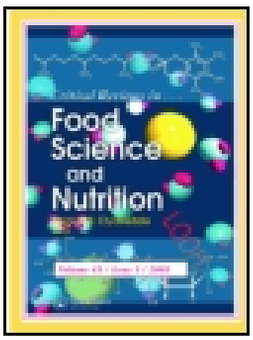

Critical Reviews in Food Science and Nutrition

ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/bfsn20

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To cite this article: Nevetha Ravindran, Sandhya Kumar, Yashini M, Rajeshwari S, Mamathi C A, Nirmal Thirunavookarasu S & Sunil C K (2021): Recent advances in Surface Plasmon Resonance (SPR) biosensors for food analysis: a review, Critical Reviews in Food Science and Nutrition, DOI: 10.1080/10408398.2021.1958745

To link to this article: https://doi.org/10.1080/10408398.2021.1958745

Published online: 30 Jul 2021.

CRITICAL REVIEWS IN FOOD SCIENCE AND NUTRITION
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REVIEW

Recent advances in Surface Plasmon Resonance (SPR) biosensors for food analysis: a review

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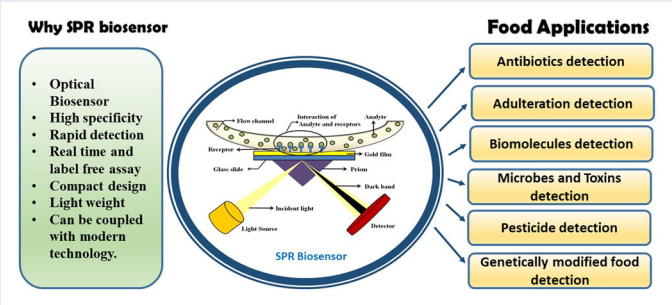
ABSTRACT

Food safety is the prime area of concern that builds trust. With the prevailing advancements, it has become facile to ensure safety in almost all aspects. Technology has grown from tedious lab techniques to modern chromatographic techniques and immunoassays, progressed with more precise and rapid sensing through the advent of Biosensors. Biosensors provide an automated technology by presenting superfast, nondestructive and cost-effective detection in food analysis. SPR biosensor is an optical biosensor known for its versatility and has wider applications in food testing and analysis. It has an optical system for excitation and interrogation of surface plasmons, and a biomolecular recognition element to detect and seize the target analyte present in a sample. The optical signal detects the binding analyte, on the recognition element, which results in a change in refractive index at the surface and modifies the surface plasmons' propagation constant. SPR aids in label-free detection of various components such as adulterants, antibiotics, biomolecules, genetically modified foods, pesticides, insecticides, herbicides, microorganisms and microbial toxins in food and assures safety. The distinct advancements of SPR in food analysis have been found and discussed. The review also provides knowledge on the advantages and the key challenges encountered by SPR.

KEYWORDS

Food analysis; food safety; label-free; optical biosensor; rapid

GRAPHICAL ABSTRACT



Introduction

Food quality and safety are a major concern among consumers. It builds pressure on the food manufacturers to meet the requirements of regulatory bodies and consumer's satisfaction. In that case, the manufacturer requires a proper food control system. Objective methods of food evaluation such as high performance liquid chromatography (HPLC), liquid chromatography–mass spectrometry (LCMS), enzyme-linked immunoassay (ELISA) and polymerase chain reaction (PCR) pose a drawback by consuming time, laboriousness and label requirement to identify the analyte. Therefore, the automation of testing methods would enhance throughput, food safety, consumer satisfaction and reduce wastages and cost. A biosensor is an automated technology that overcomes the drawbacks by presenting a rapid, nondestructive, label-free and cost-effective detection. As of late, progress in biosensors and nanotechnology for diagnostics have offered arrangements, yet generally as exploration instruments in a compact, numerous analyte diagnostics for quite a small number of samples. Present-day biosensors offer the capacity to be compact devices for regulatory bodies and the food business to check the safety use of items proposed for consumer utilization along the food supply chain. They consolidate high affinity of the biochemical interactions, bringing about high sensitivity and low detection limits. The late turn of events and studies on biosensors have pulled in much consideration since they permit continuous observation of various biomaterials' interactions (Ma et al. 2018).

An optical biosensor is a common biosensor, where optical detection occurs by using the optical field interaction

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Table 1. Comparison of SPR biosensor with other methods.

Component	Other Methods	SPR	Reference
Heavy metals	**Atomic Absorption Spectroscopy** • Destructive method • Only one sample/analysis **Inductively Coupled Plasma Mass Spectrometry** • High cost destructive method **X-ray Fluorescence Spectrometry** • Radioactive method	• Low cost • Rapid measurement technique • High sensitivity • Nondestructive	(Fen and Yunus 2013)
Food Allergens	**ELISA** • Variation occurs in some commercial kit • Detection limit – 2.5 ppm	• Highly sensitive than ELISA • Detection limit – 57.8 ng ml^{-1}	(Ashley, Shahbazi, et al. 2017)
Citrinin (Mycotoxin)	**HPLC and LC-MS** • Highly time consuming • Expensive	• Citrinin imprinted SPR - highly effective in detection & quantification • Very simple • Rapid and highly sensitive method	(Atar, Eren, and Yola 2015)
Pesticides	**LC-MS/MS** • Highly complex • Sample pretreatment required before analysis	• Nano imprinted SPR - highly precise • Less response time • Cost effective • Possess low LOD than LC-MS/MS.	(Çakır and Baysal 2019)
β-Lactoglobulin	**ELISA and LC-MS** • Variability • High cost	• β-Lactoglobulin nano-MIP SPR • Highly rapid • Real time detection • Environmental resistant	(D'Aurelio et al. 2020)
Tetrodotoxin (Fish toxin)	**LC-MS/MS, ELISA and HPLC** • High cost • Time consuming	• Highly rapid • Low detection limits compared to other techniques	(Yakes et al. 2011)

with a biorecognition element. It is simpler in configuration with low cost and skill for working and data acquisition than conventional techniques. Surface plasmon resonance (SPR) biosensor is an optical biosensor that falls into the mode of label-free detection. SPR is an optical phenomenon generated by optical coupling of thin metal. A sensible and common approach is with the aid of exciting the surface plasmon by Kretschmann. In this method, light falls through glass prism under total reflection on a metallic film vaporized onto the glass. However, in 1982 when Liedeberg and Nylander proved surface plasmon resonance as an optical biosensor, numerous advances in SPR and its programs inside the biosensing had been witnessed, especially within the closing decades. The showcasing of a gadget dependent on the SPR happened uniquely in 1990 by Biacore to recognize interactions between proteins. Later, several types of research were performed to improve the performance of the sensor (Ma et al. 2018; Karlsson 2004).

SPR biosensor is made up of two key components: optical system for excitation and interrogation of surface plasmons and biomolecular recognition element to identify and capture target analyte present in a sample where bio-molecular recognition elements are immobilized on the surface of a metal layer. It detects the binding analyte on the recognition element by means of change in refractive index within the field of surface plasmons (Miyazaki, Shimizu, and Ferreira 2017). To enhance the signal in the SPR, gold nanoparticles were used by Ferreira de Macedo et al. (2017). Some studies were also conducted to develop miniature SPR system, to make it portable (Liu et al. 2015). SPR sensors have given noteworthy focal points in label-free noninvasive detection, fast response, easy fabrication, low cost, good sensitivity, detection of multiple analytes at a time and monitors real-time biomolecular interactions. Consequently, it have been applied in biological studies, drug revelation, clinical analysis and agricultural monitoring (Ma et al. 2018; Wang et al. 2013).

Studies have explored the promising application of SPR in food analysis as it exhibits good agreement with existing methods such as ELISA, PCR, HPLC & LCMS (Wang, Dong, et al. 2018; Pilolli and Monaci 2016). A comparison of SPR biosensor with other methods is presented in Table 1. SPR is mainly used to detect biomolecules, food allergens, adulterants, microbes, toxins, antibiotic residues, pesticide and herbicide residues and genetically modified foods and characterizes their biomolecular interactions. There are reviews on different techniques employed in different domains of food analysis exclusively for allergens or mycotoxins or antibiotics which includes SPR as one of its techniques or SPR as the primary one. But this is a collective and extensive review potraying the key role of SPR in all such disciplines. It gives an overview on the principle and instrumentation of SPR and focuses mainly on the latest applications of SPR in distinct domains of food and its analysis. It also gives the future research perspective of the SPR biosensor in food analysis.

Principle

Surface plasmons (SP) are formed by exciting the free conduction electrons on a metal surface by p-polarized light. They are formed at the interface of a metal and dielectric material. These high-density electrons exhibit harmonic oscillations and the interface and, thus, are called plasmons. The metal can produce SPs efficiently, is deposited as a film on a prism, and is in contact with dielectric. The incidence

of the p-polarized electromagnetic radiation produces surface plasmons when the free electrons interact with photons present in the radiation, progressing in a wave-like manner and the metal-dielectric interface and thus can be considered as an electromagnetic wave (Kushwaha et al. 2018).

Total internal reflection occurs after a critical angle of incidence, where the incident light is assumed to be completely reflected in the same media. However, there exists a component of the incident radiation, which escapes the medium to interact with the SPs. This field is referred to as the evanescent field, formed in the medium of the lower refractive index and extending into the metal. The penetration depth of the evanescent field is a function of the wavelength of the incident light, with an exponentially decreasing intensity perpendicular to the interface. The frequency of the SPs and photons of the evanescent field, when equal, beget the resonant condition to excite the SPs. The occurrence of a dark band can characterize this event in the reflected light. This phenomenon is called surface plasmon resonance and the incident angle at the resonant condition is called the SPR angle or resonance angle (Michel, Xiao, and Alameh 2017).

The refractive index shift is proportional to the mass of the adsorbed (bound) analyte for most biological molecules (Safina 2012). The changes happening on the sensor surface due to variation in incident angle or wavelength of the electromagnetic radiation can be recorded in a sensorgram. It is a graphical depiction of the interactions occuring on the surface of the sensor, where a dip in the curve presents the occurrence of surface plasmon resonance. The SPR angle is measured in resonance units (RU) such that a shift of 1000 RU corresponds to a shift in the resonance angle of 0.1° (Hashimoto 2000). When biomolecules like proteins or DNA are used, this shift is equivalent to a change of 1 ng/mm^2 (Zhu et al. 2015). The SPR angle is dependent on parameters like metal film properties, the wavelength of the incident light and the refractive index of the dielectric material (Zamarreño et al. 2015). The shape and position of the SPR curve are influenced by the metal coated on the prism surface. Metals like silver, gold, aluminum and copper possess the potential to generate surface plasmons. Due to its reactive nature, copper is generally exempted from usage. On comparing the other metals, the dip in SPR curve is the sharpest for silver, broader for gold and the broadest for aluminum (Agrawal, Shrivastav, and Gupta 2016). Silver accounts for high sensitivity, selectivity and deeper penetration into the dielectric medium than gold, but it is preferred less due to its chemical instability (Kim et al. 2019). Using silver/gold bimetallic layers dielectric films or self-assembled monolayers (SAM) can prevent oxidation and sulfuration of silver films (Zainuddin et al. 2019; Wang et al. 2017). Most of the SPR applications sought after gold films due to chemical stability and inertness (Rodrigues et al. 2020).

The metal thickness should also be ideal. The SPR curve will be shallow and results in damping for thickness more than optimum, whereas broader for thickness lesser than optimum (Mukhtar et al. 2018). An optimum thickness of 50 nm can be used for gold films (Menon et al. 2018). The shear sensitivity of the dielectric medium's refractive index to any changes happening at the metal-dielectric interface will be exploited for bio-sensing. A shift in the resonance angle can be observed when the dielectric constant is altered. The sensor surface is sensitive to the bio-molecular interactions between the adsorbed proteins and molecules like other proteins (enzymes, antigens, cofactors and antibodies) or polymers like nucleic acids (Karlsson 2004) as the binding event occurs, the refractive index of the dielectric deviates due to the variation in mass and chemical reactions taking place on the surface of the sensor (Cennamo et al. 2018). As the interaction progresses, the binding rate gradually lowers and eventually reaches a state in which the association and dissociation process are in equilibrium (Homola 2003). The temperature of the surface can have a conspicuous impact on the refractive index. This kind of sensitivity of the SPR can be effectively employed to determine binding kinetics of biomolecules, specificity, affinity and concentration in environmental studies, food systems and the branch of biomedicine (Nguyen et al. 2015).

Modulation in SPR sensors

The categorization of the SPR sensors shall be performed depending on their sensitivity to changes in incident angle, the wavelength of the incident light, intensity, phase shift or polarization, and the angular and wavelength modulation SPR-based sensors are widespread. The principle involved is that the surface plasmon wave (SPW) propagation constant gets altered in response to any one of the modulations. The refractive index of the medium changes with respect to the propagation constant, which is recorded in the sensorgram (Guo 2012).

In angular modulation-based SPR sensors, the refractive index is a function of the varying angle of incidence of the incident light with a fixed wavelength (Zhang, Boussaad, and Tao 2003). In wavelength modulation based SPR sensors, the refractive index is a function of the wavelength of the incident light with a fixed incident angle (Homola 1997). It has an advantage over the former as the gold film can be replaced by silver as the exciting film, but the durability cannot be guaranteed (Nenninger et al. 2001). The sensitivity of the SPR sensors to changes in the refractive index increases when operated at shorter wavelengths when in angular modulation and increases with longer wavelengths when in wavelength modulation (Homola, Yee, and Gauglitz 1999).

In the intensity-modulated SPR sensor, different light sources are equipped to alter the intensity of the incident radiation. Mostly, collimated light sources, light-emitting diodes, and broadband sources are used under different light launching conditions to respond to the sensorgram. The response curve consists of a rising and decaying phase where the former can be used for high sensitivity and the latter for low sensitivity conditions. The intensity can also be improved by using nanoparticles, which impacts the operating condition and sensitivity. Compared to wavelength modulated SPR, intensity-modulated SPR is cost-effective

under industrial application conditions, and the intensity-modulated SPR can be used in the long run owing to its low operation cost.

In the phase-polarized SPR sensor, the performance of the sensor is improved by using the polarization technique. The information of the phase is derived based on spatial interference patterns using interfering of the reference beam and polarized beam (Markowicz et al. 2007). The modulation of the SPR was based on harmonics modulated frequency between the p- and s- polarization. A carrier frequency is created from a single beam of photoelastic phase modulator from which the phase quantity is extracted. It is expressed based on the modulation of the first harmonic signal (Ho et al. 2006).

Instrumentation

SPR biosensors can be used to monitor the biomolecular interactions in the food substances. In 1990, SPR sensing technology was introduced by Biacore after which many different types of instruments have been developed later (Wang et al. 2013). The SPR method is employed for various studies such as determining the solution's concentration, detecting a substance in solution, monitoring changes in the degradation of solution quality, etc. (Purwalaksana, Suendo, and Yuliarto 2019). The fundamental principle for the construction of SPR sensors is the label-free assay's resonance (Prabowo, Purwidyantri, and Liu 2018). A plasmon is defined as the oscillation of free electrons density concerning fixed ions in the metal's surface (Wammes et al. 2015). The important characteristics of SPR are sensitivity, cost-effectiveness and high specificity (Agriopoulou, Stamatelopoulou, and Varzakas 2020). The sensitivity of the SPR could be enhanced by providing secondary antibodies or nanobeads on the surface of the sensor (Wammes et al. 2015).

SPR is a sensing technique in which the incident light stimulates the resonant oscillation of electrons at the interface between positive and negative material (Ishaq et al. 2020). In SPR, a drop in the intensity of the incident light is observed at a distinct wavelength and the change in refractive index occurs when the interactions occur between the ligand immobilized on the sensor surface and the analyte present in the sample (Yakes et al. 2011). Real-time analysis of variation in the refractive index, at once, provides information about the binding of the biomolecules (Victoria 2012).

Because of its chemical stability, generally, gold is used as a noble metal in SPR based biosensors (Pechprasarn et al. 2019). The resonant oscillation of electrons is stimulated by the incident light on the metal layer (Ishaq et al. 2020). Generally, the SPR instruments are of two different configurations, namely Kretschmann and Otto configuration. Since the fabrication and usage are difficult, because of the use of high refractive index coupling, Otto type is rarely used (Zhou et al. 2019). Kretschmann configuration is widely used, and the principle involved is attenuated total reflectance (ATR) for excitation of surface plasmons (Shankaran, Gobi, and Miura 2007). The requirements for the SP excitation in Kretschmann configuration are:

1. Plane polarized incident wave in the red and infrared region should be used.
2. The thickness of the gold layer- around 50 nm.
3. Coupling prism with a high refractive index (Pechprasarn et al. 2019).

In Kretschmann configuration, the intensity of the reflected light changes and the measure of this reflected light can be used to measure the change in dielectric constant of the medium. Several new hybrid SPR sensors embracing electrochemical surface-plasmon resonance (EC-SPR), fiber-optic surface plasmon resonance (FOSPR), localized surface plasmon resonance (LSPR) and SPR imaging (SPRI) were designed by changing the different parameters like light wavelength and sensor surface (Zhou et al. 2019; Camarca et al. 2021).

The ligand immobilization on the sensor chip surface can be carried-out either internally within some systems or externally using bulk chemistry in the laboratory. SPR sensor avails several formats to monitor the binding event which produces a measurable sensor response. The most common types of format include direct detection assay, sandwich assay and inhibition assay. Inhibition assay generally quantifies smaller amounts of analyte. The serial concentrations of the analyte is used to construct a standard curve with fixed amounts of recognition elements, and unknown samples can then be measured against the standard curve. The specificity and the limit of detection (LOD) of the SPR can be enhanced by using sandwich assay format (Homola 2003; Situ et al. 2010).

The general components of an SPR biosensor are light source, detector, transduction surface – usually a gold film, glass prism, flow system and antibody/antigen binding element (Shankaran, Gobi, and Miura 2007). The light source is a major factor determining the performance, configuration, and data acquisition methodology. While designing an SPR biosensor, and the different types of light sources used are sunlight, incandescent light, a halogen lamp, light emitting diode (LED) and organic light emitting diode (OLED) (Prabowo, Purwidyantri, and Liu 2018). A tungsten lamp is used as a light source, which is connected with a collimating lens. The reflected light is again coupled and analyzed in a detector (Koubová et al., 2001). The different parameters that determine the excitation of the SPR are incident angle, the wavelength of incident light and the refractive index of the prism and medium (Zhou et al. 2019). The evanescent wave, which interacts with the plasma wave, is generated when an electron or light beam is reflected at the boundary (Wang et al. 2013). The SPR angle, apart from other parameters, is dependent on the dielectric property of the medium adjacent to the transduction surface (Shankaran, Gobi, and Miura 2007). Figure 1 gives a clear-cut idea of the instrumentation of SPR.

The carboxymethylated dextran surface provides a variety of immobilization routes via available functional groups

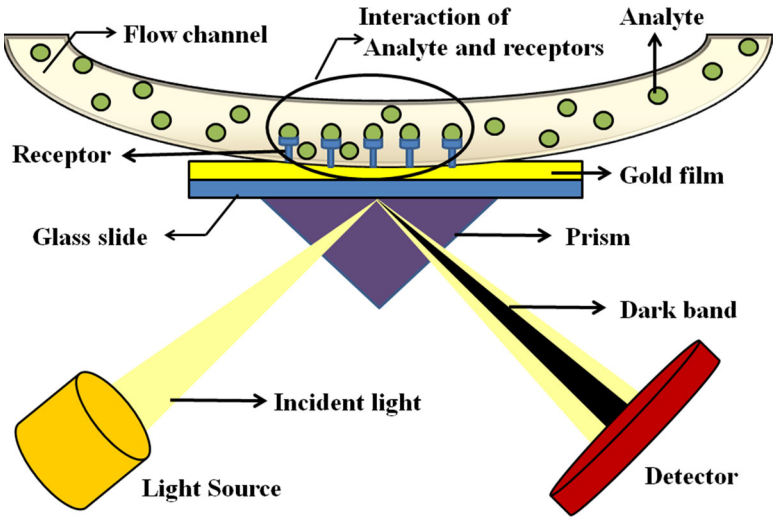

Figure 1. SPR Instrumentation.

(e.g., -NH_2, -SH, -CHO, -OH and –COOH) to analyze an extensive range of analytes, from small molecules (drugs) to large proteins and nucleic acids (Situ et al. 2010). Several approaches including reengineering light source, detection scheme and incorporating nanoparticles (NPs) promisingly offer to improve the SPR technology (Ho et al. 2017). The commercial SPR biosensors are manufactured by Biacore and updated periodically. Other establishments that manufacture SPR biosensors are Quantech and Texas Instruments (Meshram et al. 2018). Nanostructures made from noble metals have SPR effects which have been studied extensively in recent years and incorporation of those will enhance local electric field and spectral activity of the molecules in SPR. Due to this, SPR has been successfully used to detect very low level of molecules which contributed for greater application in many scientific fields (Niu et al. 2020).

Applications

SPR phenomena occur at a particular angle, modified by the introduction of the analyte on the gold film surface, thereby permitting the monitoring of binding events. Any changes in the chemical composition of the environment within the range of the plasmon field causes a change in the angle of light that resonances with the plasmons. This striking feature makes SPR an analytical tool (Yı lmaz, Saylan et al. 2017).

Detection of antibiotic residues

Antibiotics are a group of antimicrobial compounds employed to humans and animals for treating infectious diseases. The antibiotics, when applied, diffuse into the foods like milk, fish, chicken, egg, meat and honey as residues. This, in turn, will lead to several health implications such as allergy, bone marrow toxicity, hepatotoxicity, mutagenicity, carcinogenicity, reproductive disorders, nephropathy, and even anaphylactic shock in humans. Consumption of the antibiotic residues might develop antimicrobial resistance in humans, which in future cases will lead to the inefficiency of antibiotic therapy to them (Majdinasab et al. 2020). Hence, it is essential to detect the presence of these antibiotic residues in food. In the more improved world of science and technology, biosensors play a crucial role in antibiotic residue detection due to the high level of automation and its miniature size. Unlike other biosensors, SPR based biosensors, which are combined with the receptor assay, pave an innovative path for detecting antibiotics in food (Ahmed et al. 2017). A clear array of antibiotic residue detection in food by SPR biosensors is depicted in Table 2.

Antibiotics are target-specific compounds due to which molecularly imprinted polymer (MIP) biosensors can be used for detection. MIPs are a special case of polymers with a selective site for target binding. Once the target (i.e. the antibiotic) binds with the template molecule in the polymer, the cavity is filled, and it generates a signal, which is then amplified and analyzed for its presence (Tarannum, Khatoon, and Dzantiev 2020). Altintas (2018) prepared a nano MIP-SPR sensor for the detection of vancomycin in milk. When tested for vancomycin, skimmed milk samples showed a high LOD of 4.1ngmL^{-1} for direct assay (gold nanoparticle conjugated vancomycin) and 17.7ngmL^{-1} for competitive assay (between free vancomycin and AuNP-vancomycin). The sensors also had an increased level of sensitivity than that is required for antibiotic quantification. A similar kind of sensor, combining the efficiency of SPR and magnetic molecularly imprinted polymer nanoparticles (MMIP NPs), in the form of a chip was fabricated by Gao et al. (2019) to identify tetracycline in milk samples. The SPR chip was fashioned using mercaptoethylamine (MEA) for high sensitive TC detection. MEA immobilized on the chip surface adsorbed the TC-captured MMIP NPs; the higher the amount of TC captured, the higher was the SPR signal.

Enrofloxacin, sulfapyridine and chloramphenicol detection was carried out in five times diluted milk samples, where haptenized proteins acted as receptors. These proteins attached to the sensing surface covalently by a previously formed self-assembled monolayer. The LOD for enrofloxacin, sulfapyridine and chloramphenicol was found to be 1.7

Table 2. Recent applications of SPR in detection in antibiotic residues in food.

Analyte	Sample	Bioreceptor	LOD	Reference
3-methyl-quinoxaline-2-carboxylic acid	Swine tissues	Monoclonal antibody	1.4 μg kg^{-1} in swine muscle 2.7 μg kg^{-1} in swine liver	(Peng et al. 2020)
Amantadine	Animal-Derived Foods	Antibody	1.3 ng mL^{-1}	(Pan et al. 2019)
Amoxicillin	Chicken eggs	Molecularly imprinted nanofilms	0.0005 ng mL^{-1}	(Bereli et al. 2020)
Ampicillin	River water	Aptamer	1 μmol L^{-1}	(Blidar et al. 2019)
Chloramphenicol	Milk	Antibody	5.28 ng/mL	(Xia et al. 2017)
Gentamicin			2.26 ng/mL	
Enrofloxacin	Animal-Derived Foods	Antibody	1.2 ng·mL^{-1}	(Pan, Li, et al. 2017a)
Enrofloxacin	Diluted Milk	Haptenized proteins	1.7 μgmL^{-1}	(Altintas 2018)
Sulfapyridine			2.1 μgL^{-1}	
Chloramphenicol			1.1 μgL^{-1}	
Gentamicin	Milk	Antibody	4.4 ng mL^{-1}	(Xia et al. 2019)
Gentamicin	Milk	Aptamer	300 nM	(Ramalingam, Collier, and Singh 2021)
Kanamycin	Water	Chlorotetracycline coated Ag-NPs	2000 pM	(Saratale et al. 2020)
Streptomycin			120 pM	
Kanamycin	Dairy products	Aptamer	0.048 ng mL^{-1}	(Caglayan 2020)
Neomycin			0.22 ng mL^{-1}	
Sulfonamide	Animal-Derived Foods	Antibody	–	(Pan et al. 2017b)
Tetracycline	Milk	MMIP NPs	1.0 pg·mL^{-1}	(Gao et al. 2019)
Tetracycline	Honey	Aptamer	0.0069 μg/kg	(Wang, Dong, et al. 2018)
Vancomycin	Skimmed Milk	Nano MIP	4.1ngmL^{-1} (direct assay) 17.7ngmL^{-1} (competitive assay)	(Altintas 2018)

μgmL^{-1}, 2.1 μgL^{-1} and 1.1 μgL^{-1} respectively. This sensor's sensitivity was low because they were directly assayed, i.e., without any conjugation with nanoparticles, which also limited them from detecting maximum residue levels (MRLs) of some antibiotics (Altintas 2018).

An SPR immunosensor, based on immunosuppression, detected the presence of enrofloxacin in animal-derived foods, including chicken muscle, milk, pork, beef and fish. This sensor monitored the antigen-antibody reactions, i.e., the interaction between the ENRO-OVA conjugate and anti-ENRO antibody on the SPR chip surface. As the concentration of the antibody (Ab) increased, more Ab molecules bound with ligands on the chip surface. Eventually, there was a significant improvement in the SPR response, indicating the presence of ENRO (Pan, Li, et al. 2017a). Based on the similar principle of immunosuppression, a reproducible SPR immunochip was developed to detect amantadine (AM) in pure milk, chicken muscle, egg and duck samples. The binding reaction between the anti-AM absolution and the AM-BSA conjugate on the chip was monitored for detection (Pan et al. 2019). For detecting sulfonamide (SA) in animal-derived foods, an SPR immunochip was developed by Pan et al. (2017b). This sensor is based on the competition binding inhibition for SQ (a typical SA) between the SA-Ab and the SA-OVA conjugate, which was immobilized on the chip. The chip developed provided superior results when compared with other chromatographic and immunoassays for the detection of SA. Peng et al. (2020) developed an SPR based immunosensor with the monoclonal antibody 5B10 for monitoring the presence of MQCA (3-methyl-quinoxaline-2-carboxylic acid), a marker residue of Olaquindox, in the edible swine tissues. This method of SPR proved to be more efficient than the indirect competitive ELISA technique in terms of its label-free nature, LOD, sensitivity, accuracy and precision.

SPR signals were weak for detecting small molecules like chloramphenicol (CAP) and gentamicin (GEN). To overcome this, an immune inhibition assay was used by Xia et al. (2017) to fabricate an SPR immunosensor for sequential detection of CAP and GEN in milk samples. This sensor was capable of detecting CAP and GEN simultaneously using the elution strategy. In the first step, it is a common incubation of the two compounds on the chip. In the second step, i.e., the regeneration, separate detection of the analytes takes place. Regeneration involved a two-step process, where a solution primarily used, removed only GEN, while the second solution removed CAP from the chip. The sequence of unbinding from the chip generated different signals, which aided in quantifying the antibiotics. Another sensor for simultaneous detection of GEN and melamine (MEL) was developed using single-channel SPR by Xia et al. (2019). Here, both the analytes were detected rapidly with just one injection.

A label-free SPR sensor, using a specific aptamer (with high sensitivity and specificity to ampicillin) on gold chip, by pulsing electrochemical method, was developed for ampicillin (AMP) detection. When a river water sample containing AMP was injected, there was a hike in the SPR signal. This indicates that the target molecule is captured from the sample solution (Blidar et al. 2019). Research had also been done on the direct capture of tetracycline by anti-tetracycline aptamer-Apt76 in honey samples. The oriented immobilization of Apt76 on the DNA tetrahedron prevented the steric hindrance and enhanced the LOD by tenfold

Table 3. SPR in detection of Adulteration in food.

Adulterants	Food	Type of Sensor	Significant Results	Reference
Donkey meat	Meat in home sausage	SPR enhanced DNA biosensor	Fabricated SPR showed selectivity for detection of donkey gene using sandwich assay techniques	(Mansouri et al. 2020)
Glucose and fructose	Honey	Ag-graphene oxide coated fiber optic SPR probe	Probe coated with additional graphene oxide layer added sensitivity to detect adulterant at 24% and 37% for glucose and fructose respectively.	(Vikas et al. 2020)
Horse cytochrome b gene	Meat balls	Gold nanoparticle based SPR probe	Detection based on change in wavelength of color of the solution. On addition of acid, aggregation of gold nanoparticle was prevented thus producing pink color in samples with horse gene.	(Houhoula et al. 2017)
Melamine	–	SPR based optical fiber probe with electro-polymerized molecular imprinting film (MIF)	Low cost, label free, simple preparation technique with good sensitivity and specificity.	(Li et al. 2018)
Melamine	Infant Formula	Cuvette type LSPR sensor	Due to matrix effect, shift in peaks was observed with increase in melamine concentration	(Oh et al. 2019)
Melamine	Pet foods and milk products	SPR immunosensor	Sensor was highly specific to melanine and cross reaction with cyanuric acid, cyanuric chloride, and atrazine was low.	(Lu et al. 2014)
Methanol	Tequila	SPR with change in refractive index	The SPR curve generated by aged tequila is wider than white tequila and there is a larger shift in resonant angle	(Luna-Moreno et al. 2012)
Sugar	Honey	SPR sensor based on Kretschmann configuration	Pure honey has the highest refractive angle and the angle decreases with increase in adulteration along with shift in SPR curve peaks	(Zainuddin et al. 2018)
Formaldehyde	Octopus and chicken flesh	HS-SDME with Gold nano prism / Tollen's reagent sensor	Presence of formaldehyde indicated a color change due to the formation of Au-Ag np/Tollen's complex, following a redox reaction	(Qi et al. 2020)

compared with ss-Apt76-based aptasensor (Wang, Dong, et al. 2018). An aptamer based biosensor for detecting kanamycin and neomycin was developed by Caglayan (2020), by which he handed over a substitute to ELISA with his invention of SPR enhanced total internal reflection ellipsometer as the transducer. The detection limits were 0.048 ng mL^{-1} for kanamycin and 0.22 ng mL^{-1} for neomycin. Further advancements in this genre was done by Ramalingam, Collier, and Singh (2021) with the fabrication of a microfluidic paper apta-biosensor for the detection of GEN in milk samples at the rate of 300 nM in 2 mins. The sensor was reliable and quick for detection at on-farm level. Aptamer based SPR sensors were also used to detect tobramycin in drug biosensing (Spiga, Maietta, and Guiducci 2015).

Detection of adulteration

Adulteration is a common food fraud and a huge concern to the industry and consumers around the globe. Rapid procedures to perceive adulteration are released by FSSAI in India. With advanced instruments and procedures, nondestructive methods to detect adulteration in food commodities are widely employed. The SPR-based sensors working on the binding principle, can be a useful tool in assessing the food samples by binding onto specific adulterants present in them. With the advent of the latest assay techniques, adulteration can also be detected by targeting specific gene sites. Table 3 depicts the advent of SPR in detection of adulterants in food.

Many researchers have used SPR for the detection of adulteration in milk, infant foods, pet foods, pure honey, tequila and alcoholic drinks and meat sausages. Vikas et al. (2020) studied the effects of an additional layer of graphene oxide in a silver-coated fiber optic SPR probe to improve glucose and fructose sensitivity to 24% and 37%, respectively, in adulterated honey samples. Houhoula et al. (2017) carried-out a study on the adulteration of horse cytochrome b genes in meatball samples by observing a change in the wavelength of the solution with acid addition. The acid added restricted agglomeration of gold nanoparticles in the sample, thus producing pink color. The unadulterated sample produced a purple color, which absorbed the maximum wavelength at 524 nm, whereas the adulterated sample solution reported peak wavelength at $>$ 570 nm. Li et al. (2018) developed a probe based on the molecular imprinting technique. The film on electro polymerization detected melamine and the film was characterized using atomic-force microscopy (AFM) and Fourier-transform infrared spectroscopy (FTIR) techniques. SPR enhanced DNA biosensor employing sandwich DNA assays was used for the detection of sausage meat adulterated with donkey meat. This study proved to be highly selective and specific to complimentary genes and mismatch between the genes of cooked sausage and sausage adulterated with donkey meat (Mansouri et al. 2020). In another study, it was observed that with an increase in the adulteration of sugars such as sucrose, fructose and glucose in pure honey samples, the angle of incidence of SPR sensor decreased with an increase in the percentage of adulteration with a shift in the peak of the SPR curve (Zainuddin et al. 2018).

Oh et al. (2019) developed a cuvette type localized SPR sensor which can be easily handled to check melamine adulteration in infant formula such as milk powders. The developed sensor is easy to use and can be applied as a rapid detection method to check the level of adulteration in consumer samples. Luna-Moreno et al. (2012) employed an SPR technique to detect the level of adulteration of methanol in tequila and also a technique to distinguish the age of tequila.

Table 4. Recent applications of SPR in biomolecular detection in food.

Target analyte	Matrix	Bio-receptor	L.O.D/ L.O.Q	Reference
β-lactoglobulin	Bovine milk	Antibody	0.164 μg/ml	(Ashley, D'Aurelio, et al. 2018)
	–	Nano Molecularly imprinted polymer	3 ng/ml	(D'Aurelio et al. 2020)
α- casein	CIP washout samples	Nano Molecularly imprinted polymer	127 ± 97.6 ng/ml (0.127 ppm)	(Ashley, Shukor, et al. 2018)
Glycoprotein	–	Phenylboronic acid	15.56 nM	(Zhang et al. 2017)
β-lactoglobulin (milk allergen)	–	Antibody	5.54 ng/ml	(Wu et al. 2016)
Ara h1 (peanut allergen)			0.77 ng/ml	
Porcine serum albumin (pork allergen)	Pork floss, fermented ham slices, salami, air-dried sausage, Jinhua ham and raw pork	Antibody	19.81 ng/ml	(Wang, Dong, et al. 2018)
Ovalbumin (egg allergen)	Red wine	Antibody	LOD – 0.25 μg/ml Lower LOQ – 0.3 μg/ml.	(Pilolli and Monaci 2016)
Tropomyosin (Sea food Allergen)	Shellfish	Antibody	LOD – 1.0 μg/ml LOQ – 2.5 μg/ml	(Zhou et al. 2020)
Glucose	–	Enzyme	3.4 μmol l^{-1}.	(Miyazaki, Shimizu, and Ferreira 2017)
Lectin concanavalin A	–	Bovine serum albumin (BSA)-sugar conjugates	ca. 1.8 nM	(Tao et al. 2017)
Vitamin B2	Infant formula and milk samples	Molecularly imprinted polymer	0.00016 ng/ml	(Çimen and Denizli 2020)
Vitamin B9			0.00135 ng/ml	
Vitamin B12			0.00025 ng/ml	

Based on the resonant peak, it was concluded that the response peak is acquired based on the ethyl alcohol content, and thus SPR serves as a great technique to distinguish adulterated and aged tequila. Several SPR based immunosensors are developed to check the adulteration rate of melamine in milk products and pet foods (Lu et al. 2014).

One of the major food adulterants in the meat industry is infusion of formaldehyde which aids in retaining moisture to keep the food attractive for a longer period of time. Qi et al. (2020) conducted a study on headspace single drop microextraction analysis (HS-SDME) with Au nano prism/ Tollens' reagent assay on octopus and chicken flesh where color change due to formation of formaldehyde-tollens complex on redox reaction indicated the presence of formaldehyde.

Detection of biomolecules

Bio-molecules such as protein, carbohydrate, vitamin, and nucleic acid are detected or quantified using SPR biosensor. SPR approach has a great potential in monitoring interactions DNA-DNA, enzyme-substrate, protein-protein, protein-carbohydrate, ligand-aptamer, antibody-antigen and protein-lipid or lipid membranes. It characterizes the molecular interaction in food due to its ability to measure the binding affinities and kinetics between molecules (Karlsson 2004; Šakanovič, Hodnik, and Anderluh 2019). Sensorgram plays an important role in characterizing molecular interactions, and fitting the data acquired from sensorgram in kinetic models gives the values of kinetic parameters and the binding affinity. Many reviews had discussed the analysis of protein interactions, which details the SPR experimental setup and method to study affinity and kinetic measurements along with applications (Drescher, Selvakumar, and Drescher 2018; Vachali et al. 2015). SPR biosensors are used to study molecular binding dynamics from the determined thermodynamics and kinetics data (Pacheco et al. 2020). This is beneficial in the application of these molecules in food as functional ingredients and targeted drug delivery in addition to the detection of biomolecules.

Characterization of molecular interaction, for example, protein-carotenoid, is important to investigate further uptake and transport of carotenoids for targeted delivery (Vachali et al. 2015). Similarly, different kinds of molecular interactions have a significant role in the biological process. Table 4 portrays the detection of biomolecules using SPR biosensors.

Proteins

SPR possess a great emergence in protein analysis, such as detecting food allergens, milk proteins, and toxic proteins with high sensitivity, high accuracy, low cost, low sample volume, easy fabrication, and reproducibility. It is also shown for sensitive determination of amino acids, such as arginine detection, the detection was specifically based on hydrogen bond based aggregation between mercaptoundecanoic acid and arginine that leads to affect changes in local refractive index and the sensor showed a detection limit of 0.005 µM and a sensitivity of 1474 nm/µM (Fan and Wang 2020). Even the protein conjugates can be detected with an SPR sensor. Zhang et al. (2017) proposed a new fiber-optic SPR biosensor with boronic acid derivate using gold film coated with tilted fiber Bragg grating at 10° and phenylboronic acid as the recognition molecule, for the detection of glycoprotein. They reported that fiber optic SPR biosensor showed good selectivity, good regenerability and repeatability along with sensitivity up to 2.867 dB/(mg/ml) and LOD of 15.56 nM. This shows the scope of SPR biosensor for detecting glycoprotein in food, with high selectivity and sensitivity.

Food allergens are common and a major threat in food safety and consumer health. Nearly 3% of adults and 6-8%

of children was estimated to affect by food allergy. 90% of the allergic reactions caused by "big eight" allergenic foods, which includes milk, egg, peanut, soy, shellfish, fish, wheat, and tree nuts. Recent research on the detection of food allergens using SPR is gearing up due to its advantages over conventional methods (Zhou et al. 2019). Wu et al. (2016) detected allergens such as milk protein β-lactoglobulin and peanut protein Ara h1 using SPR by immobilizing the monoclonal antibody on the chip surface. Polyclonal antibodies were also injected, to increase the sensitivity. The limit of detection of β-lactoglobulin and Ara h1 using two-site antibody immunoanalytical method by SPR was 5.54 and 0.77 ng/ml, respectively, comparable to sandwich enzyme-linked immunosorbent assay. Ashley, D'Aurelio, et al. (2018) fabricated an immunoassay-based SPR sensor to identify β-lactoglobulin by optimizing pH, concentration of antibody and buffer and their results revealed better sensitivity with detection limits of 0.164 μg/ml. It was also suggested that the sensitivity could be enhanced by employing sandwich formats and amplifying with gold nanoparticles. Furthermore, the binding affinity between the polyclonal antibody and analyte was studied using Langmuir 1:1 binding model and showed dissociation constant of 2.59×10^{-9} M. Since there are still drawbacks in using antibody-based receptor because of a short lifetime and low stability in the testing environment, SPR biosensor with nano molecularly imprinted polymer were developed to detect milk allergens such as α-casein with LOD of 127 ± 97.6 ng/ml (Ashley, Shukor, et al. 2018) and β-lactoglobulin with LOD of 3 ng/ml (D'Aurelio et al. 2020). This MIP has advantages such as thermal stability and low cost and can withstand a wide pH, which overcomes antibody-based receptors' limitations.

Other allergens such as egg allergen (e.g., ovalbumin, lysozyme), seafood allergen (e.g.,tropomyosin, parvalbumin) and pork allergen (e.g.,porcine serum albumin) are also found to be detected using SPR. Pilolli and Monaci (2016) detected egg allergen (ovalbumin) in red wines by performing a purification technique combining polyvinyl-polypyrrolidone based purification and size exclusion chromatography as a pretreatment for sensitive detection and the results showed the low limit of detection of 0.25 μg/mL and lower limit of quantification of 0.3 μg/ml. It was manifested that SPR results were very close to LC-MS. In another study, Wang, Dong, et al. (2018) detected pork allergen, i.e. porcine serum albumin in pork floss, fermented ham slices, salami, air-dried sausage, Jinhua ham and raw pork using SPR biosensor with a detection limit of 19.81 ng/ml, linear range from 1.0 to 50 ng/ml and reproducibility less than 5% for both within a day and between day. It was confirmed that SPR and ELISA results were in good agreement, without any significant differences. Recently, major seafood allergen i.e., tropomyosin was quantified using SPR biosensor with gold patterned biochips in shell fish and the results reported to be L.O.D of 1.0 μg/ml and L.O.Q of 2.5 μg/ml with detection time less than 3 min (Zhou et al. 2020). Thus, SPR biosensor manifested the potential in the real-time or on-line detection of food allergens in the food and food processing environment to identify cross-contamination.

Carbohydrates

Carbohydrates, carbohydrate specific binding and glycoconjugates have been detected and studied using the SPR sensor. SPR sensor can observe the real-time carbohydrates-protein interactions by measuring the reaction rate constants and the affinity of binding. It also quantifies small molecules such as monosaccharides. The coupling of SPR with other techniques shows the ability to study detailed information about glycan molecules' structure and behavior and enhances the sensitivity of carbohydrate detection from a complex matrix (Safina 2012). Miyazaki, Shimizu, Mejía-Salazar, et al. (2017) developed an SPR biosensor for enzymatic detection of small analytes such as glucose, even at low concentrations. In this approach, a mediator-type enzyme was employed to enhance senses. Small analytes were detected through SPR signals obtained from the changes in the polarizability of mediator-type enzymes, as a result of the interaction of the analyte with the mediator-type enzyme. Glucose was detected using a glucose oxidase enzyme in this approach with the limits of detection of 3.4 μmol l^{-1}. This type of detection is highly selective for only those compounds that yield peroxide in an enzymatic reaction. In the later part, Tao et al. (2017) developed and investigated SPR-based glycan biosensors. In their study, the Bovine serum albumin (BSA) - sugar conjugates with a different number of mannose was used as ideal building blocks for sensor where it directly absorbs on gold material without any modifications. Lectin concanavalin A was detected in this sensor with a LOD of ca. 1.8 nM due to the fast binding kinetics and high affinity of BSA-sugar conjugates to lectin concanavalin A.

Vitamins

Low molecular compounds such as vitamins are also detected using SPR biosensor. Çimen and Denizli (2020) detected vitamins - B_2, B_9 and B_{12} in infant formula and milk samples using a molecular imprinted based SPR sensor. They achieved low detection limits of vitamins without significant changes in the specificity of sensor and selectively after a long period. LOD of vitamin B_2, vitamin B_9, and vitamin B_{12} were 0.00016 ng/ml, 0.00135 ng/ml, and 0.00025 ng/ml respectively. Storage stability of the sensor after three months was greater than 90% for all vitamins. They pointed out that molecular imprinted based SPR sensor shows high accuracy, storage stability, sensitivity and selectivity. In another study, a localized SPR biosensor with titanium nitride and nanostructure was developed to detect riboflavin. Titanium nitride was used as an alternative for gold for plasmonic sensing in the study. The results showed the direct absorption of riboflavin on titanium nitride nanoholes and film, and the response was higher for titanium nitride nanoholes because of high signal amplification by nanostructure (Chung 2019). Further, he introduced the novel method with biotinylated riboflavin binding protein in the titanium nitride film to detect 10000 ppb riboflavin. Prakashan et al. (2019) proposed a novel SPR based fiber-optic sensor using Au-Ag core-shell nanoparticles doped SiO_2-TiO_2-ZrO_2 ternary matrix as a sensing material to

detect Vitamin A (Retinol). SiO_2-TiO_2-ZrO_2 was selected due to its potential application in photonic devices as well as biosensors owning to have high purity and low toxicity. They reported that there were uniform shifts in transmittance intensity with an increase of vitamin A and the sensor presented good reproducibility and a low detection limit of 10 μM.

Antioxidants

Guerreiro et al. (2016) employed ultrathin molecular imprinted polymers on the sensor surface to quantify global interactions of polyphenols with proteins. Here, a complex salivary protein was immobilized and the individual polyphenol interactions with MIP supported saliva SPR biosensor exhibited specific and different binding characteristics that provide insight on the polyphenol retention. It was stated that biosensor has promising application to examine the bioavailability of polyphenols or in drug delivery application. Xu et al. (2020) inquired the interaction of amylolysis of starch with tea polyphenolic catechins using SPR. The binding studies revealed that presence of catechins reduced in vitro digestibility of gelatinized starch because of the greater degree of enzyme inhibition.

Nucleic acids

Nucleic acid detection has a substantial role in food safety. Recent researches on SPR are mainly focused on the miniaturized sensor system and enhancement of sensor performance by modification of platform/chip. Apart from SPR advantages in nucleic acid detection such as label-free, easy operation, and real-time application, there is a necessity for a high-resolution stepper motor and mechanical structure for an angle scanning system to adjust the angle. In this connection, Huang et al. (2020) developed a portable multi-angle scanning SPR for nucleic acid detection. It especially makes use of one stepping motor to rotate a belt, wherein the belt pulls the mechanical linkages of incident light and reflected light to modify the length and angle of the linkage simultaneously for accomplishing the SPR angle scanning mode that continues the incident light and reflected light equal. The overall size of the portable sensor system was 480 x150x180 mm and it possesses an angle accuracy of 0.002°, response sensitivity of 3.72×10^{-6} RIU, and an angle scanning range of 30° to 74°. In another study, a graphene-based fiber optic sensor was developed by Shushama et al. (2017) for sensing the DNA hybridization. It works on the interactions between the biomolecules and sensor by the wave created at the metal and fiber interface, which was accelerated by the introduction of graphene due to its high sensitivity to DNA. It was suggested for successful differentiation of DNA hybridization and single nucleotide polymorphisms by estimating differences of resonance wavelength and spectrum of transmitted power. Apart from the potential use of graphene in improving the sensitivity of the sensor, it was also found to reduce the other performance parameters of sensor. To increase the other performance parameters of the sensor for sensing DNA hybridization, a novel graphene-coated SPR biosensor with tungsten disulfide was proposed by Rahman et al. (2018) due to its flexibility, moderate carrier mobility, and layer-dependent electronic and optical properties and the simulation results showed tungsten disulfide was more effective compared to graphene-based SPR with a sensitivity of 95.71 deg/RIU, the quality factor of 25.19 RIU^{-1} and minimum reflectance of 3.15×10^{-2}.

In another study, Rahman, Anower, Rahman, Anower, Rahman, et al. (2017) modeled the highly sensitive MoS2-Graphene hybrid-based fiber optic SPR biosensor for DNA hybridization by using graphene, MoS_2 and Ag. In the design of sensor, the silver (Ag) was initially coated on an optical fiber substrate, followed by MoS_2 coating to increase oxidation resistance and the top layer was imposed of graphene to prevent the reaction between MoS_2 and the environment. This type of sensor accomplished high sensitivity of 105.71 deg/RIU and quality factor of 1.626 and 23.2 RIU^{-1} due to the absorption ability of graphene biomolecules and high fluorescence quenching ability of MoS_2.

Detection of genetically modified foods

The genetic composition of organisms such as plants, animals or microorganisms has been modified via way of means of genetic engineering strategies to explicit a preferred characteristic that is commonly referred to as genetically modified organism (GMO). The main contribution of this modification is to increase the throughput and to improve the nutritional and chemical properties of the resultant products. As there was the strictest legislation on GMO authorization and labeling in food by European Union, it has created a need to develop rapid detection techniques such as sensors. The main SPR strategies for GMO analysis are based on chemisorption of DNA probes onto Au chip surfaces, catalytic hairpin assembly reaction and affinity interactions between streptavidin (SA) and biotinylated probes

Using catalytic hairpin assembly reaction and AuNPs-rGO nanocomposites assisted signal amplification strategy, a novel fiber-optic SPR biosensor was developed for qualitative and quantitative detection of genetically modified foods (Chen et al. 2018). A special catalytic hairpin assembly assay made of two-base mismatches between HP1 and HP2 was employed to detect the target chain. It showed a good linear relationship with the extracted target chain from genetically modified food in the range of 0.5 to 500 nM with LOD and LOQ of 12 pM and 40 pM, respectively. In a study, Plácido et al. (2019) developed a novel label-free SPR system for quantification of genetically modified soybean by coupling biotinylated ssDNA capture probes to a sensor chip via SA-bound universal ssDNA and hybridizing to the complementary strand attached to the dextran matrix, which was observed to detect simultaneously the event-specific and the taxon-specific samples in a single chip with a detection limit of 20 pM and 16 pM respectively.

Detection of microorganisms

Food-borne illness is universal jeopardy. The vivid prevalence of food-borne diseases amidst stringent regulations and awareness programs put-forth by different countries' organizations shows the virulence of peril. Deaths incurred due to the consumption of unhygienic water and contaminated foods face headway at an alarming rate. The implications of a lack of proper nutrition and food-borne diseases are juxtaposed. According to the World Health Organization, 120000 deaths happen each year in India, with children under five years of age susceptible to food-borne diseases on a higher scale. Food is vulnerable to microorganisms like bacteria, virus, fungi and their toxins. Microorganisms like *Salmonella* spp., *Campylobacter* spp., *Listeria monocytes*, Shiga toxin-producing *Escherichia coli*, and norovirus are generally present foodstuffs. The categories of illness caused by food-borne pathogens are threefold: food infection, food-borne toxico-infections and food intoxication (Gourama 2020).

The concept of globalization led to a societal urge to try out varieties of food, and the concept of food processing harnessed preservation techniques like hurdle technology to hinder the pathogens. However, the pathogens' development of adaptive mechanisms to the processing parameters has raised the bar for achieving food safety (Woode, Daliri, and Daliri 2020). One such testimony is the study undertaken by Yang et al. (2020) where the commonly occurring antimicrobial resistant (AMR) genes and pathogens were statistically determined. The study revealed *Salmonella enterica*, *Escherichia coli* and *Shigella* as the most commonly occurring AMR pathogens in food and also that chicken happened to be a potent meat carrier of antimicrobial resistance. The techniques for detecting and quantifying food-borne pathogens include conventional techniques, molecular techniques, and immunological assays, which are either time-consuming or produce ambiguous results (Hameed, Xie, and Ying 2018). Thus, optical biosensors based on SPR principle - a label-free, pathogen-specific detection technique that is not time-intensive can opt. SPR based biosensors have the potential to quantify biomarkers like proteins, nucleic acids, adenosine triphosphate (ATP), antigens and metabolic products of the microorganisms (Jayan, Pu, and Sun 2020). A detailed discussion of the utility and limitations of different food-borne pathogen detection techniques has been discussed by Saravanan et al. (2021).

SPR based sensors are gaining demand in food applications due to higher sensitivity and real-time analysis (Wang et al. 2019). Biosensors to detect *Salmonella enteridis* (Koubova et al. 2001), *Escherichia coli* (Fratamico et al. 1998), *Camphylobactor jejuni* {Formatting Citation} were developed. Diverse applications have been proposed by Aura, D'Agata, and Spoto (2017), Bulard et al. (2015) and Xu et al. (2018) after that.

Identifying the biomarkers in microorganisms is feasible by functionalizing the sensor surface with a range of bio-recognition elements like antibodies, aptamers, enzymes, proteins, bacteriophage endolysins and Molecularly Imprinted Polymers (Jayan, Pu, and Sun 2020).

Antibody immobilized pathogens' recognition has played an indispensable role in real-time analyses (Verdoodt et al. 2017; Helali, Sawelem Eid Alatawi, and Abdelghani 2018; Raghu and Kumar 2020). The incidence of *Salmonella typhimurium* in leafy vegetables like romaine lettuce was quantified by Bhandari, Chen, and Bridgman (2019) where the *Salmonella* flagellin specific antibodies functionalized on SPR 7000 chip were used. The pre-incubation one-step sandwich assay was recommended with the highest response of order 3.2 times than the direct assay and a limit of detection (0.9 CFU g^{-1}). A novel LSPR based nano-biosensor for detection and quantification of *E. coli* O157: H7 was designed by Yaghubi et al. (2020), where spherical gold nanoparticles (S-AuNPs) were functionalized with chicken anti-*E. Coli* O157: H7 antibody. ELISA assessed the receptiveness of the nano-bio probe. Detection of the bacteria was feasible in less than 2 hours by measuring the LSPR signal's shift. The assembly's sensitivity was tested for various concentrations of the target in solution buffer to obtain a LOD of 10 CFU mL^{-1}, representing the bandwidth shift due to the changes in the refractive index. Masdor et al. (2019) developed a subtractive inhibitive assay (SIA) based on an SPR sensor for rapid detection of *Camphylobacter jejuni* from chicken samples assay was carried out with a LOD of 131 ± 4 CFU mL^{-1}.

The emergence of 2 D nanomaterials has elevated the performance characteristics of biosensing. Kaushik et al. (2019) suggested an improved assembly for effective biosensing by biofunctionalized antibodies to molybdenum disulfide (MoS_2) nanosheets via hydrophobic interactions on gold-coated fiber optical SPR biosensor. The renewed biosensor detected *Escherichia coli* with a LOD of 94 CFU mL^{-1}, efficiency and sensitivity superior to that of a conventional SPR biosensor. Mudgal et al. (2020) used an affinity layer that introduced a performance-enhanced biosensor. The sensor comprised the following layers: silver, barium titanate ($BaTiO_3$), graphene and the affinity layer was composed of nicotine, toluene and poly (trifluoroethyl methacrylate). Barium with graphene aided in performance betterment, while the graphene sensed the *Pseudomonas* with the help of the affinity layer with a LOD of 7.09 CFU mL^{-1}. The developed structure could also be applied to other microorganisms.

Aptamer-mediated biosensing can replace antibodies due to better robustness and functionality (Wu et al. 2019). An approach for aptamer functionalized LSPR sensor was worked out to reduce cost and time for pathogen detection. Oh et al. (2017) claimed to have developed an ultra-receptive and species-specific biosensing method, where the aptamer-AuNP conjugate detected *Salmonella typhimurium*, with a LOD 1.0×10^4 CFU mL^{-1} in pork meat within 30-35 minutes. This claim holds good without using pre-enrichment techniques. A portable aptamer-based LSPR plasmonic biosensor was developed by Khateb et al. (2020), accounting for suitability in real-time analysis of milk samples. The *Streptococcus aureus*-specific aptamer functionalized gold

Table 5. SPR sensors in detection of Microorganisms.

Microorganism (Analyte)	Sample Matrix	Biorecognition Element	LOD	Reference
Salmonella typhimurium	PBS solution 10% Apple juice	Det7T protein functionalized SPR chip	5×10^4 CFU mL^{-1}	(Hyeon, Lim, and Shin 2021)
Pseudomonas	Water	BaTiO3-Graphene-Affinity layer	7.09 CFU mL^{-1}	(Mudgal et al. 2020)
Listeria monocytogens	Milk	Wheat Germ Agglutinin (WGA)	3.0 log CFU/100 µl	(Raghu and Kumar 2020)
Staphylococcus aureus	Milk	Aptamer functionalized Au nanodisk	10^3 CFU mL^{-1}	(Khateb et al. 2020)
E. coli O157:H7	PBS solution	Antibody functionalized AuNP	10 CFU mL^{-1}	(Yaghubi et al. 2020)
Salmonella typhimurium	Romaine Lettuce	Antibody functionalized gold chip	0.9 log CFU/g	(Bhandari, Chen, and Bridgman 2019)
Campylobacter jejuni	Chicken	Antibodies	131 ± 4 CFU mL^{-1}	(Masdor et al. 2019)
Escherichia coli	Drinking water Orange juice	Antibody biofunctionalized MoS_2 nanosheets conjugated optical fiber	94 CFU mL^{-1}	(Kaushik et al. 2019)
Escherchia coli K12	Frozen chicken meat	Antibody	10^3 CFU mL^{-1}	(Helali, Sawelem Eid Alatawi, and Abdelghani 2018)
Escherichia coli O157:H7	Water Juice	(Magainin I-AuNP) functionalized (AgNPs-rGO) nanocomposites	5.0×10^2 CFU mL^{-1}	(Zhou et al. 2018)
Salmonella typhimurium	PBS solution	Fiber-optic probe functionalized AuNPs	128 CFU mL^{-1}	(Xu et al. 2018)
Escherichia coli	PBS solution	Antibody	1.5×10^3 CFU mL^{-1}	(Arcas et al. 2018)
Salmonella paratyphi	PBS buffer	MIP based SPR chip	1.4×10^6 CFU mL^{-1}	(Perçin et al. 2017)
Salmonella	PBS solution Chicken rinse matrix Artificially spiked chicken rinse	Antibody functionalized gold chip	2.1×10^6 CFU mL^{-1} 7.6×10^6 CFU mL^{-1} 6.8 CFU mL^{-1}	(Chen and Park 2018)
Campylobacter jejuni	PBS solution	Antibody functionalized AuNPs	Direct assay: 8×10^6 CFU mL^{-1} Sandwich assay: 4×10^6 CFU mL^{-1}	(Masdor et al. 2019)
Salmonella typhimurium	Pork meat	Aptamer functionalized AuNP	1.0×10^4 CFU mL^{-1}	(Oh et al. 2017)
E. coli O157:H7	PBS solution	Mesoporous Silica functionalized Au nanorods	10 CFU mL^{-1}	(Song et al. 2017)
Lactobacillus spp. *Streptococcus aureus*	Culture	Antibody functionalized AuNPs	105 CFU mL^{-1} 120 CFU/ml	(Verdoodt et al. 2017)
Legionella pneumophila *Pseudomonas aeruginosa* *Salmonella typhimurium*	Buffer	DNA probe functionalized AuNP	10 pg mL^{-1}	(Melaine et al. 2017)
Salmonella	Powdered milk	Capture antibody functionlized SPR chip; Detection antibody-HRP conjugate	10^3 CFU/ml	(Farka et al. 2016)
Brettanomyces bruxellensis	Wine	DNA probe functionalized AuNP	0.1 ng μL^{-1}	(Manzano et al. 2016)
Vibrio cholerae	Culture	Antibodies	43 cells mL^{-1}	(Taheri et al. 2016)
Escherichia coli O157:H7	PBS solution	Antibody	1.87×10^3 mL^{-1}	(Wang et al. 2016)
E. coli O157:H7	Hamburger Cucumber	Streptavidin functionalized (pCBAA) brushes on AuNP chip and Biotinylated antibodies	17 mL^{-1} 57 mL^{-1} 11.7×10^3 mL^{-1} 7.4×10^3 mL^{-1}	(Vaisocherová-Lísalová et al. 2016)
Salmonella sp.	Hamburger Cucumber			
Salmonella enteritidis	PBS solution	Antibody-functionalized Fe_3O_4 magnetic nanoparticles	14 CFU mL^{-1}	(Liu et al. 2016)
Salmonella typhimurium	PBS solution	Antibody	10^7 CFU mL^{-1}	(Nguyen et al. 2016)

nanodisks based sensor provided an enhanced response than the immune-based SPR sensors, with a detection limit of 10^3 CFU mL^{-1}. The microbial detection required only 120 seconds, sparing the efforts on pre-enrichment techniques. Biosensors using immobilized carbohydrate probes are of novelty {Formatting Citation}.

Antimicrobial peptides (AMPs)' ability to bind with specific bacterial species has enabled its usage in SPR biosensing. The AMPs form pores by targeting the bacterial cells' cytoplasmic membranes (Hoyos-Nogués, Gil, and Mas-Moruno 2018). A species-specific, reproducible fiber optic SPR sensor (FOSPR) employing AMP to identify pathogens was developed. For the sake of signal enhancement, silver and reduced graphene oxide (AgNPs-rGO) was used. Owing to the oxidation reaction, the conjugate was coated with gold nanoparticles (AuNPs). Magainin I was immobilized on gold nanoparticles. The sensor generated a 1.5 times better signal than the conventional sensor with a LOD of 5×10^2 CFU mL^{-1} when tested on water and juices (Zhou et al. 2018). The property of the AMPs to work as an alternative to antibiotics has attracted further studies (Qiao et al. 2020).

The invention of SPR biosensors using bacteriophage, phage receptor binding proteins, or phage-display peptides is gaining attention (Ahovan et al. 2020). As they are

naturally bacteria-ravening, possession of high specificity is apparent (Singh 2018). Hyeon, Lim, and Shin (2021) developed a novel SPR biosensor for rapid and specific detection of *Salmonella enteric* serovar Typhimurium in approximately 20 minutes. Full-length Det7 phage tail protein (Det7) was used as the bio-recognition element and was functionalized on the Biacore T200 instrument's gold surface by amine coupling. A linear range of $5 \times 10^4 - 5 \times 10^7$ CFU mL^{-1} was obtained in the buffer and 10% apple juice. The developed novel sensor overcame the drawbacks of using intact phage and mutant proteins.

Molecularly imprinted polymers is framing a niche in the field of biosensing owing to their ability to enhance sensitivity and specificity (Du et al. 2020; Kumar et al. 2020). Sener et al. (2011) employed a lysozyme imprinted SPR nanosensor to investigate the effectiveness of sensor in food matrices like chicken and egg white. For the first time, an SPR sensor chip with microcontact imprinted *Salmonella paratyphi* was made by the whole-cell. The LOD was 1.4×10^6 CFU mL^{-1} in sample buffer and was also tested with apple juice. The increased efficacy of the MIP based sensor over the conventional SPR sensors was apparent (Perçin et al. 2017).

To reduce the time and cost of detection, multiplex biosensing assays have been developed and reviewed. Improved receptivity and specificity in detecting microorganisms can be achieved by optimization (Aura, D'Agata, and Spoto 2017; Yoo, Kim, and Lee 2015). The manipulation of SPR sensors in microbial detection in food is showcased in Table 5.

Detection of mycotoxins

Mycotoxins are secondary metabolites of low molecular weight, produced from filamentous fungi, causing lethal consequences when ingested even at lower levels. Fungal species of *Aspergillus*, *Penicillium* and *Fusarium* gives rise to different mycotoxins of public concern. Foods commonly succumb to mycotoxins like aflatoxins (AF), ochratoxin (OT), zearalenone (ZEN), deoxynivalenol (DON), T-2 toxin (T-2), HT-2 toxin (HT-2), fumonisin (FB), ergot alkaloids (EA), patulin and citrinin, of which aflatoxins (B1, B2, G1, G2 & M1) are highly potent carcinogens (Eskola et al. 2020). Even though immunoassays like ELISA satisfactorily identify the minute toxin levels, biosensing methods could be used to obtain higher LOD value (Hodnik and Anderluh 2009). Table 6 displays the use of SPR sensors in mycotoxin detection.

A palm-sized SPR sensor was developed by Moon et al. (2018) for on-site aflatoxin B_1(AFB1) detection in grains. The antibody immobilized sensor surface was evaluated for its application in rice, peanut and almond. It measured a broad range of 16-200 ppb with a LOD of 2.51 ppb. Detection of AFB1 in food matrices was performed by designing a MIP-based SPR sensor. AFB1 was imprinted on gold nanoparticles (AuNPs) and was highly sensitive with a detection limit of 10 pg mL^{-1} in peanut and corn (Akgönüllü, Yavuz, and Denizli 2020). Bhardwaj, Sumana, and Marquette (2020) measured the efficiency of AFB1 antibody functionalized AuNPs in toxin detection against the conventional SPR sensor, where the former assembly had a six times lower detection limit of 0.003 nM. Wu et al. (2018) developed a highly receptive sensor exploiting streptavidin-biotin capture approach to immobilize aptamer on SPR chip with 3-minute assay time for detecting Aflatoxin B_1 and B_2, where optimization of AFB1 detection was done. Sun, Wu, and Zhao (2017) developed an aptamer-based sensor with 0.4 nM as the detection limit. Indyk et al. (2019) developed and discussed the potential of Bovine Serum Albumin and Aflatoxin M_1 functionalized SPR sensor, wherein its LOD of 0.1 ng g^{-1} was superior to European Commission's compliance regulations of 50 ng L^{-1}. The assay was accomplished in 20 min and was claimed to be promisingly stable.

A concise SPR sensor was developed and examined to quantify ochratoxin A (OTA) in coffee samples. The antibody-BSA (OTA-BSA) conjugate was immobilized on two types of nanomatrices: Chitosan (CS) and carboxymethyl cellulose (CMC) substrates with detection limits of 5.7 ng mL^{-1} and 3.8 ng mL^{-1} respectively (Rehmat et al. 2019). Similar studies were undertaken by Rehmat, Mohammed, and Anal (2018). A lab-scale SPR based sensor was developed by immobilization of ssDNA aptamer to detect OTA with a LOD of 0.005 ng mL^{-1} (Bianco et al. 2017). A strategy for simultaneous detection of mycotoxins was proposed by Wei et al. (2019). Antigen functionalized SPR chips detected AFB1, OTA, ZEN and DON. The method's ratification was performed against HPLC-MS/MS analysis with detection limit ranging from 0.59-3.26 ng mL^{-1}. Several advancements in mycotoxin detection have been performed and discussed (Agriopoulou, Stamatelopoulou, and Varzakas 2020; Joshi et al. 2016; Mahmoudpour et al. 2019; Nolan et al. 2019).

Detection of pesticides, herbicides and insecticides

Pesticides are synthetics that control bugs, weeds, organisms, and different bugs that wreck crops and incorporate an enormous number of mixes like fungicides, nematicides, bug sprays, molluscicides, rodenticides, herbicides, and plant development hormones. Pesticides are employed to restrain the undesired living species and insecticides to refrain from the insect pests. An herbicide is proposed to curb, annihilate, repulse, or relieve the development of weeds inside a yield, while fungicides are employed to restrict the growth of molds and mildew. The utilization of these mixes may present dangers to human wellbeing and the earth because their harmfulness is not continuously explicit to living beings' objective. Hence detection is very crucial to ensure safety. The improvement of quick and inexpensive determination devices for natural and biological monitoring is presently an exploration area of great intrigue. SPR biosensors have been employed generally to encounter pesticides, herbicides and insecticides due to its effortlessness, absence of prerequisite for marking and easiness of scaling down, ease, high particularity and sensitivity, and constant estimation.

Table 6. SPR sensors in detection of Microbial Toxins.

Toxin (Analyte)	Sample Matrix	Biorecognition Element	LOD	Reference
Zearalenone (ZEN)	Cereal	Aptamer functionalized SPR chip	0.08 ng mL^{-1}	(Caglayan and Üstündağ 2020)
Aflatoxin B_1 (AFB1)	Peanut Corn	MIP based SPR chip	1.04 pg mL^{-1}	(Akgönüllü, Yavuz, and Denizli 2020)
Aflatoxin B_1 (AFB1)	Wheat	Antibody functionalized Au chips Antibody functionalized AuNP chips	0.19 nM 0.003 nM	(Bhardwaj, Sumana, and Marquette 2020)
Aflatoxin M_1	Milk	Antibody functionalized gold nanorods (AuNRs)	0.11 ng mL^{-1}	(Fang et al. 2020)
α-cyclopiazonic acid	Maize Cheese	Antibodies functionalized AuNP	17 μg kg^{-1} 6 μg kg^{-1}	(Z. Hossain, Busman, and Maragos 2019)
Aflatoxin M_1 (AFM1)	Bovine milk	Bovine Serum Albumin (BSA)-AFM1 conjugate	0.1 ng g^{-1}	(Indyk et al. 2019)
Ochratoxin A (OTA)	Coffee	OTA-BSA conjugate functionalized Chitosan-Au nanomatrix OTA-BSA conjugate functionalized CMC-Au nanomatrix	5.7 ng mL^{-1} 3.8 ng mL^{-1}	(Rehmat et al. 2019)
Aflatoxin B_1 (AFB1) Ochratoxin A (OTA) Zearalenone (ZEN) Deoxynivalenol (DON)	Corn Wheat	Antigen functionalized Au chip	0.59 ng mL^{-1} 1.27 ng mL^{-1} 7.07 ng mL^{-1} 3.26 ng mL^{-1}	(Wei et al. 2019)
T-2 toxin T-2 toxin-3-glucoside (T2-G)	Wheat	Antibody functionalized AuNPs	1.2 ng mL^{-1} 0.9 ng mL^{-1}	(Hossain, McCormick, and Maragos 2018)
Aflatoxin B_1 (AFB1)	Rice Peanut Almond	Antibody functionalized Au chip	2.51 ppb	(Moon et al. 2018)
Ochratoxin A (OTA)	Buffer	(BSA-OTA Antibody) conjugate immobilized Chitosan-Au nanomatrix	2.52 ng mL^{-1}	(Rehmat, Mohammed, and Anal 2018)
Aflatoxin B (AFB)	Vinegar	Aptamer functionalized SPR chip	0.19 ng mL^{-1}	(Wu et al. 2018)
Aflatoxin B_1 (AFB1)	Buffer	Aptamer functionalized SPR chip	0.4 nM	(Sun, Wu, and Zhao 2017)
Ochratoxin A (OTA)	Sample solution	Aptamer functionalized SPR chip	0.005 ng mL^{-1}	(Bianco et al. 2017)
Ochratoxin A (OTA)	Red wine	Antibody functionalized AuNPs	0.068 ng mL^{-1}	(Karczmarczyk, Reiner-Rozman, et al. 2016)
Aflatoxin M_1	Milk	Antibody functionalized p(HEMA) based AuNPs	18 pg mL^{-1}	(Karczmarczyk, Dubiak-Szepietowska, et al. 2016)

According to a study by Shrivastav, Usha, and Gupta (2016), it was reported that a novel method has been developed for detecting profenofos using surface plasmon resonance and molecular imprinting techniques in conjunction with optical fiber technology. The sensor's operating profenofos concentration range is 10^{-4} g/l to 10^{-1} g/l, with a maximum sensitivity of 12.7 nm/log (g/l) at 10^{-4} g/l profenofos concentration. Beside high affectability and selectivity, the proposed sensor has various different benefits like invulnerability to electromagnetic obstruction, quick reaction, minimal expense, and the capacity to screen and recognize analytes online and distantly because of the probe's fabrication on optical fiber.

SPR sensors were coordinated with fabricated MIP nanofilms for sensitive, particular, quick and continuous identification of numerous pesticides, including cyanazine (SNZ), simazine (SMZ) and atrazine (ATZ). The MIP nanofilms onto the SPR gold surfaces are developed using UV polymerization reactions, which comprise of N-methacryloyl-L-phenylalanine methyl ester (MAPA) as a functional monomer, 1-vinyl imidazole (VIM) as a co-monomer, and ethylene glycol dimethacrylate (EGDMA) as a cross-linker. The real-time estimations on the SPR sensor give a recognition run from 0.10 to 6.64 nM and denoted LOD estimations of 0.095, 0.031 and 0.091 nM SNZ, SMZ and ATZ, respectively. SNZ, SMZ and ATZ imprinted SPR sensors were prepared onto the gold surface of the SPR chip and were utilized as a contender molecule to inspect the selectivity of the pesticide imprinted SPR sensors. The contender pesticide solutions (6.64 nM) were applied to both pesticides imprinted and non-imprinted SPR sensors. The pesticide engraved SPR biosensors were also ready for real-time detection of these pesticides from aqueous solutions. In the light of acquired information, the pesticide imprinted SPR sensors can be utilized as a potential choice to identify pesticides and ensure ecological and human wellbeing (Saylan et al. 2017).

An SPR affinity sensor system using molecularly imprinted nanoparticles (plastic antibodies) was found to enhance pesticide detection. Molecular imprinting-based affinity sensor was used in a study, where atrazine (the selected model pesticide) imprinted nanoparticles were attached onto the gold surface of the SPR chip and the recognition element was a polymerizable form of aspartic acid. FTIR and zeta-sizer were used to characterize the imprinted nanoparticles. To examine the atrazine recognition ability of the SPR nanosensor, atrazine, and two structurally related compounds, simazine and amitrole solutions (1.0 ng/ml), introduced the atrazine imprinted chip surface. Simazine was selected because of the similarity to the atrazine and amitrole selected due to its smaller size than atrazine, which will offer ease of entry to the imprinted nanocavities. The responses of atrazine imprinted nanosensor to atrazine, simazine and amitrole solutions at the same concentration (1.0 ng/ml) were compared to examine the selectivity and

Table 7. SPR sensors in detection of Pesticide, Herbicide and Insecticides.

Pesticide / Herbicide/ Insecticide	Type of Sensor	Detection Limit	Reference
Atrazine	Plastic antibody based SPR nano sensors	Atrazine imprinted SPR sensor was 8.167 and 25.48 times more selective for atrazine.	(Yılmaz, Ozgur, et al. 2017)
Azoxystrobin, Boscalid, Chlorfenapyr, Imazalil, Isoxathion, and Nitenpyram	(SPR) immunosensor with microarray type sensor chip	MRL – 3 mg/, 5 mg/kg, 1 mg/kg, 0.5 mg/kg, 0.01 mg/kg, and 1 mg/kg respectively	(Miyake et al. 2019)
Boscalid, Clothianidin and Nitenpyram	Immunosensor reliant on SPR	MRL- 15 – 93 ng mL^{-1}, 6.7 – 27 ng mL^{-1} and 7.3 – 62 ng mL^{-1} respectively.	(Hirakawa et al. 2018)
Cyanazine (SNZ), Simazine (SMZ) and Atrazine (ATZ)	SPR sensors was coordinated with fabricated molecular imprinting nanofilms	LOD- 0.095, 0.031 and 0.091 nM respectively	(Saylan et al. 2017)
Napropamide- Herbicide	SPR Sensor with Polypyrrole Chitosan Graphene Oxide layer	LOD- 0.1 ppm.	(Sadrolhosseini et al. 2018)
Triazophos	SPR immunosensor	LOD- 0.096 ng mL^{-1}	(Guo et al. 2018)

the imprinting efficiency. The ΔR of atrazine imprinted nanosensor against atrazine, simazine and amitrole solutions at the same concentration (1.0 ng/mL) was measured as 0.637, 0.078 0.025, respectively. Atrazine imprinted SPR sensor was 8.167 and 25.48 times more selective for atrazine concerning simazine and amitrole, respectively (Yı lmaz, Ozgur, et al., 2017).

On an analysis of 3 pesticides, boscalid, clothianidin and nitenpyram, an immunosensor that is reliant on SPR, were produced instead of the direct competitive enzyme-linked immunosorbent assays (dcELISAs). The SPR immunosensor comprised of microflow-type instrument, and its sensor chip had four channels coated with carboxymethyl dextran. The standard pesticide solutions were prepared with 10% methanol. Monoclonal Antibodies (MoAbs) were diluted to 15 μg ml^{-1} with high ion strength phosphate-buffered saline containing 0.2% BSA. The pesticides and conjugates of them were designed with the apprize blends of carboxy and bovine serum albumin were immobilized onto separate channels of a similar sensor chip. The anti-boscalid MoAb, the anti-clothianidin MoAb, and the anti-nitenpyram MoAb solutions were injected separately into the SPR immunosensor. At the point when a blend of 3 monoclonal antibodies responded to every pesticide, and three pesticides were infused into the SPR immunosensor, each channel demonstrated explicit reactivity at 15-93 ng mL^{-1} for boscalid, 6.7-27 ng ml^{-1} for clothianidin, and 7.3-62 ng ml^{-1} for nitenpyram. Recovery tests utilizing vegetables spiked with a blend of 3 pesticides indicated great outcomes: 75-90%, 88-104%, and 72-105%, separately, with a high connection to aftereffects of the dcELISAs. Therefore, SPR immunosensor would be valuable for distinguishing pesticide deposits in vegetables. The constituted SPR immunosensors were combined for the analysis of pesticides at the same time and thus, the study indicated that the developed SPR immunosensor could be applied to the simultaneous analysis of the three pesticides: boscalid, clothianidin, and nitenpyram (Hirakawa et al. 2018).

An easy, fast, and sensitive diagnostic method was proposed to trace triazophos via a noncompetitive direct SPR immunosensor. The study was characterized by SPR-based kinetic analysis. The two anti-triazophos monoclonal antibodies were immobilized on the sensor chip, and the MoAb having the relatively slow dissociation rate was used for direct immunosensing of triazophos. The biosensor assay showed high specificity and a low detection limit of 0.096 ng mL^{-1} to triazophos, with the linear detection range of 0.98–8.29 ng $mL^{-1,}$ and the sensor chip could be reused for >160 cycles. The recommendable outcomes showed that the recently created immunosensor could be utilized as a quick, advantageous, and dependable device to regularly screen triazophos and meet the maximum residue limits (Guo et al. 2018).

In another research, the PPy-Chi/GO layer was employed to quantify the napropamide dissolved in hexane in the range of 0.01 to 100 ppm utilizing the surface plasmon resonance procedure; the constraint of the sensor was 0.1 ppm. SPR setup based on Kretschmann configuration was employed. Polypyrrole- chitosan/graphene oxide layers, in the range of 8 to 60 nm thickness, were prepared by electrodeposition. The XRD pattern of the PPy-Chi/GO layer established the formation of PPy-Chi in the presence of GO. The PPy-Chi/GO composite layer (sensing layer) was attached to a high index prism. PPy-CHi/GO sensing layer could detect the napropamide and the angle shift was reported in the range of 0.012° to 2.003° and detection limits of napropamide was about 0.1 ppm (Sadrolhosseini et al. 2018).

In an examination, six pesticides such as azoxystrobin, boscalid, chlorfenapyr, imazalil, isoxathion, and nitenpyram, were distinguished by utilizing an SPR immunosensor with microarray type sensor chip. The sensor chip utilized was a glass crystal of which the surface was covered with a slim gold layer changed with alkanethiol having the carboxy group esterified by 100 N-hydroxysuccinimideu. The standard solutions of pesticides and its mixture were prepared along with samples preparation. The maximum residue limit in tomato were reported to be 3 mg/kg for azoxystrobin, 5 mg/kg for boscalid, 1 mg/kg for chlorfenapyr, 0.5 mg/kg for imazalil, 0.01 mg/kg for isoxathion, and 1 mg/kg for nitenpyram (Miyake et al. 2019).

A surface plasmon reverberation biosensor for the pesticide carbendazim is portrayed that has improved execution because of the utilization of an Au/Fe_3O_4 nanocomposite as an enhancing mark on a surface of the carboxymethyldextran-covered gold layer of the sensor. There was a linear

response in the 0.05 to 150 ng·ml^{-1} carbendazim fixation range, with LOD of 0.44 ng·ml^{-1}. This Au/Fe_3O_4 nanocomposite-based method has a lot of potential for detecting other small analytes at very low concentrations (Li et al. 2019). The list of pesticide residue detection by the SPR technique is shown in Table 7.

SPR affinity sensors

Surface plasmon resonance includes a device that binds specific target molecules from the toxins produced by a food sample to the analyte based on affinity and selectivity is termed as SPR affinity sensors. These devices employs biorecognition elements such as nucleic acids, proteins, antibodies, metabolic products, etc., to furnish necessary binding action. The surface plasmon contains a metal surface onto which the biorecognition element is immobilized. When a suitable analyte mixture is in contact with the sensor, binding occurs at the interface, thus increasing the refractive index at the contact surface (Wolfbeis et al., 2008).

SPR affinity sensors for detection of analyte and pesticides present in food samples using MIP technique was carried out by Lautner et al. (2011) and Yao et al. (2013). Scarano, Mariani, and Minunni (2016) concluded that label-free affinity sensors could be used in real matrices for in-line measurement as they are portable and have high selectivity and sensitivity for use in food analysis. MIP based sensors are used under high selectivity and high loading capacity of the analyte in food samples (Ashley, Shahbazi, et al. 2017). Bacterial isolation using WB-SELEX was studied by Ahn et al. (2018) where high binding affinity was observed with the aptamers and sensor surface. SPR biosensor based on silver, barium titanite, graphene and affinity layer was developed by Mudgal et al. (2020) aiding in the detection of Pseudomonas bacteria with high sensitivity of 220 degrees/RIU.

Apart from food analysis, SPR aptasensor is used in the medical field to detect human chorionic gonadotropin in serum samples (Chiu, Kuo, and Chen 2019). Capoferri et al. (2018) reviewed the application of affinity-based sensors to detect pesticides, and Mahmoudpour et al. (2019) reviewed toxin detection in food samples. Hybrid SPR was developed by Rahman, Anower, Hasan, et al. (2017) for sensing DNA hybridization and mathematical modeling to increase performance parameters' effectiveness. Taheri et al. (2016) studied the affinity between a recombinant surface antigen (OmpW) to detect Vibrio species as contaminants. The study proved to be highly sensitive with microbial cells with LOD 50 cells/ml.

Advantages of SPR

SPR Biosensor is 2.5 times comparatively superior compared to other types of optical biosensors (Majdinasab et al. 2020). It has potential applications in different fields like pharmaceutical research, medical diagnostics, environmental monitoring, food safety, and security (Homola 2003). SPR imaging is a tool used for the high-throughput analysis of biomolecular interactions, such as proteomic analysis, drug discovery, pathway elucidation and biomarker screening (Wang et al. 2013). It is quintessential in many chemical, physical and bio-sensing applications because of its light-weight, compact design and label-free sensing and remote sensing competency (Bhatia and Gupta 2012). They also help food industries draw confident food safety-related decisions quickly, detect potentially harmful residues in food or detect the presence of beneficial substances and vitamins in the food, etc. (Pechprasarn et al. 2019). The SPR biosensors can be used for quantitative detection of biomaterials present in food substances with high sensitivity, rapidly and will be label-free (Ishaq et al. 2020). SPR can also be used in food quality analysis for the detection of adulterants in food products. High-performance SPR sensors can be used to detect small and medium-sized analytes in concentrations even up to 1 ng/ml (Homola, Yee, and Myszka 2008). It serves as a favorable platform for quantitative analysis of therapeutic antibody concentrations or antigen-antibody binding in desired products (Ho et al. 2017). SPR sensing surfaces can be functionalized according to the user-defined task and used to quantify different glycoforms, and even monosaccharides can be quantified using modern SPR sensors (Safina 2012). SPR can be used to detect multiple mycotoxins present in foods and have potential application prospects (Wei et al. 2019). It also aids in food allergen and pathogen detection in real-time and label-free fashions using less reagent consumption. These biosensors can be employed for the analysis of food allergens in different food matrices for identifying, capturing, and quantifying target food allergens by the techniques of FOSPR, SPRI and LSPR (Zhou et al. 2019).

SPR analysis neither requires intense sample preparation (Wammes et al. 2015) nor requires frequent washing for its working (Meshram et al. 2018). The biomolecules captured on the SPR sensors' surface can be recovered and analyzed using a Mass Spectroscopy (MS); this led to rapid identification and surface characterizations of proteins even at smaller levels (Situ et al. 2010). SPR sensors have a long shelf-life compared to the chemicals used in other analysis techniques (Pechprasarn et al. 2019). On enhancing the quality of the components like the development of robust biomolecular recognition elements with high specificity and long storage life, integration of SPR sensor platforms with microfluidic devices and multianalyte, coupling with other advanced and emerging technologies and application-specific sampling systems, the capability of the SPR sensors can be improved (Shankaran, Gobi, and Miura 2007).

Limitations of SPR

The application of SPR is very less in commercial purposes due to its affordability and different factors namely cost and need for special care and maintenance. The need for the development of low cost SPR becomes the need of the hour which can provide results with high precision and accuracy (Prabowo, Purwidyantri, and Liu 2018). The major drawback of traditional SPR is simultaneous multiple allergen

detection and analysis in food samples (Zhou et al. 2019). However, it can be overcome by integration of SPR to various sensing platforms which leads to robust biomolecular recognition with high specificity and long storage life. Several studies are being carried out to enhance performance along with miniaturization of SPR sensors with low production cost and easy portability. Still, the performances of SPR biosensor in complex samples are poor, especially due to the interference of nonspecific bindings to the output signals and they have to be enhanced (Shankaran, Gobi, and Miura 2007). The recent researches of SPR in analysis of complex samples proved it has less sensitivity and analytical application which acts as a major hindrance for its further development (Chen et al. 2017). The general limitations in sensitivity and design of SPR sensors limits the application of SPR in food industry, the improvement of these parameters can make SPR a potential tool in detection of food allergens and food borne pathogens (Lan et al. 2008).

Conclusion

In recent years, SPR biosensor has shown great potential to achieve outstanding research on food analysis. SPR biosensor exhibits high throughput, versatility, robustness, real-time, high sensitivity, high affinity in molecular interactions and low limit of detection. In commercialization aspects of SPR technology, the main challenges of the current SPR-based biosensor are bulk instrument and high in cost. There is a great scope for novel development of sensor platforms and their design and novel applications in SPR, ensuring their commercial feasibility. Hence, the development of SPR biosensors could reach the market and adapt in food industries for rapid in-line and on-line food analysis.

Acknowledgements

We express our profound gratitude to Indian Institute of Food Processing Technology for supporting us to do this review study. We also thank our parents and fellow beings for their constant support and encouragement. All the co-authors have contributed equally for the article.

Disclosure statement

No potential conflict of interest was reported by the authors.

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