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Exploring sustainable forensics: silica nanoparticle powder derived from rice husk waste for aged fingermark development and the chemistry of surface interactions

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Abstract

Background Powder-based fingermark ridge visibility enhancement is a common technique employed in crime scenes due to ease of application and robustness of the application method. Pigmented powders created a contrast between the surfaces and developed ridgelines and are generally metals or metal oxides based. Previous research showed the successful development of fresh latent fingermarks using rice husk-derived silica nanoparticles on various surfaces. Nevertheless, there has been less previous evidence for the efficiency of the silica nanoparticle powder on aged fingermark development. Therefore, the aim here is to investigate the efficacy of the powder on aged fingermarks relative to commercial formulation to gauge the feasibility of having naturally derived powder as a possible alternative for field application and commercialisation.

Results Rigorous testing over a range of non- and semi-porous surfaces at varied ageing conditions revealed a minimal disparity in the performance of both powders on most surfaces. Still, silica nanoparticles exhibited superiority in terms of selectivity on silica-based surfaces. Close up analysis of developed fingermarks using electron microscope exposed clear demarcation between fingermark ridges and valleys using silica nanoparticles.

Conclusions Findings revealed that the interaction chemistry between powder particles and surface material either enhances or lowers the fingermark development capacity depending on the type of surface tested.

Keywords Green forensics, Fingerprint, Silica nanoparticle, Rice husk, Green chemistry, Sustainability

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Background

Despite the paramount value of fingermarks as an identification tool, the recovery rate of latent fingermarks found in crime scenes is not at a satiable level. Loss of fingermarks may be attributed to the destructive environment such as human acts, arson, explosion, rain and natural conditions of ageing (Dhall and Kapoor 2016). Fingermark developing products that are currently commercialised pose several disadvantages, including high cost, health hazard and non-specific interaction with ridge residue, and require the use of one brush for each powder and environmental contamination (Lee and Gaensslen 2012; Daluz 2015).

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The effectiveness of fingermark development heavily relies on the size and shape of the particles of the fingerprint powder. Smaller and rounded particles have better adherence and create a uniform layer on the ridge than large coarse particles. However, most commercial formulation still utilises fine particles in size range of one to ten micrometres (Sodhi and Kaur 2001). These particle does not have any specific affinity for fingermark residue, and this characteristic leads to non-specific interaction between fingermark residue and the particles (Becue et al. 2009).

Additionally, the powders impose health risks to the user through inhalation and secondary exposure from contaminated clothing, which was claimed to be more harmful. The health hazard from the long-term exposure to fingerprint dusting powder is caused by trace metals that lead to heavy metal toxicity (Netten et al. 1990; Maynard 2011). Topical exposure to fingerprint dusting powders can also cause skin rashes and visual impairment in the long run (Souter et al. 1992).

Nanoparticle-based techniques are not a new sight in forensic fingerprinting. Many techniques have been invented in the past, and continuing efforts have been undertaken to optimise these techniques. Ranging from multi, single and vacuum metal deposition methods, the application of nanoparticles in forensic has been growing steadily, serving as a forerunner breaking forensic grounds with nanotechnology (Ferreira et al. 2021). Functionalised nanoparticle for targeted approach has been experimented with to increase the precision of ridge development (Azman et al. 2018). These techniques offer an excellent last resort option for fingermark recovery but have limited field application owing to stringent application conditions. The precursors used for the product synthesis is significantly more costly than the commercial products available now in the market and constraints technique's robustness (Komalasari et al. 2017).

Every year, a billion tonnes of rice husk are disposed of as waste despite being recycled into livestock and other industries (Kumagai and Sasaki 2009; Rafiee et al. 2012; Ghorbani et al. 2013). Rice husk is a rich source of silica, and synthesising silica from rice husk is considerably sustainable compared to the high energy-intensive methods employed on an industrial scale. Industrial silica synthesis generally makes use of alkoxide precursors such as tetraethylorthosilicate (TEOS) and phenyltriethoxysilane (PTEOS) (Kongmanklang and Rangsriwatananon 2014). Synthesis of silica nanoparticles from agricultural waste such as rice husk, bamboo leaves, corn cobs and coconut shells have been researched, and rice husk contains the highest amount of silica nanoparticles (Noushad et al. 2012, 2014; Vaibhav et al. 2015; Pattanayak et al. 2022).

Sustainability has become a significant focus in recent years, majorly emphasising a circular economy. In line with global sustainability goals, the potentiality of rice husk waste to be upscaled into silica nanoparticles for latent fingermark development has been previously shown (Rajan et al. 2018; Nik Hassan et al. 2020). The proof-of-concept established that the spherical small-sized silica nanoparticle could develop a fingermark with somewhat improved selectivity. However, the study was limited to the development of fresh, natural fingermarks from three donors, serving as preliminary findings to support the ability of silica nanoparticles from rice husk to develop fingermarks on various surfaces (Rajan et al. 2020). The ability of the powder to develop fingermarks on various surfaces with ageing and depletions remains unexplored.

In the present study, we explored to gain comprehensive information on the performance of the powder on aged and depleted fingermarks deposited on various surfaces. This information points out the advantages and practicability of commercially using rice husk-derived silica nanoparticles, considering that fingermarks found in crime scenes are rarely fresh. Surface testing was performed in two phases to determine the interaction of the powder with fingermark residue, followed by testing the sensitivity of the powder. A variety of surfaces were selected, and fingermarks deposited on these surfaces were artificially aged under several conditions, depleted and developed using silica nanoparticles. The study revealed that the performance of the powders was not only influenced by the ageing factor and porosity of the surfaces but also the material which the surfaces were made out of. Therefore, in this study, we attempt to understand the chemistry of the surface interaction that could potentially help investigators to understand and apply the best fingermark developing powder tailored to each surface.

Methods

Rice husks (RHs) were obtained locally from Bernas in Peringat, Malaysia. Hydrochloric acid and sodium hydroxide (Merck), acetone and glacial acetic acid (Sigma-Aldrich), as well as absolute ethanol (HmbG® Chemicals), were of high purity grade and were used without further purification. HI-FI Volcano Latent Print Indestructible white powder (No.103L) was purchased from SIRCHIE Youngsville, NC, and used without modification.

As mentioned above, silica nanoparticles were synthesised from rice husk according to a published protocol (Rajan et al. 2020) as described below. The rice husk was washed and dried before being blended into a fine powder. The powder was acid digested, thoroughly washed and ashed at 700 °C for 5 h in the furnace. A total of 0.5 g ash was dissolved in 100 mL sodium hydroxide (4% w/v) to form sodium silicate, then aged in a hot air oven at 90 °C overnight to grow seed particles. The seed particles' growth was accelerated by gradually adding acetic acid (30% w/v) under strong stirring after the original concoction was volume adjusted to 100 mL and 40 mL of acetone added to it. A few drops of phenolphthalein were added to indicate pH change. The colour of the solution turned from pink to colourless, while colloid formation was observed just before the neutralisation point. The precipitated particles were separated by centrifugation, washing and freeze-drying.

Fingermark samples were collected by considering five different non-porous substrates, five semi-porous substrates, one to thirty-nine different donors for some study phases and some marks under accelerated ageing conditions. The combination of all these parameters allowed us to process a total of 573 different samples (see Table 1 for details). The donors were asked not to touch their face for natural fingermarks at least 10 min before touching the substrates. For eccrine fingermarks, the donors

Table 1 Details of the 573 samples that were considered during this study, in terms of substrate nature, donor, secretion type and ageing conditions. For each condition, triplicates were evaluated, which were further cut in half, vertically or horizontally, to compare the effectiveness of the two different powders

Surface type	Sample #	Substrate	Donor	Secretion type	Ageing conditions
Phase I – interaction of powder with different secretion types					
Non-porous	1–3	Glass	D/1	Eccrine	Fresh
	4–6	Glass	D/2	Eccrine	Fresh
	7–9	Glass	D/3	Eccrine	Fresh
	10–12	Glass	D/1	Sebaceous	Fresh
	13–15	Glass	D/2	Sebaceous	Fresh
	16–18	Glass	D/3	Sebaceous	Fresh
	19–21	Glass	D/1	Natural	Fresh
	22–24	Glass	D/2	Natural	Fresh
	25–27	Glass	D/3	Natural	Fresh
Phase II – multiple donor studies					
	28–63	Glass	D/4–D/39	Natural	Fresh
Phase III – accelerated ageing studies					
Non-porous	64–84	Metal	D/1	Natural	Fresh and aged
	85–105	Acrylic PVC	D/1	Natural	Fresh and aged
	106–126	Glass	D/1	Natural	Fresh and aged
	127–147	Soda bottle	D/1	Natural	Fresh and aged
	148–168	Tiles	D/1	Natural	Fresh and aged
Semi-porous	169–189	Painted wood	D/1	Natural	Fresh and aged
	190–210	Glossy card	D/1	Natural	Fresh and aged
	211–231	Black tape	D/1	Natural	Fresh and aged
	232–252	Keyboard	D/1	Natural	Fresh and aged
	253–273	Leather	D/1	Natural	Fresh and aged
Phase IV – depletion studies					
Non-porous	274–303	Metal	D/1	Natural	Fresh
	304–333	Acrylic PVC	D/1	Natural	Fresh
	334–363	Glass	D/1	Natural	Fresh
	364–393	Soda bottle	D/1	Natural	Fresh
	394–423	Tiles	D/1	Natural	Fresh
Semi-porous	424–453	Painted wood	D/1	Natural	Fresh
	454–483	Glossy card	D/1	Natural	Fresh
	484–513	Black tape	D/1	Natural	Fresh
	514–543	Keyboard	D/1	Natural	Fresh
	544–573	Leather	D/1	Natural	Fresh

were asked to wash their hands under running tap water and air dry before touching the substrates. Sebaceous fingermarks were collected similarly, but donors were requested to charge their fingers by touching the sides of their nose, then homogenise the secretions by rubbing their fingers together before touching the substrate. All fresh fingermarks were left at room temperature for an hour before development. *Phase II* involved testing natural fingermarks from 36 donors of both genders with varied ages.

In this study, accelerated ageing conditions were employed to artificially age fingermark samples, except for fresh fingermarks, in *Phase III* at two different temperatures (50 and 100 °C) for the duration of 1, 2 and 3 h, respectively. The exposure to a high level of heat will simulate the aged fingermarks found in the crime scene. Still, authors were cautious about accounting for the heat degradation of certain compounds that otherwise may be present in naturally aged fingermarks.

Each half of the fingermarks was developed using a SIRCHIE squirrel hairbrush by gentle stroking in one direction. The fingermarks were then reassembled from their halves and photographed using a Canon Powershot SX60 HS with 16.1 megapixel resolution using the microfocus mode set at autofocus mode. The digital images of the fingermarks were then scored using the scale shown in Table 2 adapted from Fieldhouse et al. (2016).

Scores from the silica nanoparticle powder (SNP) and white powder (WP) efficiency studies were analysed using the Wilcoxon signed-rank test to determine if there were significant differences between fingermarks developed using both powders. Wilcoxon signed-rank test was employed given that the dependant variable was ordinal data and the independent variable was matched pairs of fingermarks. Data were tested for symmetry before analysis. All statistical analyses were carried out using IBM SPSS Statistics 22.

Results

SNP powder and its characterisation studies

SNP powder worked well on most surfaces tested except hydrocarbon-derived surfaces such as poly vinyl chloride (PVC), keyboard (acrylonitrile butadiene styrene (ABS)), wood varnish, man-made leather and vinyl/PVC tape, while titanium dioxide powder (SIRCHIE) gave better development on these surfaces. The synthesised SNP powder was very fine and smooth, and the physicochemical characteristics were further investigated to determine that the powder produced is in agreement with previous findings (Rajan et al. 2020). Figure 1 displays the field emission scanning electron microscopy (FESEM) images of SNP and WP particles. The primary particle size of the working formulation obtained for SNP and WPs were ~ 270 nm and ~ 400 nm, respectively, consistent with the previous report (Rajan et al. 2020). The nanostructure of the SNP and WPs were compared using FESEM to examine the microscopic differences between these two powders. SNP particles appeared spherical and existed primarily as individual particles, while WP particles appeared as large clusters of smaller highly porous particles. Elemental analysis using energy dispersive X-ray spectroscopy (EDX) indicated the presence of oxygen and silicon at the approximate atomic ratio of 69 to 31%. There were no other elements traced confirming the purity of the powder. SNP powder was used for fingermark development without any modification.

Phase I: establishing the interaction of fingermark residue and SNP powder

Figure 2 exhibits the eccrine, sebaceous and natural fingermark from three volunteer donors developed using SNP and WPs. The three donors with different levels of fingermark residue (weak, medium and heavy) were selected to observe the interaction of the powder particle with various types of donors. Donors were requested to deposit several natural fingermarks that was developed using commercial powder. Weak donors had faint fragmented ridges; medium donor fingermark had continuous ridge patterns while the developed fingermarks from heavy donors had halo-like ring around the ridges indicating over secretion of fingermark residue. Results indicated that the latent fingermarks from different secretions were developed with different degrees of clarity. However, all the fingermarks developed had identifiable

Table 2 Fingermark grading scale system

Grade	Description
0	Fully smudged outline of a mark or no evidence of the mark
1	The presence of several ridges cannot lead to the identification
2	The major part of the mark is smudged, several ridge details are present, and analysis cannot be performed
3	A minor part of the mark is smudged, most ridge details are visible, and analysis can be performed
4	Full mark and ridge details are clearly visible; some ridgelines may be thinned or smudged but identifiable marks
5	Full mark and all ridge details are clearly visible; identifiable mark

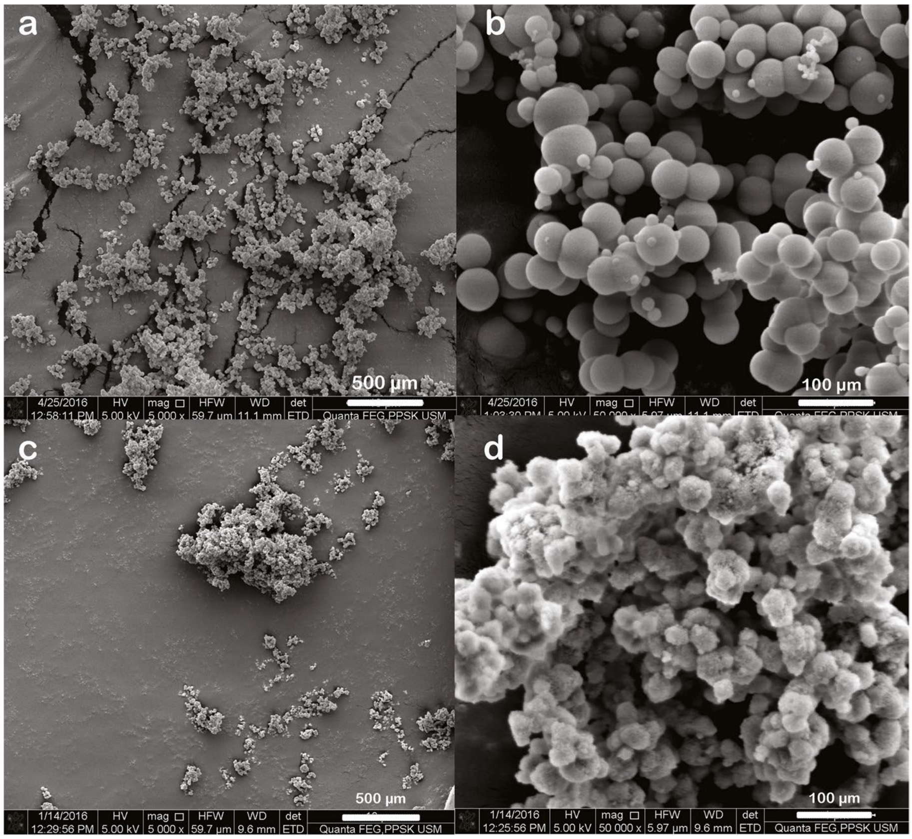

Fig. 1 SEM images SNP (**a** and **b**); SIRCHIE WP (**c** and **d**). Dispersed clusters of SNP in image (**a**) are consistent with this powder's better fingermark development capacity, while the WP particles appear to be agglomerated microstructures

ridge pattern. As expected, the powder particles were heavily distributed along the ridges of the sebaceous fingermarks and natural fingermarks, especially fingermarks of donor 2 which appeared to be highly sebaceous. Both powders have similar interaction with the ridge residue, indicating that on this surface the interaction was mainly physical adhesion of powder to the fingermark residue, and is independent of the different constituents due to the mixture of skin secretions.

Results also showed that the fingermarks developed using SNP powder was lower in contrast in comparison to the fingermark developed using WP. However, SNP exhibited increased selectivity to fingermark ridge residue as compared to WP, as such produced high clarity fingermark. WP adhered non-specifically to substrate bearing the fingermark and fingermark residue, reducing clarity. Therefore, the key difference in both powders is the selective adsorption of the particle to the ridges.

The decrease in powder's particle size increased the surface area and indirectly the number of molecules that was bound to the fingermark deposit. However, powder particle agglomeration also played a critical effect in reducing the ridge details clarity. Natural fingermark of donor 1 (medium donor) developed using commercial and proposed powder was closely inspected using FESEM (Fig. 3). The latent fingermark developed using SNP powder showed excellent ridge details with minimal background staining and better results as compared to the earlier reports due to its nanosized, minimal agglomeration and different synthetic procedures (Fig. 3a, b). Previous report suggested that silica particle of the size range 45 to 63 μm gave best results of fingermark development with the powder dusting technique (Theaker et al. 2008). Nevertheless, our findings indicated better ridge resolution achieved using 200–400 nm sized spherical silica particles. On the other hand, WP, regularly used white latent fingerprint powder composed of titanium oxide, not only adhered to the fingermark valley but also diminished the ridge details of fingermarks (SIRCHIE 2012). Excessive powdering diminishes the clarity of the minutiae visibility (Fig. 3c, d). This non-specific interaction of the WP originated presumably due to the sticky nature of the resin incorporated titanium oxide formulation.

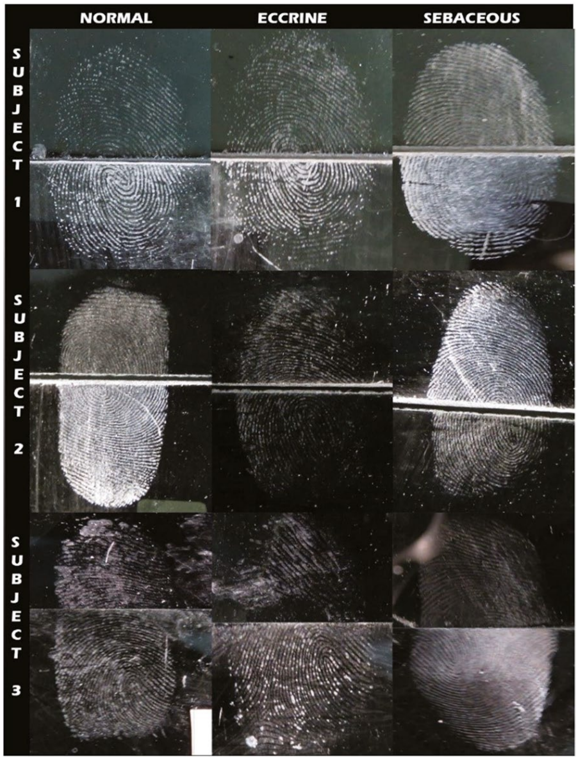

Fig. 2 Latent fingermarks of natural, eccrine and sebaceous origin from three different donors: (**a**) upper half developed using SNP, (**b**) lower half developed using WP powder. Both halves of the fingermark developed in similar manner; nevertheless, WP-developed portion shows signs of over-powdering

Phase II: SNP powder sensitivity determination studies

Multiple donor studies

Figures 4 and 5 present natural fingerprints from a diverse group of thirty-six donors, encompassing both genders and varying ages and racial backgrounds. These fingerprints were developed using SNP and WP powders, respectively. The findings in Table 3 illustrate the outcomes of Wilcoxon signed rank tests conducted on the grades assigned to the fingerprints in surface test experiments.

The statistical analysis conducted on these multi-donor studies revealed that there was no statistically significant distinction in the contrast and clarity of the fingerprints produced by SNP and WP. The performance of both powders showed no significant differentiation (p-value > 0.05), with a median score of 5.0 for WP and 4.0 for SNP. This result can be attributed to the higher contrast achieved through WP, which enhanced the visibility of ridges. However, it is important to note that SNP-developed prints still exhibited identifiable ridges, albeit with slightly reduced ridge visibility.

Depletion and split studies

Figure 6 exhibits the depletion study conducted on black glass and glossy card surfaces, an example of one of the most difficult surface owing to higher porosity and pigmented surface. Analysis of the grading score from the depletion study done on all ten surfaces revealed that SNP powder consistently performed as well as the WP (Fig. 6a, b) and, in some cases, better, except on the painted wood surface (Fig. 6c). SNPs and WPs, being white in colour, displayed excellent visibility and strong contrast when placed on dark surfaces. A Wilcoxon signed-rank test was conducted to assess the effectiveness of both SNP and WP powders in a fingerprint analysis study. The results from the Wilcoxon signed rank test revealed that, with the exception of tiled surfaces, there was no statistically significant difference (p-value > 0.05) in the quality of fingerprint development between the two powders.

However, it is worth noting that SNP powder, with a median score of 4.0, yielded significantly clearer fingerprint results than WP powder, which had a median score of 3.0 ($Z = -1.968$, $p = 0.049$). This improved clarity in fingerprints was particularly noticeable on slightly rough, non-porous surfaces, where the SNP powder demonstrated superior particle adhesion to the fingerprint residues.

Accelerated ageing studies

The findings presented in Fig. 7 **s**howcase the outcomes of fresh fingermark development using both powders on all ten different surfaces. It is evident that over-application of powder is noticeable in the fingermarks developed with both powders, particularly on non-porous surfaces. This excess powder can be attributed to heightened interaction between the powder and residue. In general, the fresh fingermarks contain a significant amount of sebaceous residue, which leads to increased interaction but lowers the clarity of the developed fingermark ridges.

Figures 8 to 9 depict the findings of the accelerated ageing study conducted on the ten surfaces. Notably, as the ageing temperature experiences a slight increase, there is an improvement in the clarity of the developed fingermarks on non-porous surfaces. However, this clarity starts to decline as the temperature and ageing time are further increased. Conversely, the fingermarks developed on semi-porous surfaces exhibited a consistent decrease in both contrast and clarity as the ageing temperature and time were increased.

The analysis of the ageing study showed that there were no significant differences in the effectiveness of WP and SNP powders, except for two specific surfaces. Notably, SNP powder significantly improved the

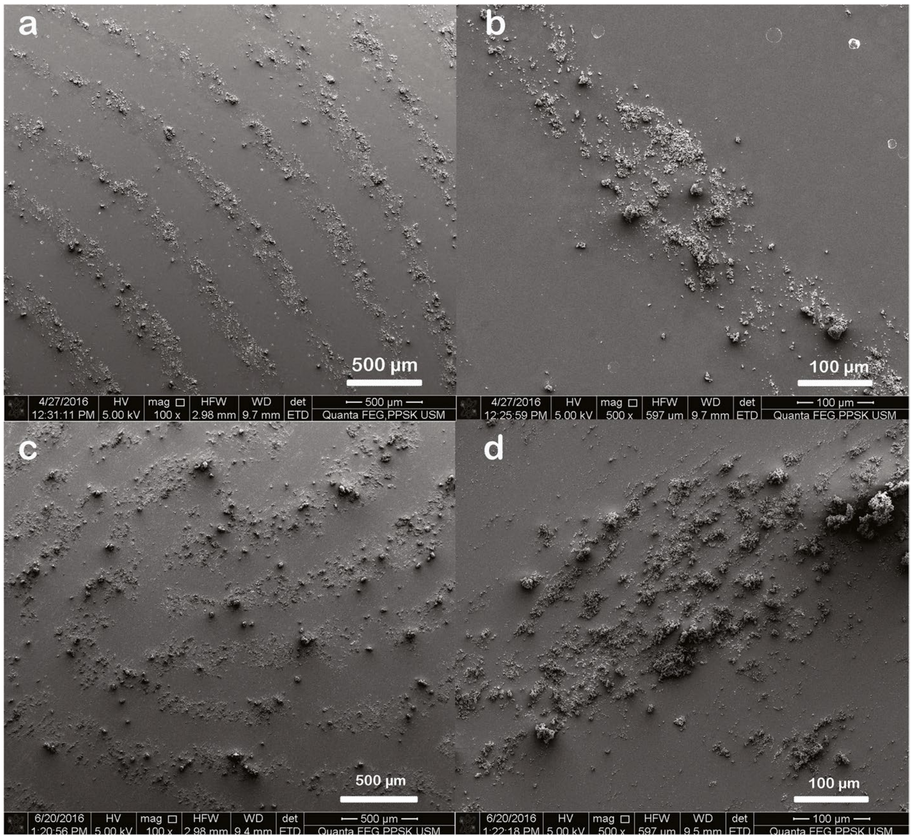

Fig. 3 SEM images of developed fingermark; SNP powder (**a**, **b**) and WP (**c**, **d**) using natural fingermark on a glass surface. In this image, there is a clear distinction between the selectivity of the powders to the ridges where the SNP exhibit clear demarcation between ridge and valley, while white powder does not

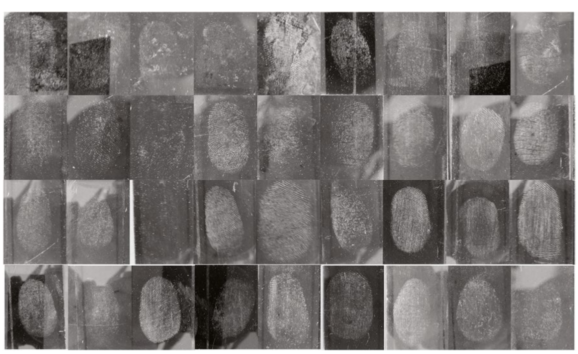

Fig. 4 Fingermarks developed using SNP powder from 36 different donors of various age, sex and race. Varied level of clarity can be observed in these marks depending on the varied fingermark residue coming from different donors

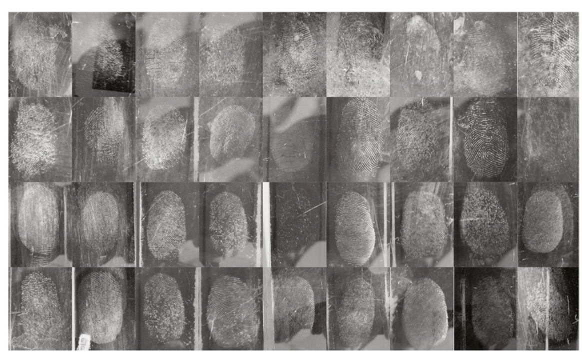

Fig. 5 Fingermarks developed using WP from 36 different donors of various age, sex and race. Compared to the SNP-developed fingermark, WP produces higher contrast in the developed marks; however, overpowdering can be observed along the ridge lines

quality of fingermarks on painted wood ($Z = -2.271$, $p = 0.023$) and soda bottles ($Z = -2.546$, $p = 0.011$) in terms of clarity and contrast. The results indicate that SNP powder performed comparably to, and in some instances, even outperformed commercial WP powder. Imaging techniques revealed that SNP powder exhibited specific and selective interactions with fingermark residue when compared to WP powder.

Table 3 Wilcoxon signed rank test results for fingermark scores in depletion and ageing studies on each surface

Material	Negative rank	Positive rank	Ties	*Sig./p* value	*Z*	*N*	Median SNP	Median WP
Multiple donor studies								
Glass	16	9	11	0.324	−0.987[b]	36	4.0	5.0
Depletion studies								
APVC	5	13	12	0.142	−1.470[a]	30	2.0	2.0
Tiles	4	11	15	0.049	−1.968[a]	30	4.0	3.0
Black glass	7	8	15	1.000	0.000[c]	30	4.0	4.0
Metal	13	8	9	0.112	−1.588[b]	30	2.0	2.0
Painted wood	15	9	6	0.213	−1.245[b]	30	3.0	4.0
Soda bottle	7	17	6	0.121	−1.550[a]	30	4.0	4.0
Black tape	5	12	13	0.089	−1.699[a]	30	0.0	0.0
Glossy card	7	7	16	1.000	0.000[c]	30	3.0	3.0
Leather	3	9	18	0.059	−1.889[a]	30	1.5	1.0
Computer keyboard	13	11	6	0.823	−0.224[a]	30	3.0	3.0
Ageing studies								
APVC	8	6	7	0.974	−0.032[b]	21	3.0	3.0
Tiles	5	1	15	0.084	−1.725[b]	21	5.0	5.0
Black glass	5	1	15	0.084	−1.730[b]	21	5.0	5.0
Metal	4	5	12	0.623	−0.491[a]	21	3.0	2.0
Painted wood	11	5	5	0.023	−2.271[b]	21	4.0	5.0
Soda bottle	11	2	8	0.011	−2.546[b]	21	4.0	4.0
Black tape	4	9	8	0.167	−1.381[a]	21	2.0	0.0
Glossy card	6	8	7	0.360	−0.915[a]	21	4.0	4.0
Leather	2	6	13	0.135	−1.496[a]	21	1.0	0.0
Computer keyboard	3	8	10	0.053	−1.935[a]	21	2.0	0.0

[a] Negative ranks: SNP < WP

[b] Positive ranks: SNP > WP

[c] Ties: SNP = WP

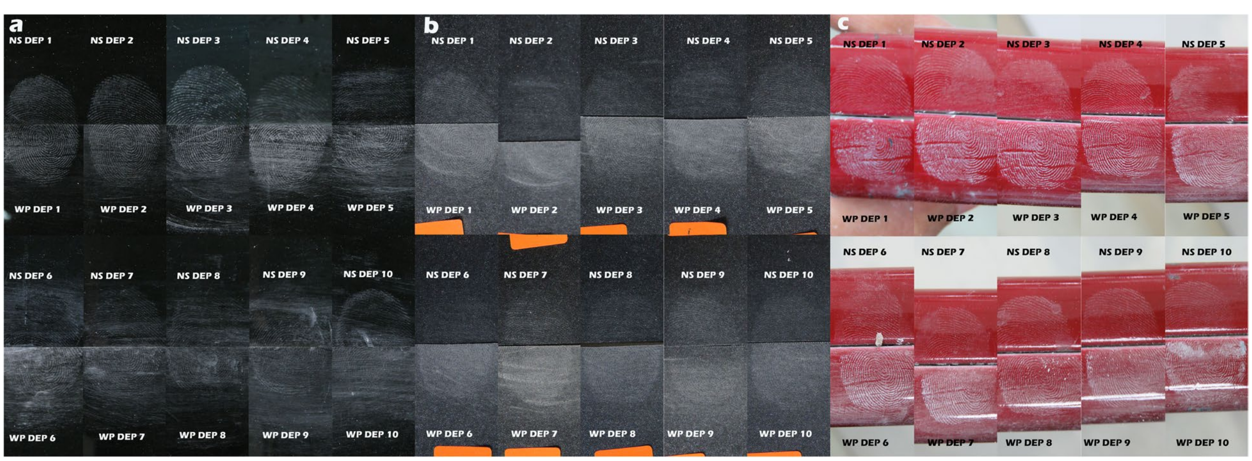

Fig. 6 Depletion studies on (**a**) black glass and (**b**) glossy surface. SNP and WP were effective to develop ridges with identifiable details up to depletion nine on black glass surface and glossy card surface

Fig. 7 Fingermarks developed on various surfaces using SNP and WPs. SNP and WP performed comparably on surfaces except WP was more effective on keyboard while SNP was better on the black tape

Discussion

In this study, our primary objectives were twofold. Firstly, we aimed to comprehensively assess the performance of silica nanoparticles in the development of latent fingermarks, particularly in scenarios involving aged and depleted fingermarks on a variety of surfaces. Secondly, we sought to evaluate the practicality and advantages of employing silica nanoparticles derived from rice husk waste in the realm of forensic science.

The findings of this research unequivocally address these objectives, providing invaluable insights that contribute significantly to the field of forensic science. We explored the use of silica nanoparticles in real-world forensic scenarios, where fingermarks discovered at crime scenes are seldom pristine. Through systematic surface testing, multiple donor studies, depletion and split studies, as well as accelerated ageing experiments, our study encapsulated a wide spectrum of conditions and scenarios that forensic experts encounter in their investigations.

Our multiple donor studies, encompassing a diverse group of thirty-six donors representing various genders, ages and racial backgrounds, yielded results of paramount importance. The statistical analysis conducted on these multi-donor studies revealed that there was no statistically significant distinction in the contrast and clarity of the fingerprints produced by silica nanoparticles (SNP) and commercial white powder (WP). The performance of both powders showed no significant differentiation (p-value > 0.05), with a median score of 5.0 for WP and

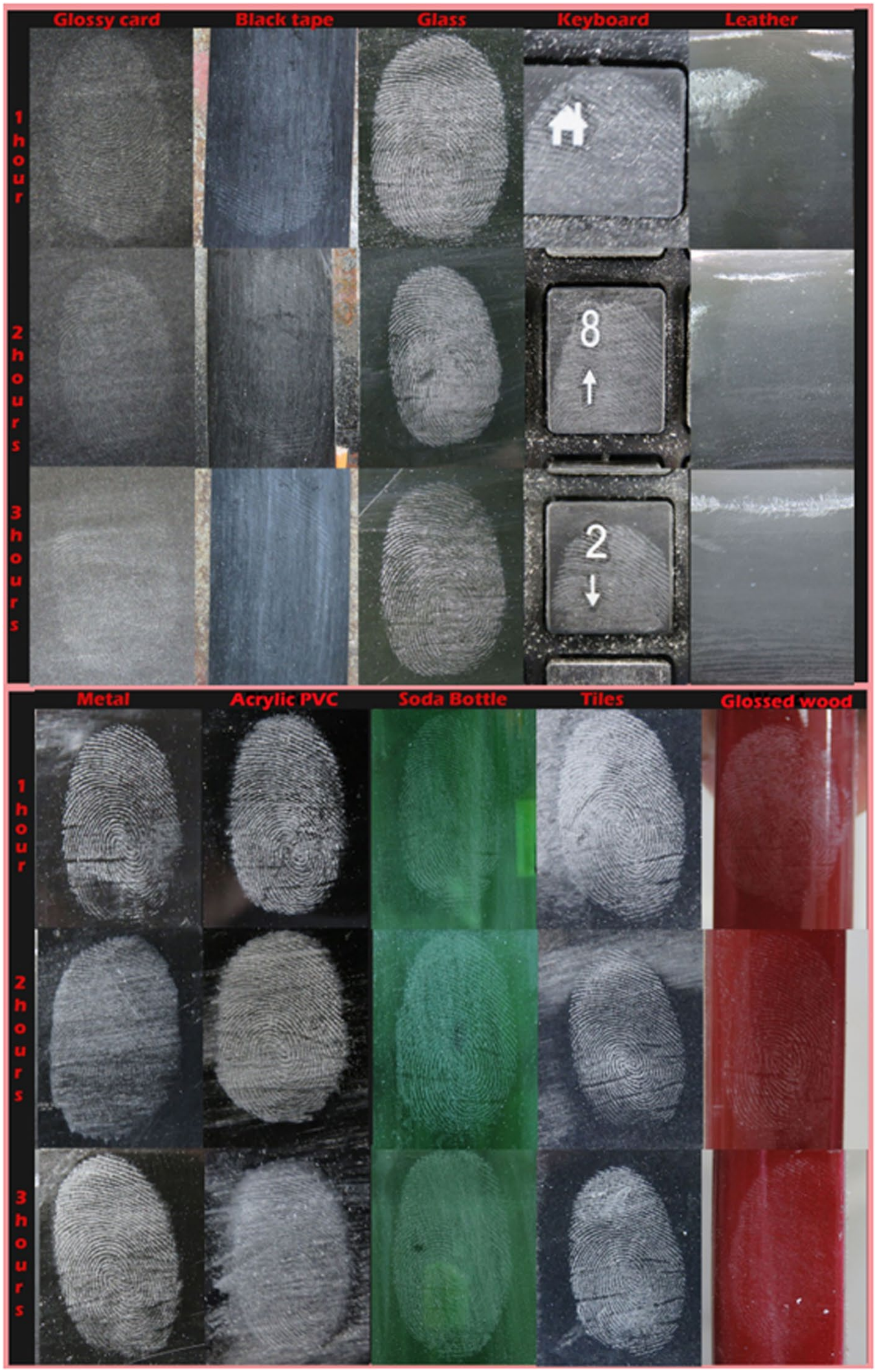

Fig. 8 Fingermarks developed on various surfaces using WP after aged at 100 °C for 1, 2 and 3 h. WP was effective in developing aged fingermarks on all surfaces except leather, glossy card and black tape

4.0 for SNP. These findings suggest that SNP is a practical and viable alternative to WP for developing fingermarks in diverse scenarios, reinforcing its applicability in real-world forensic contexts.

Our depletion and split studies further underscored the practicality of SNP. These experiments revealed that SNP consistently performed as well as, and in some instances, better than WP, particularly

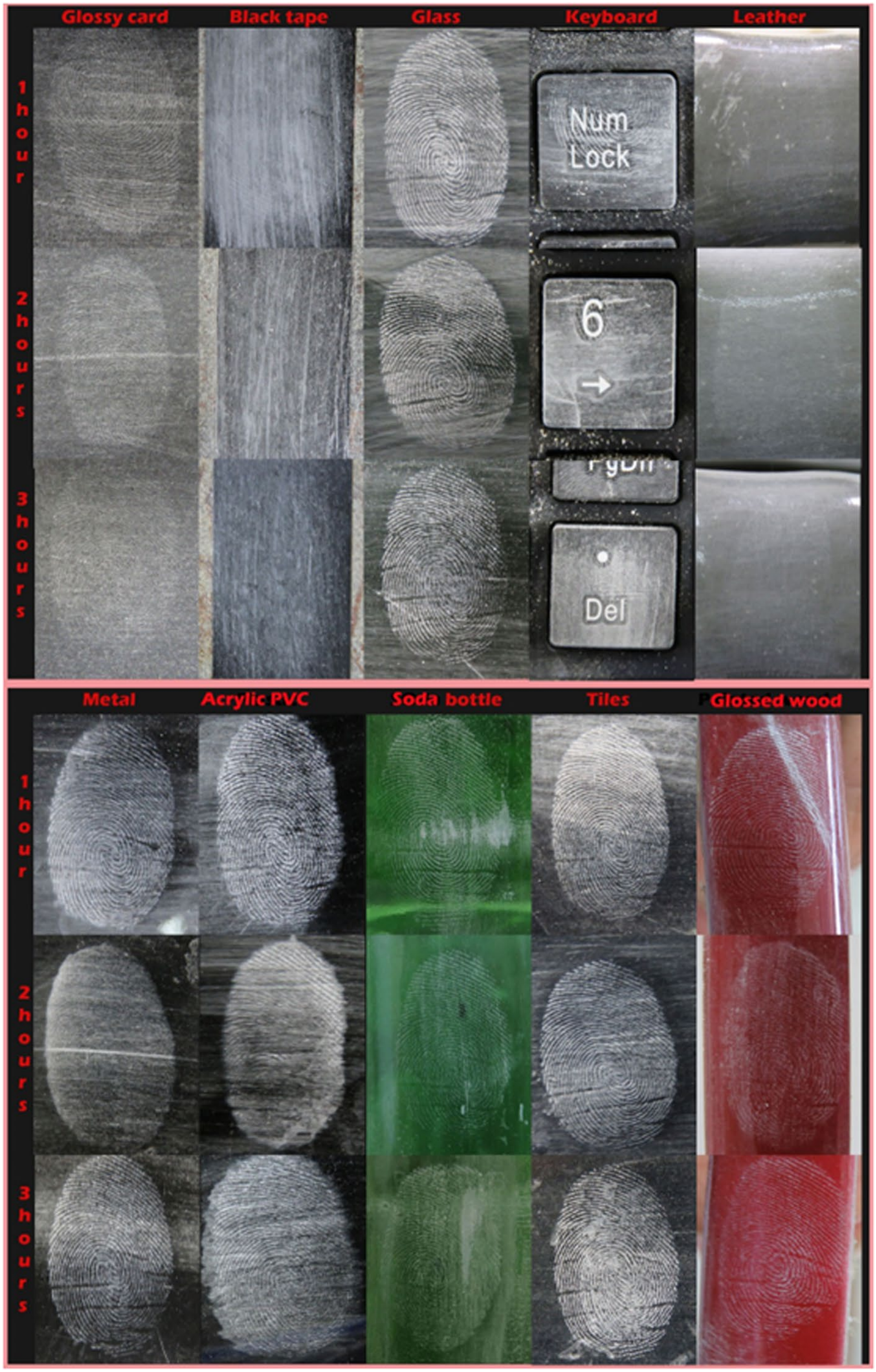

Fig. 9 Fingermarks developed on various surfaces using SNP powder after aged at 100 °C for 1, 2 and 3 h. SNP was effective in developing aged fingermarks on all surfaces except leather. It also had limited efficiency in developed fingermarks aged for 2 h and above at 100 °C on black tape and glossy card

on challenging surfaces. With the exception of painted wood, where SNP exhibited superior performance, there was no statistically significant difference (p-value > 0.05) in the quality of fingerprint development between the two powders. These findings emphasise the versatility and effectiveness of SNP across a range of surfaces and conditions, strengthening its standing as a reliable forensic tool.

The results from our accelerated ageing studies shed light on the behaviour of SNP in comparison to WP when faced with different ageing temperatures and times. The findings revealed that SNP exhibited specific interactions with fingermark residue that improved the quality of fingermarks, especially on slightly rough, non-porous surfaces. While there were no significant differences between SNP and WP on most surfaces, SNP significantly outperformed WP on painted wood and soda bottles, confirming its effectiveness in challenging scenarios.

In addition to its forensic utility, the environmentally friendly aspects of our approach, which repurposes biowaste into effective forensic materials, align with the principles of green chemistry and sustainability. This not only contributes to the field of forensic science but also supports ongoing efforts to reduce environmental impact.

Interaction of SNP powder with different surfaces

The depletion and ageing studies uncovered that SNP powder was particularly effective at developing fingermarks left on silica-based surfaces compared to other types of surfaces. Notably, SNP powder particles exhibited a selective affinity for adhering to the ridges on these silica-based surfaces when compared to WP. Conversely, WP performed well on tile surfaces, and both powders demonstrated similar performance on the remaining surfaces, as shown in the ageing studies.

This distinction in performance can be attributed to the presence of oxygen atoms in silica and titanium dioxide molecules, which have the ability to form hydrogen bonds with fingermark residues as well as hydrocarbon compounds. The tested surfaces can be primarily categorised into two types: silica-based and hydrocarbon-based surfaces. Silica-based surfaces encompass materials like glass, soda bottles and tiles, while all other surfaces fall under the category of hydrocarbon-based surfaces. For example, the metal from vehicle bodies is classified under the latter category since the first layer that comes into contact with the powder is the acrylic polyurethane paint on the surface of the metal.

Silica-based surfaces

Regular glass panes and soda bottles are generally made out of the mixture of sand, soda ash, limestone and some other minerals, with sand being the primary component. On the other hand, tiles are made out of sand by adding components like clay, talc, feldspar and dolomite (Frizzo et al. 2020) and they have less than 0.3% porosity (Monfort et al. 2014). In these surfaces, the Si–OH or Si=O functional groups in the hydrated silica or silicon dioxide promote hydrogen bonding with water, amino acids and sebaceous components of the fingermark residue, along with van der Waals forces. Hence, the SNP powder particles become attracted to the fingermark residue rather than the surface, forming a clear demarcation of the ridges.

Hydrocarbon-based surfaces

The other surfaces examined in this study consisted of hydrocarbon-based materials. For instance, automotive paint and the steering wheel cover were constructed from acrylic polyurethane (PU), while electrical black tape and number plates were composed of acrylic PVC polymer. External electronic keyboards are typically crafted from ABS, and glossy wood paint contains acrylic or vinyl resins. Lastly, glossy art cards are generally coated with a polymer to achieve a metallic shine.

On these surfaces, hydrogen bonding between the Si–OH and Si=O groups and the surface materials led to a reduction in the clarity of the developed marks. Even though more particles were deposited on the ridges, a thin layer of powder was also present on the surfaces, creating a halo-like appearance around the ridges. This halo effect became more pronounced as the fingermark residue depleted, affecting the overall clarity of the developed marks.

Optimal particle size for fingermark development

The observation indicated that SNP particles with a mean particle size below 200 nm were not only too lightweight to effectively adhere to fingermark residue but also resulted in faint fingermark development. These findings were in line with prior reports on the multi-metal deposition (MMD) technique, where the use of a physical developer was necessary to enhance the visibility of fingermarks developed with gold nanoparticles (GNPs) of 20 nm size (Becue et al. 2012). Moreover, it is essential to note that smaller-sized SNPs can pose a greater health risk to users, as they have the potential to easily penetrate face masks and enter the lungs, which can lead to silicosis syndrome upon prolonged exposure (Merget et al. 2002).

Health and safety

Titanium oxide is a compound classified as substance that causes long lasting harmful effects to aquatic life, with carcinogenic effects. Studies were carried out to determine the effect of titanium oxide to human and preliminary findings acknowledged the carcinogenic property of this compound (Sha et al. 2011; Shah et al. 2017). Amorphous silica was classified as a substance that causes acute toxicity through inhalation or ingestion (UN 2017). However, some findings suggested that the toxicity effect of silica was enhanced in the crystalline form and there were no conclusive proof that confirms the toxicity of amorphous silica (Merget et al. 2002). Another study presented that silica was an essential mineral for bone growth and prevents osteoporosis (Price et al. 2013). Although amorphous may cause toxicity by inhalation, titanium oxide was suggested to have long lasting adverse effects. The inhalation of powder particle can be avoided by using suitable personnel protective equipment.

Conclusions

The synthesis and characterisation of spherical silica nanoparticles from rice husk, as well as their initial application in latent fingermark development, have been previously published. Building upon this foundation, our current study extends the application of these silica nanoparticles to the challenging task of developing aged, depleted fingermarks and fingermarks contributed by multiple donors. This advanced research not only reaffirms the potential of these nanoparticles for latent fingermark enhancement but also highlights their effectiveness in addressing complex forensic scenarios. As we continue to explore the practical applications of eco-friendly nanomaterials, this study offers a significant contribution to the field of forensic science by providing a sustainable and reliable solution for latent fingermark development across a broader spectrum of real-world cases.

Abbreviations

PU	Acrylic polyurethane
ABS	Acrylonitrile butadiene styrene
EDX	Energy dispersive X-ray spectroscopy
FESEM	Field emission scanning electron microscopy
GNP	Gold nanoparticles
MMD	Multi-metal deposition
PTEOS	Phenyltriethoxysilane
PVC	Poly vinyl chloride
RH	Rice husk
SNP	Silica nanoparticle powder
TEOS	Tetraethylorthosilicate
WP	White powder

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Author's contributions
All authors contributed to the design of the study. RR performed the experiments. RR and NF analysed the data. All authors discussed and wrote the manuscript. All authors read and approved the final manuscript.

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Declarations

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The authors declare no competing interests.

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References

Azman AR, Mahat NA, Wahab RA et al (2018) Novel safranin-tinted Candida rugosa lipase nanoconjugates reagent for visualizing latent fingerprints on stainless steel knives immersed in a natural outdoor pond. Int J Mol Sci 19:1–17. https://doi.org/10.3390/ijms19061576

Becue A, Moret S, Champod C, Margot P (2009) Use of quantum dots in aqueous solution to detect blood fingermarks on non-porous surfaces. Forensic Sci Int 191:36–41. https://doi.org/10.1016/j.forsciint.2009.06.005

Becue A, Scoundrianos A, Moret S (2012) Detection of fingermarks by colloidal gold (MMD/SMD) – beyond the pH 3 limit. Forensic Sci Int 219:39–49. https://doi.org/10.1016/j.forsciint.2011.11.024

Daluz HM (2015) Fundamentals of fingerprint analysis. CRC Press, Boca Raton FL, First Edit

Dhall JK, Kapoor AK (2016) Development of latent prints exposed to destructive crime scene conditions using wet powder suspensions. Egypt J Forensic Sci 6:396–404

Ferreira RG, Paula RBA, Okuma AA, Costa LM (2021) Fingerprint development techniques: a review. Rev Virtual Quim 13:1278–1302. https://doi.org/10.21577/1984-6835.20210083

Fieldhouse S, Oravcova E, Walton-Williams L (2016) The effect of DNA recovery on the subsequent quality of latent fingermarks. Forensic Sci Int 267:78–88

Frizzo RG, Zaccaron A, de Souza NV, Bernardin AM (2020) Pyroplasticity on porcelain tiles of the albite-potassium feldspar-kaolin system: a mixture design analysis. J Build Eng 31:101432. https://doi.org/10.1016/j.jobe.2020.101432

Ghorbani F, Younesi H, Mehraban Z et al (2013) Preparation and characterization of highly pure silica from sedge as agricultural waste and its utilization in the synthesis of mesoporous silica MCM-41. J Taiwan Inst Chem Eng 44:821–828. https://doi.org/10.1016/j.jtice.2013.01.019

Komalasari I, Krismastuti FSK, Elishian C et al (2017) Straightforward fabrication of black nano silica dusting powder for latent fingerprint imaging. AIP Conference Proceedings 1904. AIP Publishing

Kongmanklang C, Rangsriwatananon K (2014) Hydrothermal synthesis of high crystalline silicalite from rice husk ash. J Spectrosc 2015:1–5

Kumagai S, Sasaki J (2009) Carbon/silica composite fabricated from rice husk by means of binderless hot-pressing. Bioresour Technol 100:3308–3315. https://doi.org/10.1016/j.biortech.2009.02.001

Lee HC, Gaensslen RE (2012) Lee and Gaensslen's: advances in fingerprint technology. CRC Press, Boca Raton, Florida, Third Edit

Maynard AD (2011) Don't define nanomaterials. Nature 475:31. https://doi.org/10.1038/475031a

Merget R, Bauer T, Küpper H et al (2002) Health hazards due to the inhalation of amorphous silica. Arch Toxicol 75:625–634. https://doi.org/10.1007/s002040100266

Monfort E, Mezquita A, Vaquer E et al (2014) 8.05 - ceramic manufacturing processes: energy, environmental, and occupational health issues. In: Hashmi S, Batalha GF, Van Tyne CJ, Yilbas B (eds) Comprehensive Materials Processing. Elsevier, Oxford, pp 71–102

Netten C Van, Teschke KE, Souter F (1990) Occupational exposure to elemental constituents in fingerprint powders. Arch Environ Heal 45:123–127. https://doi.org/10.1080/00039896.1990.9935936

Nik Hassan NF, Che Hamzah CN, Rajan R, Zakaria Y (2020) Agriculture waste recycling technologies: recycling of nanosilica powder from bamboo leaves and rice husks for forensic applications. In: Bergmann CP, Makhlouf ASH, Ali GAM (eds) Topics in Mining, Metallurgy and Materials Engineering Waste Recycling Technologies for Nanomaterials Manufacturing, 1st Editio. Springer, pp 295–324

Noushad M, Ab Rahman I, Che Zulkifli NS et al (2014) Low surface area nanosilica from an agricultural biomass for fabrication of dental nanocomposites. Ceram Int 40:4163–4171
Noushad M, Rahman IA, Husein A et al (2012) A simple method of obtaining spherical nanosilica from rice husk. Int J Adv Sci Inf Technol 2:28–30
Pattanayak DK, Muduli J, Sahu SS et al (2022) Production and characterization of amorphous silica nanoparticles from coconut shell and coir. Lett Appl NanoBioScience 11:4040–4049. https://doi.org/10.33263/LIANBS114.40404049
Price CT, Koval KJ, Langford JR (2013) Silicon: a review of its potential role in the prevention and treatment of postmenopausal osteoporosis. Int J Endocrinol 2013:1–6
Rafiee E, Shahebrahimi S, Feyzi M, Shaterzadeh M (2012) Optimization of synthesis and characterization of nanosilica produced from rice husk (a common waste material). Int Nano Lett 2:1–8. https://doi.org/10.1186/2228-5326-2-29
Rajan R, Zakaria Y, Shamsuddin S, Hassan NFN (2018) Nanocarbon powder for latent fingermark development: a green chemistry approach. Egypt J Forensic Sci 8:2–10. https://doi.org/10.1186/s41935-018-0091-5
Rajan R, Zakaria Y, Shamsuddin S, Nik Hassan NF (2020) Robust synthesis of mono-dispersed spherical silica nanoparticle from rice husk for high definition latent fingermark development. Arab J Chem 13:8119–8132. https://doi.org/10.1016/j.arabjc.2020.09.042
Sha B, Gao W, Wang S et al (2011) Cytotoxicity of titanium dioxide nanoparticles differs in four liver cells from human and rat. Compos Part B Eng 42:2136–2144. https://doi.org/10.1016/j.compositesb.2011.05.009
Shah SNA, Shah Z, Hussain M, Khan M (2017) Hazardous effects of titanium dioxide nanoparticles in ecosystem. Bioinorg Chem Appl 2017:1–12. https://doi.org/10.1155/2017/4101735
SIRCHIE (2012) 103LD HiFi volcanic latent print powder, indestructable white, safety data sheet. Youngsville, NC
Sodhi GS, Kaur J (2001) Powder method for detecting latent fingerprints: a review. Forensic Sci Int 120:172–176
Souter FCG, Van Netten C, Brands R (1992) Morbidity in policemen occupationally exposed to fingerprint powders. Int J Environ Heal Res 2:114–119. https://doi.org/10.1080/09603129209356739
Theaker BJ, Hudson KE, Rowell FJ (2008) Doped hydrophobic silica nano- and micro-particles as novel agents for developing latent fingerprints. Forensic Sci Int 174:26–34. https://doi.org/10.1016/j.forsciint.2007.02.030
UN E, (2017) Globally harmonized system of classification and labelling of chemicals (GHS), 7th edn. United Nations Publications, Herndon, VA, USA
Vaibhav V, Vijayalakshmi U, Roopan SM, Mohana Roopan S (2015) Agricultural waste as a source for the production of silica nanoparticles. Spectrochim Acta - Part A Mol Biomol Spectrosc 139:515–520

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