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## (Bio)polymer-Based Powders As Hidden Treasures in Dactyloscopy

### بودرات البوليمر (الحيوي) كنز مخفي في مجال التعرف على بصمات الأصابع

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### Abstract

Different chemical, physical, and physico-chemical methods with addition of optical methods have been used for decades for the development of latent fingerprints (LFPs), even though the choice of a method depends on various factors (type and structure of the surface, external conditions, donor etc.). However, a universal system has not yet been fabricated, while many of those already used are toxic to the humans and the environment. Recently, researchers designed formulations based on (bio)polymeric materials and their specific properties, suitable for targeted interaction with fingerprint (FP) sweat and lipid residues.

Some research groups produced fluorescent properties of particular polymeric materials to map sweat pores, while others encapsulated/incorporated dyes, pigments, etc. into polymeric matrix to obtain formulations of desired color and properties. Additionally, polymer micelles have become interesting due to their amphiphilic properties and the ability to incorporate compounds which could enable multi-colored emission brightness. Nevertheless, (polymeric) nanomaterials are currently of a great importance in material science world, due to specific optical and electronic properties convenient for interaction with FP residues found on different (multi-colored, electroconductive, etc.) substrates.

This paper focuses on (bio)polymer-based systems used to develop LFPs, different approaches of research groups and future possibilities to create the optimal system for specified purpose.

**Keywords:** Forensic sciences, Latent fingerprints, Physical systems, (bio)polymer powders.

### المستخلص

تم استخدام طرق كيميائية وفيزيائية وفيزيائية كيميائية مختلفة مع إضافة الطرق البصرية لعقود من الزمن لتطوير بصمات الأصابع الكامنة (LFPs)، وعلى الرغم من أن اختيار الطريقة يعتمد على عوامل مختلفة (نوع وبنية السطح، المظهر الخارجي الشروط، الجهة المانحة إلخ). فإنه لم يتم تصنيع نظام عالمي بعد، في حين أن العديد من تلك المستخدمة بالفعل سامة للإنسان والبيئة. وفي الآونة الأخيرة، صمم الباحثون تركيبات تعتمد على مواد بوليمرية (حيوية) وخصائصها المحددة، مناسبة للتفاعل المستهدف مع بقايا العرق والدهون في بصمات الأصابع.

وأنتجت بعض المجموعات البحثية خصائص الفلورسنت لمواد بوليمرية معينة لرسم خريطة لمسام العرق، بينما قامت مجموعات أخرى بتغليف/دمج الأصباغ والمواد الملونة وما إلى ذلك في مصفوفة بوليمرية للحصول على تركيبات من اللون والخصائص المطلوبة. بالإضافة إلى ذلك، أصبحت مذيلات البوليمر مثيرة للاهتمام بسبب خصائصها المزدوجة والقدرة على دمج المركبات التي يمكن أن تمكن من سطوع الانبعاث متعدد الألوان. ومع ذلك، فإن المواد النانوية (البوليمرية) لها حاليًا أهمية كبيرة في عالم علوم المواد، نظرًا لخصائص بصرية وإلكترونية محددة ملائمة للتفاعل مع بقايا بصمات الأصابع الموجودة على ركائز مختلفة (متعددة الألوان، وموصلة للكهرباء، وما إلى ذلك).

تُركز هذه الورقة على الأنظمة القائمة على البوليمر (الحيوي) المستخدمة لتظهر بصمات الأصابع الكامنة، والأساليب المختلفة لمجموعات البحث والإمكانات المستقبلية لإنشاء النظام الأمثل لغرض محدد.

**الكلمات المفتاحية:** علوم الأدلة الجنائية، بصمات الأصابع الكامنة، الأنظمة الفيزيائية، بودرة البوليمر (الحيوية).



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## 1. Introduction

Since 1891, one of the main approaches used in criminal investigations to identify individuals is forensic examination of fingerprints (FPs), also known as dactyloscopy [1-3]. The fingerprint (FP) represents an impression deposited by the friction ridges of a human fingertips, and consists of secretion of sweat and sebaceous glands in the form of papillary line traces. By excluding injuries, diseases and other factors that can artificially contribute to the modification or change, papillary lines are perpetual and do not change over time [4-7]. Therefore, FPs are unique for each person (including identical twins) [8]. Friction ridge details are generally described in a hierarchical order at three levels, namely level 1 (FP basic pattern), level 2 (minutiae points) and level 3 (pores and ridge shape) [9-11]. On the other hand, FPs are also classified as patent, plastic and latent, and therefore different approaches and techniques are being employed for their manipulation [5, 12, 13]. Latent fingerprints (LFPs), are imperceptible to the naked eye and they are often more challenging, because they should be developed prior to collection and further processing, so for that purpose different chemical, physical, and physico-chemical, assisted with optical methods are being applied [9, 13-15].

The intensity and quality of LFPs depends on substrate properties (type, color, structure, etc.) and environmental conditions, but also sex, age, ethnicity, metabolism of the donor and its daily routine (nutrition, smoking, medication, cosmetics, etc.) are of great impact [16-18]. Optical methods are non-destructive and often employed to visualize or enhance the FPs with alternative light sources [19-21], while chemical methods are developing LFPs by specific chemical reaction between the species and the FP residues, resulting in the formation of (stable) compounds and/or complexes [22-24]. Fur-

thermore, physical methods are based on physical interactions between some components (usually powders, pigments, dyes, and/or their mixtures) and FP residues. Since almost all chemical methods are detrimental to the environment and humans, the physical systems took advantage, especially during crime scene investigations [25-27]. The type of physical formulation that will be employed depends on various factors, but the properties of the substrate that contains FPs are probably the most important [28-30]. Therefore, when deploying new systems and approaches for visualization of LFPs, researcher groups should comply with the instructions proposed by professional and internationally recognized organizations. International Fingerprint Research Group (IFRG) proposed complex and informative Guidelines for the assessment of fingerprint detection techniques, with four main research phases for assessment of novel systems. Phases include system development from initial concept through to final casework implementation, so each following phase is more complex than the previous one (e.g. requires more donors, substrates, external conditions, etc.) [31].

Considering all aforementioned problems, while highlighting the toxicity as the main shortcoming of many systems, researches are making efforts in developing novel formulations that will satisfy both cost-benefit and eco-friendly demands. In this review paper, the focus will be on (bio)polymer-based powder formulations used for detection and enhancement of LFPs, especially on current achievements and capabilities for improvement in forensic investigations.

## 2. Powder dusting

The most recognized method for developing FPs is powder dusting, where powder particles adhere to the sweat and lipid FP residues by physical in-



teractions. Powder method is used extensively on non-porous substrates such as glass, metal, painted surfaces, etc. [32, 33], but it does not always detect older FPs due to the lack of residues, as a consequence of the surface properties and external factors [34, 35]. Regular (e.g. black and gray powder), luminescent (fluorescent), metallic (magnetic, silver) and thermoplastic powders are routinely employed in forensic practice. The structure, brightness, color and other characteristics of the substrate will influence the choice of the powder (e.g. black powder for brighter and gray/silver for darker surfaces), and it is always desirable to use less- or non-destructive system. The powder is applied with a brush (e.g. squirrel hair, zephyr, marabou brush or magnetic stick – depending on the type of powder) to the object with traces, which results in the visualization of the FP image. Afterwards, the papillary line traces can be photographed, collected and further examined [7, 9, 13, 32]. However, it is very difficult to produce a universal FP powder that will match all demands. Experts indicate that currently no powder can be used for all tasks, so each FP should be treated independently, based on conditions found at the crime scene [36].

### 3. Sweat pore mapping

The development of LFPs increasingly demands improved sensitivity and quality of detection, where poroscopy can have a great impact, as the study of sweat pores present on the papillary ridges of the skin. Lee and associates (2014) were one of the first researchers in the field of mapping sweat pores using polymeric materials. Research group indicated that a polymer showing immediate fluorescence and color change in response to a small amount of water may be used for visualization of FPs [37]. In recent years, several other research groups have used polymeric materials to map sweat pores on

FPs. A simple and efficient sweat pore mapping method based on fluorescein and poly(vinyl pyrrolidone) (PVP) composite film was developed by Pyo et al. (2015). The composite film displayed a fluorometric turn-on response upon contact with a small quantity of water secreted from human sweat pores, allowing precise mapping of sweat pores on a fingertip. Each pore was reflected in a discrete fluorescence microdot, where the coordinates of the microdots can be extracted and the patterns can be exploited for identification of individuals [38]. Additionally, the same group of authors reported a new method for mapping sweat pores using a water-soluble polymer matrix film. They determined that local adsorption of water secreted from sweat pores on human fingertips creates discrete microdimples on the hydrophilic polymer film, and these microdimples can be clearly visualized by a conventional optical microscope [39]. Furthermore, Park and associates (2016) reported that incorporation of headgroups composed of hygroscopic ions such as cesium or rubidium and carboxylate counter ions enables polydiacetylenes (PDAs) to undergo a blue-to-red colorimetric transition, as well as a fluorescence turn-on response to water. The authors reported four hydrochromic systems namely: hygroscopic headgroup PDA, inkjet compatible and temperature dependent hydrochromic PDA, fluorescein hydrophilic polymer PVP, and carbon nanodots. It was determined the small quantities of water secreted from human sweat pores were found to be sufficient to trigger fluorescence turn-on responses of the hydrochromic PDAs, allowing precise mapping of human sweat pores [40].

### 4. Incorporation of fluorescent materials

Fluorescent powders are extensively employed for detection and enhancement of LFPs on some unconventional substrates, such are multi-colored



surfaces, skin, firearms, etc. [41]. Therefore, researchers are constantly trying to improve these formulations in order to increase their sensitivity and selectivity, and to obtain better contrast and overall quality. The electrodeposition of bilayer systems based on conjugated and fluorescent polymers was reported by Costa et al. (2020) to develop LFPs from stainless steel. The first layer of polypyrrole was electrodeposited onto the surface containing an LFP and the second layer of a fluorescent poly( $\alpha$ -terthiophene) was electrodeposited onto the first layer. Such bilayer systems showed fluorescent properties and could be applied for the visualization of LFPs on stainless steel, due to the high definition of images under both visible and ultraviolet (UV) light (with development of level 1 and 2 ridge details) [42].

Barros et al. (2020) proposed fluorescent developers based on starch, a high abundant natural polymer. The small amount of organic (benzazole) dye incorporated into starch matrix (under aqueous and ethanolic conditions) considerably reduced a risk of toxicity, while prepared developers also exhibit high photophysical stability due to the excited-state intra-molecular proton transfer mechanism and a high Stokes shift, which enables preservation for a long time. It is evident that proposed system is cheaper, more environmentally friendly and less harmful, with promising results in developing fresh and non-fresh FPs on porous and non-porous substrates [43]. Similar group of authors reported the entrapment of four different benzazole derivatives into starch matrix, which showed intense fluorescence emission in the violet-green region while developing FPs from various porous and non-porous substrates [44].

On the other hand, silicates have drawn high interest as matrix materials due to multiple advantages, such are easy surface functionalization, low toxicity, high stability, and good optical properties

[45]. Li et al. (2022) proposed the phosphorescent polymer/silica-based FP powder, prepared by one-step pyrolytic synthesis. The carbonized polymers gel-like solid with intense green room-temperature phosphorescence emission were obtained by dispersing carbonized polymers in silica gel. Obtained polymer-silica powders show bright blue fluorescence and strong green phosphorescence under ambient conditions, by visualizing minutiae points of LFPs on diverse substrates without background interference [46].

Furthermore, conjugated polymers have high potential due to possible structure modification. Poly(*p*-phenylene vinylene) (PPV) nanoparticles (NPs) in aqueous colloidal solution were proposed by Chen et al. (2017). The FPs were deposited onto three different types of adhesive tapes (both visible and LFPs), as well as cover glass and aluminum foil (only LFPs), then immersed into prepared colloidal solution and rinsed with deionized water. Developed FPs excited with 365 nm UV light showed good fluorescence and clear visualization of level 1 and 2 ridge details on all selected substrates, assisted by digital magnification. Prepared PPV solution showed good steadiness up to 6 months [47]. Based on these results, a similar group of authors proposed a combination of conjugated polymer NPs and cyanoacrylate fuming for enhanced fluorescent visualization of FPs. The PPV NPs possess a better developing capability than rhodamine 6G when excited by 365 nm UV light [48].

## 5. Polymer micelles

Lately, polymer micelles have aroused a high interest due to their amphiphilic properties and ability to interact with both sweat and lipid residues present in FPs. Wang et al. (2021) used a commercial conjugated polyelectrolyte based on polyfluorene derivative to prepare polymer micelles in water, while



multi-color emission was achieved by encapsulating small organic dyes. Most organic fluorophores usually sustain significant aggregation-caused quenching at dry state, thus limiting the emission brightness. However, the encapsulation of dyes into micelles enabled bright emission whether in micelles solution or in solid film. Polymer micelles showed blue emission under UV irradiation, while encapsulation of coumarin 6 and Nile red exhibited green and red emissions, respectively. Prepared micelles were applied onto glass, aluminum, and stainless steel (with smooth and rough surface), and results showed high-definition FP images, with no background interference due to fluorescence and/or overlapping of FPs [49]. Furthermore, Zou and collaborators (2022) proposed the off-on fluorescent polymer micelles based on poly((ethylene glycol)-co-(4-formyl-3-hydroxyphenyl)). Hydrazine was used as a crosslinking agent, due to the reaction with aldehyde groups of amphiphilic copolymer, to form polymer micelles. The crosslinking induced emission can be associated with the occurrence of both aggregation-induced emission and excited state intramolecular proton transfer, contributing to the high-contrast fluorescence properties of micelles. Polymer micelles were thoroughly investigated on FPs deposited onto glass sheets, while their application capabilities were examined on no-background substrates (Büchner funnel, leather, plastic wrap, and aluminum foil) and background substrates (cans, wood, white paper, and paper coupons). Prepared polymer micelles showed high selectivity, good photostability, and excellent long-term steadiness, while developed FPs exhibited bright yellow-green fluorescence, with visualization of 1–3 level ridge details of LFPs [50].

## 6. Polymer nanomaterials

The field of nanotechnology spreads out its application in many scientific disciplines, due to spe-

cific properties of nanomaterials (NMs), i.e. NPs. Particles size up to 100 nm, selective binding to FP residues obtained through the surface functionalization of the particles, and optical properties that facilitate FP observation after development, represent the most important factors for their application in forensic examinations of FP [51–53].

Prompt visualization of LFPs at the crime scene is important for law enforcement agencies. In that manner, fluorescent systems could enable high fluorescence with maximum brightness and contrast. Abdollahi et al. (2022) developed multi-color photoluminescent functional polymer NPs based on fluorescent dye oxazolidine and copolymer matrix of methyl methacrylate (MMA)/2-(dimethylamino)ethyl methacrylate (DMAEMA). Seven different formulations of polymer NPs have been prepared to develop LFPs deposited onto different porous, semi-porous and non-porous substrates, which were observed under visible and UV light. Developed FPs observed under visible light showed white color, while under UV light FPs exhibit intensive purple color. Results demonstrated high sensitivity and contrast, as well as the visualization of FPs ridge details from level 1–3 [54]. Similar research was reported by Abdollahi and Dashti (2023), who synthesized photoluminescent amphiphilic NPs by doping of the hydroxylated oxazolidine derivative (OXOH) to the surface of MMA/DMAEMA. The results showed high fluorescence and sensitivity, accompanied with good visualization of FPs deposited onto multiple substrates (as explained in the research of Abdollahi et al. (2022) [54, 55].

Dusting, spraying and vapor deposition techniques have been routinely used for the development of LFPs. However, shortcomings regarding the synthesis procedure, post-treatment process and toxicity have limited application of many systems, and therefore electrospun polymer (nano)materials





are proposed to circumvent these downsides. Gal et al. (2023) proposed novel NMs, i.e. four fluorophores based on electrospun PVP polymer doped with fluorescent 10-ethyl-10H-phenothiazine-carboxylate salts (lithium, sodium, potassium, and calcium salt). The advantages of electrospun polymers are their small pore size, high porosity and large surface area, which enable higher sensitivity and interaction with LFP residues. Proposed polymer systems showed multiplane properties – transfer of LFPs from different substrates (glass, polymer, metal, banknote, ceramic, and wood) onto electrospun mats and simultaneous development of LFPs on the initial surface. The images were taken under visible and UV (254 nm) light, and fluorophores visualized ridge details from level 1–3 [56].

On the other hand, Zhang et al. (2022) used the mechanism of fluorescence resonance energy transfer and reported aptamer functionalization and high-contrast reversible dual-color photo-switching fluorescence of polymeric NPs. The results indicated that prepared NPs (named LBA-RPFNPs) displayed good water dispersibility, quick photo-reversibility, satisfying photo-reversibility and good long-term steadiness, as they target lysozyme in LFP residues [57].

Some authors reported nanophosphor formulations, which can exhibit luminescence. Narasimhamurthy et al. (2021) developed Bovine Serum Albumin (BSA) functionalized  $Gd_2O_3:Eu^{3+}$  (5 mol %) nanopowders dispersed in a poly(vinyl alcohol) (PVA) matrix. Prepared nanocomposites were used to detect and preserve sebaceous FPs, previously deposited onto different non-filtrating and filtrating surfaces, then recorded under UV light (254 nm). LFPs were visualized with  $Gd_2O_3:Eu^{3+}$  (5 mol %) $@BSA$  stain following powder dusting, and then PVA film was used to preserve developed FPs for a long period. The results on non-filtrating surfaces

showed well defined ridges with visualization of level 2 details, clearly visible under UV light. Similarly, visualization of LFPs on filtrating substrates indicated clear images of FPs, with good contrast and without background interference. However, as confirmed by SEM analysis, one of the biggest achievements of the research was the observation of ridge details level 1–3 on FPs preserved for 1 year [58].

Furthermore, quantum dots made a breakthrough into many scientific branches due to specific optical and electronic properties [59]. They can exhibit light of various wavelengths when exposed to some alternative light source, and currently one of the most interesting type are carbon dots (CDs). Milenkovic and collaborators (2019) prepared N-doped CDs using the hydrothermal treatment of PVP as N source, then used to develop LFPs deposited onto tweezers and a plastic bag. Prepared N-CDs exhibited bright fluorescence under UV illumination by visualizing ridge patterns [60]. Ding and associates (2021) developed bifunctional composite powder with magnetic properties and intense fluorescence. Powder marked as  $Fe_3O_4@SiO_2-CD$  showed high contrast, sensitivity and selectivity over traditional methods for visualization of FPs [61]. Additionally, Lin et al. (2022) designed Nile blue-derived CDs encapsulated into polyethyleneimine polymer dots (CPPDs). Prepared CPPDs visualization of ridge 1–3 level details of LFPs on multiple substrates, with high quality, resolution and fluorescence [62].

## 7. Bio-based (polymer) formulations

Many commercial (especially chemical) systems for development of LFPs are marked as detrimental to humans and the environment. In that manner, researchers opted for application of easily procurable, non-toxic and cheap bio-based components. Some authors used commonly available materials as pow-



ders, such are minerals, food powders, cosmetics, plant materials, etc. [63]. A couple research groups used turmeric powder due to its eco- and cost-friendly properties. Garg and associates (2011) investigated the application of turmeric powder on sebaceous FP deposited onto 9 different porous, semi-porous and non-porous substrates. The results indicated that powder adheres selectively to the FP residues, developing ridge patterns and details [64]. These results were confirmed by Dhunna and collaborators (2018), who used turmeric powder to develop LFPs deposited onto aluminum foil [65].

Plant materials brought attention due to easy dusting, and possible application as sole materials or in combination with other components. Some research groups explored the utilization of henna powder (*Lawsonia inermis*), which contains the red-orange pigment lawsone. Chauhan and Udayakumar (2017) successfully employed henna powder to develop FP deposited onto white paper sheets [66]. Additionally, Anand et al. (2017) examined henna powder on different porous and non-porous substrates with LFPs. The best results were obtained on non-porous substrates (especially aluminum foil), while porous and rough substrates aggravated the development of complete FP image and ridge details [67].

Furthermore, new powders based on marine algal biomass were prepared by Passos et al. (2021), to investigate their potential for enhancement of LFPs. Natural and sebaceous FPs were deposited onto the glass surface and developed with prepared (bio)powders. The best results were obtained with the powder prepared from biomass of *Spirulina* sp. due to good visualization and satisfying contrast between the surface and the prints [68]. On the other hand, some research groups investigated the potential of natural recycled material for the visualization of LFPs. Said and collaborators (2021) used powders prepared from eggshells and clamshells to

develop eccrine, sebaceous and natural LFPs deposited onto 5 non-porous substrates. The results showed that both bio-powders successfully developed FP with visualization of ridge patterns when compared with commercial (white) powder, which was additionally confirmed when testing aged FPs. Exposure to sunlight, submersion in water and burial in soil where used to investigate the influence of external factors, and it was determined that LFPs could be recovered from all non-porous substrates (except painted wood) [69].

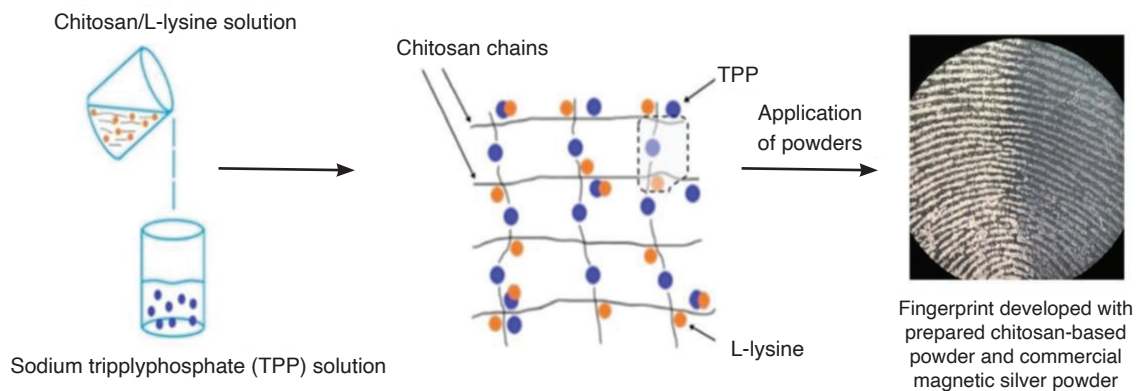
According to the results reported by Hejjaji et al. [70], Vučković and co-workers prepared dextran- and chitosan-based bio-formulations [26, 71]. When compared, chitosan-based formulations showed better results than dextran-based systems probably due to smaller particle size and better binding to the FP residues. Briefly, chitosan-based conjugates were prepared for detection and enhancement of dry and sebaceous LFPs from glass and rubber substrate. Results showed that obtained bio-based powder formulation exhibited satisfying properties for the visualization of sebaceous LFPs deposited onto non-porous substrates [71].

Most recently, Li et al. (2023) proposed an eco-friendly and low-cost FP powder based on polydopamine nanospheres. Prepared powder was used to develop LFPs deposited onto permeable and non-permeable surfaces. Both level 1 and 2 ridge details were observed on colored lid, cambered cup, paper, wood, and leather, while FPs aged more than 90 days could be visualized on non-permeable surfaces [72].

## 8. Discussion and conclusions

Methods and techniques for developing LFPs are constantly improving in order to satisfy numerous demands, but the most important are the ease of application, price and toxicity. This review





**Figure 1**– Preparation process of chitosan-based conjugates and visualization of fingerprint with prepared chitosan-based powder (left-half of the fingerprint) and commercial magnetic silver powder (right-half of the fingerprint).

paper encompassed the reports/publications of newly proposed (bio)polymer-based formulations for detection and enhancement of FPs. Biopolymer-based systems are highly suitable due to their non-toxicity, but biodegradability represents a problem for storage of products and visualized FPs. Synthetic polymeric materials are interesting due to possible functionalization which can ameliorate the interaction with FP residues. Therefore, the combination of natural and synthetic polymers could help to produce the material with desired properties. However, fluorescent polymer nanoformulations gave the best results under UV illumination, due to easy application on diverse substrates, nanosized particles, generating different colors, as well as high sensitivity, selectivity and quality of images.

Based on the results obtained from all publications presented in this review paper, it can be concluded that although there is no universal system or formulation for development of FPs, polymer-based systems offer satisfying and improved visualization of ridge patterns on various substrates compared to those commercially employed. This allows the advancement of the research of application of polymeric materials in various forms in the field of forensic trace analysis. The choice of method depends on conditions in a specific case, so the

operators need to recognize and opt for the most suitable approach. Additionally, different laboratories should communicate, share experiences and knowledge with the aim to find the best practical solution, considering the standardization processes, as well as the necessity to comply with the guidelines suggested by the IFRG.

### Conflict of interest

The authors declare no conflicts of interest.

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