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\textbf{ABSTRACT}

Pesticide residues in food products cause human health concerns through food contamination, thereby necessitating their rapid and facile detection. Although surface-enhanced Raman scattering (SERS) technique can rapidly and reliably detect pesticide residues, its application in food safety diagnostics is restricted by its high expense, low scalability, and low reproducibility of the necessary sensors. Herein, we present a low-cost, large-scale, and highly reproducible nanofabrication route for SERS nano-sensors, based on the thermophoresis-assisted direct deposition of plasmonic core-shell structured Ag-SiO\textsubscript{2} nanoparticles produced in the gas phase, on temperature-controlled inexpensive glass substrates. The high-performance SERS substrates were fabricated at a laboratory production rate of 100 samples/hour, demonstrating the scalability and cost-effectiveness of our aerosol manufacturing strategy. Our highly sensitive SERS substrates rapidly and quantitatively detected pesticide residues in fresh orange, indicating their practical applicability for food safety diagnostics.

1. Introduction

Pesticides are widely used in modern agricultural practices to protect crops and food products from pests, diseases, and weeds, thereby improving food quantity and quality. According to a report from the Food and Agriculture Organization of the United Nations, the pesticide use in the world has grown from \(\sim 1.68\) million tons in 1990 to \(\sim 2.66\) million tons in 2020, distributed as 45.5% in America, 28.5% in Asia, and 20.7% in Europe [1]. With the increasing use of pesticides to meet the food productivity demands of growing populations, concerns regarding their adverse effects on ecosystems are also increasing [2,3]. It has been reported that just less than 0.1% of pesticides applied to crops, reach the targeted pests, and the remainder enters the ecosystem, contaminating soil, water, and air, thereby potentially adversely affecting non-target organisms [4]. Human health concerns caused by food contamination due to pesticide residues have recently increased [5]. According to the Pesticide Action Network (PAN) Europe, in 2018, approximately half of the food sold on the European Union (EU) market contained pesticides that were mostly often detected in fruits for European products (67% of fruits had pesticides) [6,7]. In addition, Brazil, a major global food supplier, has authorized 475 new pesticides since 2019, and 31% of which are not approved by the European Commission. This uncontrolled pesticide use results in the contamination of the exported food products, giving rise to global health risks [8].

Pesticide residues cause health problems owing to their potentially toxic nature. According to the numbers of exposures and the time taken for symptoms to develop, the toxicity of pesticides to human beings may be acute and chronic [9,10]. Despite these consequences, the risk to human beings is considered acceptable when the pesticide residue level in and/or on crops, food, and fruits is compliant with legal limits (maximum residue level or local tolerance limit) [11]. Therefore, monitoring and detecting pesticide residues on and/or in crops and food...
products is essential for ensuring food safety and human health. Compared with 2015–2017, the detection frequency and intensity of highly toxic pesticide residues on fruits and vegetables sold in EU increased dramatically by 8.8% in 2019, as highlighted in a report from the PAN Europe [12].

Detection techniques for pesticide monitoring require key capabilities like excellent sensitivity and reproducibility. Since the 1960s, pesticide residues in food products have been detected extensively using conventional chromatographic-based techniques, such as gas chromatography (GC), liquid chromatography (LC), chromatography combined with mass spectrometry (MS), and high-performance liquid chromatography (HPLC) [13]. However, chromatography-based techniques are subject to many restrictions, including sample preparation, sample transferring to the laboratory, long waiting times for readout, the requirement of sophisticated laboratory instruments, and well-trained operators [14]. Immunoassays, electrochemical detection, and capillary electrophoresis techniques enable fast detection; however, these methods are limited by their inherent drawbacks, such as solution instability and short storage time [15]. Fluorescent probes are another promising method for the rapid on-site detection of pesticide residues, offering simple operation and fast response times. However, this fluorescent method is limited by insufficient detection limits, poor anti-interference ability against environmental factors, such as pH and free ions, and the requirement to label non-photophysically active pesticide molecules [16,17]. Enzyme-based biosensors are user-friendly and time-saving pesticide detection tools based on enzymes that can recognize and react with a target analyte to produce chemical or physical signals. However, these enzymes are expensive to produce, and their instability in non-aqueous environments limits their application of these biosensors [18,19]. Over the past two decades, surface-enhanced Raman scattering (SERS) has emerged as a promising technique for pesticide detection [15,20–23]. Compared to conventional techniques, apart from excellent sensitivity, SERS also offers high selectivity due to the fingerprinting above. The selection of multiple pesticides using Raman signals of analytes, direct detection of pesticides without any labelling techniques, simultaneous detection of multiple pesticides using the same SERS sensor, fast detection times, simple protocols, in-situ sampling, portability, and reduced costs [21,24–27]. The detection accuracy of specific substances is not affected by the detection range due to the specific SERS fingerprinting spectra of each substance. The development and applications of SERS sensors for pesticide detection in both simple and complex matrices have been widely discussed in previously published reviews [14,15,21,28]. The choice and/or fabrication of SERS substrates is the most critical aspect in experimentally testing the performance of these sensors [29]. For example, the flexible SERS substrates show promising label-free and on-site detection capabilities of pesticide residues in a convenient and facile manner [30–32]. An ideal SERS substrate requires high sensitivity, large-scale uniformity, good stability, good signal reproducibility, and a low fabrication cost [33–36]; however, high fabrication costs, low-scalability, and poor batch-to-batch reproducibility are three major challenges hindering the practical applicability of existing SERS substrates [20,34,37,38].

To address these challenges, we herein propose a novel strategy for the large-scale and low-cost fabrication of highly reproducible SERS sensing substrates. Flame spray pyrolysis (FSP), a versatile and reproducible flame aerosol technology [39–42], was employed to synthesize and thermophoretically deposit, within seconds, homogeneous plasmonic core-shell Ag-SiO$_2$ nanoparticles (NPs) on inexpensive glass substrates in a single step. The surface uniformity, limit of detection, long-term stability, spot-to-spot reproducibility, sample-to-sample reproducibility, and batch-to-batch reproducibility of the fabricated SERS substrates were investigated and verified. We then proceeded to benchmark their performance against several state-of-the-art commercially available SERS substrates, and demonstrated their feasibility in a realistic and practical application scenario, namely the detection of pesticide residues in fruit juices. By combining high performance and low fabrication costs, our developed SERS substrates have the potential of being widely employed in a variety of practical applications related to food safety surveillance.

2. Materials and methods

2.1. Chemicals

25 mL of 2-ethylhexanoic acid (99%, Sigma-Aldrich) and 25 mL of acetonitrile (>99.5%, Sigma-Aldrich) were mixed to prepare the solvent solution. Then, 3.372 g of silver acetate (99%, Alfa Aesar) was added in the solvent solution, and the reflex at the temperature of 110 °C was done for 1.5h to promote the dissolution of silver precursors. Next, 0.478 g of tetraethyl orthosilicate (98%, Sigma-Aldrich) was added to the precursor solution to achieve the 6% SiO$_2$ mass fraction in the final produced Ag-SiO$_2$ NPs. Varying the SiO$_2$ content in the Ag-SiO$_2$ NP films can tune their plasmonic coupling extinction by controlling the interparticle distances [43–45], thus optimizing the SERS performance [46–48]. Our previous study revealed that a 6% SiO$_2$ content in the final produced Ag-SiO$_2$ NPs leads to optimal SERS performance [47].

2.2. Scale-up fabrication of SERS sensing substrates in one-step

Flame spray pyrolysis (FSP) [43,49,50] was used to synthesize and subsequently deposit plasmonic Ag-SiO$_2$ NPs on temperature-controlled cover glasses. A syringe pump (New Era Pump Systems, Inc.) was used to feed the precursor solution into a capillary (KeI F 7750–12, HAMILTON) at a liquid feed rate of 5 mL/min. The precursor solution was atomized into fine droplets by oxygen (>99.5%, Strandumllen AB) at a gas flow rate of 5 L/min. The pressure drop of the dispersed oxygen across the capillary gap was kept constant at 1.5 bar to maintain a stable liquid atomization. The atomized droplets were ignited using a pilot flame to form a spray flame, where homogeneous NPs were synthesized in the gas phase [51,52]. The pilot flame was formed by premixed methane (>99.5%, AGA Gas AB) and oxygen (>99.5%, Strandumllen AB), which were fed at gas flow rates of 1.5 L/min and 3.2 L/min, respectively. The gas flow rates of the various gases were controlled using mass-flow controllers (Bronkhorst). A brass deposition holder was placed 22 cm above the flame reactor and its temperature (16 °C) was controlled using a water bath (CORIO CD-200F, JULABO). For each deposition experiment, 10 small round cover glasses (12 mm in diameter, 0.16–0.19 mm in thickness, pure white glass of hydrolytic class 1, MENZC- B0012ORAC20, Epedra) were attached on the bottom of the deposition holder using a Thermal Pad (APT2560, thermal conductivity 6 W/mK, ARCTIC). Freshly formed NPs were directly deposited on the 10 small round cover glasses via thermophoresis. The deposition time, t$_d$ = 40 s, was controlled using a movable metal shield plate, which protected the cover glasses from nanoparticles contacting.

2.3. Nanoparticle and film characterizations

The morphology and size of the generated NPs were determined using a transmission electron microscopy (TEM, Talos 120C G2, Thermo Fisher Scientific). During the TEM measurements, magnifications of 50,000x and 90,000x were used to obtain high-resolution images. For sample preparation, the produced NPs were dispersed in ethanol at a concentration of ~ 0.5 mg/mL and the suspension solution was sonicated for ~ 5 min. Two drops of the solution were then placed on a carbon film-coated copper grid (S160-4, Agar Scientific) and dried at room temperature.

The surface and cross-sectional morphologies of the NP films were characterized by a scanning electron microscopy (SEM, Aquilos 2 Cryo-FIB, Thermo Fisher Scientific). An Everhart-Thornley SE detector (ETD), a high voltage of 10 kV, and various magnifications of 1,000x, 5,000x, 30,000x and 50,000x were used during the SEM measurements.

The scanning transmission electron microscopy (STEM) images and the elemental mapping images in Fig. 1 were acquired using an
aberration-corrected transmission electron microscope (Thermo Fisher Scientific™ Themis Z, 300 kV operation voltage) equipped with a SuperX energy-dispersive X-Ray (EDX) detector (Thermo Fisher) and Quantam post-column Gatan Image Filter (Gatan Inc.). Elemental maps were generated using the hypermodal data fusion method, which combines co-registered low-loss electron energy loss spectroscopy (EELS), core-loss EELS, and EDX datasets via weighted matrix concatenation, as described by Thersleff et al [53–55].

A compact desktop powder X-ray diffractometer (Rigaku MiniFlex) was used to conduct X-ray diffraction (XRD) measurements. The 2-theta
angle ranged from 10° to 80° with a speed of 1°/min and a step width of 0.01°. Diffractions analysis was performed using the PDXL2 software to obtain the crystallite sizes of the particles, which is based on the Scherrer equation for the line broadening of the peak, i.e., the peak width is inversely proportional to the crystallite size [56].

2.4. Deposition of target molecules on SERS substrates

To evaluate the performance of the SERS substrates, Rhodamine 6G (83697, Sigma-Aldrich) was used as the Raman reporter. Specifically, 2 µL R6G/ethanol solution were dropped on the SERS substrates for R6G adsorption on the film surfaces. 10⁻⁴, 10⁻５, 10⁻⁶, 10⁻⁷, 10⁻⁸, and 10⁻⁹ mol/L R6G/ethanol solutions were used for sensitivity determination measurements, and 10⁻⁴ mol/L R6G/ethanol solution was employed for the batch-to-batch reproducibility tests.

One pesticide chemical, parathion-ethyl (>99.6%, 45607, Sigma-Aldrich), was dissolved into fresh orange juices at volume fractions of 100 ppm, 10 ppm, 1 ppm, and 0.1 ppm, which corresponded to mass fractions of 141, 14.1, 1.41, and 0.141 ppm, respectively. 2 µL of the solution were dropped onto the SERS substrates for further SERS measurements.

2.5. 2D Raman mapping measurements

The SERS maps were collected with a DXRxi Raman Microscope (Thermo Scientific, Waltham, MA, USA), using a 532 nm laser, a 10x objective lens, and a 50 µm slit. An acquisition time of 0.025 s and laser power of 2.5 mW were set. The whole area of the substrate covered with the sample was mapped, using a step size of 100 µm and 2 accumulations at each location.

The spectral data were analyzed with data analysis software developed in-house using a combination of Delphi RAD Studio (Embarcadero Technologies, USA) and Python (Python Software Foundation) [57]. The spectra were baseline-corrected using a rolling circle with a radius of 100. A rectangular area of 10 × 22 spectra was then selected as the central part of the map. The SERS spectra included in this region were considered for further analysis. The mean intensity and the standard deviation of the mean were calculated and the mean spectrum was extracted.

2.6. SERS measurements using a portable Raman spectrometer system

SERS measurements were carried out using a portable Raman spectrometer system (Ocean Insight), which included a 532 nm laser with adjustable output powers, a Raman-coupled fiber probe, a Preconfigured QEPRO for 532 nm Raman, and a Raman SERS substrate holder. Data acquisition and analysis were performed using the OceanView version 2.0. A laser power of 5 mW, an integration time of 1 s, and two scans were performed during the SERS measurements of the R6G molecules. Accordingly, for parathion-ethyl molecules, a laser power of 20 mW, an integration time of 5 s, and two scans were used.

2.7. Statistical analysis

The data preprocessing parameters, analyzed number (n) of samples, and repeat times are listed for each experiment as described in the above-mentioned parts of the Experimental Section. The quantitative data are presented as mean values ± standard deviation of the mean (SD). All statistical analyses of the Raman and XRD data were performed using the Origin software. All schematics were constructed using the Microsoft PowerPoint software.

3. Results and discussion

3.1. Scale-up fabrication of SERS sensing substrates in one-step

In one step, flame aerosol technology was employed to generate plasmonic NPs, which were deposited as homogeneous films on inexpensive glass substrates [50]. To explore the one-step large-scale fabrication of SERS substrates, round glass substrates were attached to a temperature-controlled deposition holder using a thermal pad and placed above a flame (Fig. 1a-b, and Supplementary Information, SI, Fig. S1a-b). Using this method, 10 SERS-sensing substrates were fabricated with a short deposition time of 40 s (SI, Fig. S1c). This process allowed the rapid large-scale fabrication of SERS substrates (Fig. 1b).

In this study, we achieved a laboratory production rate of 100 substrates/hour, highlighting the large-scale potential of this nanomanufacturing process.

The flame-synthesized Ag₂SiO₄ NPs were homogeneous, and their fractal morphologies were observed by TEM and STEM (Fig. 1c-d). The high crystallinity of the generated NPs is shown in the bright-field STEM image (Fig. 1e) and corresponding selected area electron diffraction (SAED) patterns (Fig. 1f). The plasmonic Ag₂SiO₄ (6 wt% SiO₂) NPs consisted of spherical Ag NPs (crystal size 12 nm, see SI, Fig. S2 for crystallinity analysis) held together by the nano-thin SiO₂, that acted as a dielectric spacer to tune the SERS hotspots (Fig. 1e, right image) [46,47,58]. The presence of SiO₂ surrounding the core Ag NPs was verified using high-angle annular dark-field (HAADF) STEM and elemental mapping imaging (Fig. 1g-l) in which the elemental composition including Si, O, C, and Ag is shown.

3.2. Surface uniformity of SERS sensing substrates

Flame aerosol NP deposition resulted in a highly uniform 3 µm thick and highly porous particle film (97% porosity, see SI for calculations), as shown in the side-view (Fig. 2a) and top-view (Fig. 2b) SEM images. High uniformity is crucial for achieving robust and reproducible sensor signals from SERS substrates, and is often one of the primary reasons for poor batch-to-batch reproducibility. To benchmark the SERS sensing capability of the developed films, the rhodamine 6G (R6G) dye deposited on the NP surfaces was used as the Raman reporter, because this molecule has a high Raman cross sections due to resonant enhancement [37]. The high porosity of the NP film is crucial in this case, as it augments the accessible surface area for R6G molecules to reach the hotspots, where the inserted Raman reporter molecules contribute the most to the SERS signals [59–61]. The nanoparticle film retained its morphology after R6G deposition and infusion within the nanoparticle film, as shown in the SEM images in Fig. 2c (top view) and 2d (side view).

3.3. 2D Raman mapping for spot-to-spot reproducibility, sample-to-sample reproducibility, and batch-to-batch reproducibility of SERS sensing substrates

To demonstrate the spot-to-spot reproducibility of the developed SERS substrates, we conducted 2-dimensional (2D) Raman mapping analysis using a Raman micro-spectroscopy and a low-concentration Raman reporter. Two µL 10⁻⁴ mol/L R6G/ethanol solution were pipetted in the middle of the SERS substrate and allowed to dry before mapping acquisition. The whole area of the substrate covered with R6G molecules was mapped using a step size of 100 µm, giving rise to ~10,000 Raman spectra (Fig. 3a). Although our substrate surface was uniform, the distribution of R6G molecules was inhomogeneous owing to the coffee ring effect [62]. To avoid this effect, a rectangular area at the center areas of each substrate (10 × 22 mm, containing 220 SERS spectra) was selected for further data analysis (Fig. 3b, and SI, Fig. S3).

The reproducibility standard deviations (RSD) of the surface sensor response of 5 substrates was ≤ 6.39% (Fig. 3f, see SI, Fig. S4 for the
The applicability of the developed SERS sensors was examined using a portable Raman spectrometer system. We collected the R6G (2 µL of 10⁻⁴ M R6G/ethanol solution) SERS spectra from 11 detection points on the same substrate from edge to center (Fig. 4a), and their intensity values at 611 cm⁻¹ were plotted (Fig. 4a,b). SERS tests using a portable Raman spectrometer system also indicated good spot-to-spot reproducibility (Fig. 4b, and see SI, Fig. S5) and batch-to-batch reproducibility (Fig. 4c and SI, Fig. S6).

The chemical stability of the SERS-sensing substrates directly determines their commercialization and affects the storage time from manufacturing to effective usage (shelf-life). Our previous study has indicated that such flame-made substrates retain their performance for up to 12 weeks when stored under ambient conditions, owing to a protective nano-thin coating made of amorphous SiO₂ [47]. To further explore the stability of our SERS-sensing substrates, we stored them under ambient conditions for 9 months. After this period, the Raman intensities at 611 cm⁻¹ remained above 80% of the intensity of freshly prepared substrates, indicating an acceptable long-term stability (Fig. 4c, see SI, Fig. S7 for the raw SERS data of 9 months storage). When compared to other Ag-based SERS substrates, such stability for several months of Ag-based SERS substrates storage under ambient conditions is quite unique [63]. Typically, Ag-based SERS substrates require storage under inert conditions (e.g., N₂ sealed), explaining why the more expensive Au is preferred as a SERS plasmonic material. The signal deterioration was attributed to the potential oxidation of the core Ag nanoparticles indicating that the SiO₂ coating made here is not hermetic. Such oxidation may be prevented via the hermetic coating of the core Ag nanoparticles using an enclosed flame reactor [43,64-66].

To assess the SERS performances of the manufactured substrates, we performed sensitivity measurements using a portable Raman spectrometer. R6G molecules were dissolved in ethanol at molar concentrations from 10⁻³ to 10⁻⁴ M, and 2 µL solutions were dropped on the SERS substrates for the Raman tests. Upon measuring the signals of different R6G concentrations (Fig. 4d) using a portable Raman spectrometer and plotting the intensity at a Raman shift of 611 cm⁻¹, our SERS sensors achieved a lowest detectable R6G concentration of 10⁻⁸ M (Fig. 4e). A polynomial fit was implemented on the Log-Log plot of the averaged peak intensities at 611 cm⁻¹ as a function of the R6G molar concentration. Our SERS sensors exhibited an enhancement factor of 10⁶-10⁸ using R6G as the analyte [47].

### 3.5. Benchmarking against commercial products

To benchmark the performance of our developed SERS substrates, we compared their SERS sensing capability for R6G with 4 commercially available SERS substrates (from Silmeco®, SERSitive®, ATOID®, and Ocean Insight®, see Fig. 5a-b, Table S1-2, and Fig. S8-11 for details). Fig. 5c shows the sensor response of all SERS substrates, with the lowest detectable concentration values ranging from 10⁻⁷ to 10⁻⁹ M, demonstrating that the sensor response of our SERS substrates was within this range. The low-cost, large-scale, and reproducible fabrication of SERS substrates via FSP renders them suitable for broad employment in practical applications. In addition, our SERS substrates offer a large active area (five times more than the other products listed in SI, Table S1, and Fig. 5a-b), thus enabling the detection of large-volume
samples. Further scale-up can be achieved using either small glass substrates or large deposition holders. For example, employing glass substrates with an 8 mm diameter enables the deposition of 25 samples in each run and further reduces the direct material cost.

3.6. Rapid and quantitative detection of pesticide residues in fruit juice

To explore the feasibility for practical applicability in food safety diagnostics, we used our fabricated SERS substrates to detect pesticide residues in fruit juices. A pesticide, parathion-ethyl, was dissolved in fresh orange juices at various concentrations (range 0.1–100 ppm). These fresh orange juices were directly squeezed from oranges and used
on the same day. Initially, 2 µL of the orange juice was dropped on the SERS substrate and allowed to dry before the Raman measurements (Fig. 6a). Raman peaks corresponding to parathion-ethyl were observed (Fig. 6b, and SI, Fig. S12-13, Table S3), indicating pesticide presence down to 0.1 ppm, demonstrating a SERS sensing performance comparable to that reported in the literature (SI, Table S4). This lowest detectable concentration of 0.1 ppm is below the tolerance level of parathion-ethyl on agricultural commodities, which is 1 ppm according
to the Pesticide Data Program from the Agricultural Marketing Service of U.S. Department of Agriculture [67]. The concentration-sensor response curve (Fig. 6c) verifies that our SERS sensors can effectively enable the rapid and quantitative detection of pesticide residues in fruit juices, thus highlighting them as suitable candidates for food safety diagnostics. The presence of common substances in orange juices such as ascorbic acid, glutathione, hydrogen peroxide and peroxynitrite was not detected when the unspiked orange juice sample was measured (Fig. 6b,
This sensing method only needs small amount of squeezed orange juice. This sensing method only needs small amount of fresh orange and large scalability. The innovation in the detection performance here that the flame nanoparticle synthesis and deposition method is attractive and promising for fabricating commercial C SERS substrates, owing to its advantages of short fabrication time, low cost, high reproducibility, and large scalability. The innovation in the detection performance here is highlighted by the utilization of a realistic sample such as fresh orange juice, demonstrating the ability of SERS sensing for label-free detection of pesticide residues in a low pH level between 3.3 and 4.2 (freshly-squeezed orange juice). This sensing method only needs small amount of sample (2 µL) and has high potential for fast and easy-to-use on-site detection of pesticide residues (point-of-consumption detection).

4. Conclusions

In this paper, we have reported a fast, single-step, low-cost, large-scale, and highly reproducible method for manufacturing SERS-sensing substrates. Within less than one minute, plasmonic Ag-SiO$_2$ nanoaggregates were deposited on several glass substrates for the rapid and large-scale fabrication of uniform SERS sensing films. The manufactured substrates exhibited good surface uniformity, high detection sensitivity, good spot-to-spot reproducibility, and high batch-to-batch reproducibility. Their ability to perform rapid and facile SERS tests in practical applications was verified through experiments using a Raman reporter and a portable Raman spectrometer system. The rapid and quantitative detection of pesticide residues in orange juices was performed using the fabricated SERS substrates, demonstrating their practical applicability in food safety diagnostics. Compared to other commercial products, our SERS-sensing substrates exhibited similar performances at a lower fabrication cost, rendering them suitable for broad employment in practical applications. Our study demonstrates that the flame nanoparticle synthesis and deposition method is attractive and promising for fabricating commercial C SERS substrates, owing to its advantages of short fabrication time, low cost, high reproducibility, and large scalability. The innovation in the detection performance here is highlighted by the utilization of a realistic sample such as fresh orange juice, demonstrating the ability of SERS sensing for label-free detection of pesticide residues in a low pH level between 3.3 and 4.2 (freshly-squeezed orange juice). This sensing method only needs small amount of sample (2 µL) and has high potential for fast and easy-to-use on-site detection of pesticide residues (point-of-consumption detection).

CRedit authorship contribution statement

Haipeng Li: Conceptualization, Nanoparticle synthesis and film deposition, Data curation, Data collection, Formal analysis, Methodology, Visualization, Writing – original draft, and Writing – editing. Elodie Dumont: 2D Raman mapping and related data analysis, Visualization, and Writing. Roman Slipets: 2D Raman mapping and related data analysis, Visualization, and Writing. Anja Boisen: 2D Raman mapping and related data analysis, Visualization, and Writing. Thomas Thersleff: Elemental mapping, Visualization, Methodology and Writing. Georgios A. Sotiriou: Conceptualization, Formal analysis, Funding acquisition, Methodology, Resources, Visualization, Writing – original draft, and Writing – editing. All authors reviewed the results and approved the final version of the manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Section S1: Scale-up fabrication of SERS sensing substrates in one-step (Fig. S1-2, calculation of film porosity). Section S2: 2D Raman mapping for spot-to-spot reproducibility and batch-to-batch reproducibility (Fig. S3-4). Section S3: Spot-to-spot and batch-to-batch reproducibility test of SERS sensing substrates (Fig. S5-6). Section S4: Stability test of SERS sensing substrates (Fig. S7). Section S5: Estimation of fabrication cost and comparison with commercial products (Table S1-2, Fig. S8-11). Section S6: Rapid detection of pesticide residues in fruit juice (Fig. S12-14, Table S3-4). References.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2023.144023.

References


