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## Recent developments and applications of nanomaterial-based lab-on-a-chip devices for sustainable agri-food industries

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## ABSTRACT

**Background:** Lab-on-a-chip (LOC) devices have attracted considerable scientific attention due to their ability to incorporate multiple complex analytical processes onto a single chip. Such miniaturised devices can reduce most large-scale laboratory processes to small chips.**Scope and approach:** This review discusses the recent developments and applications of nanomaterial-based LOC devices for sustainable food and agricultural industries. First, we present a brief introduction to this topic. We then highlight the applications of nanomaterial-based LOC devices in the food and agriculture industries. In the subsequent section, we discuss the advantages and disadvantages of such devices in food screening. Finally, we conclude the review by providing the future perspectives of this promising field for detecting and monitoring important analytes in food and agricultural products.**Key findings and conclusion:** Due to the miniaturisation of the entire assay, a minute sample is needed to perform the complete analysis quickly, thereby increasing the efficiency of the overall process. Thus, by exploiting the unique electrical, optical, and physical properties of the nanomaterials onto such LOCs, several properties of the sensing process can be improved, including the ability to selectively label the target analytes and thereby improve the overall sensitivity of the process. Such nanomaterial-based LOC devices have considerable potential in identifying nucleic acid, proteinic, and cellular components from complex food and agricultural samples with high specificity and, therefore, can be applied in the continuous monitoring of multiple agri-food analytes to ensure sustainability and food safety.

## 1. Introduction

With rapid urbanisation and constant advancements in science and technology, the previous few decades have seen the agricultural and food sector change being transformed by a large degree. Though science has positively contributed towards the improvement and efficiency of production to meet the rapidly rising demands of food with the growing population, there is a higher price to pay owing to the health complications with the excessive use of chemicals such as pesticides and contaminants. Because of the presence of hazardous elements within the environment and the ecosystem, there exists a massive threat to food

safety (Hu et al., 2019). In addition, food wastage has tremendously increased over the past decade. According to Beg et al., more than 1.3 billion tons of food gets wasted yearly (Baig, Al-Zahrani, Schneider, Straquadine, & Mourad, 2019), which is only expected to increase in the coming years. Moreover, with unmonitored food contamination, food spoilage because of bacterial or other external agents also leads to the wastage of food materials. Food spoilage also produces deleterious effects on human and animal health and harms the country's economy (Sridhar et al., 2022). Therefore, rapid detection and prevention of food contaminants are crucial to ensure agricultural, food, and health safety. Rapid identification of agri-food biomarkers is crucial to expedite

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analysis. It also facilitates the development of reliable and advanced detection systems capable of achieving hastened response rates and delivering precise outcomes during the detection process. This imperative not only promotes time efficiency, but also establishes a foundation for state-of-the-art detection methodologies.

In the recent decade, there has been an increase in the demand for the development of inexpensive analytical methods that are sustainable and eco-friendly. Conventional methods like chromatographic and spectroscopic techniques are commonly used to rapidly detect toxic compounds in food and agricultural products (Guo, Feng, Fang, Xu, & Lu, 2015). Despite the convenient compatibility and reliable results, these methods rely on skilled professionals and costly equipment. Therefore, the scientific community has now geared toward developing better and novel methods to help overcome the persistent complexity of agri-food systems. New methods involving microfluidic chips, micro-pumps, and microvalves have recently been introduced (Sridhar et al., 2022). These inventions have paved a path toward the development of lab-on-a-chip (LOC) technology that permits automatic biosensing process control and is relatively inexpensive, portable, and easy to handle. LOC is a small-scale device that permits multiple laboratory processes to function over a miniaturised portable system. The size of such devices ranges from a few centimetres to millimetres and consists of microfluidic channels holding small volumes of target analyte samples ranging from nano to microliters (Sridhar et al., 2022). Thus, a LOC platform is a unique diagnostic tool that helps analyse and treat DNA or protein-related samples.

Any LOC platform generally integrates two fundamental fields: microfluidics and molecular biology (Elveflow, 2022). This platform is widely accepted within the scientific community owing to its unique advantages like inexpensive availability, small sample requirements, easy portability, and easy integration with conventional detection methods like polymerase chain reaction (PCR) (Zhu, Fohlerová, Pekárek, Basova, & Neužil, 2020) and enzyme-linked immunosorbent assay (ELISA) (Roy, Mohd-Naim, Safavieh, & Ahmed, 2017; Safavieh, Ahmed, Sokullu, Ng, & Zourob, 2013), among others. A brief discussion of the various conventional methods available for detecting the different agri-food-related biomarkers has been provided in the supplementary file. Incorporating nanomaterials with LOC offers further benefits like enhanced sensitivity during the detection process and more straightforward labelling of the target analyte owing to the unmatched electrochemical, physical, and optical properties of the nanomaterials (Roy et al., 2022). Processes like DNA sequencing or detection of any biomolecule, which are done on a laboratory scale, are easily carried out over a single chip. Because of integration over a single chip, the cost of the overall process dramatically reduces, alongside a significant increase in the efficiency of the detection method. Therefore, studies involving LOC devices must demonstrate enhanced sensitivity, selectivity, and specificity during detection.

The microfluidic technology used for developing LOC devices permits the development of several multiple microchannels within a small chip. The microfluidic elements on the chip can independently perform actions like storing reagents, mixing fluids, and detecting the analytes. Studies on LOCs have shown the interconnection of fluidic microchannel networks, mixers, pumps, and detectors that can perform the reactions without requiring skilled laboratory technicians (Jung, Han, Choi, & Ahn, 2015). With the integration of nanomaterials into LOC devices, their biosensing capacity will increase owing to the unmatched and versatile physiochemical properties of nanoparticles, including their small size and large surface area, which provide them with improved sensing capabilities (Sarker et al., 2022). Also, nanomaterial-based LOC systems will reduce the sample volume needed and permit the simultaneous detection of multiple analytes (Naveen Prasad et al., 2022). Therefore, nanomaterial-based LOCs will permit the development of user-friendly point-of-care testing devices that can be produced in hefty amounts for their easy commercial availability (Junsheng Wang et al., 2017). Several studies have focused on fabricating smartphone-based

chips, paper-based microfluidics and similar monitoring systems for detecting pathogens, toxins, additives, and biomarkers in agri-food products and derivatives (Kotsiri, Vidic, & Vantarakis, 2022).

To better study and understand the recent developments in nanomaterial-based LOC devices and their application for sustainable food and agricultural industries, we searched the Scopus database using the following keywords: 'lab-on-a-chip + food'. We deeply analysed the recently published data from the last ten years (from 2013 to 2023). The search results are presented in Fig. 1. As can be inferred from the figure, although several research works have focused on developing and studying LOC devices, very few works have addressed the application and potential of such devices towards the detection and monitoring of food markers (Sridhar et al., 2022). One can appreciate the importance of discovering and detecting food biomarkers, as they can find applications across the detection of microbial and chemical contaminations, allergens, toxins, adulteration, and GMO identification to establish the geographical traceability of food products (Agrawal et al., 2013). However, most of these reports have provided extensive discussions on the development of LOC devices alone and have not shed much light on the working mechanisms of nanomaterial-based LOCs, specifically those for sustainable food and agricultural industries. Therefore, to better understand the advances in developing various nanomaterial-based LOCs for sustainable food and agricultural industries, we have provided an in-depth review of the recent developments in this field. We first discussed the different types of nanomaterials used in developing LOC devices. In the next section, we provided a brief discussion of the currently available methods for food screening. We then highlighted their mechanism of action, followed by applications of such LOC devices in sustainable food and agricultural industries. Next, we discussed the advantages and disadvantages of such LOC devices for food screening. Subsequently, we highlighted the industrial challenges of nanomaterial-based LOCs and their possible solutions to overcome such limitations. Finally, we presented the future perspectives of this rapidly growing field. We believe that this review will aid in bridging the gap between nanoscience and food sciences and give a compressive overview of the persistent gaps in the field for the research community to develop better and novel ways to ensure safety in the global agri-food industries.

## 2. Design concept and fluid dynamics of LOC devices

LOC devices, being easily portable, are promising candidates for applications in developing point-of-care devices. These can efficiently analyse and detect different agri-food analytes and biomarkers on-site or within a laboratory setting to ensure food quality and safety. In addition, such systems require very low volumes of samples and reagents, making the process very efficient and inexpensive. LOC devices are tools based on several networks of wells and channels supported over a polymer substrate attached to a micro or nanostructured functionalised surface that helps to analyse target molecules from a given complex sample.

Several factors, like fluid flow, patterns, composition, and designs, affect the overall efficiency of a LOC (Calado, dos Santos, & Semiao, 2016). Multiple fabrication methods have been studied to develop different LOC devices, including inkjet printing, wax screen printing, laser treatment, photolithography, plasma oxidation, and plotting (Sridhar et al., 2022). Usually, within microfluidic devices, fluids are present within microchannels. They are regulated via non-mechanical (gravity-driven flow, capillary action flow, osmosis, surface tension flow) or mechanical (pressure-driven systems, centrifugal microfluidics, acoustic streaming) methods (Iakovlev, Erofeev, & Gorelkin, 2022). Passive flow or non-mechanical techniques usually use microchannel systems or natural effects. They, therefore, do not need intricate mechanical structures or additional power sources (Xu, Wang, Li, & Oh, 2020). As displayed in Fig. 2, there are multiple novel pumping methods in microfluidic devices, which include (but not limited to) semi-open gravity-driven overflow microfluidic flow supply system or

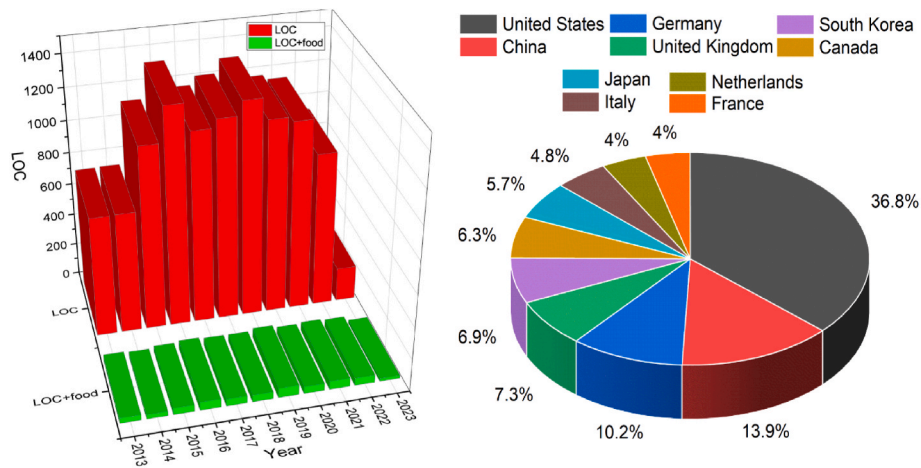


Fig. 1. (a) Trends in LOC-related papers annually over the last decade (2013–2023, inclusive). (b) Trends in the percentage of LOC-related papers published by different countries from 2013 to 2023. Data was collected from Scopus, as accessed on March 2023.

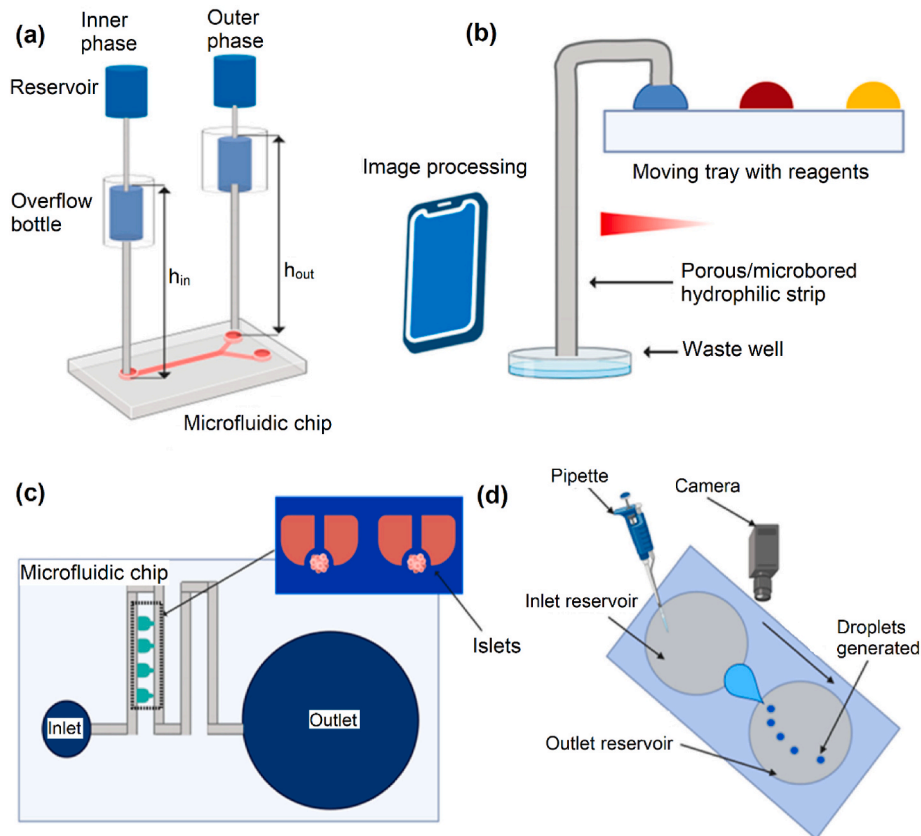


Fig. 2. Novel pumping methods for microfluidic devices. (a) Semi-open gravity-driven overflow microfluidic flow supply system (b) Gravity-driven microfluidic siphon (c) Microfluidic array to study Langerhans pancreatic islets (d) Schematics of droplet microfluidic system. Adapted with permission from ref. (Iakovlev et al., 2022), copyright (2022) MDPI.

gravity-driven microfluidic siphon, or autonomous pressure-driven analytical microfluidic systems. Hence, these systems are inexpensive, can function autonomously, and are easily portable. On the contrary, active flow or mechanical fluid systems use an external miniaturised mechanical system self-contained within the LOC. Unlike passive fluid systems, mechanical systems can be controlled at multiple steps and thus are highly reliable. Such external sources include electromagnetic or syringe pumps and valves, among others.

Recently, the applications of autonomous microfluidic systems that use capillary flow as their primary driving force have been widely

studied (Berthier, Dostie, Lee, Berthier, & Theberge, 2019). For instance, paper or porous material-based substrates are used to develop capillary flow-driven LOC devices (Altschuh et al., 2022). Such systems are used in pregnancy tests and COVID-19 rapid antigen testing kits. However, because of the non-uniformity of the porous membrane used in such devices, such systems do not generate reproducible capillary flow results. Therefore, incorporating techniques like microfabrication during the development of LOC devices has allowed for uniformity in the capillary flow (Yasuga, Shoji, Koizumi, & Kawano, 2023, p. 526). Patterning agents also determine the design of the device development.

Usually, polymer-based patterning agents are used for LOC development owing to their high binding power and versatile properties like high durability and the ability to give high-resolution patterns. However, such polymer-based LOC devices are comparatively expensive and complicated and involve external pumps for liquid transport and mixing the samples. Therefore, paper-based devices are preferred because of their cheap and easy availability (Scott & Ali, 2021). Besides, papers have inherent desirable features like hydrophilicity and porous structure, confer higher flexibility and inexpensive availability in terms of usage. Also, the conventional micro-paper analytical devices are made using hydrophilic and/or hydrophobic structures based on paper substrates. Paper-based LOC devices are broadly categorised into two or three-dimensional analytical devices. Usually, two-dimensional microfluidic devices are developed by decorating hydrophobic borders to create microfluidic channels. In comparison, three-dimensional microfluidic devices are formed by piling two-dimensional microfluidic layers on top of each other by different methods, like folding or 3-D printing (Pang, Zhu, Wei, Meng, & Wang, 2022).

Studies have demonstrated that two-dimensional microstructure patterns over hydrophilic materials like glass or silicon with straight channels result in capillary flow, while intricate microstructures like arrays function as capillary pumps (Roy, Wei, Ying, Safavieh, & Ahmed, 2016). Three-dimensional microstructures have recently obtained capillary flow (Verma & Pandya, 2022). For instance, Duong and colleagues developed a cheap 3D printed microfluidic device using a solvent bonding method based on 8 bars of poly (methyl methacrylate) (PMMA) and acrylonitrile butadiene styrene (ABS) thermoplastic materials and developed a 3D printed microfluidic device (Duong & Chen, 2019). A spray coating method was used for high-strength bonding between PMMA and ABS substrates, followed by UV exposure and a post-annealing step. Multiple studies have also been performed to understand further the fabrication of a hybrid microfluidic mixer based on specific element analysis. For instance, a 48–53% mixing efficiency of two substrates has shown high potential in additive manufacturing. Therefore, further studies on highly efficient devices are required to develop highly specific and selective LOC devices successfully.

### 3. Materials used in the development of LOC devices

Several materials are used in developing LOC devices and can be broadly categorised into organic, inorganic, or composite materials. For instance, silica has been a promising platform for developing LOC devices among inorganic materials due to its excellent chemical and physical properties, including high stability and biocompatibility. However, silica-based LOC device fabrication is comparatively more expensive (Sengupta & Hussain, 2019). Therefore, glass has been preferred for fabricating LOC because of its inexpensive availability and optically transparency (Gimenez-Gomez et al., 2016). Other inorganic materials like low-temperature cofired ceramic are also used to develop three-dimensional LOC devices with electrical circuits and fluid networks (Joseph, Varghese, Teirikangas, Vahera, & Jantunen, 2019). These are prepared by simultaneously firing multilayered ceramic substrates with high-conductivity thick film electrodes at temperatures below 1000 °C.

Compared to inorganic materials for LOC fabrication, organic materials like polymers, hydrogel, and paper substrates are preferred because they are a cheaper alternative and are easy to fabricate (D. Liu et al., 2022). Among polymers, usually, elastomers like polydimethylsiloxane (PDMS), polyethylene glycol diacrylate (PEGDA), polycarbonate, and polymethyl methacrylate (PMMA) are used for the fabrication of microfluidic devices, including LOCs (Morbioli, Speller, & Stockton, 2020). Paper-based LOC devices, like those based on cellulose or nitrocellulose sheets, have been rampantly used because of their cheap and easy accessibility and biocompatible nature (Ardalan, Hosseinifard, Vosough, & Golmohammadi, 2020). However, these do not show excellent mechanical properties; therefore, further research is

needed to improve their application towards LOC fabrication. Similarly, hydrogels are promising alternatives for developing LOC devices (Bhusal et al., 2022). Primarily, hydrogels are cross-linkable polymers that can swell and adsorb water but cannot dissolve in water; thus, their bonding remains a crucial challenge that needs to be addressed. Therefore, introducing nanomaterials for LOC development and successful application in agri-food sensing is necessary to overcome these challenges. We have also provided a detailed overview of the different kinds of nanomaterials used towards LOC development for application in agri-food industries in the supplementary file.

### 4. Applications of nanomaterial-based LOC devices in sustainable food and agricultural industries

Nanomaterial-based LOC devices can be potentially applied in detecting target analytes like pathogens, biomolecules, other small molecules, and metal ions within the food samples to ensure food and agricultural safety (Mohamad, Teo, Keasberry, & Ahmed, 2019). For instance, in a recent study by Pungjunun and colleagues, bismuth nanoparticles (BiNPs) modified screen-printed graphene electrode modified over a paper-based sensor was developed that could simultaneously help detect metal ions like lead and tin from a given sample using a portable potentiostat (Pungjunun et al., 2020). The possible interferences from the two target analytes were reduced using hexadecyltrimethylammonium bromide (CTAB), which permitted excellent metal ion detection sensitivity. Also, oxalic acid was added as an electrolyte during voltammetric analysis. The researchers noted that easily oxidising intermetallic species were formed within the BiNPs and the target metal analytes. In addition, the BiNPs enhance the total electroactive surface area and the electron transfer rate. The authors recorded the linear detection range as 10–250 ng/mL for both the metal ions, while the LOD was recorded as 0.26 ng/mL and 0.44 ng/mL for tin and lead, respectively. Furthermore, the researchers successfully applied this sensor to detect the two metal ions in canned food samples simultaneously and obtained similar results. Therefore, this promising research can be applied to detect metal ions like lead and tin from food samples, including agricultural products, to ensure food safety.

Specific LOC devices, especially those based on gas sensors that employ cavity resonance shifts to study molecule concentrations, permit enhanced sensitivity and real-time detection of multiple analytes. However, their sensitivity is easily perturbed by minute changes in the external environment, like fluctuations in temperature conditions (Radhakrishnan, Mathew, & Rout, 2022). This shortcoming can be overcome, as understood by Wang et al., who studied the changes in the optical conductivity of graphene as induced by gas (Wang, Chen, Geng, Hong, & Li, 2019). The developed silicon slot fibre structure increased the optical interaction within the graphene-based silicon device, demonstrating a linear increase in the central reflection with increasing gas concentration. The group investigated the changes in temperature over the effective refractive indices of TE<sub>0</sub> and TE<sub>1</sub> modes available in the resonator, which could be delinked following the modal linear independent responses. Based on this strategy, the researchers developed a sensor to detect nitrogen dioxide with high sensitivity of 0.02 ppm, and the LOD was noted to be 0.5 ppm. Thus, this device can be employed for monitoring gases like nitrogen dioxide that aid in plant growth and development and therefore is beneficial for the agricultural industry.

Recently, studies have also been done to develop LOC devices and other small molecules like dopamine (Q. Liu et al., 2020), L-lactic acid (Abhyankar, Gunjal, Kapadnis, & Ambade, 2022), and cystatin (Tremblay, Goulet, Vorster, Goulet, & Michaud, 2022) that play fundamental roles in monitoring plant health and, thus, the development of agricultural products. For instance, Fishlock and colleagues developed a simple microfluidic electrochemical paper-based LOC platform that could detect dopamine, creatinine, and uric acid from blood samples (Fishlock, Bhattacharya, & McLaughlin, 2020). The researchers developed a 3-electrode-based ePAD with electrodes on a wax-derived microfluidic

channel within a capillary flow device. A working electrode was developed over Whatman filter paper over which laser-induced graphene was developed. First, the filter was treated with a commercial flame-retardant spray, followed by treatment with a carbon dioxide infrared laser to develop a highly conductive, 3D graphene network that demonstrated an enhanced surface area and good conductivity. Two electrodes were patterned using a laser near the waxy microfluidic channel developed, while the third electrode was prepared using inkjet silver and was later treated with commercial bleach to generate Ag/AgCl pseudo reference electrode. To collect the sample, a novel blood sampling design was prepared based on ePAD cassette that allows rapid sampling of the target molecules even when present in low concentrations. Though this study was performed to detect analytes from clinical samples, a similar approach can be developed to detect the same analytes from agricultural products.

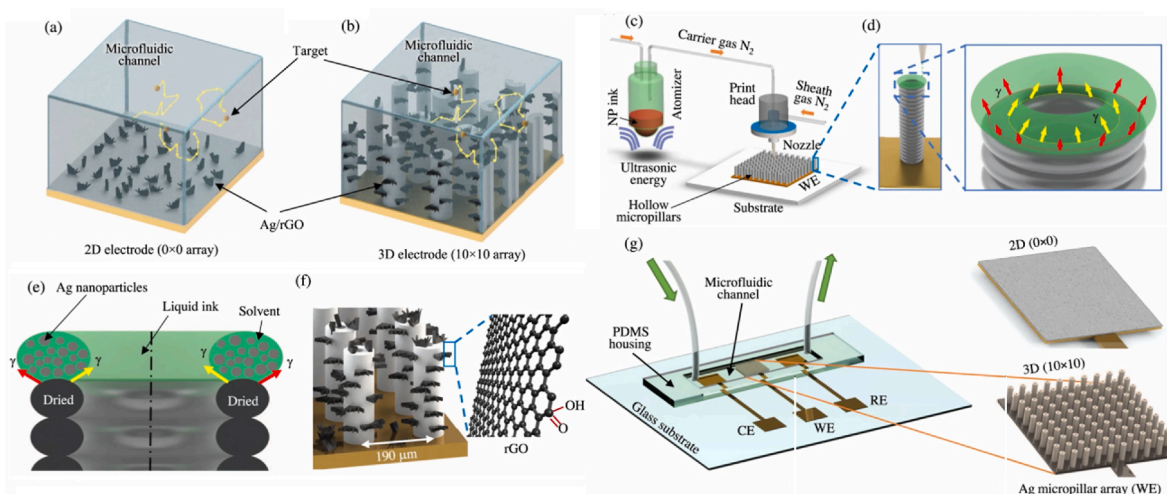
Yang et al. also developed a laser-engraved sensor that detected tyrosine and uric acid molecules (Yang et al., 2020). This device was developed over polyimide that could quantify the biomolecules and also consisted of a temperature and respiratory monitor alongside a microfluidic channel based on a vector engraving within the device. The authors recorded a high sensitivity of  $0.61 \mu\text{A} \mu\text{M}^{-1} \text{cm}^{-2}$  for tyrosine and uric acid. The LOD was noted to be  $3.5 \mu\text{A} \mu\text{M}^{-1} \text{cm}^{-2}$  and thus could be potentially employed for detecting small molecules in agricultural samples to monitor food safety. In a similar study, Stojanovic and team developed a microfluidic chip to detect ascorbic acid with high sensitivity and selectivity (Stojanovic et al., 2021). The xurography method developed a microfluidic channel consisting of several silver electrodes. A graphene sheet was placed within the electrode gaps that permitted high conductivity and electron transfer. Also, owing to the enhanced electrical properties of graphene, including increased electron transfer rate, electrical conductivity, and surface-to-volume ratio. This further allows for reduced detection limits and improved linear range. The team noted that as the concentration of ascorbic acid increased, the conductivity of the sensor also increased, and the parameters could easily be recorded. In the presence of graphene, a 5.28% increase in the linear response range was noted, and the LOD was shown to reduce by over 12%. Therefore, such a graphene-based LOC device can also be potentially developed to detect various analytes within agricultural systems (Arshad, Nabi, Iqbal, & Khan, 2022).

Pathogens like bacteria and viruses wreak havoc and destroy several

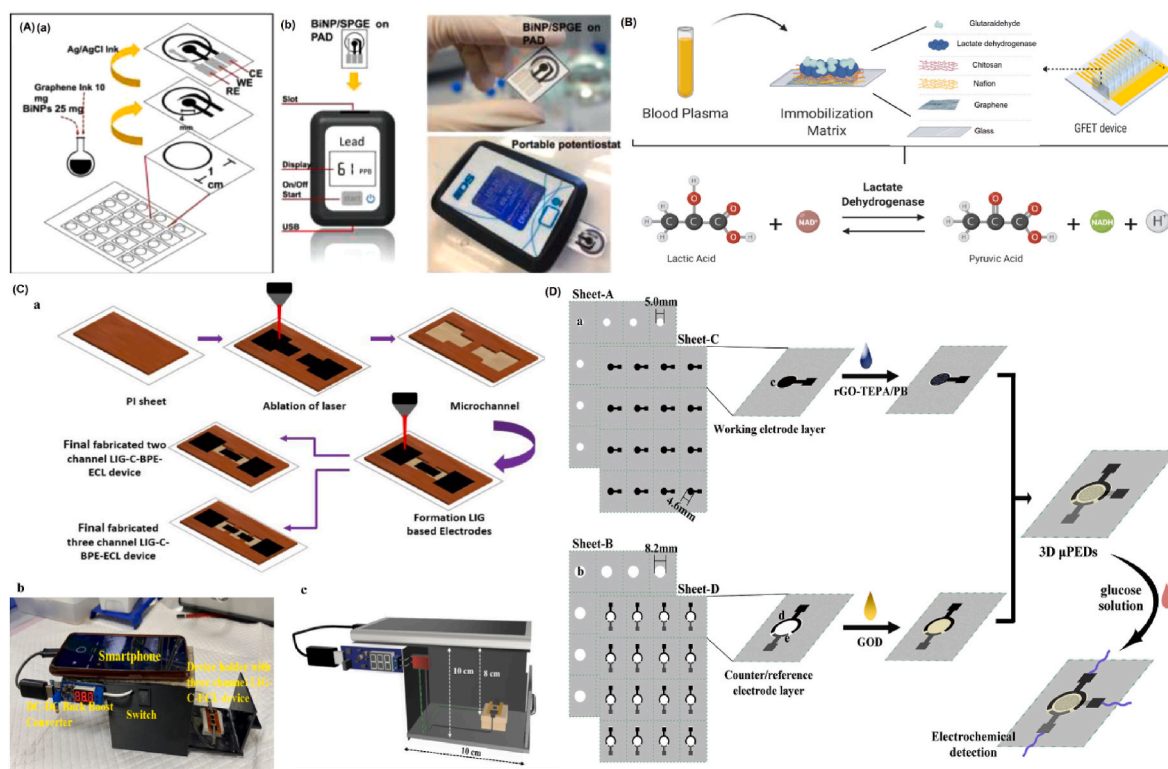
plants, affecting the agricultural and food industries (Safavieh, Ahmed, Ng, & Zourob, 2014). Therefore, the fabrication of rapid, inexpensive, selective, and highly sensitive LOC devices that can simultaneously detect multiple pathogens within a sample is necessary to avoid adverse effects on human health and the country's economy. Several LOC devices have been developed in recent years to combat this. For instance, Ali and colleagues developed a 3D printing-based gold micropillar array of electrodes alongside an aerosol jet nanoparticle (Ali, Hu, Jahan, et al., 2021a,b) (Fig. 3). They then functionalised this system using rGO nanoflakes and immobilised antigens on the surface of the electrode using the EDC/NHS chemical system. The developed nanocomposite showed an enhanced surface area and allowed for an increase in the diffusion of electroactive species. This electrochemical transduction system allowed for the detection of antibodies by forming antigen-antibody complexes over the surface of the 3D electrode. This device could thus be potentially applied to detect multiple plant viruses.

In another study, Schuck and colleagues developed a LOC device that can detect lactate molecules by functionalising the graphene surface with lactate dehydrogenase enzyme using reagents like chitosan and glutaraldehyde that allowed for an increase in the selectivity and stability of the sensor (Schuck, Kim, Moreira, Lora, & Kim, 2021) (Fig. 4). The researchers also confirmed that the sensor permitted selective detection of glucose molecules even in the presence of interfering molecules like uric acid, and ascorbic acid. The developed system remained stable for over 50 days and, therefore, can be used in point-of-care applications to detect such biomarkers in agricultural and food samples. In another study by Devi and colleagues, an electrochemical immunosensor was developed to detect cysteine C molecules (Devi & Krishnan, 2020). A glassy carbon working electrode was modified using GO-chitosan nanocomposites that were then used to develop a paper-based microfluidic substrate. This paper-based substrate was developed based on screen printing using carbon ink printed on wax-enveloped paper. Then Ag/AgCl paste was applied over the developed paper and compared with the modified glassy carbon electrode. The researchers observed that the paper-based system displayed a high sensitivity of  $15.5 \mu\text{A}/\text{ng}$  and LOD of  $2.6 \text{ pg}/\mu\text{L}$ .

Another device, based on laser-induced graphene (LIG), was used to fabricate PDMS-based microfluidic bio-cells (Rewatkar, Kothuru, & Goel, 2020). The team used LIG within a PDMS via the soft lithography method, and this was placed within a linker solution containing



**Fig. 3.** Architecture of 3D printed silver micropillars decorated with graphene nanoflakes. (a and b) Schematic representations of Brownian motion of target molecules for 2D and 3D electrodes within the microfluidic chamber. (c) 3D printer with an ultrasonic atomiser and a print head. (d) The mechanism of hollow-micropillar formation with concentric toroid-shaped rings of metal nanoparticle ink printed layer-by-layer. (e) A cross-sectional view of the printing process is shown in (d) where surface tension provides the fluid dynamic stability required to keep the ring in a stable state. (f) A schematic showing the silver micropillars with rGO flakes attached to their surfaces. (g) Schematic of the dopamine sensing device including the PDMS housing, a microfluidic channel, a 3D printed micropillar array (from (c)), and tubes for injection and removal of dopamine. Adapted with permission from ref. (Ali, Hu, Yuan, et al., 2021a), copyright (2021), Nature.



**Fig. 4.** Development and working of different kinds of LOC devices for detection of various agri-food biomarkers. (A) (a) The fabrication of bismuth nanoparticle-modified screen-printed graphene electrode for simultaneous detection of lead and tin (b) and the complete device into a portable potentiostat. Adapted with permission from ref. (Pungjunun et al., 2020), copyright (2020), Elsevier. (B) Schematic of the study performed to detect the concentration of lactate in the samples that were injected in the microchannels over the immobilisation matrix while measuring the transfer characteristics of the graphene-based device. Adapted with permission from ref. (Schuck et al., 2021), copyright (2021), MDPI. (C) (a) Fabrication flow for two and three-channel laser-induced graphene (LIG) based closed bipolar electrode (BPE) Electrochemiluminescence (ECL) device (b) Real image of 3D printed miniaturised portable ECL image capturing platform having a black box with device holder, DC to DC buck-boost converter and smartphone, (c) Schematic of ECL system for the sensing of various analytes (vitamins B<sub>1,2</sub> and C). Adapted with permission from ref. (Bhaiyya, Pattnaik, & Goel, 2021), copyright (2021), Elsevier. (D) Preparation of 3D paper-based microfluidic electrochemical biosensor for glucose detection (a, hydrophilic zone (5 mm); b, hydrophilic zone (8.2 mm); c, carbon working electrode; d, carbon counter electrode; e, Ag/AgCl reference electrode). Adapted with permission from ref. (Cao, Han, Xiao, Chen, & Fang, 2020), copyright (2020), Elsevier.

1-ethyl-3-(3-dimethyl aminopropyl) carbodiimide that allowed the binding of glucose oxidase and lactase on the electrode. Separately, PET sheets were used to develop microfluidic structures that displayed a fluid rate of 200  $\mu\text{L}/\text{min}$ , a power density of 13  $\mu\text{W}/\text{cm}^2$  of the sensor, and the linear range of the sensor was noted to be 10–40 mM. The device is easy to operate and allows rapid processing and detection of multiple analytes simultaneously, and these can be applied for detecting and monitoring agricultural and food products. In another study by Manish et al., a 3D-printed ECL system was developed based on LIG (Bhaiyya, Rewatkar, Salve, Pattnaik, & Goel, 2021). Graphene was developed upon polyimide sheets followed by injection of hydrogen peroxide and luminol inside the microchannels for H<sub>2</sub>O<sub>2</sub> detection. When the voltage was applied, ECL signal generation occurred over the electrode that was monitored using a smartphone. Then glucose oxidase was applied over the BPE anode for glucose detection. The LOD of the sensor was noted to be 0.13  $\mu\text{M}$  for glucose and 5.87  $\mu\text{M}$  for H<sub>2</sub>O<sub>2</sub> molecules. In another similar study by the same group, the same BPE-ECL-LIG-C multiplex sensing system was developed that allowed for the simultaneous detection of vitamin C, B12, and H<sub>2</sub>O<sub>2</sub> and demonstrated an excellent linear range for detection (Bhaiyya, Pattnaik, & Goel, 2021) (Fig. 4). The LOD for vitamin C was noted to be 0.10 nM, for vitamin B12 was 0.95 M, and for H<sub>2</sub>O<sub>2</sub>, the LOD was noted to be 0.30 M. Cao and colleagues reported the fabrication of paper-based microfluidic chips-based LOC devices (Cao et al., 2020) (Fig. 4). They used 3D aldehyde functionalised  $\mu\text{PAD}$  that was developed via periodate oxidation. The working electrode was based on a Prussian blue deposited rGO-tetraethylenepentamine composite. The working electrode was

functionalised with aldehyde and was used for glucose oxidase (GOD) immobilisation. The system displayed a LOD of 25  $\mu\text{M}$ , an excellent linear range of 0.1–25 mM, and high sensitivity. The electrode modification permits the electron exchange between the electrode surface and GOD and therefore causes an amplified signal generation. Therefore, such a multiplex LOC system can be developed to detect various target analytes for agricultural and food monitoring simultaneously.

Table 1 provides a detailed overview of the different nanomaterial-based systems used to detect multiple agri-food-based analytes.

#### 4.1. Microfluidic and nanomaterial-based LOC devices for on-site edible oil quality assessment

Edible oil is an important human food and agricultural product due to its nutritional value, antioxidant property, and economic importance. In 2021–2022 alone, it accounted for about 210 million metric tons of consumption globally, which included palm oil (35%), soybean oil (29%), rapeseed oil (14%), sunflower seed oil (10%), peanut oil (3%), and olive oil (2%) among others (Statista, n. d). China leads the world's consumption of edible oil with 19%, followed by India (11%) and Indonesia (9%) (ILibrary, n. d).

Edible oils are extracted from oil-rich fruits, seeds, grains, and nuts and therefore are nutrient-dense and a major source of unsaturated fatty acids (oleic acids) and promoters for digestion and absorption of fat-soluble vitamins such as A and D (Zhao et al., 2021). Thus, they are essential in cooking and their consumption provides various health benefits such as reducing the risk of heart diseases and cancer (Teasdale

**Table 1**

Different nanomaterials-based systems used for the detection of several analytes that can be potentially applied towards the development of LOC systems to ensure agri-food safety.

Nanomaterial used	Mode of detection	Analyte detected	Sample matrix	Detection range	Limit of detection	Ref.
Bismuth nanoparticle	Bismuth nanoparticle-modified screen-printed graphene electrode on a paper analytical device	Tin and lead	Canned food samples (mushrooms and bamboo shoots)	10–250 ng/mL	0.26 ng/mL for tin and 0.44 ng/mL for lead	Pungjunun et al. (2020)
Graphene	Graphene-based lactate device	Lactic acid	Buffer solution and plasma	0 mM–7.5 mM	–	Schuck et al. (2021)
Graphene oxide/chitosan nanocomposite	Graphene oxide-chitosan based electrode	Cystatin C	Serum samples	1–10 mg/L	0.0078 mg/L	Devi and Krishnan (2020)
Laser-induced graphene	Laser-induced graphene based open bipolar electrodes	Glucose	Buffer solution	1 $\mu$ M–100 $\mu$ M	0.138 $\mu$ M	Bhaiyya, Rewatkar, et al. (2021)
Laser-induced graphene	Two and three channel laser-induced graphene based open bipolar electrodes	Vitamin B <sub>12</sub>	Buffer solution	0.5–1000 nM	0.109 nM	Bhaiyya, Pattnaik, and Goel (2021)
Laser-induced graphene	Two and three channel laser-induced graphene based open bipolar electrodes	Vitamin C	Buffer solution	1–1000 $\mu$ M	0.96 $\mu$ M	Bhaiyya, Pattnaik, and Goel (2021)
Reduced graphene oxide (rGO)	Prussian blue deposited rGO-tetraethylene pentamine modified paper working electrode	Glucose	Buffer solution	0.1 mM–25 mM	25 $\mu$ M	Cao et al. (2020)
rGO nanoflakes	Aerosol jet nanoprinted rGO-coated 3D electrodes	Antibodies to SARS-CoV-2 spike S1 protein	Buffer solution	$2.8 \times 10^{-15}$ M	$1.0 \times 10^{-15}$ m to $30 \times 10^{-9}$ m	Ali, Hu, Jahan, et al. (2021a)
Graphene	Microfluidic channel placed between the silver electrodes	Ascorbic acid	Apples and oranges	–	–	Stojanovic et al. (2021)
Ti <sub>3</sub> C <sub>2</sub> MXene solution and AuNP	Liquid metal based microfluidic sensor	Glucose	Buffer solution	0.21 mM	0.5–4.0 mM	(Y. Zhang et al., 2022)
AuNP	AuNP mediated fluorescence detection on a microfluidic device	Heavy metal (mercury) and pesticide (ziram)	Buffer solution	0.6 $\mu$ g/L (mercury) and 16 $\mu$ g/L (ziram)	0.6 $\mu$ g/L to 200 $\mu$ g/L	Lafleur, Senkbeil, Jensen, and Kutter (2012)
Polyaza functionalised graphene oxide nanomaterial	PDMS polymeric substrate with microfluidic channels for the nanosensor electrodes	<i>E. coli</i>	Water samples	–	$10^3$ – $10^8$ cfu/mL	Rose et al. (2021)
Chromium and gold	Impedance based microfluidic biosensor	<i>Salmonella</i>	Cell culture	300 cells/mL	–	(J. Liu et al., 2019)

et al., 2022). However, inadequate oil storage conditions, such as a lack of protection against sunlight and heat exposures, may result in thermal and oxidative decompositions of unsaturated fatty acids, thus forming lipid peroxides and free radicals (Gertz, Aladedunye, & Matthäus, 2014). As a result, the quality and shelf life of edible oils may deteriorate, causing partial loss of their health-related beneficial effects (Zhou, Zhao, Lai, Zhang, & Zhang, 2020). Therefore, monitoring the quality of edible oils in real time is paramount to ensure the highest consistency and safety for consumption.

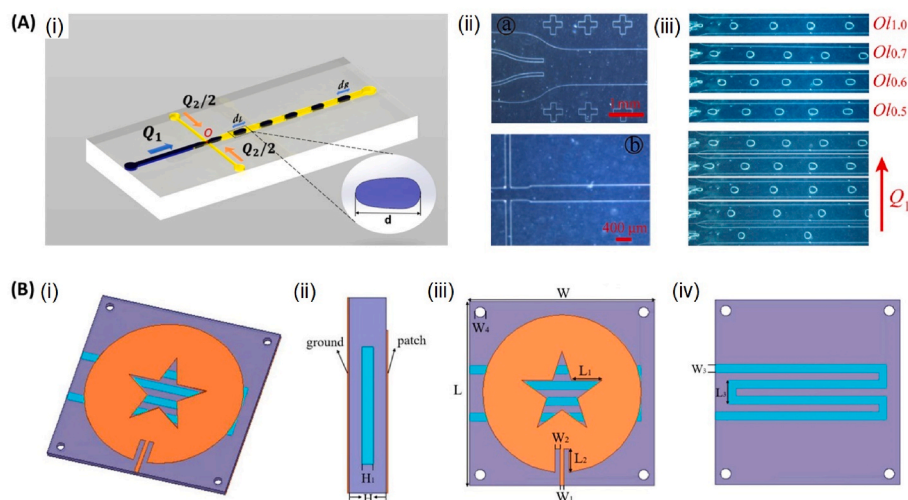
Phenolic compounds in oils, such as oleuropein, hydroxytyrosol, tyrosol, and tocopherol, are important indicators of high edible oil quality (Mikołajczak, Tańska, & Ogrodowska, 2021). On the other hand, free fatty acidity and peroxide value are among the major indicators of hydrolysis, oxidation, and polymerisation in oils, and therefore the changes in their thermal and oxidation stability indices are under intense scientific investigation (Nunes, 2014). Phenols in olive oil, soybean oil, and coconut oil exhibit strong antioxidant activity, which is shown to prevent the formation of lipid peroxides and act as free radical scavenging (Paradiso, Flammini, Pittia, Caponio, & Di Mattia, 2020). In contrast, high acidity and hydroperoxide contents are shown to lower oxidative stability and be toxic to humans because of the formation of secondary oxidation products (epoxides, ketones, alcohols, aldehydes) (Choe & Min, 2006). Noteworthy is that the moisture content (Ragni, Berardinelli, Cevoli, & Valli, 2012) and adulteration (i.e., mixing high-quality edible oils with low-quality ones (Przykaza et al., 2021)) may also reduce the overall quality of edible oils.

#### 4.1.1. Polymer-based microfluidic LOC devices for testing edible oil quality

Polymer-based microfluidic LOC devices are a promising trend in edible quality sensing since they allow small sample volumes to be processed efficiently with high reaction rates and ease-of-sample

handling and manipulations. PDMS is the base component of these devices, which is suitable for flexible moulding of an unlimited number of designs and can be prepared with minimal technical expertise. Several studies have designed and manufactured PDMS-based microfluidic LOC devices to test the quality of edible oils by considering their different physical aspects, such as viscosity, interfacial tension, and dielectric permittivity. For instance, in one study, Deng and colleagues developed a microfluidic chip that generates water-in-oil droplets to assess the quality of olive oil and frying soybean oil based on droplet sizes (Deng et al., 2018). Two types of microfluidic devices were fabricated in one chip: hydrodynamic focusing for adulterated olive oil and cross-shaped for frying soybean oil (Fig. 5).

In the chip, generated droplet sizes varied depending on the viscosity and interfacial tension changes in the oil adulteration and frying oil degradation. Results revealed that, for olive oil, the water-in-oil droplets reflect the changes in oil viscosity after adulteration, and for frying oil, the water-in-oil droplets reflect the changes in both oil viscosity and oil interfacial tension. In another study, Bianco and colleagues developed a microfluidic microviscometer to measure rheology and material properties of oils in extensional capillary flows (Bianco et al., 2018). The microviscometer consisted of a microfluidic channel with an oleophobic coating and was attached to an optical microscope with custom software. The coating was to decrease oil adsorption during the flow along the microchannel length. The device was used in testing commercial and home-made extra virgin olive oil, sunflower oil, commercial frying oil, and home-made mixture of oils. Apart from obtaining the absolute values of dynamic viscosity, with the chip oils in mixtures were successfully separated and identified. The success of the latter was attributed to the efficient correlation with microfluidic viscosities of known oils. In addition to above viscometric microfluidic LOC devices, Ramos and team introduced another microfluidic device, this time with two



**Fig. 5.** PDMS-based microfluidic LOC devices designed and developed for testing edible oil quality. (A) Evaluation of edible oil quality based on viscosity and interfacial tensions: (a) Schematic of the microfluidic device for measuring oil quality:  $Q_1$  and  $Q_2$  are the rate of the inner (deionised water) flow and sheath flow (oil sample), respectively.  $W$  is width of the microchannel, and  $d$  is the steady-state length of the droplet.  $d_L$  and  $d_R$  are the longitudinal dimensions of droplets at front and end positions in the field of view, respectively.  $O$  is the centre point of the system. (b) Micrographs of types of microfluidic devices: Top panel, hydrodynamic focusing ( $h = 80 \mu\text{m}$ ); bottom panel, cross-shaped ( $h = 40 \mu\text{m}$ ). (c) Water-in-oil droplets generated at different conditions: At same flow rates, droplets enlarge as adulteration increases ( $Q_1 = 0.04 \text{ mL/h}$ ,  $Q_2/2 = 0.2 \text{ mL/h}$ ). Droplets in  $O_{0.5}$  increase as  $Q_1$  increases. Adapted with permission from ref. (Deng et al., 2018), copyright (2017), IFST. (B) Microwave sensor loaded with star-slotted patch for edible oil quality inspection: (a–c) The structure of the microwave sensor which is composed of three layers: The slotted circular radiation patch on the top layer, the dielectric substrate in the middle, and the copper-plated ground on the bottom, respectively. (d) The shape of the microfluidic channel in the middle layer, with cross-section size of  $3 \text{ mm}$  by  $0.4 \text{ mm}$ . Adapted with permission from ref. (Han et al., 2022), copyright (2022), MDPI.

the middle, and the copper-plated ground on the bottom, respectively. (d) The shape of the microfluidic channel in the middle layer, with cross-section size of  $3 \text{ mm}$  by  $0.4 \text{ mm}$ . Adapted with permission from ref. (Han et al., 2022), copyright (2022), MDPI.

compartments: one to extract phenolic compounds of extra virgin olive oil using alkaline aqueous solution and other to develop reaction product using Folin–Ciocalteu reagent (Chávez Ramos, Olguín Contreras, & del Pilar Cañizares Macías, 2020). In the process, different lengths and width of the microchannels, flow rates, solution pH, and Folin–Ciocalteu reagent concentrations were used to investigate the efficiency of the chip. Results revealed that longer microchannels provide high order of phenol extraction.

More recently, Han et al. introduced a new microfluidic microwave sensor loaded with a star-slotted patch for detecting the quality of edible oils (Han et al., 2022). The working principle of the sensor relied on the rationale that relative oil dielectric permittivity can be used to assess the quality of edible oils. Based on this rationale, the structural design of the sensor composed of three layers: top layer with slotted circular radiation patch, middle layer with dielectric substrate, and bottom layer with copper-plated ground. The middle dielectric substrate also composed of three layers: epoxy resin on top and bottom, and PDMS with microfluidic channel in the middle. Simulations and experimental results showed that the sensor can effectively distinguish as-received rapeseed oil and soybean oil from their heated counterparts by measuring the resonance frequency offset of the input reflection coefficient. This was due to fact that heating of edible oils not only changes their dielectric constant but also has a greater impact on their dielectric loss factor.

#### 4.1.2. Paper-based microfluidic LOC devices for testing edible oil quality

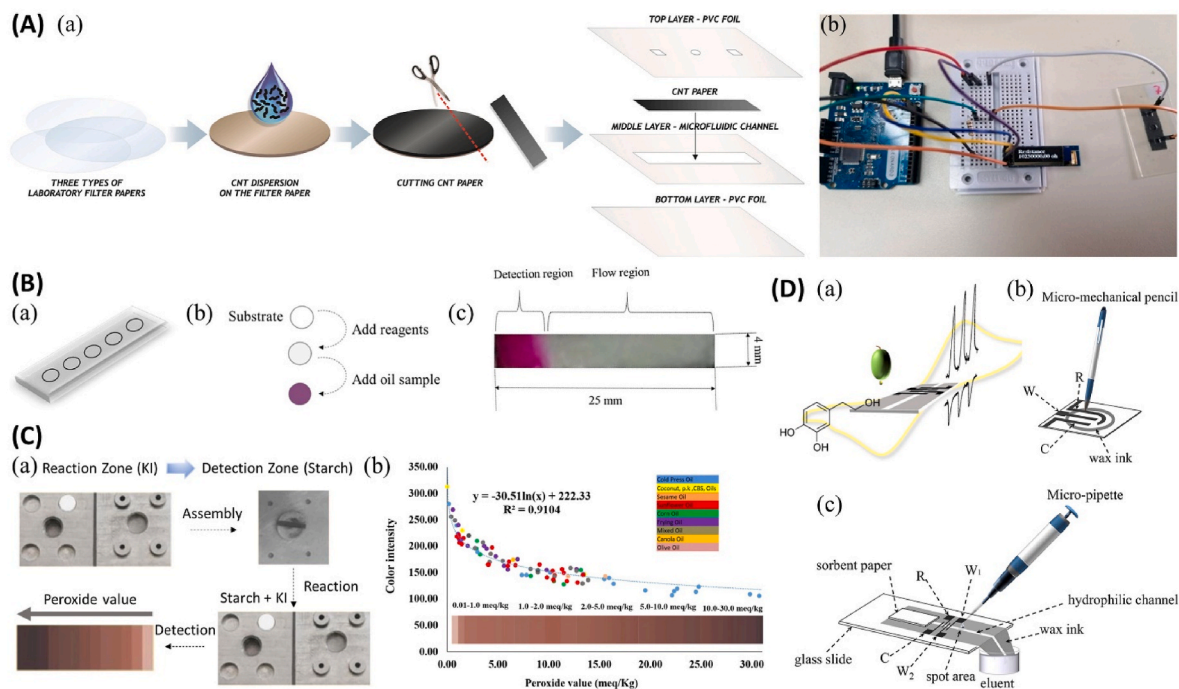
Paper-based microfluidic LOC devices developed for the detection of adulterants in many food products is reviewed and discussed by Ray et al. for their cost-affordability, specificity, and sensitivity features (Ray et al., 2022). Indeed, paper-based microfluidic platforms have emerged as an attractive alternative to PDMS-based microfluidic platforms due to added features such as fabrication cost and time, tunable porosity and fiber surface characteristics, external pumping-free flow of fluid, and remarkable applicability for large-scale, multilayer testing. Moreover, the overall trending of paper-based microfluidic LOC devices is supported by facts that they can be lightweight and portable, requiring small sample volumes for processing through capillary action and gravitational forces, and space saving for handling and storage. They can also be effectively designed, by chemical modifications and plasma etching (Samara, Deliorman, Sukumar, & Qasaimah, 2021) or wax printing and heating (Younas et al., 2019), to create hydrophobic barriers. As a result, with regard to food testing, these devices are shown to

offer results comparable to standard PDMS-based microfluidic LOC devices (Ray et al., 2022). In relation to their use in edible oil quality assessment, however, their use is still in infancy stage.

One example is the microfluidic platform developed by Radovanović et al. (Radovanović et al., 2021). The platform was assembled using a filter paper, after which its surface was coated with multi-walled carbon nanotubes (Fig. 6). Thanks to the plausible electrical conductivity of carbon nanotubes, it was shown that the platform is effective in determining the percentage of extra virgin olive oil in a mixture with high blend sunflower oil at different concentrations, by measuring the variation in electrical resistance. In the device, cost-effective polyvinyl chloride foils were used to laminate the filter paper for easy handling and sample acquisition, and electronic circuit was created for fast and straightforward determination of electrical resistance and portability (Fig. 6).

Another example is the microfluidic chip developed by Muthukumar et al. for the detection of palm oil contamination in sunflower oil and free fatty acid content—defined as the amount of fatty acids not bound or esterified to triglyceride molecules and measured as the percent of oleic acid in  $100 \text{ g}$  of oil—in oil samples (Muthukumar, Kapoor, Balasubramanian, Vaishampayan, & Gabhane, 2018). Similar to standard pregnancy test kits, the chip was prepared in the form of circular discs and rectangular channel strips where a double-sided adhesive tape was used for support and stability (Fig. 6). Circular discs were then used for spot testing of adulterated oil and free fatty acid by standard titrimetric procedure using phenolphthalein (Brown & Campbell, 1968), and their content was visually observed through colorimetric detection and quantitatively analysed through imaging. Results, by change in color from white to pink, revealed  $10\% \text{ (v/v)}$  detection limit of palm oil contamination in sunflower oil, and that free fatty acid contents can be detected qualitatively by varying phenolphthalein concentration. Moreover, the color change was stable for 6 days allowing the user to analyse data conveniently. The chip was also integrated to a smartphone for on-site image analysis. Similarly, in another colorimetric study by Ghohestani et al., a microfluidic device was assembled using two layers of filter papers stuck on each other to facilitate the sample transport based on vertical flow (Ghohestani, Tashkhourian, & Hemmateenejad, 2023). The reaction (potassium iodide) and detection (starch) zones were vertically aligned in a cylindrical metallic mold to measure the peroxide value—defined as the amount of hydroperoxides formed during the initial stage of lipid oxidation and measured as the





**Fig. 6.** Paper-based microfluidic LOC devices designed and developed for testing edible oil quality. (A) Detection of olive oil blends: (a) Schematic of main fabrications steps in the realization of the microfluidic platform using filter paper and multi-walled carbon nanotubes. (b) The electronic device (left) for fast measurement of electrical resistance and percentage of olive oil content in oil blends based on manufactured prototype device (right). Adapted with permission from ref. (Radovanović et al., 2021), copyright (2021), Elsevier. (B) Detection of adulteration in sunflower oil: (a) Schematic of microfluidic device for detection of adulteration in sunflower oil using spot colorimetric assay. (b) Steps involved in spot colorimetric assay. (c) Lateral flow assay results for 50% palm oil contamination in sunflower oil. Adapted with permission from ref. (Muthukumar et al., 2018), copyright (2021), Elsevier. (C) Colorimetric determination of peroxide value in vegetable oils: (a) Schematic of procedure using a paper-based microfluidic device. (b) Correlation between peroxide value obtained using the standard method and obtained using color intensity of colorimetric method. The brown band represents sensor's color changes. Adapted with permission from ref. (Ghohestani et al., 2023), copyright (2022), Elsevier. (D) Quantification of *ortho*-diphenols in extra virgin olive oil: (a) Scientific illustration of the proposed method. The layouts of (b) a paper-based electrochemical cell with pencil-drawn electrodes, together with (c) a paper-based fluidic system. R, C, W1 and W2 are reference, counter and working electrodes, respectively. Adapted with permission from ref. (Dossi et al., 2017), copyright (2017), Elsevier. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

milliequivalent of active oxygen per kilogram of oil, meq O<sub>2</sub>/kg—of edible oils by iodometric titration (Fig. 6). With the method hydroperoxides present in oil react with iodide ions (potassium iodide) to produce iodine molecules. Iodine molecules then react with the starch and produce color change based on the peroxide value. Impressively, in the study total of 9 different oil samples were used for method validation, including cold press oil, coconut oil, sesame oil, sunflower oil, corn oil, frying oil, canola oil, olive oil, and mixed oil. Whereas the calibration curve between the color intensity and peroxide value was derived from total of 80 oil samples. As a result, a gradient of brown to purple color was visually observed with increase in peroxide value, with peroxide value detection range of 0.01–30.0 meq/kg and limit of 0.015 meq/kg. The reaction and detection zones of device were stable for months at room temperature and under dark and thus, highly suitable for on-site detection of edible oil peroxide value.

Another interesting example is the microfluidic device developed by Dossi et al. for discrimination of mono-phenols from *ortho*-diphenols in extra virgin olive oils (Dossi et al., 2017). The device consisted of dual electrode detector that was pencil drawn at the end of a paper microfluidic channel which was surrounded by hydrophobic barrier (Fig. 6). In the device, compounds were subjected to comigration via capillary flow under on-plate separation conditions. The selective detection of *ortho*-diphenols, which are known to be in abundant amounts in extra virgin olive oil, was then achieved by using two different working electrodes: Upstream pencil-drawn working electrode, where a potential is applied for the oxidation of both diphenols and mono-phenols; and downstream pencil-drawn working electrode, where a potential is applied for the reverse process involving only diphenols. The latter

configuration was based on the fact that *ortho*-diphenols undergo reversible oxidation at less positive potentials than those required by monophenols. Overall results of the study revealed that, different *ortho*-diphenol contents can be found in extra virgin olive oil with up to 40.8 mg/kg compared minimal traces in regular control olive oil (2.46 mg/kg) and no traces in sunflower oil (negative control). These results proved that the method can be effectively adopted for the rapid discrimination of edible oils.

Analytically, the content of lipid oxidation products, moisture, adulteration, and phenolic compounds in edible oils is determined with superior accuracy using various spectroscopy and chromatography methods (N. Zhang et al., 2021). Examples include Fourier transform near-infrared spectroscopy to measure the acidity and peroxide value, time-domain reflectometry to measure the moisture, liquid and gas chromatography to measure adulteration, and UV spectroscopy to measure total phenols. Although these methods certainly have their quantitative merits, they require expensive instrumentation that cannot be used in the field, lengthy protocols and analysis, and trained personnel. Therefore, the microfluidic LOC devices reviewed herein hold great promise for field applications as they are cost-effective, robust and simple, quantitative with high sensitivity, adaptable to a hand-held device, and expandable to include various analytical capability.

On a concluding note, edible oils could be also prone to microbial contamination during their production by small-scale farmers, which will reduce their quality. A recent study by MacArthur and colleagues revealed presence of *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Escherichia coli*, and *Pseudomonas aeruginosa* in palm oil samples that

were randomly collected from seven regions of Ghana, thus indicating potential health hazards for consumers (MacArthur, Teye, & Darkwa, 2021). In tested oils, the presence of yeast, fungi, and extremely pathogenic bacteria such as *Salmonella* and *Shigella* species were absent. In the study, standard methods and procedures were applied to isolate and identify fungi and bacteria based on their cultural, morphological, and biochemical characteristics. Therefore, future directions should include expanding microfluidic LOC devices to target the detection and monitoring of biological contamination in edible oils to protect the consumer.

### 5. Advantages and disadvantages of nanomaterial-based LOC devices for food screening

As discussed in the previous sections, LOC devices permit narrowing down large bulky laboratory equipment to tiny chips. Promising inherent advantages like lesser consumption of reagents, faster detection, excellent biocompatibility, portability, reduced sample size requirement, inexpensive availability, and quick automation process during the detection of target bodies have permitted the application of nanomaterial-based LOC devices for food and agricultural screening (Arshad, Mohd-Naim, Chandrawati, Cozzolino, & Ahmed, 2022). In addition, because of the incorporation of nanomaterials, these devices provide a larger surface area to volume ratio and therefore allow for better complete sample analysis compared to other analytical methods. The LOC systems are microelectromechanical systems that permit complete microanalysis and multiple laboratory processes over a single chip (James, Mannoor, & Ivanov, 2008).

Also, with the integration of nanomaterials on LOC devices, the overall sensitivity of the system increases. Nanomaterials are often used as detectors or modifiers in detection systems to improve the performance of the LOC system (Kuchmenko, Dorovskaya, Bosikova, Smetankina, & Bitukova, 2021). Furthermore, nanomaterials have also been used as microchannel modifiers to achieve easier pretreatment of the samples for better sensitivity, selectivity, and reproducibility of the device (Chen, Yin, & Wu, 2021). Additionally, nanomaterials allow enhancement in microanalytical processes and applications in filtering, sorting, and capturing processes. Few studies have also demonstrated other promising advantages of nanomaterial-based LOC devices, like developing tissue nano scaffolds and drug delivery systems (Harish et al., 2022). Reduced preparation time and lower consumption of the reagents are also significant advantages of nanomaterial-based LOC devices.

However, a few shortcomings towards developing nanomaterial-based LOC devices still need to be addressed. For starters, nanomaterials, even those based on inert noble metals, may have toxic effects (Naikoo et al., 2022). Also, the manufacturing process of the LOC systems is indirect and requires skilled professionals and technicians and expensive instruments and starting materials (Azizipour, Avazpour, Rosenzweig, Sawan, & Ajji, 2020). Furthermore, monitoring physical and chemical parameters at the micro and nanoscale becomes tedious and requires high precision to achieve the desired and effective results. Also, other factors like poor signal-to-noise ratio affect the detection process and reduce the overall sensitivity and selectivity of the process (Haji Mohammadi et al., 2021). Therefore, further work is needed to develop novel nanomaterial-based LOC systems that employ user-friendly, inexpensive, non-toxic nanomaterials that can be produced at an industrial scale and quickly commercialised. Further discussions on the current industry challenges and their possible solutions for developing LOC devices for agri-food applications have been provided in the supplementary file.

### 6. Integrating artificial intelligence (AI) and internet of things (IoT) with LOC devices: exploring the future of the agri-food industry

Apart from the many advantages of LOC systems discussed in the

previous sections, LOCs can also be automated and optimised with the help of AI and IoT. With the integration of sophisticated robotics and automation, human interference in the detection and analysis process can be significantly reduced, and the rates of human errors can be eliminated (Jiang, Jokhun, & Lim, 2021). Furthermore, by relying on pre-programmed and easily customisable or personalised systems, the time taken to analyse multiple samples simultaneously can be significantly reduced – thereby allowing rapid and highly accurate detection of several food samples simultaneously (Ahmed, Saaem, Wu, & Brown, 2014). Therefore, the researchers can invest their time in protocol development and data analysis. In addition, incorporating self-learned algorithms in research workflows will permit better data accessibility and scrutiny for researchers worldwide, allowing large-scale, high-quality research collaboration (Zare Harofte, Soltani, Siavashy, & Raahemifar, 2022).

On any platform, like with LOC device, high throughput imaging is sometimes necessary for highly accurate, sensitive, and specific imaging of target molecules over a large scale (De Stefano, Bianchi, & Dubini, 2022), including their size, morphology, interaction, and composition of the sample. With the integration of AI, the rapid analysis and highly sensitive identification and characterisation of complex samples can be quickly made. This was seen in the work by Schaumburg and colleagues; integrating smartphones and microfluidics was possible for efficient point-of-need testing (Schaumburg et al., 2022). As illustrated in Fig. 7, the team developed a free platform called the ‘appuente’ that could easily integrate microfluidic chips with smartphones and the cloud. The mobile application interface was user-friendly and provided the analyte analysis, control, smart imaging, processing, and data reporting details. The platform also included a web application for the point of need testing developers who could customise the mobile application and manage the data available. The team used this application to carry out three different tests: a colourimetric test, an elongation assay to check pesticide toxicity, and a lateral flow immunoassay that could detect leptospirosis.

Similarly, another recent study by McRae et al. developed an innovative diagnostic platform that could perform multiplex measurements using integrated microfluidic cartridges that functioned as a point-of-care device (McRae et al., 2022). The image analysis of multiple cell-based oncology tests revealed fluorescent signals that could be recorded via an AI-linked universal diagnostic system. Therefore, such AI and IoT-linked LOC systems can be developed to improve the efficiency in detecting multiple agri-food biomarkers within complex samples.

### 7. Conclusions and future perspectives

As discussed in the previous sections, nanomaterials-based LOC systems are promising in detecting and monitoring various plant and agricultural product analytes. Multiple studies have shown that incorporating nanomaterials enhances the system sensitivity apart from conferring miniaturisation capabilities to the devices (Kim et al., 2021). Also, nanomaterials, when applied as detectors or microchannel modifiers, help improve the versatility of the system and thus improve the overall applicability of conventional LOC devices. Carbon nanomaterials like carbon nanotubes and graphene have demonstrated excellent optical and electrical activity and thus have been extensively studied for their application to develop LOC devices (Sengupta & Hussain, 2019). Thus, employing nanomaterials allows for unique ways of functionalisation to improve the sample pretreatment and separation processes within LOC devices. Also, with the incorporation of microfluidic channels, novel pathways are possible for the development and characterisation of nanomaterials, like the research studies involving the synthesis of quantum dots and nanowires using microfluidics (Richard, McGee, Goenka, Mukherjee, & Bhargava, 2020). Apart from this, nanomaterials are also used in the microanalytical process enhancement in LOC. That is, they filter, sort, or enrich the target analyte, thus allowing for the



**Fig. 7.** Integration of IoT and AI with microfluidic devices. (a) Elements of a Smart Diagnostics platform. Adapted with permission from ref. (McRae, Rajsri, Alcorn, & McDevitt, 2022), copyright (2022), MDPI. (b) A 3D render of the LFA parts (top panel) and its cassette (bottom panel). (c) Screenshots of the custom mobile app created using 'appuente', at six running steps: (1) Start new test, (2) scan ID code, (3) add sample, (4) chronometer, (5) automatic imaging and (6) result report. (d) Real examples of LFA chips for positive (top) and negative (bottom) samples. Adapted with permission from ref. (Schaumburg et al., 2022), copyright (2022), Nature.

sensitive detection of target molecules. Therefore, nanomaterial-based LOC devices offer excellent applicability in detecting and monitoring multiple target analytes from agricultural and food samples.

Though LOC systems have emerged as an excellent platform to detect multiple food analytes and allow rapid testing of agricultural products, a few challenges in this rapidly evolving field still need to be addressed. For starters, better and more efficient signal amplification methods must be investigated to develop such susceptible devices. For this purpose, CRISPR/Cas systems can be incorporated into LOC devices (Ngamsom et al., 2022). Also, including microfluidic control systems within LOC systems will help in increasing the accuracy of the detection method. In addition, the presently available LOC devices are bulky due to the requirements of external peripherals, such as pumps, and require highly skilled professionals and expensive instruments. Therefore, more studies must be done to reduce the footprint of external peripherals supporting LOCs, making LOC devices more suitable for resource-limited settings in underdeveloped and developing countries. In addition, smartphone-based LOC systems can be developed, including applications with easy-to-navigate interfaces that translate data into understandable signals. For LOC devices to become widely accepted, more studies are needed to develop rapid, easy-to-use, inexpensive, user-friendly, multiplexed, eco-friendly systems that can be produced on an industrial scale and can be made commercially available in the agri-food market for on-site detection of complex analytes in food and agricultural industries.

#### CRediT author statement

Fareeha Arshad: Conceptualization, Investigation, Methodology, Visualization, Writing - original draft, Writing - review & editing; Muhammedin Deliorman, Pavithra Sukumar and Mohammad A. Qasaimeh: Investigation, Methodology, Writing - original draft; James Salveo Olarve, Gil Nonato Santos: Investigation, Methodology, Writing - original draft; Vipul Bansal: Visualization, Writing - review & editing; Minhaz Uddin Ahmed: Conceptualization, Investigation, Methodology, Writing - review & editing, Project administration, Funding acquisition and Supervision.

#### Declaration of competing interest

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

#### Data availability

No data was used for the research described in the article.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.tifs.2023.04.010>.

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