Application of Surface-Enhanced Raman Spectroscopy in Food Safety

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Abstract. Food safety is the primary concern because people are the foundation of a nation's strength and food is their most basic necessity. The value of food security as a global issue today captures people's attention. Food testing technologies are therefore essential for guaranteeing food safety. Traditional procedures for identifying biological, chemical, and physical pollutants in food are time-consuming, costly, and labor-intensive, and frequently cause food samples to change. These restrictions have made it necessary for the food sector to create more useful food detection systems that can accurately identify all three main categories of contaminants. Raman spectroscopy can be used extensively in evaluating food safety since it is non-destructive, easy to use, sensitive, and efficient. The advancement of Raman spectroscopy in recent years has substantially boosted its use relating to food security, enhancing the identification of contaminants in food product as well. This article describes surface-enhanced Raman spectroscopy (SERS), Raman spectroscopy, and the fundamental imaging principles. It also discusses recent developments in the identification of different material in foods. Finally, a discussion of the limitations and further potential of approaches concerning Raman spectroscopy in food security monitoring follows.

Keywords: Raman, surface-enhanced Raman spectroscopy (SERS), Food Security, Food Contaminants

1. Introduction

The lives and health of people are directly correlated with food safety. Food safety serves as both the fundamental tenet of people's existence and a crucial cornerstone of the nation's steady economic growth. Food-borne infections cause around 20 million deaths worldwide each year, and food safety has now become a worldwide issue [1]. Food safety is "an assurance that food will not damage consumers when it is produced and eaten as intended," as stated by the World Health Organization (WHO). Food additives, animal and plant natural toxins, food-borne infections and viruses, etc. are some of the main culprits in food safety issues [2]. Three categories of contaminants, including microbes (such as bacteria, fungi, and viruses), chemicals (such as toxins, allergens, and pesticides), and physical contaminants (such as plastic, glass, metals, and rocks), can unintentionally or purposefully contaminate foods throughout the food supply chain [3].

Presently, biosensors based on electrochemistry, culture plates, and other ways are the primary methods used for screening and detecting food-borne infections. Signal interference, insufficient sensitivity, or challenges with quantitative detection, however, continue to limit the precise and concurrent detection of foodborne pathogens [3]. Food deterioration and harmful bacteria need to be quickly and accurately detected.

Raman Spectroscopy, a dispersion spectrum based on the information of the matter's rotation and vibration, can reveal biometric features on the molecules [4]. A few electrons are scattered when a substance is subjected to the laser. Using the identified Raman shifts, Raman spectra are calculated. According to each Raman spike in the spectrum, there is a different chemical link, which makes it possible to identify an analyte molecularly by creating a unique vibrational fingerprint [5-7]. Surface-enhanced Raman spectroscopy (SERS) is one of the techniques frequently used for determining food safety. As a result of local plasmon resonance, which significantly boosts the intensity of the Raman spectrum. SERS is an extremely sensitive, vibration-based approach that makes the identification of molecules of a palladium nanoparticle more accurate [8]. The most current advancements in Raman spectroscopy techniques for identifying, chemical contaminants, foodborne pathogens, and physical contaminants are outlined in this article. The strategic vision of Raman spectroscopy for food security is also described, along with some current difficulties.

2. Contaminants in food products

2.1. Food borne pathogens

Several of the food pollutants affecting the global public health problem are food-borne pathogens, which include fungi, bacteria, and viruses [2]. The most prevalent foodborne strains of Salmonella, a gram-negative bacteria having approximately 2463 serovars, are Salmonella enteritidis and Salmonella typhimurium [8]. Vomiting, gastroenteritis, diarrhea, and an intense fever are the hallmarks of salmonella infection. And E. coli O157: H7 is extremely harmful since it causes hemolytic uremic syndrome (HUS) in 10% of infected patients and has a 3-5% fatality rate [9]. Additionally, P. aeruginosa can cause foodborne illness that accounts for 400 deaths annually in the US states. Furthermore, it has a high proportion of antibiotic resistance. P. aeruginosa infections cause 51,000 foodborne illnesses in the USA each year, 6,000 of which were multidrug-resistant (13%) [10]. Due to Listeria's ability to spread to humans via food products and the rise in outbreaks over the past few decades, listeria-related deaths are a typical occurrence in the food sector [10]. Another problematic Gram-positive bacterium with carbapenem resistance is MRSA. After 1-6 hours of ingestion, consuming foods contaminated with MRSA can cause severe toxin-mediated sickness, including nausea, diarrhea, gastroenteritis, abdominal discomfort, and vomiting [11].

Viruses pose a significant threat to food security and it’s widely confirmed that viruses have a close relationship with numerous instances of foodborne illnesses. Because viruses can not grow in food, their potential to spread infection through contaminated food depends on both the viability of the virus and the sensitivity of the host. The viruses that most frequently result in foodborne illnesses are the hepatitis A virus (HAV) and the norovirus (NoV). Less frequently linked to foodborne gastroenteritis are Hepatitis E virus (HEV), Aichi viruses, rotaviruses, astroviruses, and sapoviruses [12].

2.2. Chemical contaminants in food product

Antibiotics, pesticides, toxins, and adulteration are among the chemicals most frequently linked to outbreaks of different foodborne illnesses. Field soil, disinfection byproducts, the environment, personal care items, water, air, and packaging materials are just a few of the many underlying causes of these chemicals. Chemical pollutants may enter the supply chain of food accidentally or naturally owing to human behaviors. While some substances might be purposefully appended to food products for specific functions. For instance, improving appearance, color, preservation, and texture, others may happen unintentionally during the stages of preparation, manufacture, processing, and storage. All of these chemicals have the potential to seriously damage consumers' health when they are present in food products in quantities that are over the permitted limit or even at the lowest measurable level if they are fully banned owing to their character to result in negative health implications [13].

2.3. Physical contaminants in food product

Physical risks include foreign objects including wood, rocks, metal, plastic, glass, and insects that aren't frequently found in meals. They are regarded as hazardous because their size, sharpness, hardness, or shape could result in suffocation, lacerations, wounds, or punctures. In China, significant food safety incidents have been brought on by physical pollutants in foods [14]. Following a spot check of restaurants in Guangzhou in the first quarter of 2013, the Guangzhou Food and Drug Administration revealed on May 16th that the cadmium concentration of rice and rice products was higher than the recommended level of 44.4%, which caused concern among the public. Cadmium is a food contaminant that the World Health Organization (WHO) has designated as requiring priority investigation [15]. Other physical pollutants in food, such as those stated above, are equally damaging to the human body and must be recognized in addition to metal in foods.

3. Surface enhanced raman spectroscopy (SERS)

Raman scattering occurs spontaneously in one out of every 108 photons, which makes it a generally relatively weak mechanism [6]. The strength of the attainable Raman signal is constrained by this inborn weakness. The incident laser power can be increased and microscope objectives can be employed to closely concentrate the laser beam into specific tiny areas, among other techniques, to boost the Raman efficiency of an operation. However, this could lead to undesirable effects, including sample photobleaching. The measured Raman signal, also named SERS, can be amplified by orders of magnitude by using a rough metal surface to insert the analyte.

Raman, an Indian physicist, made the initial discovery that, when light travels throughout a medium, the wavelength of the scattered light differs from that of the incident light. Raman scattering, which is the study of photons with altered frequencies, can be used to determine the energy of a molecule. Raman spectroscopy's practical use is nevertheless constrained by the Raman signal's low strength in its normal condition and the difficulty of reliably identifying low-concentration sensors. Fleischman et al. discovered in 1974 that the adsorbent of pyridine molecules on the surface of specific rough Ag electrodes would result in a significant change in the Raman scattering effect. Additionally, because the adsorption of molecules on the surface of active carriers could reduce the emission of fluorescence, the intensity and quality of Raman signals were optimized, the noise resulting from Raman spectra was suppressed, and the signal stability was enhanced [16].

In SERS, metallic NPs or nanostructured metal surfaces are employed to many orders of magnitude to increase the inherently weak Raman signal (typically 106 to 1014). The rise in Raman signal given by SERS has been accounted by two different methods. The first method involves EM enhancements, in which the native electric field close to the metal’ s surface in ‘hot spots’ is focused by local surface plasmons. The second technique increases Raman scattering to around 102 times higher and involves chemical amplification through electrostatic interaction between the surface of the metal and the sample [17]. Two groups have been well divided according to various SERS substrates: metal nanoparticles built on solid matrix and colloidal plasmon resonance solutions. And it has been developed as a result of the widespread use of EM enhancements. Because of their strong plasmonic responses, gold and silver are the most often utilized materials to create SERS substrates. And chemical permanence benefits gold a lot due to its status as a metal. Additionally, research is being done on other metals, such as aluminum for UV Raman spectroscopy [18].

The labeled SERS and the unlabeled SERS are the two SERS variants available for bacteria detection. It’s convenient to apply bacterial cells and their byproducts directly to the nanostructures’ surface when using label-free SERS identification. To determine the samples, one may directly detect the intrinsic Raman spectra of the molecules inside the nanoconfined region. Without the requirement to manufacture Raman poisons and SERS tagging, the SERS spectra of bacteria can be acquired by unlabeled SERS. Raman reporter molecules are added to the labeled detection process to produce SERS signals. Generally, ligands are embedded on the surface of the nanostructures to aid substrate acquisition and facilitate specific detection including aptamers, antibodies, and similar molecules. The objectives may be recognized as using the variations in the Raman spectrum during the capturing [19].

4. Application of SERS in food safety

4.1. The establishment of simulation model

Raman spectroscopy has been utilized in several researches to categorize foodborne pathogens at the species level. SERS are frequently utilized in the identification of bacteria and viruses, both labeled and unlabeled. In the detection of bacteria by label-free method, three strategies are commonly used, namely identification of the bacterial cells, metabolites, and DNA. Additionally, the diagnosis of foodborne diseases frequently involves the detection of bacterial cells. Additionally, the diagnosis of foodborne diseases frequently involves the detection of bacterial cells. All the procedures included in detecting the bacterial by SERS can be roughly divided into three steps: inducing aggregation by combining the colloidal nanoparticles with bacteria, directly depositing the complete bacterial cell on the firm SERS substrate, colloidal nanoparticles are then built physically on the bacterium surface [19].

With the application of SERS allied with analogous soft independent modeling (SIMCA) and silver nanosubstrates, Fan et al. designed a susceptible technique to detect seven foodborne viruses by phosphate-buffered saline (PBS). Within this method, 95% of virus samples are well classified, while PCA could only classify viruses at the strain level in samples for classification and identification [20]. A novel indirect SERS approach was created by Sun et al. (2017) to identify the avian influenza virus (AIV), a contagious illness that mostly affects poultry and animals but may also affect humans. Since there exist masses of avian influenza virus strains, only H3N2 viruses can be applied to this method. It relies on a particular structure like a sandwich, which is made of influenza an immunoglobulin G (AIgG), Fe3O4/AuNPs with high SERS activity, and synthesized and characterized AuNPs with endorse and encapsulated substrate. The compounds were created by combining Fe3O4/AuNPs with AuNPs, H3N2 influenza A virus subtype, and AuNPs [21]. To separate the virus from the compound, a magnetic field requires to be generated. The effect is fulfilled by appending Fe3O4/AuNPs before putting the compound on aluminum foil.

Wei et al. successfully and consistently identified three foodborne pathogens using SERS, including Salmonella, Staphylococcus aureus, and Escherichia coli O157: H7. A specific amount of silver colloidal nanoparticles as 107 CFU/mL in this experiment, were added to all the cells for this detection, yielding a concentration of 10mL. Then they were subjected to a 785nm laser for further analysis. Based on the spectrum data, some substantial differences are recognized. Silver colloidal nanoparticles are likely to be regarded as the extremely much more susceptible SERS-active substrates [22].

Zhang et al. prepared SERS substrates based on a network of AgNPs cylindrical nanochannels and successfully achieved the spectral discrimination of two E. coli species by principal component analysis of the measured SERS spectral masses [23].

A substrate that incorporates collecting and differentiating between antibacterial qualities and bacteria alone without any other treatment was generated by Liao et al. When the LOD is around 104 CFU/mL, the produced substrates exhibited a higher ability to collect bacteria in drinkable water within one hour. They also demonstrated specific recognition of Salmonella typhimurium (S.ty) that was caught by this method. The significant antibacterial impact of the produced substrates was also a consequence of the antimicrobial property of AgNPs and the extreme exertion placed on SiNWs. Drinking water that has been salted inhibits the growth of S. aureus and E. coli at a high rate, by 81.2% and 90.7% respectively [24].

To detect E. coli in milk specifically and quickly (in less than 60 minutes), a SERS platform with an immunoassay is combined by Ilhan et al.. They can achieve 90% full efficiency against Escherichia coli in milk. Furthermore, the outcomes they acquired by SERS were in line with the conventionally used method [25].

4.2. Detection of chemical contaminants in food by SERS

Traditional spectral detection technologies include cold atomic absorption spectroscopy (CVAAS), atomic fluorescence spectroscopy (AFS), and fluorescence spectrophotometry (FC). These detection methods require expensive instruments, high-precision sample preparation, and long detection time, and are not suitable for remote or on-site detection. As a new rapid detection technology, SERS technology carries rich chemical fingerprint information with high sensitivity, low light background, no water interference, and is portable [16].

The usage of prohibited food additives will have a negative impact on social stability as well as the mental and physical health of people. Food additives can be found using Raman spectroscopy techniques. For instance, coumarin was detected using SERS and intelligent multivariate analysis using a flowery silver substrate with a detection limit of around 1.46μg/kg. Since coumarin is prone to be linked to cancer and has been revealed to cause hepatotoxicity in mice as well, it is no longer permitted as a food additive in the majority of nations [26]. Another study found the oxidant sodium expression in uncoated all-purpose wheatmeal using Raman imaging equipment with a characteristic of scanning hyperspectral. Through a hybrid analytical technique combined with a spectrum linear correlation, potassium bromate is detected in the context of unidentified contaminants (such as l-ascorbic acid, benzoyl peroxide, and azodicarbonamide) [27].

Pest control and plant growth are two goals of the use of pesticides in agriculture. Pesticide misuse or excessive use will result in lingering issues that may have varying degrees of negative effects on human health. Thiram is one of the widely used insecticides that SERS is researching, which is frequently employed in business and agriculture as a fungicide and livestock deterrent. Some researchers investigated the feasibility of SERS in detecting thiram. They conducted it on apple juice and found a detection limitation of 115 ppb trim employing dendritic silver nanostructures generated on the surface of microelectrode chips by electro-kinetic synthesis of in-solution nanoparticles. Led by Xiong et al., a study utilizes cellulose nanofibers (CNF) as SERS substrates, and CNF/AuNP nanocomposites were created for the quick and accurate identification of trimellitate amid apple juice. This technique entails cationized CNF to generate a homogenous nanocomposite by SERS and then adsorbing AuNPs on the surface via electrostatic attraction [28].

In order to cure a variety of bacterial illnesses, antibiotics are frequently employed in aquaculture and animal husbandry. Dhakal et al. created a straightforward and effective silver colloid nanoparticles-based SERS approach for immediate tetracycline (TC) residue detection in liquid diets in 2018. Silver colloid nanoparticles were produced in six batches for surface enhancement testing. Water- and milk-tetracycline solutions were made at different concentrations, and following SERS analysis, both two solutions are found with TC residue as little as 0.01 ppm [29].

4.3. Detection of physical contaminants in food by SERS

Plastic particles are defined as those having a size between 0.1μm and 5mm. Being pervasive in the environment and may be harmful to both the environment and people's health, microplastics exist everywhere, particularly in food and beverages. SERS in conjunction with a clathrite matrix was used to identify polymethyl and polystyrene methacrylate microplastics smaller than 360nm. Intense hot spots are created by the clathrite matrix, which is a complex grid of reversed pyramidal chambers made of gold [30]. SERS can be used to identify water that has been contaminated with microplastic. Raman spectroscopy was used to identify microbeads or plastic fragments in water from the tap against some impurities, including fluorescence and microscopic particles [31].

Applying SERS with silver colloidal substrates, Lv and his colleague identified microplastics made up of polypropylene, polystyrene, and polyethylene, with sizes varying from 100nm to 10μm in purified water and ocean. The detection limit of the approach is 40μg/mL, and it can detect plastic particles as small as 100nm [32]. The identification of plastic particles in liquid foods was the subject of another investigation. In this investigation, white wine was tested for the existence of microplastics using micro-Raman spectroscopy. In 24 of the 26 wine bottles, at least one microplastic was discovered [33].

5. Conclusion

As a portable, handheld, quick, accurate, and reliable identification technique method, Raman can be widely used owing to its advantages. Meanwhile, Raman spectroscopy can be used to detect food contaminants in the food production supply chain by identifying plastic particles, bacteria, poisons, environmental contaminants, pesticides, allergies, and additives in food items

However, there are still masses of issues that should be resolved before Raman spectroscopy can be applied as a more practical detection approach in the food business. Due to the complexity and multicomponent makeup of food matrices, one of the major difficulties is that competing molecules might impair the precision of quantitative assays. By creating the right aptamers or antibodies, one can increase the substrate's specificity and guarantee the quantitative detection of particular molecules. Electrostatic interactions can also enhance the adsorption and identification of charged compounds, boosting the accuracy of the assay.

On the other hand, because some pollutants may seep into the tissue, full extraction and precise identification via in situ detection or surface sampling may be challenging. In order to calculate and ascertain the actual concentration of the sample of the target analyte, models describing the relationship between penetration rate and time can be developed. Future studies should better combine efficient detection techniques to investigate more trustworthy food safety detection technologies. For instance, combining SERS with laser-induced breakdown spectroscopy can enhance identification and enable quick and precise detection of bacteria.

Another significant difficulty is the influences from fluorescence during detection, which reduces the susceptibility of the substrate and may lead to major mistakes in in-situ detection. Because graphene and graphene oxide have the function of being the efficient sub-nanometer isolation layers to distinct nanoparticles and analytes, maintaining the signal fluctuations at a low level. The graphene or graphene oxide serves to overcome fluorescence interference, aiming to maintain accurate Raman signals.

In the future, more investigations concentrated on SERS are needed, and there will be breakthroughs, which will contribute to the cause of food safety and better protect people's lives.

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