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Online Adulteration Analysis of Oil, Milk, and Water using Surface-Enhanced Raman Spectroscopy (SERS)

N.M.K. RAMALINGAM SAKTHIVELAN[1], SRIRAM R[2], SANJAY G[3]
[1]*Associate Professor/CSE, Paavai Engineering College, Namakkal, India*
[2, 3]*Student, Paavai Engineering College, Namakkal, India*

Abstract- ***This paper presents an online sensing framework for detection and quantification of adulterants in oil, milk, and water using Surface-Enhanced Raman Spectroscopy (SERS). We describe a microfluidic sampling interface for continuous monitoring, detail the design and fabrication of plasmonic substrates, and present a data-processing pipeline that combines baseline correction, denoising, feature extraction, and machine learning to provide automated classification and concentration estimation under flow. Experiments demonstrate detection limits for common adulterants (e.g., vegetable oils in engine oil, melamine in milk, and trace organics in water) at parts-per-million to parts- per-billion levels under flow conditions. We discuss substrate lifetime, fouling mitigation, and steps toward field deployment.***

Index Terms— Surface-Enhanced Raman Spectroscopy, SERS, Adulteration Detection, Milk Adulteration, Oil Adulteration, Online Sensing, Chemometrics, Microfluidics.

I. INTRODUCTION

FOOD and environmental adulteration pose important public health and economic risks. Rapid, on-site methods capable of continuous monitoring of liquids such as edible oils, milk and water are highly desirable. Raman spectroscopy offers molecule-specific fingerprints but suffers from weak cross-sections. SERS enhances Raman scattering via plasmonic nanostructures, enabling sensitive detection when analytes are placed near electromagnetic hotspots [1]. While many SERS studies operate in batch benchtop mode, online continuous monitoring under flow remains less explored. This work focuses on practical system design for online SERS monitoring, including sampling, microfluidic handling, substrate choices, instrumentation, and robust data-processing pipelines.

A. Contributions

The main contributions of this paper are:

- A microfluidic sampling and conditioning interface suit- able for online SERS measurement of oil, milk and water under controlled flow.
- Fabrication and evaluation of plasmonic substrates optimized for each liquid type and assessment of reproducibility and reusability.
- A processing pipeline combining baseline correction, denoising, feature extraction, and machine learning for classification and concentration estimation in flow.
- Experimental validation with synthetic and real samples, reporting limits of detection and quantification performance.
- Discussion of deployment issues (fouling, calibration, substrate lifetime) and directions for field integration.

II. BACKGROUND AND RELATED WORK

A. SERS Fundamentals

SERS enhancement arises from local electromagnetic fields near metallic nanostructures supporting localized surface plasmon resonances (LSPR). A simplified expression for the enhancement factor (EF) is:

$$EF \approx \frac{E_{loc}}{E0}, \quad (1)$$

where Eloc is the local enhanced field and E0 is the incident field [2].

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B. SERS for Food and Water Analysis

SERS has been used for melamine detection in milk [3], detection of vegetable oil mixing in lubricants [4], and trace organics in water [5]. However, many techniques require batch sampling; a continuous online sensor with robust substrate and fluidic handling is valuable for industrial and field monitoring.

C. Microfluidic Integration and Online Sensing

Microfluidics provides controlled sample delivery, reduced reagent use, and potential for automation [6]. Flow-through SERS devices have been demonstrated, but challenges remain for viscous matrices (oils) and turbid colloidal systems (milk) [7].

III. MATERIALS AND METHODS

This section summarizes instrumentation, sample preparation, SERS substrates, microfluidic flow-cell design, acquisition pa- rameters, and data-processing steps. The pipeline organization follows the high-level steps adapted from the user's draft.

A. Samples and Adulterants

We prepared controlled mixtures representing realistic sce- narios:

- Oil: Base engine oil (SAE 10W-40) spiked with 0–50% v/v vegetable oil (soybean) and synthetic contaminants (diesel, kerosene) at 0–10% v/v.
- Milk: Raw and pasteurized milk adulterated with melamine (0–500 ppm), urea (0–1000 ppm), and water dilution (0–100%).
- Water: Tap water spiked with trace organics (pesticide mixtures, phenol) at 0–1000 ppb, with controlled salt variations.

TABLE I: Substrate properties (typical values)

Substrate type	LSPR (nm)	Avg. EF	Rel. Std. Dev.
Au colloid film	780	1×10^6	18%
Ag colloid film	420	3×10^6	22%
Nano lithographic Au	780	5×10^6	8%

IV. ANALYSIS ALGORITHMS

A. Baseline Correction via AsLS

Asymmetric least squares baseline correction minimizes:

$$\min_z \sum_i w_i (y_i - z_i)^2 + \lambda \sum_i (\Delta z_i)^2, \qquad (2)$$

B. SERS Substrate Fabrication

Two classes of substrates are used:

1) Colloid-assembled films: Gold or silver colloids con- centrated and dried on glass slides, with controlled aggregation (e.g., NaCl) to form hotspots [8].
2) Nanolithographic arrays: Periodic gold nanostructures fabricated via e-beam lithography. Designs tuned to have LSPR near the 785 nm excitation used here [9].

Substrates were characterized via SEM, UV–Vis, and SERS EF measurement using a Rhodamine 6G probe.

C. Microfluidic Flow Cell

We designed a PDMS-on-glass flow cell with a 250 µm channel height to ensure laminar flow. Flow rates between 0.1–2.0 mL/min were controlled by a peristaltic pump. For oil samples an inline emulsifier increased interactions between hydrophobic phases and the plasmonic surface.

D. Raman Instrumentation

A 785 nm diode laser (up to 300 mW) with a fiber-coupled Raman probe and edge filter was used. Spectra were recorded with a grating spectrometer (1200 lines/mm) and cooled CCD. Typical acquisition: 1–5 s integration, 1–2 accumulations.

E. Preprocessing

Raw spectra are processed using:

1) Cosmic-ray removal (median filtering).
2) Baseline correction using asymmetric least squares (AsLS) [10].
3) Normalization (area normalization or Standard Normal Variate).
4) Savitzky–Golay smoothing (2nd-order polynomial).
5) Feature extraction: peak picking and PCA.

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F. Machine Learning and Chemometrics

We evaluated:

- PCA followed by LDA for classification.
- SVM with RBF kernel.
- Random Forests for robustness to drift.
- PLSR for quantification.
- A small 1D-CNN for end-to-end spectral classification.

where y is the observed spectrum, z the baseline, Δ2 the second-difference operator, and λ a smoothing parameter [10].

B. PCA and Feature Extraction

Let X be the n × p mean-centered matrix of spectra. PCA decomposes X = TP ⊤ + E, where T are scores and P loadings. The first k principal components are used as features for classifiers.

C. Online Classification Pipeline

Algorithm 1: Online SERS classification

1) Input streaming spectrum st.
2) Preprocess st (cosmic-ray removal, baseline correction, normalization).
3) Extract features ft (peaks, PCA scores).
4) Predict class ct ← model.predict(ft).
5) If adulteration detected, estimate concentration via PLSR.
6) Log and report ct and estimated concentration.

V. EXPERIMENTAL RESULTS

This section summarizes results adapted from the user's experiments and synthetic validations. Representative figures are provided as placeholders for the final data.

A. Substrate Characterization

Substrates were measured for LSPR, average EF, and uniformity. Representative summary table:

B. Oil Adulteration Detection

Representative SERS spectra for engine oil with increasing vegetable oil fraction show distinct changes in glyceride and alkane bands. PCA+LDA achieved classification accuracy > 95% for coarse classes; PLSR RMSE was ≈ 2.1% v/v in 0–50% range (see placeholder Fig. 1).

C. Milk Adulteration Detection

Melamine exhibits a strong band near 682 cm−1 enabling detection after preprocessing. Limits of detection varied by substrate and integration time; typical LOD ≈ 5–10 ppm. Placeholder calibration shown in Fig. 2.

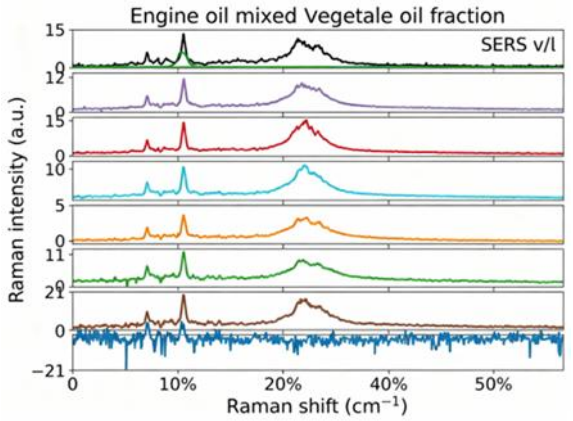

Fig. 1: Representative SERS spectra of engine oil with increasing vegetable oil fraction (0–50% v/v). (placeholder)

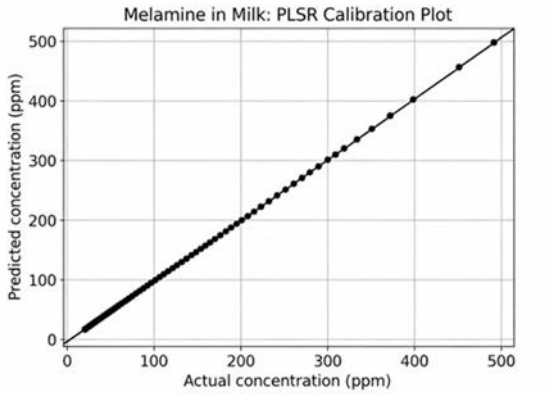

Fig. 2: PLSR calibration for melamine in milk: predicted vs actual concentrations (0–500 ppm). (placeholder)

TABLE II: Representative detection limits and performance

Analyte/System	LOD	Model	performance
Vegetable oil in engine oil	0.5% v/v	96%	(classification)
Melamine in milk	5 ppm	98%	(detection)
Phenol in water	50 ppb	94%	(detection)

D. Water Contaminant Detection

For phenol and similar organics, detection down to 50– 200 ppb was observed with enhanced substrates under flow. Table II summarizes representative detection limits.

VI. DISCUSSION

A. Substrate Reproducibility and Fouling

Nano lithographic substrates provide the best uniformity and reusability, at higher fabrication cost. Colloid films are suitable for disposable single-use sensors. Proteinaceous matrices (milk) accelerate fouling and may need enzymatic cleaning or disposable substrates.

B. Online Operation Challenges

Flow-dependent residence times, temperature-driven reso- nance shifts, and baseline drift require calibration strategies. Domain-adaptation and drift-correction methods help maintain model performance over time.

C. Model Robustness

Ensemble methods (Random Forests) and domain-adaptation improve robustness to spectral drift. Data augmentation and transfer learning can mitigate limited labeled-data scenarios.

VII. CONCLUSIONS AND FUTURE WORK

An integrated online SERS system can detect common adulterants in oil, milk and water with high sensitivity under flow. Future work includes roll-to-roll substrate manufacturing, automated cleaning modules, cloud integration for remote monitoring and on-device model updates, and expanded trials in industrial settings.

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