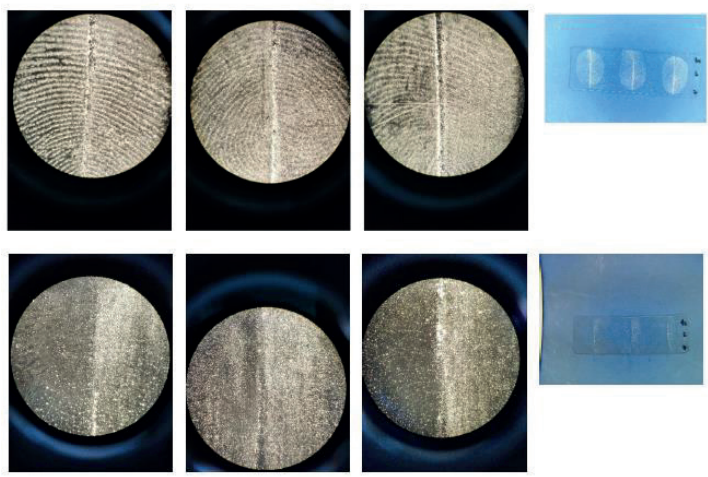
Development of latent fingerprints 7 days after deposition – June 13, 2019

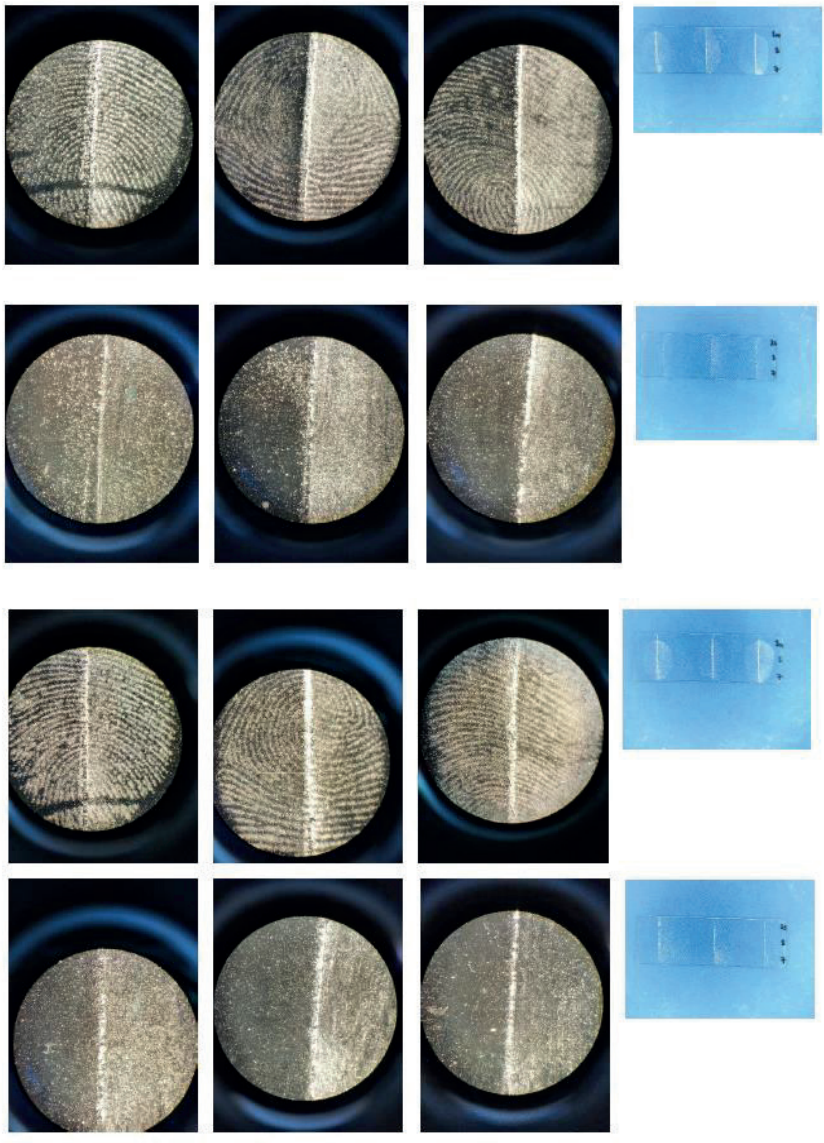
Sebaceous and dry fingerprints of three different donors, kept under dry (desiccator) and humid (laboratory) conditions, developed 7 days after deposition, with samples S(1/0/0), S(1/1/0), S(1/0/1), S(1/1/1) and pure dextran powder (as a control sample). Figures S8 and S9 represent prints on glass (non-porous) surface kept under dry and humid conditions, respectively, recorded using dark-field (Figures S8 and S9, a)) and bright-field (Figures S8 and S9, b)) contrast techniques. In the far right column all developed fingerprints photographed using a magnifying glass (magnification ×5) with a 12 MP camera (aperture *f*/2.2, pixel size 1.22 μm) were shown.

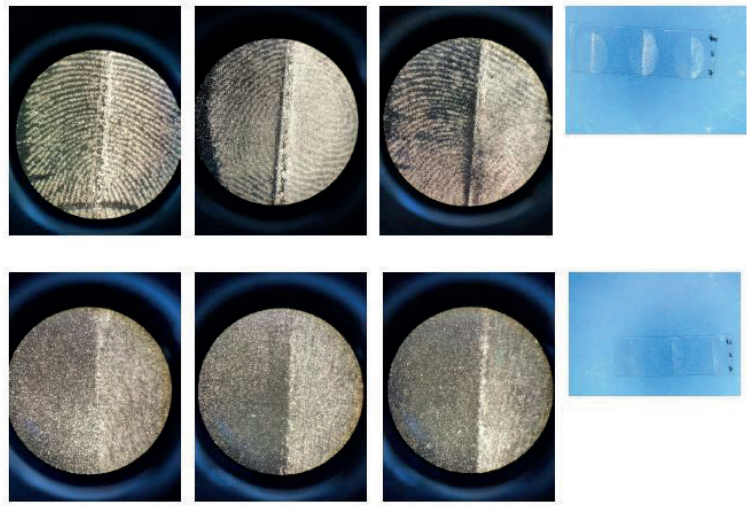
Fingerprints developed on glass (non-porous) surface (dry (desiccator) conditions)

Fingerprints were recorded with the optical microscope, using:

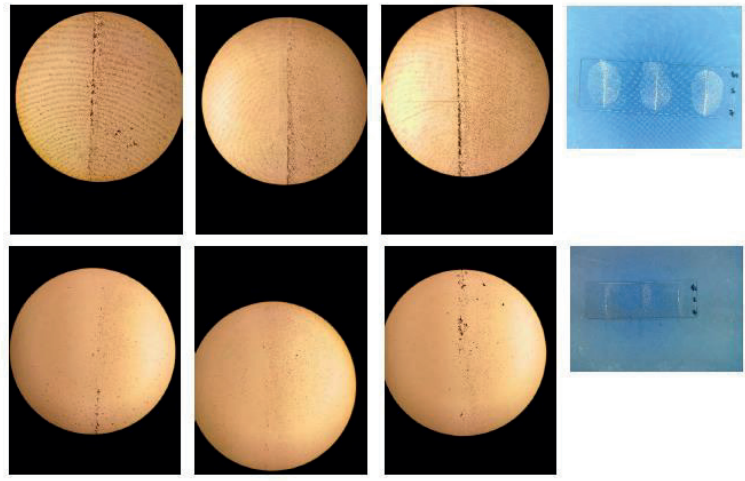
a) Dark-field contrast technique

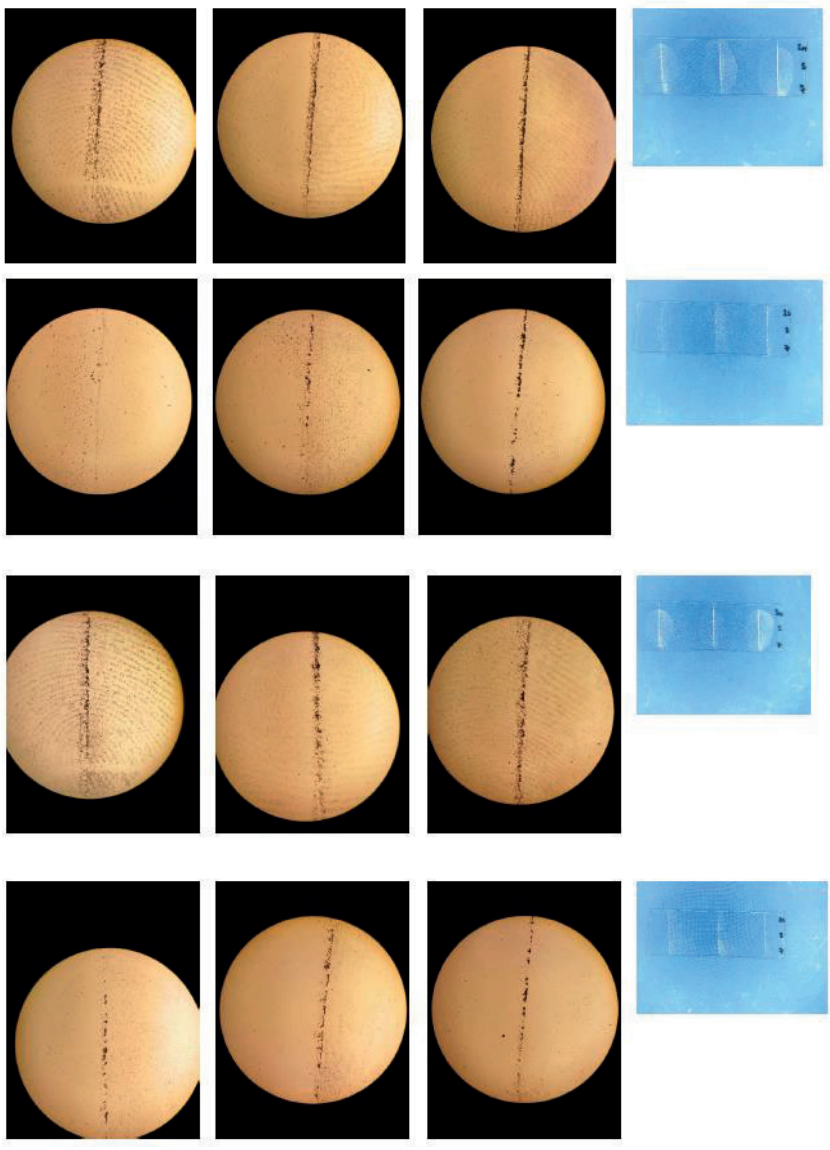






b) Bright-field contrast technique





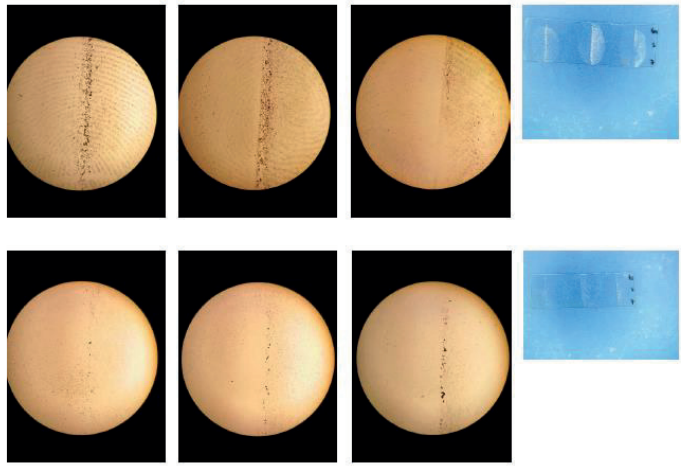
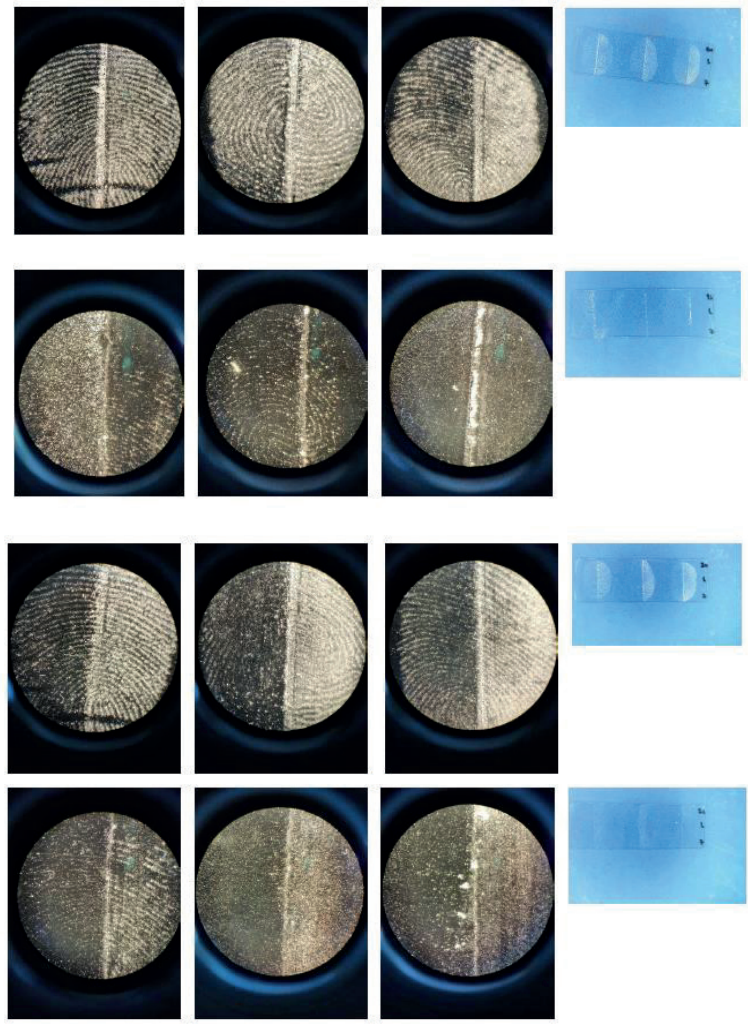


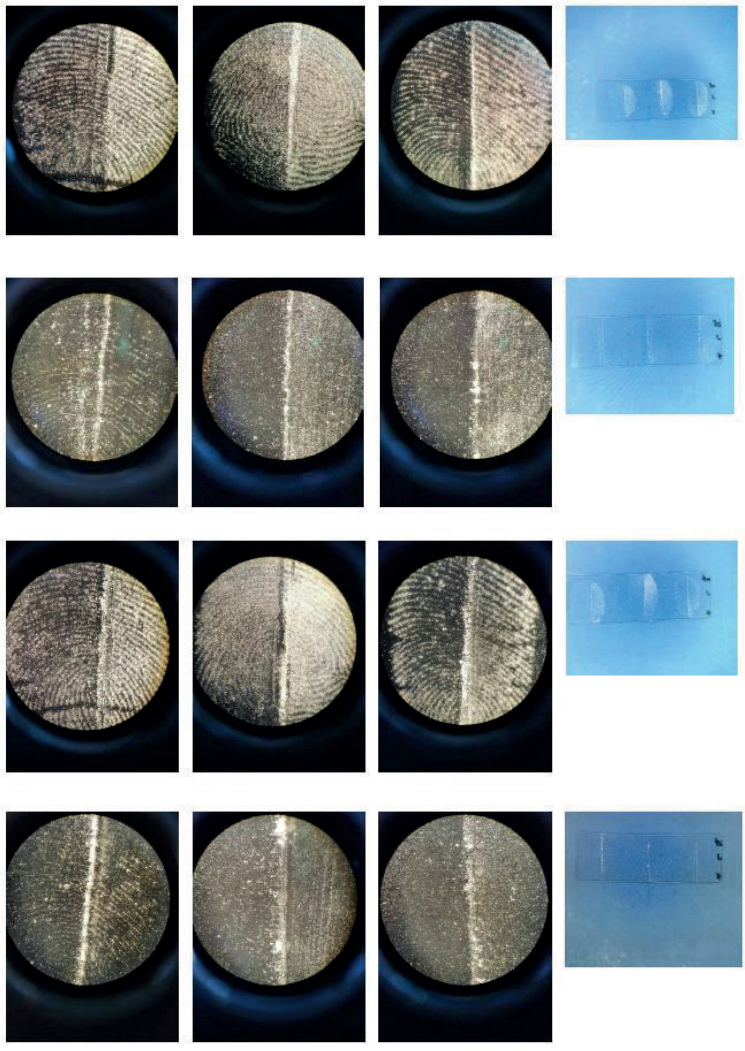
Figure S8. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under dry conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

Fingerprints developed on glass (non-porous) surface (humid (laboratory) conditions)

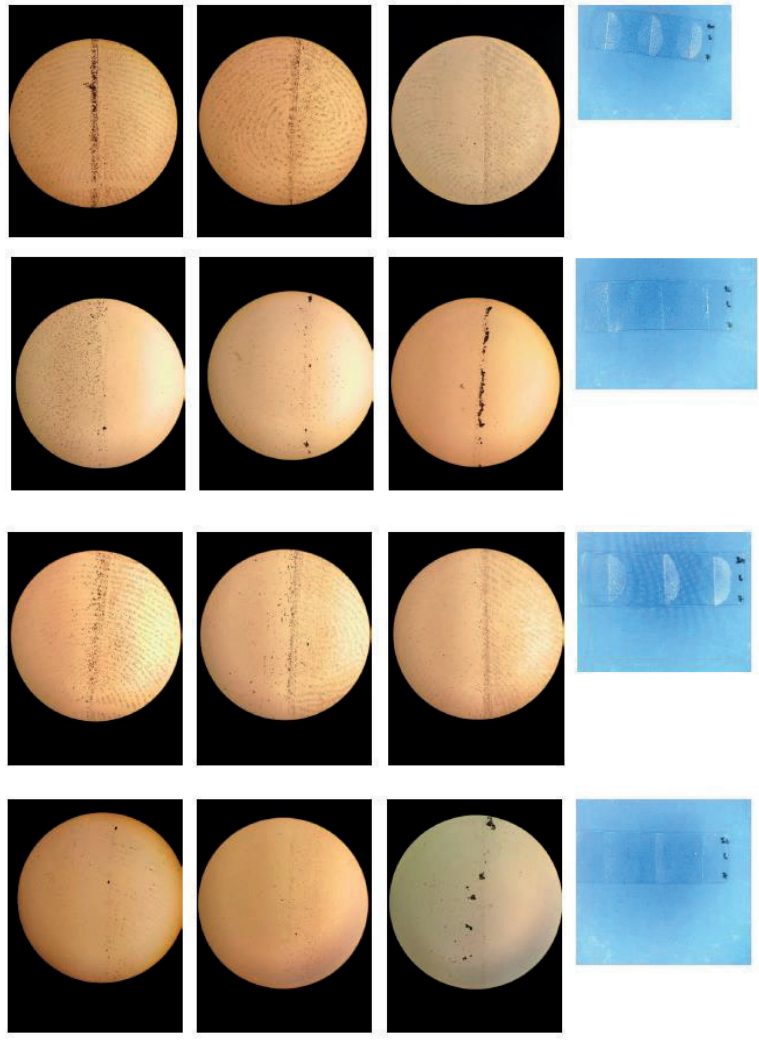
Fingerprints were recorded with the optical microscope, using:

a) Dark-field contrast technique





b) Bright-field contrast technique



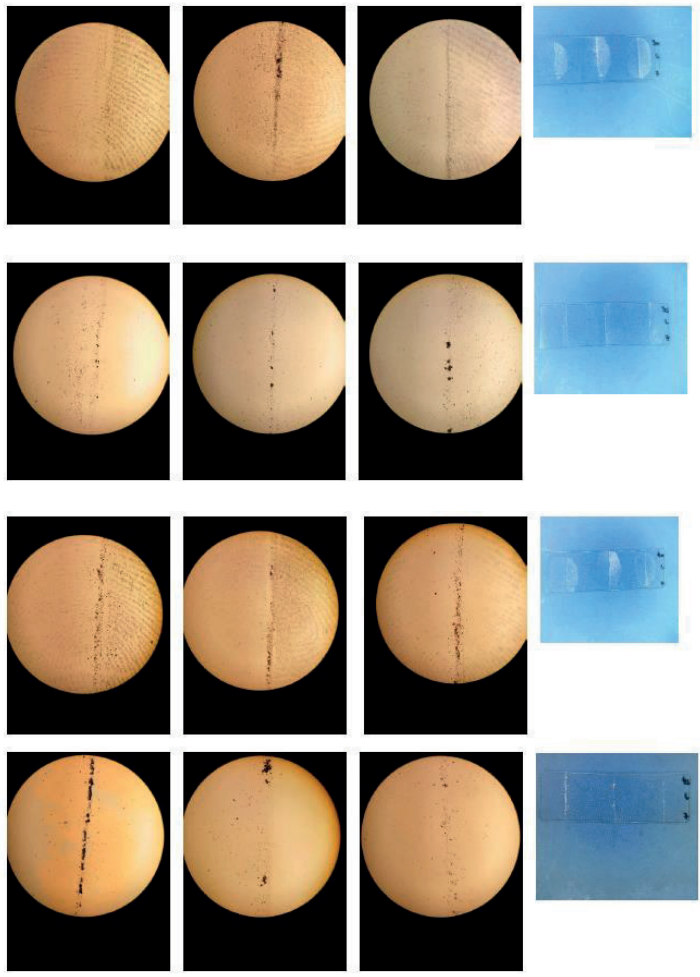


Figure S9. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under humid conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

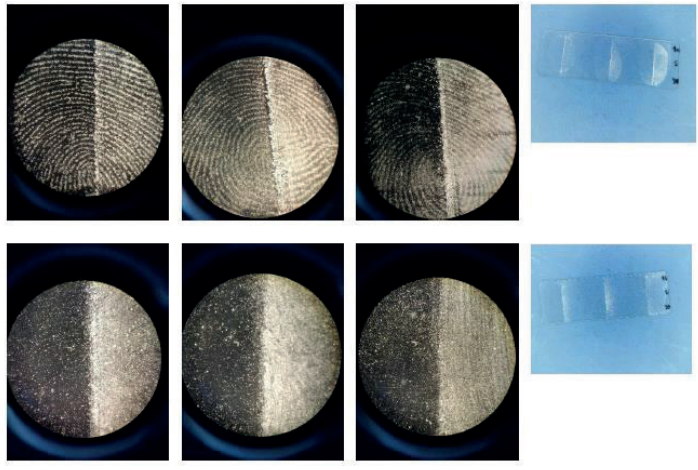
Development of latent fingerprints 1 month after deposition – July 6, 2019

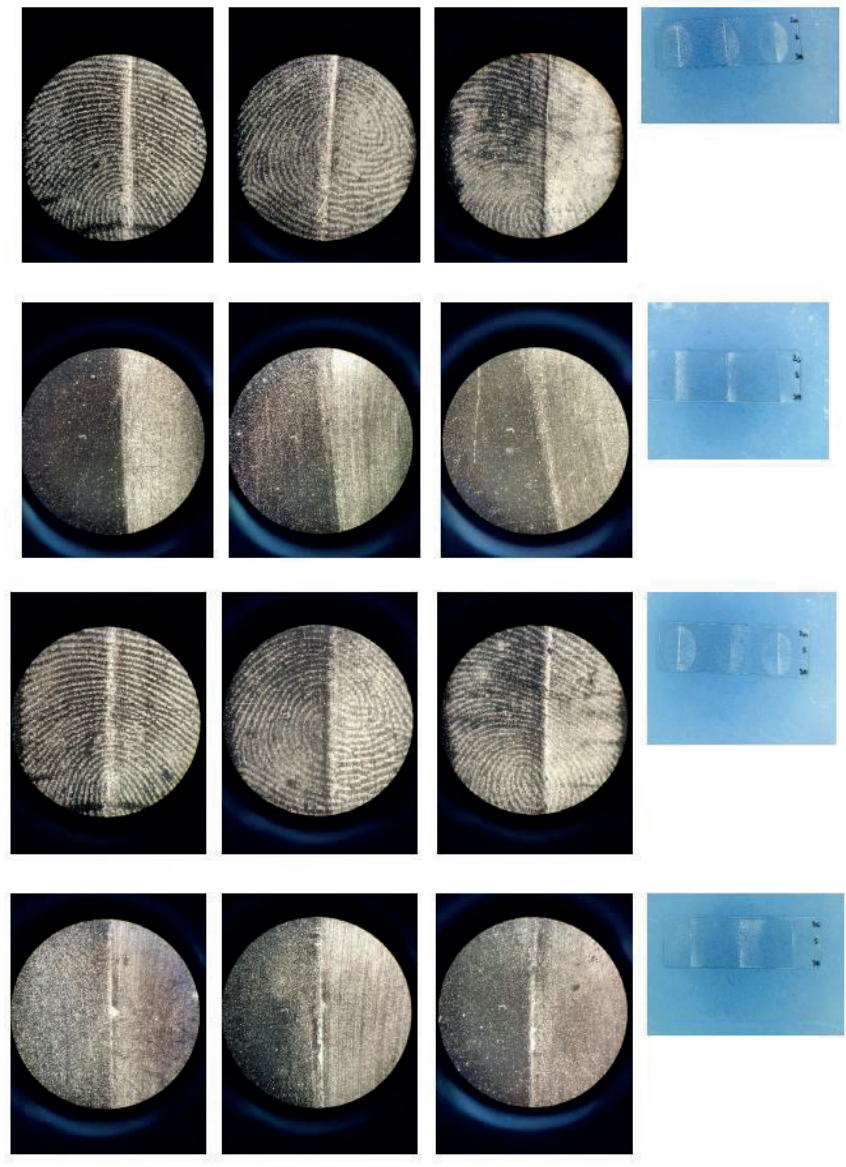
Sebaceous and dry fingerprints of three different donors, kept under dry (desiccator) and humid (laboratory) conditions, developed 1 month after deposition, with samples S(1/0/0), S(1/1/0), S(1/0/1), S(1/1/1) and pure dextran powder (as a control sample). Figures S10 and S11 represent prints on glass (non-porous) surface kept under dry and humid conditions, respectively, recorded using dark-field (Figures S10 and S11, a)) and bright-field (Figures S10 and S11, b)) contrast techniques. In the far right column all developed fingerprints photographed using a magnifying glass (magnification ×5) with a 12 MP camera (aperture *f*/2.2, pixel size 1.22 μm) were shown.

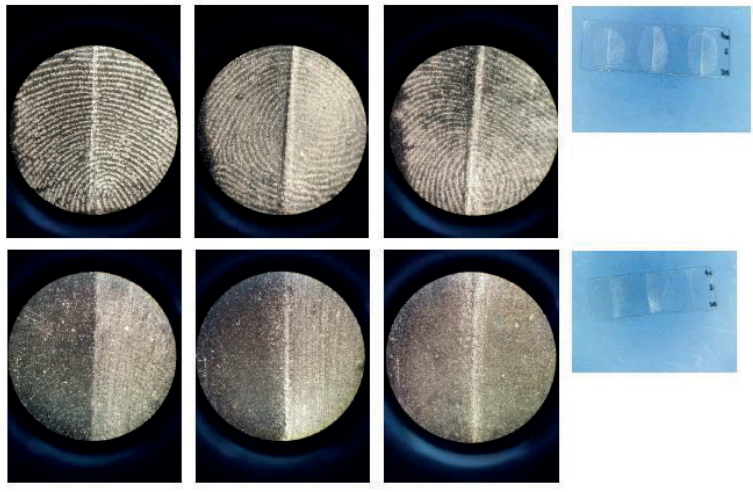
Fingerprints developed on glass (non-porous) surface (dry (desiccator) conditions)

Fingerprints were recorded with the optical microscope, using:

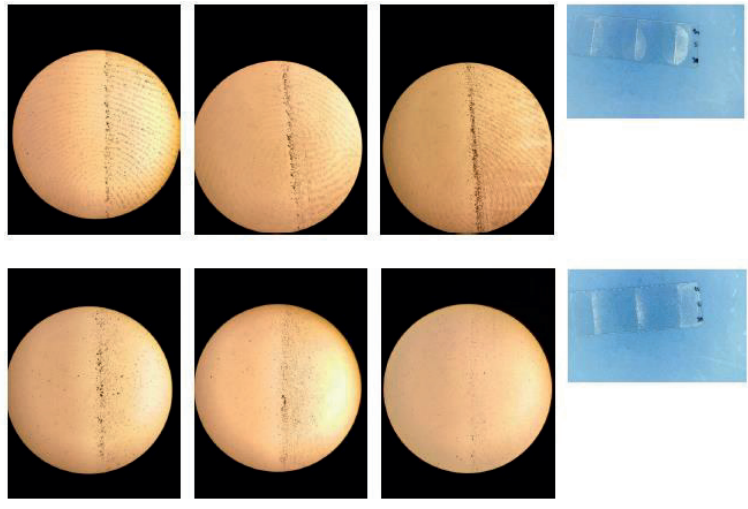
a) Dark-field contrast technique

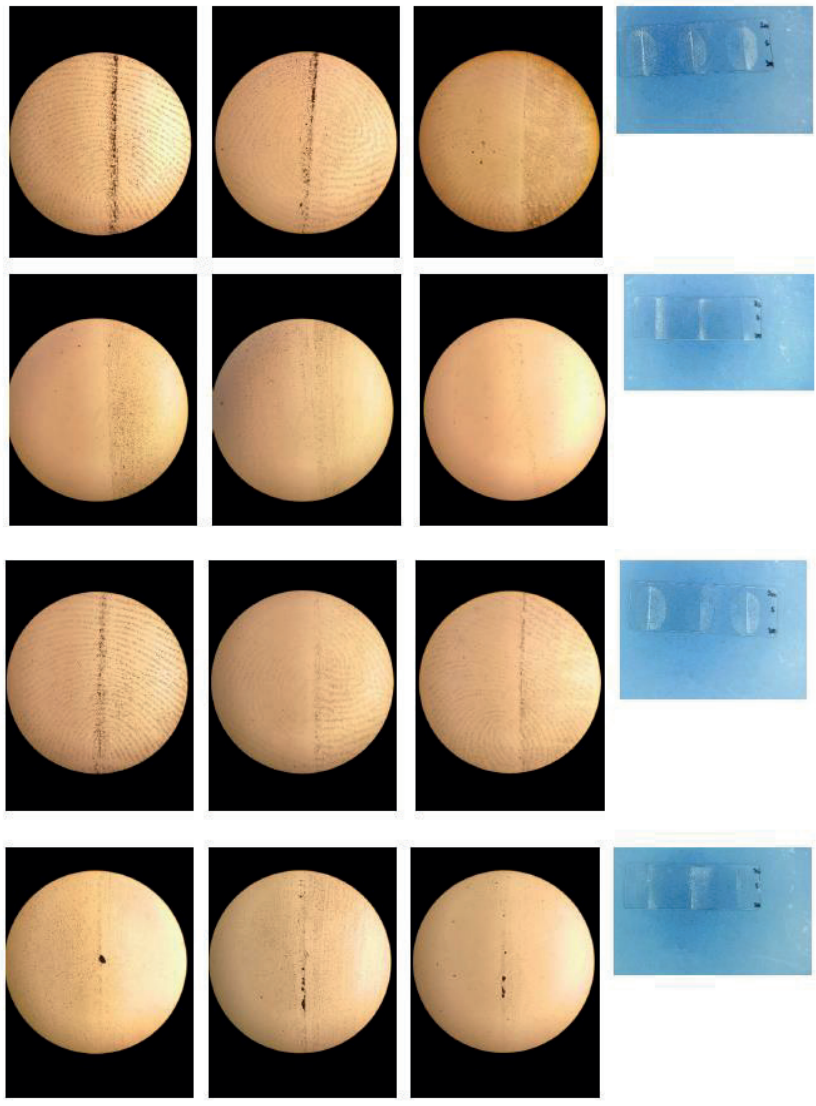






b) Bright-field contrast technique





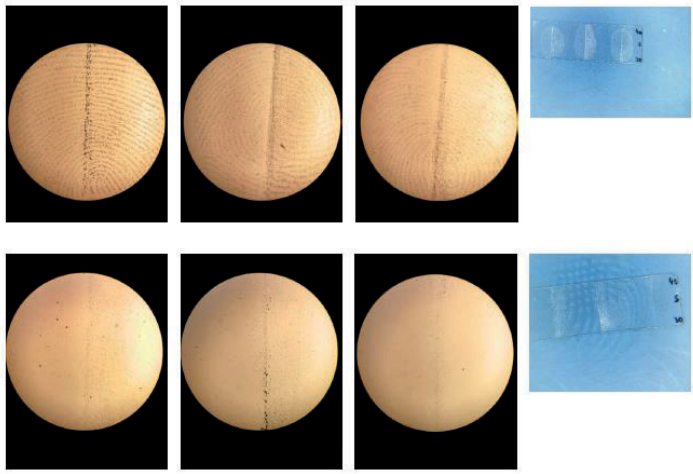
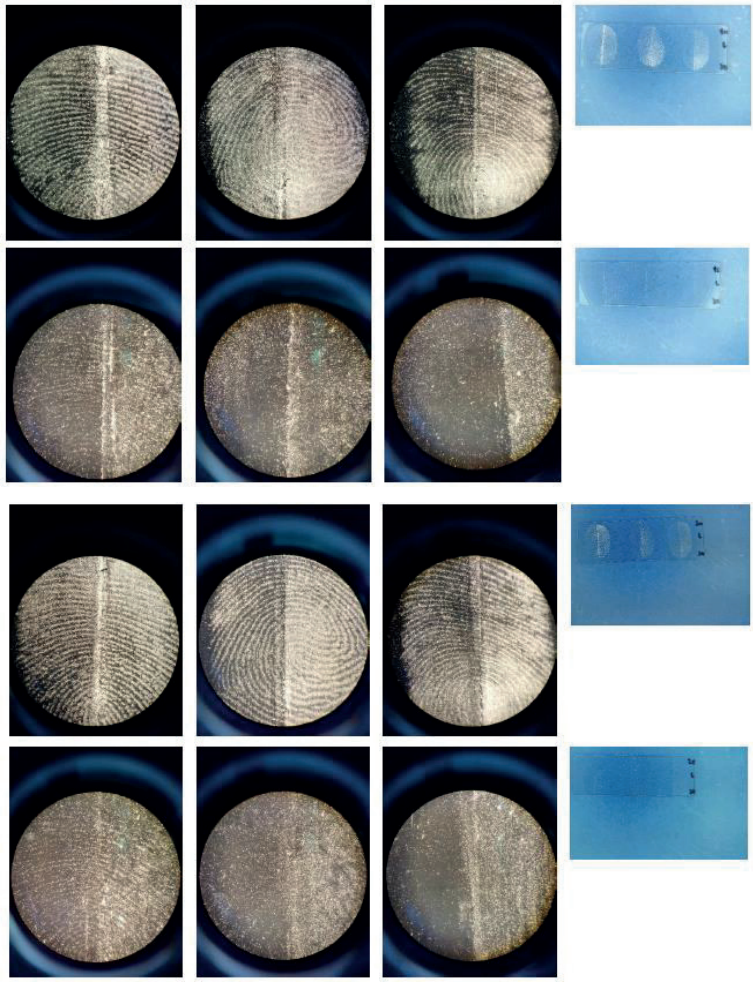


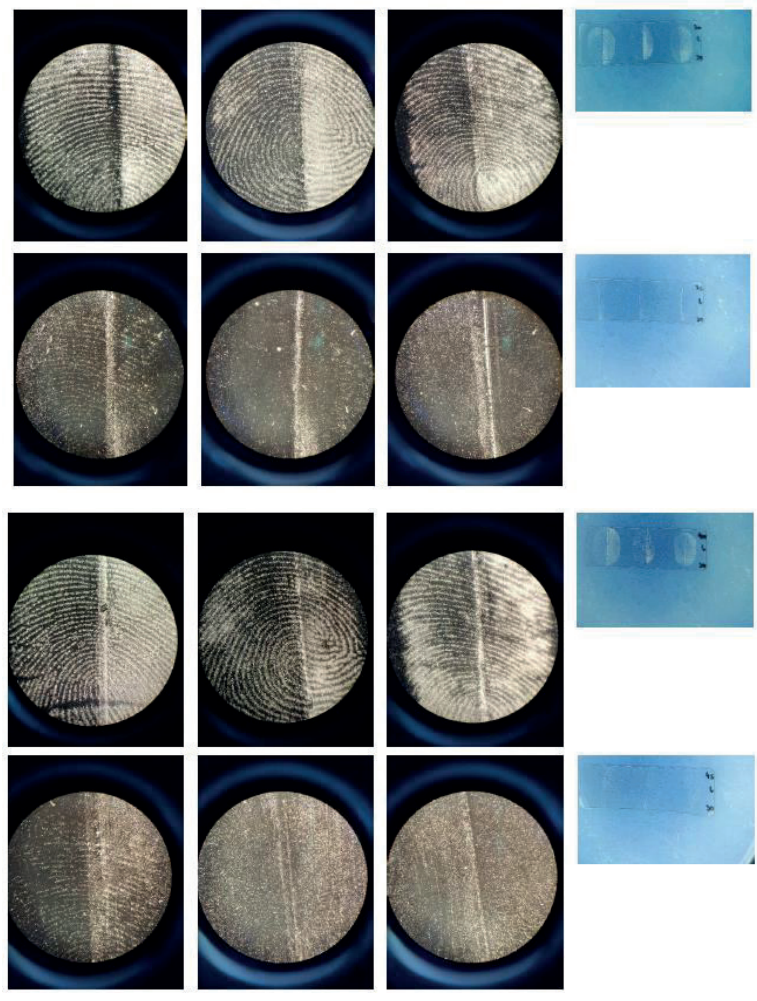
Figure S10. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under dry conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

Fingerprints developed on glass (non-porous) surface (humid (laboratory) conditions)

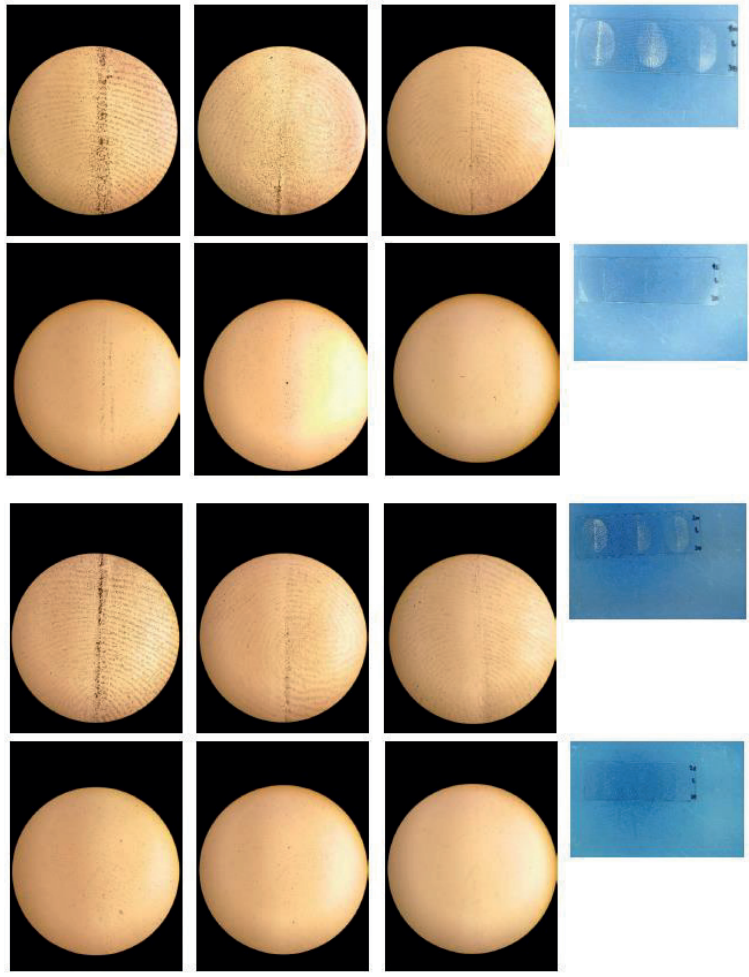
Fingerprints were recorded with the optical microscope, using:

a) Dark-field contrast technique





b) Bright-field contrast technique



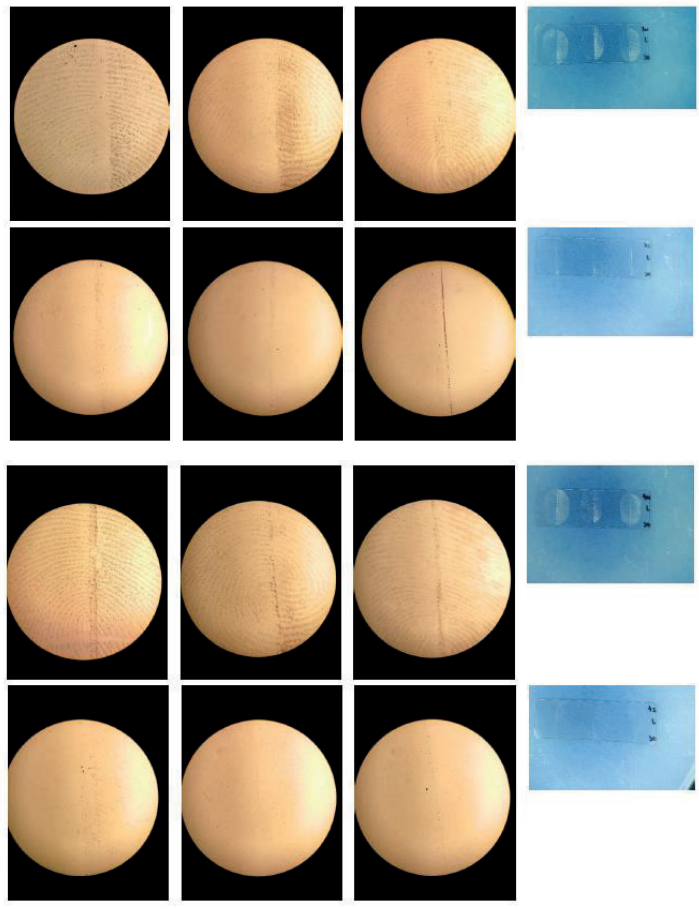


Figure S11. Sebaceous and dry fingerprints on glass (non-porous) surface, kept under humid conditions, developed with aforementioned powdered samples, then recorded using: a) dark-field and b) bright-field contrast technique.

Conclusions

Fingerprints developed on porous (paper) and semi-porous (rubber) surface

As already stated, when developing latent fingerprints on white paper surface with prepared powders, it was not possible to accomplish satisfactory contrast between the fingermark and the background, due to the white powder color. Additionally, visualization of fingerprints on rubber surface was poor, since that surface contains many bulges and indentations, thus disabling the binding of the prepared micropowders to the fingerprint residues. Therefore, additional research in order to improve this system should be conducted, with the aim at expanding application of these micropowders on different surfaces.

Fingerprints developed on non-porous (glass) surface

When observing dry fingerprints, it was evident that their development was poor and satisfactory results were not obtained with none of the applied powders. On the other hand, visualization of sebaceous fingermarks was far better, since all powder samples have developed whole fingerprint image, with papillary lines and minutiae points, as well. When comparing fingerprints kept under humid conditions with those kept under dry conditions, it was possible that higher humidity enabled better adhesion of powder samples to fingerprint residues (i.e. salts) after specific period of time, and, therefore, the visualization of those marks was significantly enhanced[2]. When comparing prepared micropowders and pure dextran powder, all samples show better performances in visualizing latent fingermarks. This may be associated with smaller diameter size of prepared powders' particles, which was confirmed by optical microscopy (smaller particles better adhere and bind to sweat and lipid fingerprint residues). Additionally, when applied with a brush, prepared powdered samples bonded with fingerprint residues and did not remain in the interpapillary space, when compared with the control sample (where "overfilling" of the traces was noticeable). When comparing prepared micropowders, sample S(1/0/0) showed as good results as S(1/1/1) and even better results than samples S(1/1/0) and S(1/0/1), which was very promising, since that sample contained only dissolved dextran powder precipitated with methanol, without initiator and/or crosslinker. It is already well known that KIO_4 and MBA showed toxic, detrimental and irritating effect, while MBA is also potentially carcinogenic[3,4]. Therefore, very satisfying visualization of sebaceous fingerprints was achieved on glass surface, and with relatively cheap and less harmful dextran-based powdered system.

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