Versatile Polyaniline-Based Polymers in Food Industry

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Intrinsically conducting polymers (ICPs) have been widely studied in various applications, such as sensors, tissue engineering, drug delivery, and semiconductors. Specifically, polyaniline (PANI) stands out in food industry applications due to its advantageous reversible redox properties, electrical conductivity, and simple modification. The rising concerns about food safety and security have encouraged the development of PANI as an antioxidant, antimicrobial agent, food freshness indicator, and electronic nose. At the same time, it plays an important role in food safety control to ensure the quality of food.

Keywords: polyaniline ; intrinsic conducting polymer ; active and intelligent food packaging

1. Biodegradable Food Packaging

Biodegradable food packaging is a current trend due to the rising awareness of environmental sustainability, and it is further encouraged by governmental policies, such as the Plastic Tax (European Union), Plastic Packaging Tax (United Kingdom), and Climate Change and Principle-Based Taxonomy (Malaysia). It was recorded that about 584 million tonnes of plastic waste was generated, and packaging waste accounted for over 50% of global plastic waste. Therefore, the replacement of single-use plastics with biodegradable plastics is deemed to overcome plastic pollution in both terrestrial and aquatic ecosystems [1][2]. Biopolymers such as starch, cellulose, and chitosan are widely studied for the replacement of synthetic polymers in food packaging. Nonetheless, these biopolymers are often associated with drawbacks such as hydrophilicity, poor mechanical properties, weak thermal resistance, intrinsic electrical insulation, susceptibility to wet environments, and high cost of production [3][4][5]. PANI is able to improve upon the chemical and mechanical properties of biopolymers so that they can be more feasible for applications in food packaging [6][7][8]. At the same time, studies have shown that biopolymer matrices retain their biodegradability behaviour after the incorporation of polyaniline (PANI) [9][10][11][12]. In fact, the addition of PANI improves the biodegradability of low-density polyethylene film (LDPE) via oxo-biodegradation [13]. The combination of PANI with biopolymers not only contributes to the packaging industry, but also has great potential in a broad array of applications, including sensors, supercapacitors, solar cells, dye removers, and optical devices [14][15][16][17][18].

The hydrophobicity of PANI and the hydrogen linkage interactions between PANI and the matrix reduce the free hydrogen groups for the binding of water molecules [19][20]. Therefore, the incorporation of PANI into chitosan film reduces water solubility and water vapour permeability by approximately 30% and, at the same time, lowers the transmission of light (transparency), allowing the chitosan film to be used in the packaging of foodstuffs that are easily oxidised under light [21]. The strong interaction between PANI and chitosan also improves the processability and tensile strength of biofilms [22][23].

Additionally, the encapsulation of biodegradable polymers by PANI and the cationisation of PANI are used for the enhancement of thermal stability. PANI is able to enhance the thermal stability of whey protein isolate (WPI), starch, cellulose nanofibre (CNF), cellulose nano-whiskers (CNWs), and polylactic acid (PLA) films [24][25][26][27][28]. PANI may also exhibit electrical conductivity and anti-static properties in intrinsically insulating biopolymers, making it a good material for anti-static packaging. Although the electrical conductivity of PANI-based biopolymers is often lowered when compared to that of pristine PANI, nonetheless, they are still suitable for application as anti-static materials. However, the addition of PANI into highly oriented CNF and PLA results in the weakening of tensile strength [9][26]. It is important to note that the poor processability, solubility, and fusibility of PANI are major drawbacks that have to be overcome with suitable blending routes for the preparation of homogenous biodegradable packaging films [29].

2. Active Food Packaging

PANI is a potential antioxidant material due to its radical scavenging capability [30][31][32]. In the form of emeraldine salt, PANI possesses nitrogen atoms that are capable of electron transfer [33][34][35]. Specifically, the stabilisation of peroxyl radicals depends on the donating ability of the hydrogen atoms [36]. Its antioxidant activity is expected to greatly contribute to active food packaging applications, since it is reported that PANI has a comparable antioxidant activity to that of well-known antioxidants, such as catechin and ascorbic acid [37][38].

In addition, PANI has been proven to be active against various fungi and bacteria, such as *Aspergillus niger, Escherichia coli*, *Pseudomonas aeruginosa*, *Bacillus cereus*, *Salmonella typhimurium,* and *Staphylococcus aureus* [39][40][41][42]. Its quaternary ammonium structure renders it strong antibacterial and antifungal properties. The biological activity of PANI arises from its electrical conductivity, which could mediate contact on the surface of a bacterial cell via electrostatic adherence [43][44][45]. So, it is more active against Gram-negative bacteria. In addition, the hydrophobic benzene ring on PANI interacts with the membrane core of bacteria, causing membrane permeabilisation. Membrane disruption eventually leads to cell lysis due to the leakage of cellular components and the potential breakdown of the membrane. In addition, PANI may induce oxidative stress on microorganisms through the production of hydroxyl radicals (H_2O_2), leading to the Fenton reaction. In this reaction, free ions accelerate the formation of H_2O_2, causing cell destruction [39].

Several studies on the antioxidant and antibacterial activities of PANI have been conducted for the purpose of active food packaging [22][46][47]. PANI was proven to improve the mechanical properties, electrical conductivity, and antimicrobial activity of pure chitosan film [23]. PANI-coated PMMA/CNC showed 45% inhibition of DPPH after 240 min and was active against *B. cereus* and *S. typhimurium* [48]. On the other hand, chitosan/PANI exhibited slight antioxidant strength. PLA/PANI film strengthened with CuO and ZnO also performed well as an antibacterial film against *S. aureus* and *E. coli*. The film was proven to slow down the microbial growth (total aerobic bacteria and acidophilus bacteria) in orange juice, thus preserving the quality of orange juice and resulting in a longer life span [49].

3. Intelligent Food Packaging

Microbial growth and metabolism are major causes of food spoilage, and they result in the formation of amines, sulfides, alcohols, aldehydes, ketones, and organic acids with unpleasant and unacceptable off-flavours [50][51][52]. Trimethylamine (TMA), dimethylacetamide (DMA), decomposed urea, and amino acid are released in the form of ammonia during food spoilage by bacteria [53]. Therefore, ammonia and TMA are common marker gases for indicating food spoilage [54][55][56]. They are usually released during the spoilage of high-protein foods, but can also be released by spoiled vegetables, including spinach, seaweed, and corn [57][58].

Moreover, *E. coli* is a very common food-borne bacterial pathogen found in the gut of cattle. *E. coli* often leads to food-borne illnesses such as haemolytic uremic syndrome and haemorrhagic colitis. In addition, due to the extreme climatic change, COVID-19 pandemic, and Russia–Ukraine war, people are alarmed about global food insecurity and are concerned about solutions for tackling food waste [59]. According to the Food Waste Index Report 2021 by the United Nations (UN), in 2019, approximately 931 million tonnes of food waste (17% of global food production) was generated. Therefore, it is crucial to develop a rapid and reliable technique for the detection of *E. coli* in foodstuffs. An effective and simple colourimetric sensor can be utilized to detect the presence of these gases within food packaging to monitor the quality of perishable foods in a real-time manner and reduce food waste [54].

As a stimulus-responsive polymer, PANI is able to conduct electricity and is sensitive to pH changes through protonation and deprotonation of its central axis. It has been widely studied as a colourimetric indicator of food freshness [60][61][62]. Starch/PANI film was studied as an ammonia sensor for the purpose of the indication of food spoilage. During food decomposition, ammonia vapour was released, and its interaction with PANI changed it from a green emeraldine salt into a blue emeraldine base. This starch/PANI colourimetric sensor exhibited a limit of detection (LOD) as low as 245 ppm and a relative standard deviation (RSD) of 8.72% [24]. PANI was also specifically applied to food samples as a real-time food freshness indicator. PANI/TPE showed a good linear response to TVB-N within the concentration range of 25.2 mg to 100 g and was able to indicate the spoilage of live red drum (*Sciaenops ocellatus*) fish through its colour change from emerald green to peacock blue [63]. A similar study was also performed to evaluate the freshness of tilapia, where doped PANI film acted as a colourimetric sensor, even under chilled conditions (4 °C), and was able to be recycled up to three times [64]. On the other hand, *E. coli* in milk and butter was detected by PANI through colour changes due to its interaction with the acidic product of *E.coli*'s glycosidic pathways, such as succinate, acetate, lactate, and malate [65].

Furthermore, the changes in PANI's electrical conductivity due to gases released during food spoilage make it a simple yet effective food freshness indicator. This is due to the flexible polar bond rotation, which eventually modifies the PANI chains through the structure of complex charge transmission, resulting in AC conductivity [66]. There was a more recent study in which a PANI/silver nanowire/silk composite was applied as a resistometric microsensor for the detection of TMA, and it was capable of indicating the freshness of pork. The high sensitivity (LOD: 3.3 μg/L), good stability, and repeatability (up to five cycles) made this composite a great potential freshness indicator for pork [67]. In addition, in a PLA/ZnO/CuO film, PANI played a significant role in the estimation of the shelf life and expiration date of orange juice, where it yielded an accuracy higher than 90%. As the spoilage of the juice began, gases were released, and they applied pressure to the smart film as they accumulated in the packaging. The pressure applied caused a change in the electrical conductivity of the film, thus indicating food spoilage [49]. However, the greatest challenge of PANI in intelligent food packaging is that the electroconductivity of PANI is easily affected by humidity, leading to the false detection of target molecules [68]. Moreover, the lack of an evaluation of its toxicity and the imposed risk of the migration of substances are stumbling blocks on the path toward PANI-based food packaging [69][70].

4. Food Safety Control

Antibiotic contamination from the wastes of hospitals, the pharmaceutical industry, and human and animal excretion often occurs in an aquatic environment, where prolonged unintended exposure towards these antibiotics promotes antibiotic resistance in humans [71][72][73]. Therefore, a PANI-nanofibre-coated U-bend optical-fibre-based sensor is a useful tool for monitoring β-lactam antibiotics in food. During the enzymatic hydrolysis of β-lactam, protons and acidic by-products, such as penicillinoic acid (from penicillin) and ampicilloic acid (from ampicillin), are released, leading to a pH change that converts PANI from the form of an emeraldine base into that of an emeraldine salt, which is measured by the increase in wave absorbance at 435 nm. This optical fibre sensor is able to detect penicillins and cephalosporins in food, including milk, chicken, and water, with an LOD as low as 0.18 nM [74]. PANI nanowires have also been synthesised into molecular-imprinted (MIP) sensors, where the nanowires were electro-polymerised onto a gold electrode and then imprinted with chloramphenicol as a molecular template. It turned out that a PANI MIP sensor was able to detect chloramphenicol at levels as low as 10^{-7} mM [75]. In addition, magnetic mesoporous PANI coated with hydrophilic monomers and casein for solid-phase extraction coupled with HPLC was able to simultaneously determine a wide array of antibiotics in milk samples with recoveries as close as 100%. These included doxycycline, oxytetracycline, trimethoprim, and penicillin G [76]. Similarly, PANI/GO was used for the electrochemically controlled SPME of antibiotics—specifically, tetracyclines—in milk samples, with recoveries ranging from 71% to 104% [77].

Non-steroidal anti-inflammatory drugs (NSAIDs) are common veterinary medicines, and NSAID residues in animal-originating foodstuffs adversely affect human health, causing cardiovascular diseases, gastrointestinal ulceration, kidney toxicity, and platelet aggregation inhibition [78]. Solid-phase extraction (SPE) with a PANI/PAC nanofibre mat was used in the extraction of NSAIDs, as the amino and benzene ring groups of PANI rendered it a strong affinity towards NSAIDs via a hydrogen bond, π–π interaction, its acid–base function, and its hydrophobicity. This showed practical feasibility with meat and egg samples and was able to detect a wide array of NSAIDs (ibuprofen, naproxen, diclofenac, carprofen, ketoprofen, tolfenamic acid, and salicylic acid) with LODs and recoveries in the ranges of 0.6–12.2 µg kg^{-1} and 85.18–107.31%, respectively [79]. Other similar studies on the extraction of NSAIDs using PANI have also been done in recent years [80][81][82].

Heavy metal ions, such as lead, chromium, cobalt, and copper, are severely toxic to human health, even at low dosages [83][84][85]. In order to separate, enrich, and detect metal ions at trace levels, SPE is one of the most selective and sensitive techniques. Electrically or magnetically assisted SPE eliminates the use of eluent by changing the surface of the conducting sorbent, thus simultaneously improving the extraction efficiency. Due to its reversible redox and electroactivity properties, PANI is an excellent candidate as the conducting sorbent of electrically assisted SPE [86]. Moreover, the abundance of imine and amine functional groups in PANI also enhances the adsorption of heavy metal ions [87][88]. A PANI nanofibre–graphene oxide (GO) sorbent was developed to extract Pb^{2+}, and it achieved an LOD, RSD, and reproducibility of 0.04 µgL^{-1}, 1.97%, and 2.51%, respectively [89]. Another similar study was performed by using SiO_2-coated GO/PANI/Polypyrole (PPy) in magnetic SPE of Cr(III) and Pb(II) with LODs of 4.808 and 3.401 ngL^{-1}, respectively [83]. In addition to SPE, free-standing PANI composites have also been studied in the adsorption of heavy metals from aqueous samples [90][91][92][93]. Although volume changes in PANI—either swelling or shrinking—may occur during electrochemical cycling, thus adversely affecting its stability, most of the PANI-based sensors reported for food safety control have exhibited reliable stability with good reproducibility [81][84][89].

5. Electronic Noses

The discrimination of aromas is important for the determination of the freshness, quality, and safety of food. The π-conjugated PANI chains capable of electron delocalisation make it suitable for application as a sensitive layer for gas sensors. A HCl-doped PANI electronic nose system’s potential effectiveness was proven in detecting and distinguishing several aromas, and it showed the best sensitivity towards grapes (112%) [94]. In another study, an electronic nose with PANI-layered gold-interdigitated microelectrodes (IDEs) was also able to analyse artificial aromas found in gummy candies by detecting aromas with concentrations as low as 900 ppb, as it had good reversibility (97.6%) [95][96]. Another similar study in situ involved the polymerisation of PANI onto a graphite-interdigitated electrode to monitor the release of aromas (apple, strawberry, and grape) from gummy candies. It showed that an electronic nose with camphor sulfonic acid (CSA)-doped PANI had the best sensitivity towards artificial aromas [97]. On the other hand, a PANI/functionalised single-wall carbon nanotube was developed into an electronic nose for the detection of ammonia vapours to monitor the freshness of beef [98].

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