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# Comprehensive review on the integration of self-healing polymers with smart food traceability: scope, application and challenges

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Food traceability is a key component in ensuring food safety, quality, and supply chain transparency. The growing global population and increased food demand highlight challenges in ensuring safe food. The invention of smart food traceability systems (such as RFID, barcode, and blockchain systems) has been a significant milestone in the food industry. However, the development of an effective food traceability system is affected by various challenges, such as the possibility of damage or scratches occurring during the food supply chain. The integration of self-healing technologies with smart food traceability systems represents a promising advancement in food technology. This comprehensive review explores various self-healing mechanisms (mostly covalent interactions such as hydrogen and imine bonds) utilized in food processing and discusses the development of self-healing-based biopolymers for food applications. It also focuses on the development of photo-cross-linkable polymeric films, self-healable RFID tags, and techniques for electronic tracking using data matrix codes and wireless-based detection systems. Limited studies have been conducted on the use of embedded symbology techniques for food traceability, thereby providing a good scope for future research in this field. This comprehensive review of current technologies offers valuable insights for researchers, manufacturers, marketers, and consumers to develop customized smart food traceability systems that incorporate self-healing functionalities for a wide range of applications.

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## Sustainability spotlight

Considering the global economy, the production of plastics and paper has created a significant surge in the utilization of fossil fuels and forests, respectively. Conventional methods of packaging are typically single-use in nature, more susceptible to damage, and have an adverse effect on the environment. Thus, production of sustainable food packaging with inbuilt self-healing functionalities is a viable solution for minimizing reliance on non-renewable resources. Embedding self-healing polymers with smart traceability technologies, such as RFID, sensors or QR codes, can provide real-time information on the food quality and food transparency to customers. Thus, this study completely aligns with the UN Sustainable Development Goals (SDGs) by reducing food and packaging waste (SDG 2, SDG 12, and SDG 13), promoting safe food (SDG 3) and innovative approaches in industry (SDG 9).

## 1. Introduction

The rapid growth of the global population poses a significant challenge to existing food production and distribution systems. Ensuring the safety, authenticity, and quality of food within increasingly complex supply networks has become a global priority. Recently, there has been a growing emphasis on traceability in the food industry due to increasing incidents of fraud and food safety issues. These types of scandals may pose a threat to consumers' health and trust, in addition to economic losses.<sup>1</sup> In recognition of these challenges, the World Health Organization (WHO) has recently released the 'Global Strategy for Food Safety 2022–2030', which outlines a comprehensive

approach to emerging technologies, new challenges, and innovative methods to strengthen food safety systems and enhance laboratory facilities for foodborne disease surveillance.

Prominent regulatory authorities, including the United States Department of Agriculture (USDA) and the Food and Drug Administration (FDA), play a pivotal role in overseeing food safety.<sup>2</sup> These agencies are empowered to regulate food products and ingredients, including their labeling, which can help mitigate instances of food fraud. In India, while no mandatory traceability system exists, the government has collaborated with various state, central, and private organizations to develop traceability systems within the food supply chain. Key stakeholders include the Food Safety and Standards Authority of India (FSSAI), GS1 India, the Agricultural and Processed Food Products Export Development Authority (APEDA), the Fruit Products Order (FPO), the National Bank for

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Agriculture and Rural Development (NABARD), Reliance Industries, and the ITC's e-Choupal initiative.<sup>3</sup> To advance and transform its food industry and supply chain, India must strengthen its existing national food regulations and establish an efficient traceability framework. Despite the enactment of food safety laws, challenges related to food fraud and contamination persist, continuing to threaten the integrity of the food supply.

Conventionally, traceability data have been obtained through documented records or by measurements performed with various instruments, targeting the physical, chemical, biochemical, molecular, biological, and/or microbiological characteristics of foods.<sup>4</sup> Recently, advancements in digital traceability technologies have led to the production of various smart systems such as Radio Frequency Identification (RFID), blockchain and Near-Field Communication (NFC). The incorporation of blockchain technology into food logistics has revolutionized global food marketing and distribution. Blockchain offers a decentralized yet cohesive framework for tracking goods throughout the supply chain, addressing critical issues such as food recalls and safety.<sup>5</sup> These systems have reduced food losses, increased efficiency and quality, and improved sustainable development. However, current food traceability systems face several challenges, including managing products with multiple ingredients sourced from diverse suppliers, regions, or countries; the inherent complexity and variability of supply chains; and the high costs and technical sophistication of devices required to collect, analyze, and manage traceability data.<sup>6,7</sup> Even physical tag fragility may lead to damage or loss of traceability data during logistics and handling. Therefore, existing traceability systems still have gaps in linking food chain records, as well as issues with inaccuracies, record errors, and delays in obtaining critical data, which are vital in addressing foodborne outbreaks.<sup>8</sup>

An ideal smart food traceability system should address these challenges by enabling an efficient, reusable and cost-effective system that can trace food products (location, ingredients, and packaging) throughout the supply chain from production to consumption.<sup>9</sup> Therefore, developing materials with inherent self-healing capabilities could substantially enhance the reusability and efficiency of physical identifiers. Such systems would have the inbuilt ability to restore structural and functional integrity after sustaining damage. Incorporating self-healing ability into barcodes and RFID devices enhances reusability, durability, and sustainability within food traceability systems.<sup>10</sup> These materials could significantly reduce costs, resource consumption, and waste in the supply chain. Thus, self-healing technologies are a novel area of research that complements digital traceability advances and addresses a fundamental technical challenge currently hindering the food industry.

The adoption of a robust traceability systems aligns with several United Nations Sustainable Development Goals (SDGs) by ensuring safer and more reliable food supplies (SDG 2: Zero Hunger), reducing foodborne illnesses (SDG 3: Good Health and Well-being), food waste reduction enabled by precise recalls and quality control (SDG 12: Responsible Consumption and Production) and by fostering technological innovation in supply

chains (SDG 9: Industry, Innovation, and Infrastructure). Additionally, the sustainability dimension of self-healing materials is addressed by promoting resource-efficient and reusable packaging solutions (SDG 13: Climate Action). Thus, these digital traceability systems, with the incorporation of self-healing ability, represent a significant leap in the direction of attaining sustainable development goals.

Despite growing research on blockchain and other digital traceability tools, research based on self-healing materials tailored for food traceability is limited.<sup>11</sup> This work consolidates existing knowledge on advanced traceability technologies (RFID, barcode, and wireless technologies) and discusses the challenges involved. The various mechanisms of self-healing and their commonly found applications are also discussed. Furthermore, this review provides a critical perspective on the potential application of self-healing technologies within food traceability systems, identifies key challenges, and outlines potential avenues for future research in the emerging domain of self-healing and traceability. This review paper will help pave the way for future researchers to achieve a paradigm shift in traceability systems and move towards a sustainable future.

## 2. Food traceability and its significance in the global economy

Traceability is the ability to access any or all information regarding an item throughout its entire life cycle *via* recorded identifications.<sup>12</sup> Traceability systems enable food manufacturers to provide detailed information about their foods (such as the species, geographical origin, and conditions of transport and storage), transparency, and regulatory mandates, based on consumers' demands. The food supply chain is critical for ensuring the safety of food products, encompassing stages from farm production to processing, distribution, and retail, ultimately reaching consumers or food service establishments. As complex processing and supply chains are involved in the food industry, many risk points are introduced that must be controlled.

Foodborne diseases annually impact around 7.7% of the global population (out of 7.8 billion people) and are responsible for 7.5% of worldwide deaths (from a total of 56 million).<sup>13</sup> It has been reported that 3 out of 10 children under the age of 5 die from foodborne illnesses. However, due to widespread under-reporting to health authorities, the actual burden of foodborne illness remains significantly underestimated. Contamination events occur at various stages of the food supply chain, including processing, distribution, retail, and catering (Food Standards Agency, 2019).<sup>14</sup> Such contamination can lead to outbreaks of foodborne illnesses, posing significant public health threats. Notable incidents, such as the deliberate adulteration of infant formula with melamine in China<sup>15</sup> and outbreaks of *Escherichia coli* contamination linked to Chipotle Mexican Grill restaurants in the United States,<sup>16,17</sup> have garnered attention from industry leaders and researchers, prompting detailed investigative studies. These studies reveal the limitations of conventional traceability systems; thus, an



effective food safety management system is critical for mapping potential hazards, including physical, biological, and chemical threats. The growing integration of advanced technologies, in conjunction with cutting-edge computational and simulation models,<sup>5,18,19</sup> offers significant potential to enhance hazard identification throughout the supply chain (Fig. 1).

Food fraud is defined as “a collective term encompassing the deliberate and intentional substitution, addition, tampering, or misrepresentation of food, food ingredients, or food packaging, including false or misleading claims about a product, for economic gain”.<sup>19</sup> The Food and Agriculture Organization (FAO) identifies three essential elements inherent in all cases of food fraud: intentionality, deception, and the pursuit of undue advantage (FAO, 2021). Buyuktepe *et al.* (2023) applied explainable AI (XAI) techniques to predict fraud risks using data from the Rapid Alert System for Food and Feed (RASFF) and economically motivated adulteration (EMA) databases.<sup>21</sup> Results showed that data quality strongly affects predictive accuracy, but discrepancies and incomplete reporting remain critical challenges, reinforcing the need for harmonized regulations and interoperable global data platforms.

Food Traceability Systems (FTS) are critical for integrating traceability activities with food logistics and improving communication among supply chain partners. However, the ease of use remains a significant concern. Developing an effective traceability system requires increased engagement from stakeholders, including industry participants, government agencies, and researchers. There is an urgent need to create smart food traceability systems that are user-friendly and capable of enhancing food safety, reducing foodborne illnesses, and ensuring food security. Research efforts should focus on designing and developing specialized smart traceability systems tailored for different food supply chains, utilizing Internet of Things (IoT) technologies, cloud computing, and sensing technologies. The extensive data gathered from critical control points across the food supply

chain can be transmitted *via* the internet to cloud servers for real-time analysis. These technologies can optimize the recall process, reduce associated costs, increase transparency, and enhance the overall quality of food supply chain management by mitigating information asymmetries and strengthening coordination among network partners.<sup>22</sup>

The rising prevalence of foodborne illnesses and food insecurity highlights the urgent need for the development of advanced food traceability systems that are rapid, accurate, reliable, cost-effective, and capable of performing multifaceted analytical functions.<sup>23</sup> Thus, an effective food traceability system plays a crucial role in ensuring food authenticity, safety, and quality throughout the supply chain, minimizing risk of food-borne illness, fraud and contamination.<sup>8,24</sup>

### 3. Recent advancements in food traceability

The rising incidence of fraud and counterfeiting in the food sector, particularly in high-value products such as ham, wine, cheese, and extra virgin olive oil, has led to significant economic losses, unfair competition, and detrimental impacts on brand reputation.<sup>25</sup> By integrating advanced technologies, tracking, tracing, and identification of products across the supply chain, counterfeiting and fraud can be effectively detected and prevented. Techniques such as barcoding, blockchain technology, RFID, Near Field Communication (NFC), and printed graphic identifiers represent promising and potentially reliable methods for detecting and identifying instances of food fraud. This section provides an in-depth discussion of recent technological advancements that have contributed to improved food traceability.

#### 3.1 Radio frequency identification (RFID)

Recent advancements in Radio Frequency Identification (RFID) technology, including the integration of data loggers and

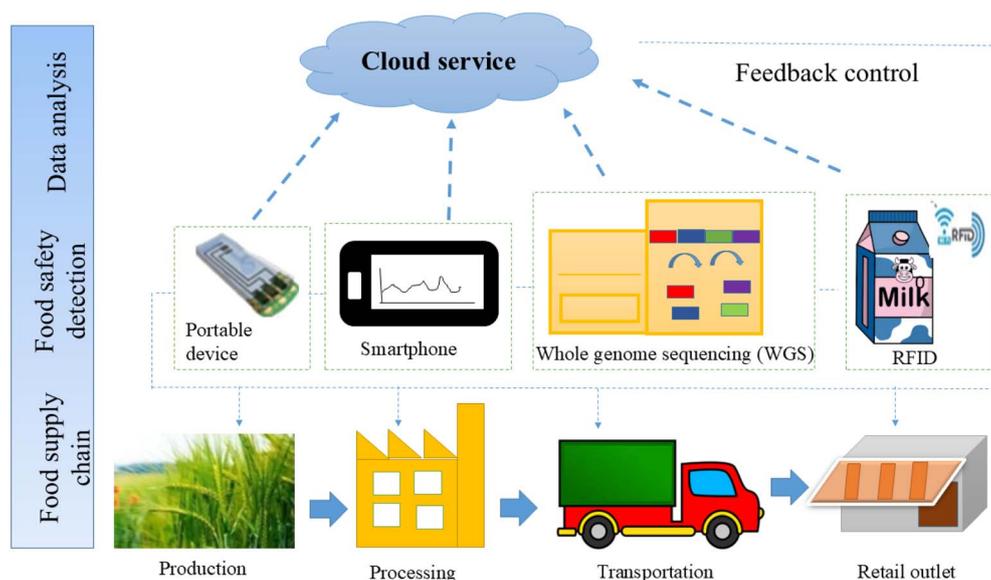


Fig. 1 Schematic overview of a smart food traceability system. Adapted and modified from ref. 20. Copyright © 2020 Taylor & Francis.



sensors, have significantly expanded its application in food traceability systems. RFID tags, particularly those equipped with temperature loggers, enable real-time monitoring and recording of the transportation and distribution conditions of perishable goods. Such insights are crucial for optimizing supply chain logistics and supporting intelligent inventory management practices such as the first-expired-first-out (FEFO) approach.<sup>26,27</sup>

**3.1.1 Key components and functionality.** An RFID system typically comprises three primary components: a reader, a transponder (RFID tag), and an antenna, supported by application software. The RFID tag contains a coupling element and a chip for unique identification, with the antenna facilitating the transmission of radio frequency signals between the tag and the reader.<sup>28</sup> RFID tags are categorized based on their communication mechanism and power source into active and passive tags. Active RFID tags are powered by an internal energy source, such as a battery, which limits their operational lifespan to the battery's capacity. In contrast, passive RFID tags lack an internal power source and instead derive their energy from the electromagnetic field generated by the RFID reader. When activated, RFID tags facilitate data exchange by transmitting stored information to the reader, which then processes and transfers the data to a connected computer system for further applications.

**3.1.2 Applications in the food industry.** RFID tags are extensively employed in the food industry for enhancing product identification and traceability throughout the supply chain, allowing faster recall of unsafe or expired foods.<sup>29</sup> They have been used to monitor 30 cold chain logistics routes by recording and transmitting temperature data to safeguard the quality of perishable goods<sup>9</sup> and they have improved automation and efficiency in enterprise management. Tags designed for cold chain logistics often feature enhanced protection against condensation, freezing, or heat, ensuring reliable data transmission and durability under harsh conditions.

A traceability system integrating RFID and IoT sensors was proposed by Alfian *et al.*, 2020, to monitor environmental parameters of perishable foods during transportation and storage.<sup>30</sup> RFID technology was employed to track and identify the distribution of low-quality products along the supply chain, while IoT sensors measured critical environmental parameters. Additionally, the study incorporated a machine learning algorithm based on XGBoost that was used in tandem with the RFID gates to identify whether goods are shipped or received, optimizing the tracking process. The XGBoost system was used to classify product movement direction and filter out false-positive tag reads. Based on previously reported studies, temperature and humidity were found to be important parameters for monitoring the agricultural food system.<sup>31</sup> Electronic Product Codes (EPCs) associated with each RFID-tagged item were transmitted through an EPC Information Service (EPCIS) to a host computer, thereby ensuring accurate, secure, and real-time traceability, as shown in trials involving the kimchi supply chain in Fig. 2(a).

Similarly, another traceability system, *i.e.*, the RFID-based Cattle/Beef Traceability System (RCBTS), was developed using

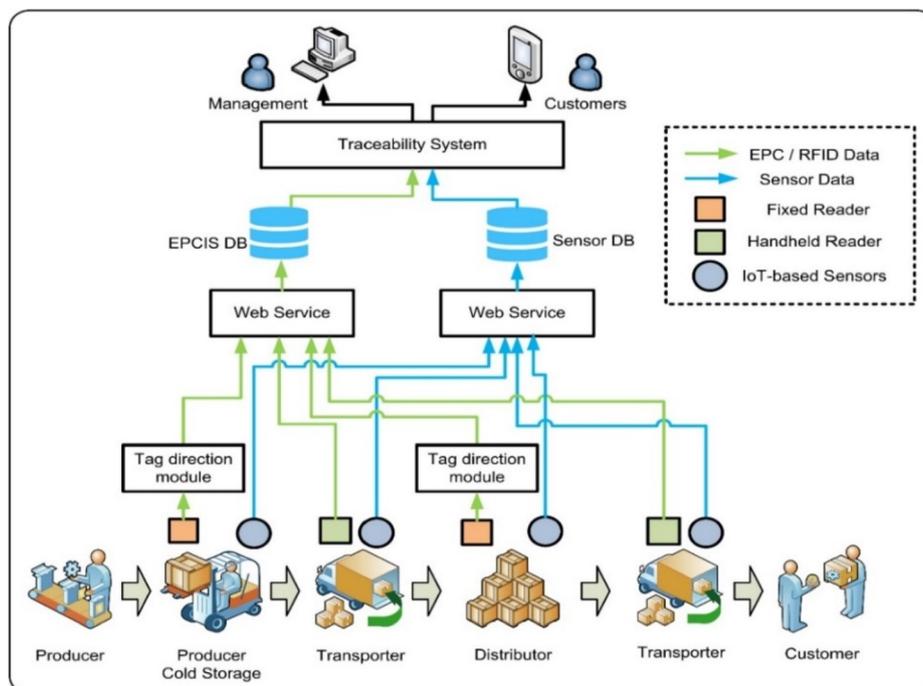
a Personal Digital Assistant (PDA) to facilitate real-time traceability management throughout the cattle/beef production process.<sup>20</sup> The system integrates RFID technology, a PDA, and a barcode printer to collect, process, and transmit traceability data across the cattle/beef supply chain. RCBTS provides a comprehensive framework for cattle and beef identification and traceability, covering the entire production flow from cattle breeding to the final product, as illustrated in Fig. 2(b). System trials demonstrated that the RFID-based traceability system significantly enhanced the automation, efficiency, and convenience in cattle/beef enterprise management.

**3.1.3 Challenges in RFID.** Even though several research studies have been conducted on RFID sensor technology, it still faces key challenges, including the use of food-safe materials, restricted energy harvesting and limited reading range, multi-tag collisions, security and privacy risks, recycling issues and high cost.<sup>32</sup> As research on polymer-based sensors was focused on developing sensors with smart functionalities, they lack food safety credentials. Thus, future research must be directed to the use of organic oils and biocompatible conductive inks and dyes as temperature-sensing materials because they do not compromise food safety. Another challenge was found in passive RFID tags, as they are limited by their short read ranges and can even create signal propagation issues. The functioning of RFID devices is also compromised by multi-tag collisions, which can reduce system efficiency, often requiring advanced algorithms such as binary tree or hybrid protocols for reliable identification. RFID also raises risks related to unauthorized data access, privacy leaks of supply chain information, and tampering of traceability records. Moreover, RFID tags generate waste, such as adhesives, chips, pieces of metal and conductive inks, which can impact the recycling process of other food packaging.<sup>33</sup> High cost is another concern in RFID technology as the unit price of RFID tags varies from 0.11\$ to 0.20\$ (USD per unit). Unit prices can be lowered by 17–33%;<sup>34</sup> however, improved fabrication techniques and materials, such as affordable substrates, metals, and conductive inks, are essential for lower tag prices. Further, the current economy emphasizes sustainable technology; thus, recyclable and reusable RFID tags will be beneficial for the green economy.

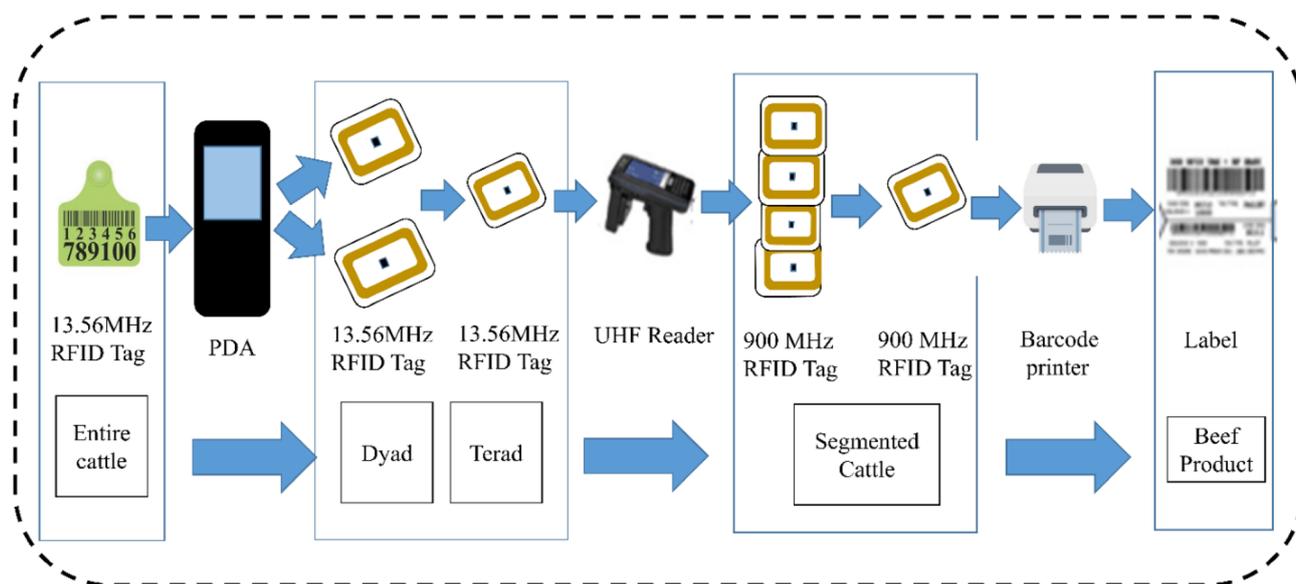
### 3.2 Blockchain in traceability systems

The implementation of blockchain in food chain applications is still in the early stages, but research interest in blockchain technology (BCT) dramatically increased in the 2020s.<sup>35</sup> A blockchain is a distributed database that is composed of orderly blocks that are chained together. Each block works like a ledger to store data. Decentralization is a defining characteristic of blockchains that enables them to be applied in supply chain management, including food supply chains. A study was reported in 2016 in which RFID was integrated with BCT to track the agri-food supply chain.<sup>36</sup> The model integrates a centralized database for supply chain stages with blockchain for environmental data, enhancing transparency in the agri-food sector. This approach ensures food safety, sustainability, and quality, while promoting low environmental and social impacts. By





(a)



(b)

Fig. 2 (a) A traceability system based on RFID and IoT-based sensors. Reprinted (reproduced) with permission from ref. 30. Copyright © 2019 Elsevier. (b) The process of data transformation and transmission between traceable units in RC BTS. Adapted and modified from ref. 20. Copyright © 2012 Elsevier.



integrating food traceability technologies (barcodes, QR codes, RFID, and IoT, *etc.*) with blockchain, their effectiveness can be improved and costs reduced.<sup>37</sup>

To demonstrate the practical benefits of blockchain-based food traceability systems within actual food supply chains, several companies have developed and implemented pilot programs. Walmart and Kroger pioneered the adoption of blockchain technology in their supply chain operations, initially focusing on applications for Chinese pork products and Mexican mangoes.<sup>38</sup> The blockchain technology reduced the time required to trace mango sources and transport pathways from farm to retail store in a few seconds. However, since each company uses its own system, it becomes challenging to implement a unified traceability system for all stakeholders across the supply chain.

BCT is in its initial stages of development; therefore, it requires a great deal of financial assistance to implement it in supply chains. Moreover, as the blockchain network becomes more complex, computational power increases, and a lot of energy is required.<sup>37</sup> Further, BCT is considered to be an immature technology as it is unable to store large amounts of data, and it is even found to have a negative effect on protecting privacy, as all the information about the supply chain will be available on the distributed ledger.<sup>37</sup> By resolving these issues, it will help BCT to be implemented in real-time scenarios of food supply chains.

### 3.3 Printed graphic identifiers

Printed Graphic Identifiers (PGIs) are visual barcodes or symbols printed on products, packaging, or documents that enable data storage and automatic retrieval through optical scanning technology. According to the International Electrotechnical Commission (IEC) and the International Organization for Standardization (ISO), a barcode (1D, 2D, or 3D) is a machine-readable format used for encoding information. It is represented by patterns or regions of varying reflectance on an object's surface, which are interpreted as binary digits ('1's and '0's) by an optical scanner. The scanner transmits the encoded information to a system for storage and processing.<sup>39</sup>

Printed Graphic Identifiers (PGIs) provide several advantages, including cost-effectiveness, rapid and accurate machine readability, and support for various symbologies (numeric, alphanumeric, and binary), which offer differing levels of error detection and correction. Additionally, PGIs can be produced on durable materials, such as printed media or metals, and are compatible with a wide range of optical scanning equipment.<sup>39</sup> This section examines the different types of PGIs employed in traceability systems.

**3.3.1 Linear barcode symbology.** Linear barcodes are among the most widely utilized forms of Printed Graphic Identifiers (PGIs), encoding information in a single dimension through varying widths of parallel light and dark patterns (bars and spaces).<sup>40</sup> These barcodes typically include guard bar patterns at both ends, which assist scanners in detecting the start and end of the code, enabling bi-directional scanning.<sup>39</sup> The operational mechanism of barcodes is analogous to a laser

beam scanning a horizontal cross-section of the vertical bars. As the beam moves across the symbol, it measures the relative durations spent on dark bars and light spaces, which are then decoded into individual characters using a lookup table.<sup>41</sup> To prevent data loss from damaged barcodes, 1D barcodes are often paired with the corresponding alphanumeric characters for manual readability. Common 1D barcode symbologies include UPC, EAN, Code 39, Code 93, Code 128, ITF-14, Codabar, GS1-DataBar, and MSI Plessey,<sup>42</sup> with examples illustrated in Fig. 3. The primary advantages of linear barcodes are their simplicity and the cost-effectiveness of the reader technology. However, their major limitation is low data storage capacity, restricting their role to serving as indices that link to external databases, and poor printing quality, which means that excessively small sizes or low resolution can adversely affect readability.<sup>43</sup>

**3.3.2 Two-dimensional (2D) symbology.** Two-dimensional (2D) barcodes, with their higher data storage capacity, can encode significantly more information than their one-dimensional (1D) counterparts by utilizing matrices of geometric patterns such as rectangles, dots, or hexagons.<sup>39,42,45</sup> These barcodes are categorized into two primary symbology types: stacked and matrix. Stacked symbols, such as Code 49 and PDF417, are arranged as rectangular blocks comprising multiple rows, each resembling a linear barcode. In contrast, matrix symbols, including DataMatrix, Maxicode, QR codes, and Aztec codes, consist of arrays of binary (black or white) cells organized in a grid.<sup>39</sup> The 2D barcodes enable high-density information storage within a compact footprint. Notably, Quick Response (QR) codes can store even larger volumes of data using four encoding modes: numeric, alphanumeric, byte/binary, and kanji. The decoding process for 2D symbologies requires scanning devices capable of reading data in both vertical and horizontal dimensions simultaneously.<sup>46</sup>

**3.3.3 Data matrix (DM) code.** Data Matrix (DM) codes are commonly used for labeling small items, goods, and documents. These codes can store up to 3116 numeric characters, 2335 alphanumeric characters, or 1555 bytes of binary data.<sup>39,47</sup> The U.S. Electronic Industries Alliance (EIA) specifically recommends DM codes for labeling small electronic components. Similar to QR codes, DM codes exhibit high fault



Fig. 3 Different types of barcodes: (a) EAN-13, (b) Code 39, (c) PDF-417, and (d) data matrix code. Reprinted (adapted) with permission from ref. 44. Copyright © 2006 Elsevier Ltd.



tolerance and rapid readability due to their encoding in a true digital format, which enhances error resistance, even under low-contrast printing conditions. Utilizing the Reed–Solomon error correction algorithm, DM barcodes of the  $10 \times 10$  GS1 size (nominal dot size –  $40 \mu\text{m}$ ) can still be recognized despite up to 20% physical damage, a capability absent in traditional barcodes.<sup>47</sup>

**3.3.4 Application of PGIs in food research.** The multi-technology integration approach by combining PGIs, RFID, and blockchain technologies represents a promising approach to resolving food traceability issues. A study was proposed by Wahab *et al.* in which QR codes and blockchain technology were integrated for halal meat traceability.<sup>45</sup> The QR code data consists of origin, breed, rearing and slaughtering conditions. These data are securely transmitted to the blockchain network at each stage to maintain a continuous, tamper-proof record of the product's journey. The researchers found that this dual approach shows positive potential for enhancing transparency, restoring consumer trust, and improving compliance rates. However, limitations included high implementation costs, standardization requirements, and infrastructure needs.

Another study was reported to develop a traceability system for fresh tuna loin quality based on various handling temperatures using quick response (QR) codes.<sup>48</sup> Sampling involved the selection of 19 respondents and detailed procedures for temperature-monitored collection, processing, and statistical analysis using One-Way ANOVA, supplemented by the design and expert validation of a QR code traceability system. The research validated QR code effectiveness for accessing real-time quality information, with laboratory testing confirming compliance with Indonesian national standards for histamine content, total plate count, and *Salmonella* detection.

**3.3.5 Limitations of PGIs.** Despite their advantages, Printed Graphic Identifiers (PGIs) have notable limitations. Linear barcodes offer low storage capacity, typically below 100 characters, while even data matrix codes are capped at 2335 alphanumeric characters. Additional drawbacks include dependence on specific symbologies and the requirement for line-of-sight readability. Barcodes must be scanned individually and often rely on manual tracking, making them prone to human error. Moreover, their machine readability is significantly compromised if symbols are damaged by adverse environmental factors such as moisture or mechanical abrasion.

## 4. Different approaches to self-healing and its integration into food traceability

Accidental damage to the material can be self-healed with different approaches, including physical, chemical, or physicochemical processes. This section discusses the classifications of self-healing mechanisms and details the different processes involved. The self-healing mechanism in printing technology for food traceability using printed graphic identifiers is also discussed.

### 4.1 Mechanisms of self-healing

Self-healing mechanisms can be generally classified as extrinsic and intrinsic, wherein the extrinsic mechanism requires the presence of a healing agent that is incorporated into the matrix to heal the damage, and the intrinsic mechanism mainly takes place with the help of external stimuli (UV, heat, or relative humidity). The intrinsic mechanisms are crucial for modern food packaging, enabling materials to repair themselves and maintain barrier integrity, mechanical properties, and ultimately food quality and safety.

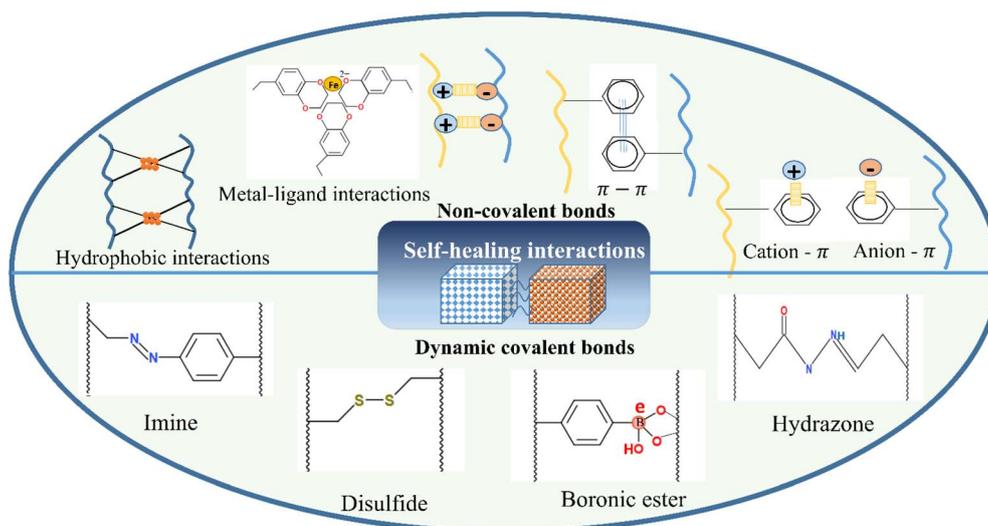
In food packaging, self-healing materials are developed using dynamic covalent and dynamic non-covalent interactions. Dynamic non-covalent bonds have a lower mechanical strength but can heal a material with little to no energy consumption at ambient temperature,<sup>49</sup> whereas covalent bonds exhibit higher stability, and bonding usually occurs in the presence of external stimuli (force, light, and heat).<sup>50</sup> Some typical dynamic covalent and non-covalent bonds used in self-healing are discussed in the following sections and are depicted in Fig. 4(a).<sup>49</sup>

**4.1.1 Dynamic non-covalent interactions.** Dynamic non-covalent interactions consist of hydrogen bonding and dipole–dipole, electrostatic, hydrophobic, van der Waals, and ionic interactions, which can be incorporated into diverse biochemical and chemical processes (Table 1). These interactions provide a great deal of flexibility and reversibility, which is affected by pH value, molecular density, light irradiation, and temperature.<sup>54</sup>

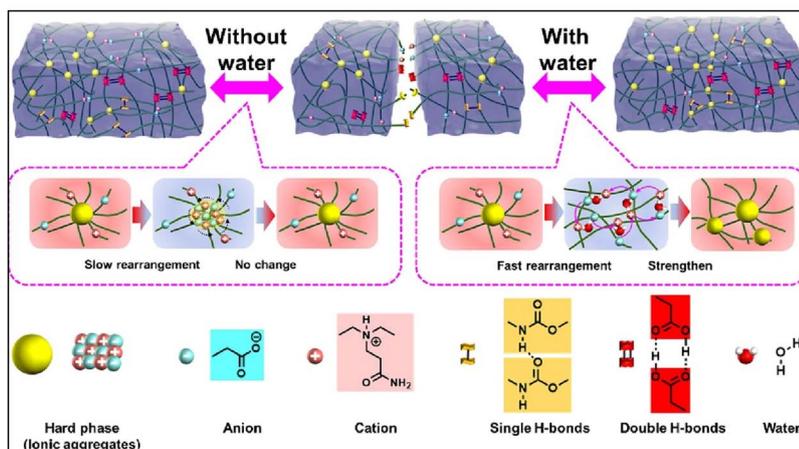
**4.1.1.1 Hydrogen bonding.** Hydrogen bonding is the strongest non-covalent interaction, occurring mainly between hydrogen and highly electronegative atoms such as fluorine, nitrogen, and oxygen. It plays a vital role in protein-ligand interactions, protein stability, and self-healing polymers based on chitosan, carrageenan, proteins, and starch. In polymer matrices, hydrogen bonds influence self-healing efficiency, with bond energies ranging from 5 to 30  $\text{kJ mol}^{-1}$ , they are much weaker than covalent bonds ( $\sim 345 \text{ kJ mol}^{-1}$ ),<sup>55</sup> but are quickly restored once broken.

The cellulose structure has abundant hydroxyl groups exposed and has an innate ability to facilitate hydrogen bonds. Polyvinyl alcohol (PVA) is a suitable candidate for binding with cellulose using hydrogen bonds because it has abundant hydroxyl moieties, which contribute to better self-healing ability.<sup>56</sup> Numerous studies demonstrating the self-healing properties of PVA and cellulose-based hydrogel have been published.<sup>62</sup> According to a study by Liu *et al.*,<sup>14</sup> the mechanical properties of nanocomposite hydrogels were improved by the electrostatic interaction of cellulose nanocrystals (CNC), quaternary ammonium xylan (QAX), and a suitable amount of PVA (20 wt%). With the aid of hydrogen bonds and the entanglement of long polymer chains, a cellulose-based optimized weight ratio of PVA and CNC of 60 : 1 produced a self-healing efficiency of 37.03% at room temperature without the need for any external stimulation.<sup>63</sup> However, hydrogen bonds are found to have weaker strength, and the self-healing materials made from hydrogen bonds exhibit weaker mechanical properties. Therefore, cross-linking agents can be used to improve

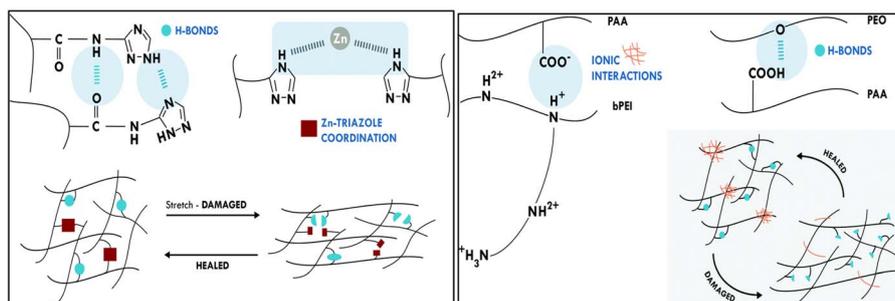




(a)



(b)



(c)

(d)

Fig. 4 (a) Representation of reversible dynamic covalent and non-covalent interactions for the preparation of self-healing hydrogels.<sup>49</sup> (b) Molecular behavior in the self-healing process based on the rearrangement of H-bonds and interactions, exhibiting distinct molecular behavior during healing in the absence and presence of water. Reprinted (adapted) with permission from ref. 51. Copyright © 2024 Elsevier. Schematic of (c) a combination of hydrogen bonds and metal–ligand coordination in poly(isoprene) (IR). Reprinted (reproduced) with permission from ref. 52. Copyright © 2017 The Royal Society of Chemistry. (d) The combination of hydrogen bonds and ionic interactions in bPEI/PAA/PEO. Reprinted (adapted) with permission from ref. 53. Copyright © 2019 American Chemical Society.



Table 1 Summary of dynamic non-covalent interactions and their application in self-healing

Non-covalent interaction	External stimulation for healing	Strength	Sustainability	Application	References
Hydrogen bond	Room temp	Moderate	High	Cellulose/PVA hydrogels, starch, protein films	55 and 56
Ionic interaction	Room temp/RH	Moderate	High	Zwitterionic polyurethanes, polyelectrolytes, PEI/PAA	57, 58 and 53
Host-guest	Room temp/water	Moderate	High	Cyclodextrin-adamantane coatings/1-menthol	59
Electrostatic	Room temp	Moderate	High	Layer-by-layer PEM films	60
Metal coordination	80 °C	High	Moderate	PAA/HEC hydrogels, Zn-triazole elastomers	61, 52 and 64

mechanical strength. For example, the fabrication of a hydrogel with non-cytotoxic and antibacterial qualities using aluminum(III) ions ( $Al^{3+}$ ) and carboxymethyl chitosan nanoparticles (CMCS NPs) to crosslink with polyacrylic acid (PAA) chains through triple dynamic non-covalent interactions (hydrogen, coordination, and electrostatic interactions) was used to study the synergistic effect of multiple non-covalent mechanisms. This mechanism of strong bonding achieved desirable mechanical properties (fracture elongation of 1930%, fracture strength of 190.9 kPa) and self-healing abilities (98.8% of strain, healing efficiency 92.9% of stress, 25 °C, 24 h of healing), and was not visibly affected by surface erosion or aging (e.g. alkali, salt solutions, and acid).<sup>64</sup>

**4.1.1.2 Ionic interactions.** Carboxypyridyl zwitterions, thio-betaine ions or imidazolium ions are common structures found in self-repairing materials that rely on ionic interactions.<sup>62</sup> The ionic interactions in amphiphilic ions occur due to unique water sensitivity, which dissociates negative and positive ions under humid conditions.<sup>58</sup> The associations between positive and negative ions are re-formed with the application of heat and drying treatment for the processing of self-healing materials. In the case of thermal transition, such as glass or melting transitions, the amphiphilic ions will re-form from water-sensitive weak dynamic non-covalent bonds to a permanent network. Recently, reversible ionic interactions and H-bonding have also been used to produce the self-healing capabilities of zwitterionic polymers. A study was reported on novel zwitterionic multi-shape-memory polyurethanes (ZSMPUs) from thio-betaine that was based on *N*-methyldiethanolamine (MDEA), hexamethylene diisocyanate (HDI) and 1,3-propanesultone (PS).<sup>58</sup> Thio-betaine forms an ionic bond with the permanent dot of polyurethane, and the glass transition temperature of the polymer acts as a molecular switch to exhibit shape-memory properties. Additionally, because amphoteric ions are sensitive to water, PU films that were split into two sections successfully re-bonded into a single piece at 30 °C and 80% RH, although a visible healing interface remained. However, after drying at 50 °C for 2 hours, the interface disappeared entirely, and the tensile strength and fracture strain of the repaired PU film were comparable to those of the original. A similar study related to polyurethane elastomers (PU-n) was reported in which tertiary amine groups and carboxyl groups were

introduced to PU-n to form H-bonds and intermolecular ionic interactions. This contributed to the formation of microphase separation structures, which resulted in a maximum tensile strength of 20.61 MPa and toughness of 59.02 MJ m<sup>-3</sup>.<sup>51</sup> The self-healing ability of PU-n occurs at room temperature and is significantly accelerated by water because the dissociation of hard phases enhances the molecular mobility, as depicted in Fig. 4(b). Following self-healing with water, increased molecular movement and further ionization of ionic groups strengthen the hydrogen bonds and ionic interactions, leading to more pronounced microphase separation. Consequently, the mechanical strength improves, and the self-healing efficiency reaches 104.72%. In ternary polymeric complexes of branched poly(ethylenimine) (bPEI)/poly(acrylic acid) (PAA)/poly(ethylene oxide) (PEO), the combined effect of hydrogen bonds and ionic interactions has been found to produce exceptionally high tensile strength (27.4 MPa) and self-healing efficiency (96%) at ~90% relative humidity conditions at room temperature,<sup>53</sup> as depicted in Fig. 4(c).

**4.1.1.3 Host-guest interactions.** According to Huang and Wang, the physical insertion and combination of the guest and host moieties result in host-guest interactions.<sup>65</sup> This results from hydrophobic interactions as well as the complementary size and shape characteristics of the host and guest molecules. Crown ethers, cyclodextrin (CD), cucurbiturils, cuproaromatics, and so on, are examples of common host molecules.<sup>62</sup> CD is used extensively because it is non-toxic, simple to produce, and contains a hydrophobic cavity to bind hydrophobic guests. Self-healing characteristics are demonstrated by the dynamic interactions between the host and guest molecules. Because it is easy to synthesize and structurally resembles a cyclic oligosaccharide with a cavity, cyclodextrin is frequently utilized as a host molecule.<sup>62</sup> Adamantine, ferrocene, and 1-menthol are examples of guest molecules that can interact with cyclodextrins in a host-guest manner. It was discovered that damaged coated regions expanded under the external action of water and self-healed within 20 minutes after  $\beta$ -CD grafted chitosan (CD-g-CS) was immobilized with 1-menthol through host-guest interactions.<sup>59</sup> The self-healing effectiveness of 1-menthol-containing coatings was found to be 59.49%, roughly 13.18% greater than that of the control coatings.



Table 2 Summary of dynamic covalent interactions and their application in self-healing

Covalent interaction	External stimulation for healing	Strength	Sustainability	Applications	References
Imine/Schiff	Heat, pH	High	Moderate	Chitosan starch hydrogels, gelatin/green tea based hydrogel	69 and 70
Acylhydrazone	pH, temp	High	Moderate	Phenolic pectin hydrogels, PU elastomers	71 and 72
Diels-Alder	30 °C	Very high	Low	Bio-derived BCPs, furan-modified pectin/maleimide-modified chitosan	73 and 74

**4.1.1.4 Electrostatic interactions.** Another effective non-covalent interaction is reversible electrostatic interaction, which can be used to form self-healing films, aerogels, and hydrogels. It is initiated between charged ions and polymers, polyampholytes, zwitterionic fusions, and polyelectrolytes. Layer-by-layer (LBL) assembly of polydopamine (PDA) and sodium alginate (SA) results in polyelectrolyte multilayer (PEM) films with self-healing ability.<sup>60</sup> In the reported study, (PDA/Alg) m PEM multilayer films were supplemented with nanocapsules made from chitosan and capsaicin (CAP). After mechanical scratching, the ions penetrate and free chains transfer in artificial seawater solutions, demonstrating self-healing characteristics in the electrostatic interaction between Alg and PDA.<sup>60</sup> Itaconic acid (IA), which is negatively charged, and positively charged chitosan (CS), were assembled to generate self-healing biomass aerogels.<sup>66</sup> The aerogel showed excellent mechanical strength and a very quick rate of self-healing because of its very high levels of electrostatic interaction. Upon disintegration, the aerogel restored its mechanical functionality and structural integrity in 30 seconds at room temperature by wetting the damaged surface.

**4.1.1.5 Metal coordination bond.** Metal coordination bonds have a substantially higher binding energy than other non-covalent bonds. The benefits of metal coordination bonds are that they are easily bindable, adaptable, and dynamically reversible. In order to produce a reversible supramolecular structure and accomplish a self-healing function, a metal ion can be introduced into a polymer matrix through coordination between metal ions and organic ligands.<sup>67</sup> Hussain *et al.* (2018) reported a study in which Fe<sup>3+</sup> was loaded onto a network of polyacrylic acid (PAA) and hydroxyethyl cellulose (HEC) to fabricate a self-healing hydrogel.<sup>61</sup> Due to hydrogen bonding, the polyacrylic acid and ethers possess abundant carboxyl groups, and the hydroxyl groups on the hydroxyethyl cellulose offer a significant amount of mechanical strength. The synergistic action of ion-dissipative coordination bonds between Fe<sup>3+</sup> and the carboxylic group of acrylic acid, in addition to hydrogen bonds, helps to increase the mechanical strength of hydrogel materials. When a hydrogel breaks, the free Fe<sup>3+</sup> ions diffuse to the vicinal interface, where they interact with the oxygen in the HEC backbone and the carboxyl groups of the PAA network. High tensile strength (1.35 MPa), widespread fracture strain (1660%), high toughness (8.8 MJ m<sup>-3</sup>), and exceptional self-healing efficiency (87%) were demonstrated by the HEC/PAA-

Fe<sup>3+</sup> hydrogel without the need for external intervention.<sup>61</sup> According to another study, a high-performance elastomer was developed by using stronger Zn–triazole coordination into an unvulcanized *cis*-1,4-polyisoprene (IR) matrix and employing several weaker hydrogen bonds, as illustrated in Fig. 4(d). The elastomer achieved good healing efficiency of 72% at 80 °C and good mechanical characteristics of 21 MPa.<sup>52</sup>

**4.1.2 Dynamic covalent interactions.** Dynamic covalent polymer networks undergo a self-healing mechanism when sufficient energy is supplied in the form of irradiation or heat or by an increase in the change of reactants, *e.g.* condensation reactions.<sup>68</sup> This type of bond provides excellent solvent stability and thermal stability. A summary of dynamic covalent interactions is listed in Table 2.

**4.1.2.1 Imine bonds.** Imine bonds, which are reversible covalent bonds, are formed through the facile condensation of aldehydes or ketones with primary amines. Variations in the aldehyde and amine groups within the material influence the strength of the covalent interactions and the self-healing properties of the resulting hydrogels. Liu *et al.* (2020) synthesized a chitosan hydrogel crosslinked with dialdehyde debranched starch (DADBS) *via* dynamic Schiff-base linkages between aldehyde groups in DADBS and amino groups in chitosan.<sup>69</sup> This hydrogel demonstrated rapid gelation within 30 seconds and exhibited excellent self-healing capabilities. The mechanical properties of such hydrogels can be controlled by adjusting the reaction temperature and the molar ratio of aldehyde to amino groups. Similarly, Xu *et al.* fabricated self-healing hydrogels using chitosan and natural vanillin as a cross-linking agent.<sup>75</sup> Chitosan was dissolved in aqueous acetic acid (5% w/v) and mixed with different concentrations of vanillin dissolved in anhydrous alcohol. It was found in the study that 0.3 g of vanillin gelled with 5% chitosan solution in 6 minutes and demonstrated optimal self-healing of around 5 h, without tackiness or an overly rigid structure. In this system, the self-healing ability was primarily attributed to the reversible formation of Schiff-base bonds, while hydrogen bonds provided stability at room temperature. An optimal aldehyde-to-amino group ratio of 7 : 1 was found to balance self-healing efficiency and mechanical performance.<sup>75</sup> Another study reported that hydrogels were synthesized from gelatin and green tea. It exhibited self-healing properties due to the synergistic effects of covalent (Schiff-base and imine bonds) and non-covalent (hydrogen bonding) interactions.<sup>70</sup> The study involved the



development of a self-healing hydrogel by oxidizing green tea polyphenols with sodium periodate and crosslinking them with gelatin in aqueous solution under alkaline conditions, and these interactions contributed to a self-healing efficiency of 53.4%, which could heal within 2 h. These studies underscore the critical role of dynamic covalent bonds and non-covalent interactions in the design of hydrogels with tunable mechanical properties and self-healing capabilities.

**4.1.2.2 Acylhydrazone bonds.** Acylhydrazone bonds are formed through the reaction between aldehyde and hydrazine functional groups under mildly acidic conditions (pH 4–7) or in catalytic environments at elevated temperatures. These bonds exhibit greater stability than imine bonds.<sup>65</sup> Chen *et al.* reported the synthesis of a degradable hydrogel in a mild NaHCO<sub>3</sub> solution by cross-linking poly(*N,N*-dimethylacrylamide-*stat*-4-formylphenyl acrylate) [P(DMA-*stat*-FPA)] with pectin acylhydrazide (pectin-AH).<sup>76</sup> This hydrogel demonstrated self-healing properties attributed to dynamic phenolic and acylhydrazone bonds and featured a microporous structure conducive to controlled drug release. Qiao *et al.* designed a self-healing hydrogel using dialdehyde-terminated polyethylene glycol (PEG-CHO) and adipic dihydrazide-modified alginate (ALG-ADH), which provided aldehyde and hydrazide functional groups, respectively, to facilitate dynamic acylhydrazone bond formation.<sup>71</sup> This hydrogel achieved a self-healing efficiency of 84.4% within 6 h under ambient conditions, driven by the reversibility of acylhydrazone bonds and multiple hydrogen bonding interactions.

**4.1.2.3 Diels–Alder (DA) reactions.** The Diels–Alder (DA) reaction, a click electrocyclic process between a conjugated diene and dienophile pairs, is characterized by its excellent selectivity, high yield, and minimal side reactions. Despite its advantages, self-healing mechanisms based on the DA reaction often require prolonged durations and elevated temperatures to be effective. For instance, bio-derived block copolymers (BCPs), such as poly(lactic acid)-*block*-poly(2,5-furandimethylene succinate) (PLA-*b*-PFS), utilize furan groups in the PFS block for crosslinking with bis(maleimide) triethylene glycol, as demonstrated<sup>73</sup> by Cai *et al.*, 2019. These materials achieved over 50% self-healing efficiency at room temperature within five days at lower crosslinking densities, while optimized crosslinking enabled 96.3% efficiency in just 5 min at 30 °C. Moreover, integrating the DA reaction with other reversible interactions has been shown to improve self-healing properties. For example, Li *et al.* (2021) reported the development of a hydrogel composed of furan-modified pectin and maleimide-modified chitosan that exhibited a two-step self-healing mechanism: initial electrostatic interactions followed by DA-based network formation.<sup>57</sup> In the study, pectin was chemically modified with furfural to introduce conjugated diene groups, and chitosan was modified with 6-maleimidocaproic acid to provide dienophile sites. The hydrogel produced displayed outstanding self-healing properties under mild conditions and was able to support a load of 500 g without sustaining any damage. The healing analysis was conducted for 5 h at 37 °C, and the two pieces joined seamlessly within that time.

## 4.2 Self-healing based biopolymers in food packaging applications

Food packaging materials are susceptible to damage during logistics and transportation, potentially compromising food quality.<sup>67</sup> Incorporating self-healing properties into these materials enables the restoration of functionality after damage, thereby reducing maintenance costs and enhancing the reliability, safety, and service life of polymeric materials.<sup>77</sup>

**4.2.1 LbL-based self-healing films.** The layer-by-layer (LbL) fabrication of polyelectrolyte multilayer films (PEMs) is a proven strategy to impart self-healing ability using various interactions, including  $\pi$ - $\pi$  interactions, hydrogen bonding, host-guest interactions, and covalent bonding.<sup>78</sup> For instance, an edible PEM coating fabricated from chitosan (CS) and carboxymethyl cellulose sodium (CMC) *via* electrostatic deposition exhibited self-healing functionality driven by ionic bonds (COO<sup>-</sup> and NH<sub>3</sub><sup>+</sup>) and hydrogen bonding.<sup>63</sup> This coating achieved an 87.4% self-healing efficiency and demonstrated effectiveness in preserving fresh-cut apples, highlighting its potential for freshness-keeping applications.<sup>63</sup>

Similarly, coatings developed using host-guest interactions through LbL assembly involved alternating layers of carbon nitride (C<sub>3</sub>N<sub>4</sub>), poly(ethylenimine)- $\beta$ -cyclodextrin (PEI- $\beta$ -CD), and poly(acrylic acid)-adamantanamine (PAA-AD).<sup>79</sup> Ultrasonication facilitated the formation of a homogeneous suspension of PEI- $\beta$ -CD-C<sub>3</sub>N<sub>4</sub> and prevented C<sub>3</sub>N<sub>4</sub> agglomeration. These coatings, applied to bananas, effectively inhibited ethylene production or degradation, supporting their use in freshness-keeping packaging. Additionally, a study demonstrated the application of a LbL assembly to create an edible coating using chitosan (CS) and sodium alginate (SA). The coating self-healed within 5 minutes upon water stimulation and exhibited recovery rates of tensile strength (19.89 MPa), oxygen permeability (95%;  $4.78 \pm 0.82 \text{ cm}^3 \text{ m}^{-2} \text{ day}^{-1} \text{ atm}^{-1}$ ), and water vapor permeability (63%;  $40.56 \pm 7.60 \text{ g m}^{-2} \text{ day}^{-1}$ ).<sup>80</sup> Furthermore, this (SA/CS)<sub>3</sub> coating effectively delayed strawberry deterioration, emphasizing its potential in food preservation.

**4.2.2 Structural modification based self-healing films.** The enhancement of self-healing and functional properties of polymers and films through the incorporation of specific additives and structural modifications has been reported in numerous studies. Recently, a study was conducted to fabricate films from banana peel by incorporating emulsified beeswax. The emulsified beeswax was mechanically agitated with an iced alkaline urea solution of LiOH/urea (7 and 12%, respectively). The results showed that the use of aminolysis-treated films enhanced the self-healing efficiency (~77%) of the films.<sup>81</sup> The aminolysis treatment was provided by introducing NH<sub>2</sub> groups using 1,6-hexane diamine on a cellulose-based matrix of banana-peel-based film. Aminolysis treatment improved the printability of film by enhancing its surface morphology, resulting in more favorable water contact angles that promote better ink absorption and increased surface energy.

A self-healing, mechanically robust polymer, poly(etherthiureas) (PETU), was blended with small amounts of



commercial cationic antibacterial agents, such as poly(ethylene imine) (PEI) or cetyltrimethylammonium bromide (CTAB). These additives improved both the self-healing efficiency and mechanical robustness of PETU while effectively eliminating *E. coli* (Gram-negative) and *S. aureus* (Gram-positive) on its surface.<sup>82</sup> Another innovation involved multilayer films created *via* alternate deposition of polyacrylic acid (PAA) and PEI polyelectrolytes by using a robotic dipping system under ambient conditions. These films exhibited high glass transition temperatures, superior elastic modulus, and exceptional oxygen barrier properties. An eight-bilayer PEI/PAA film demonstrated a complete restoration of its oxygen transmission rate (OTR <math>0.005\text{ cm}^3\text{ m}^{-2}\text{ day}^{-1}\text{ atm}^{-1}</math>) after ten stretching–healing cycles, achieving self-healing in high-humidity environments (>97% RH) within 10 minutes.<sup>83</sup>

A study was conducted to develop cellulose-based films by dispersing natural wax emulsions into an iced alkali-urea aqueous solution containing dissolved cotton linter pulp to create a uniform cellulose-wax suspension. The self-healing was analyzed through annealing at 150 °C because this allowed the wax to migrate over the film surface and thereby enhancing self-healing without compromising the mechanical strength (122 MPa) for about 10 scratching/annealing cycles.<sup>84</sup> These films exhibited increased water resistance (contact angle of 120°) and flexibility (11%). Similar study related to soy protein isolate (SPI)-based films combined with PEI (polyethyleneimine) and metal ions ( $\text{Cu}^{2+}$  or  $\text{Zn}^{2+}$ ) demonstrated superior self-healing and antibacterial properties. In the study, an SPI/PEI solution was uniformly dispersed, and  $\text{CuSO}_4$  or  $\text{ZnCl}_2$  solutions were added dropwise under stirring for 8 h. The highly branched structure and abundant amine groups of PEI enhanced the mechanical strength by disrupting the ordered structure of the SPI and forming extensive hydrogen bonds. The addition of  $\text{Cu}^{2+}$  further improved coordination bonds and antimicrobial activity, yielding tensile strain and stress of 81.78% and 10.09 MPa, respectively, which recovered to 90.17% and 105.57% after self-healing at 25 °C for 10 h (ref. 85) (Fig. 5(a)).

Low-temperature self-healing has been achieved using cellulose nanocrystals (CNC), hexamethylene diisocyanate, and dibutyltin dilaurate. Dynamic hydrogen bonding and metal–ligand covalent coordination conferred a rapid self-healing rate (99% within 1 h at 5 °C) along with excellent mechanical properties, including high elongation (230%), toughness (2538 MJ  $\text{m}^{-3}$ ), tensile strength ( $25.49 \pm 0.02$  MPa), and thermo-mechanical stability<sup>86</sup> (Fig. 5(b)). The self-healing mechanism of cephalopods has been emulated using ion doping and nanoclay complexation to create a coating capable of rapidly repairing surface scratches in water. By incorporating  $\text{CaCl}_2$ -derived counterions and montmorillonite nanobricks into polyelectrolyte multilayers through layer-by-layer self-assembly, the non-covalent polymer network was enhanced.<sup>87</sup> The strong electrostatic interactions between ions and nanoclay improved polymer chain mobility, enabling complete surface repair of scratches within 10 seconds.

**4.2.3 Challenges in large-scale implementation.** Analysing the research conducted on bio-based self-healing films highlights the challenges of implementing these systems on

a commercial scale. Chitosan/CMC-based polyelectrolyte multilayer coatings face hurdles related to cost and scalability. Precursors such as PEI and 1,6-hexanediamine enhance the self-healing efficiency but are not a sustainable solution, as they are not suitable for contact with food. Moreover, as the films are developed from synthetic or non-biodegradable materials, this raises environmental concerns. Similarly, host–guest interaction-based LbL coatings, although effective in extending the shelf life of fruit, also confront biocompatibility and regulatory hurdles.

Across these different systems, a recurring challenge is balancing functional enhancement, environmental sustainability, consistent performance under real-world logistics, and consumer acceptance. Especially, as biopolymers rely on additives such as wax or surface modification (aminolysis). Therefore, these studies emphasize that achieving robust self-healing films in food packaging not only requires innovation in material design but also must focus on resolving interconnected issues of cost, regulatory compliance, processing, and consumer safety. These factors must be resolved for the transition from laboratory innovation to reliable, cost-effective commercial technologies in the food industry.

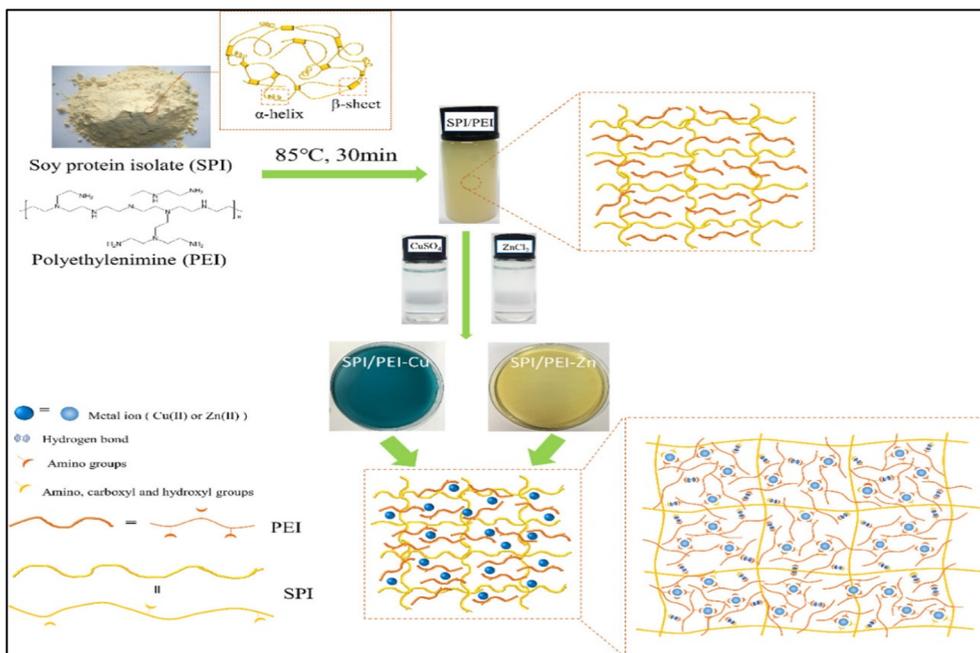
### 4.3 Self-healing inks utilized in 3D printing for food traceability applications

The 3D printing technique, formally known as additive manufacturing (AM), is a fabrication method that builds objects layer-by-layer from raw materials, contrasting with subtractive manufacturing processes, such as machining, which remove material to create objects (ASTM, 2012).<sup>88</sup> This method minimizes waste by utilizing only the exact amount of material required for production, thereby reducing costs and conserving resources.<sup>89</sup>

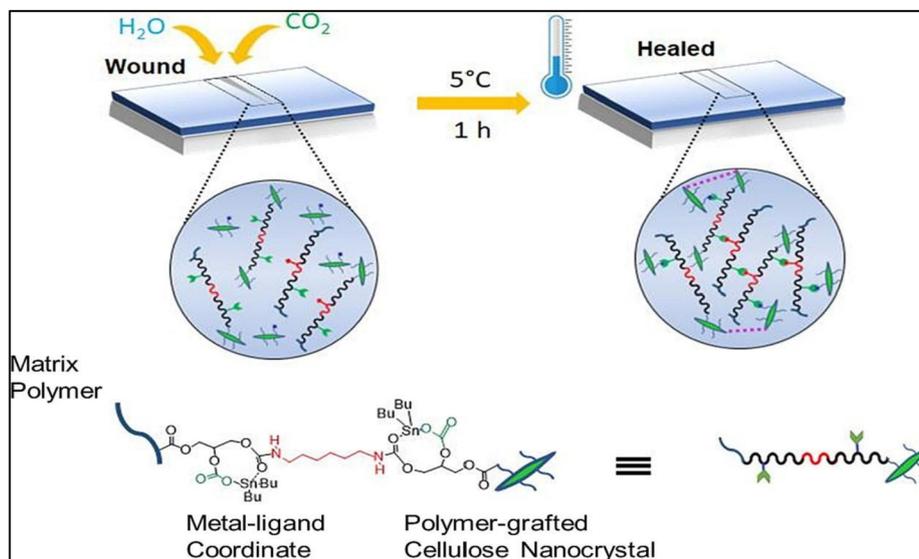
Additive manufacturing of smart materials facilitates the production of highly customizable printed components, with applications spanning soft robotics, shape-memory actuators,<sup>90</sup> advanced soft machines,<sup>91</sup> tissue engineering, and biomedical devices.<sup>92</sup> Commonly employed 3D printing techniques include vat photopolymerization, material extrusion, and powder bed fusion.<sup>93</sup> A study was reported to develop photoreactive resins with varying amounts of thermally reversible Diels–Alder cross-linkers for high-resolution stereolithography 3D printing.<sup>94</sup> By optimizing the dynamic cross-link content, specifically a polyacrylate network of 1.8 mol%, an excellent balance of 99% self-healing efficiency and strong shape stability was achieved after thermal repair, enabling durable, reprocessable printed materials suitable for sustainable manufacturing applications.

In additive manufacturing, incorporating Covalent Adaptable Networks (CANs) has established a novel paradigm for creating smart materials with self-healing and shape-memory capabilities. Printed objects with dynamic covalent adaptable networks can self-repair under specific stimuli such as light, temperature, or pH changes, while remaining stable under ambient conditions.<sup>95</sup> These networks exhibit either associative or dissociative behavior. A recent study reported the development of polyhydroxyurethane (PHU) based luminogens that





(a)



(b)

Fig. 5 (a) Fabrication process and schematic of the SPI/PEI-based films. Reprinted (reproduced) with permission from ref. 85. Copyright © 2019 American Chemical Society Ltd. (b) Schematic of self-healing achieved at low temperature in cellulose nanocrystal (CNC)/polymer nanocomposites. Reprinted (reproduced) with permission from ref. 86. Copyright © 2022 Elsevier Ltd.

exhibited shape memory, fluorescence and rapid self-healing properties.<sup>96</sup> The PHU-based paper composite samples were synthesized using a catalyst-free reaction between bis(6-membered) cyclic carbonates and amines at room temperature, followed by thermal curing at elevated temperatures (90 °C–105 °C). Further fluorescent patterns were encoded using light-mediated ink-free screen-printing. Due to the presence of

polyfunctional cyclic carbonate and amines, the composite exhibits programmable fluorescence, shape memory, and rapid self-healing (over 95% healed in 30 s at 160 °C) owing to thermally triggered transcaramoylation reactions. These embedded patterns are erasable, exhibit temperature-sensitive fluorescence, and are suitable for anticounterfeiting labels in food traceability applications.



#### 4.4 Self-healing integrated food traceability technologies

##### 4.4.1 Embedded symbology to enhance food traceability.

Food traceability has been made simpler for primary agro-food with the introduction of identification labels such as serial numbers or bar codes, which are attached either to the packaging or product. The Traceable Resource Unit (TRU) is an important component for implementing a traceability system.<sup>97</sup> It contains unique and precise data that accompanies the TRU throughout the supply chain.<sup>12</sup> Various identification technologies are used to identify different TRUs, including RFIDs and barcodes.<sup>46</sup> Embedded symbology technology helps to imprint QR codes or barcodes onto food products, thereby acting as a carrier of product information and facilitating sustainable traceability. Although this technology is an environmentally friendly process, it is sensitive to harsh environments and thereby susceptible to scratches and cuts. Thus, developing an embedded symbology technology with self-healing properties can be considered as a solution to this limitation.

Limited studies have explored the use of embedded symbology techniques for food traceability. Mc Inerney *et al.* (2011) investigated the potential application of electronic tracking in poultry by inkjet printing Data Matrix (DM) barcodes directly onto the beaks of broiler chickens in a live commercial environment.<sup>47</sup> The study aimed to identify a commercial ink capable of producing a durable barcode that was resistant to physical abrasion, as part of an integrated individual animal traceability system in a commercial setting. In the initial attempts, barcodes printed very well with 100% readability within a period of 10 s. However, after two days, it was found in visual inspection that the complete structure of the barcode eroded, as shown in Fig. 6(a). It appears that the hard keratinous surface of the beak did not support the retention of any commercially available ink types, even under the gentlest abrasive conditions, such as preening. Thus, the abrasion on the barcode may make it unreadable. This study proposes developing an ink that can adhere to the beak, and self-healable inks may provide such a solution.

Recently, a method was developed that utilizes 3D printing to embed information inside food, with a decoding system used to decode the information inside the 3D printed food using backlight illumination and a simple image processing technique.<sup>100</sup> Cookie dough was used as the target food to be embedded with an edible tag using black food-grade material. This study determined a way to embed edible tags, whether with air space inside the food or with secondary materials, to generate a specific pattern inside the food without changing the food geometry or adding any artificial materials. While the proposed study represents a novel approach to address durability limitations in embedded symbology systems, there is a constraint in the application to meat due to shrinkage during post-processing. This compromises the readability and scanning accuracy of embedded identification tags in meat-based products.

Another study explored the feasibility of printing on starch-based films for potential applications. Corn and potato starch films were developed using 50% glycerol, and Quick Response

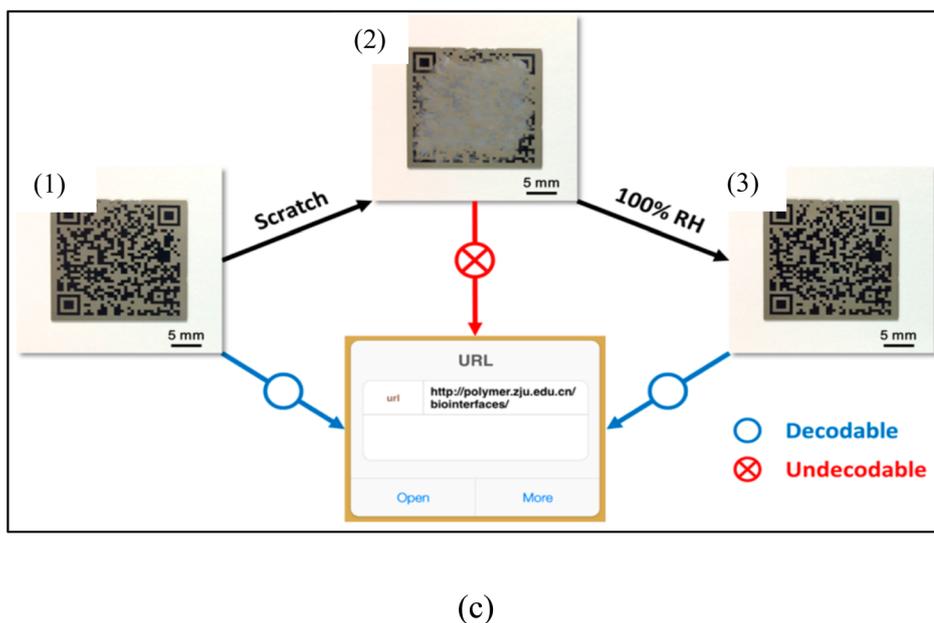
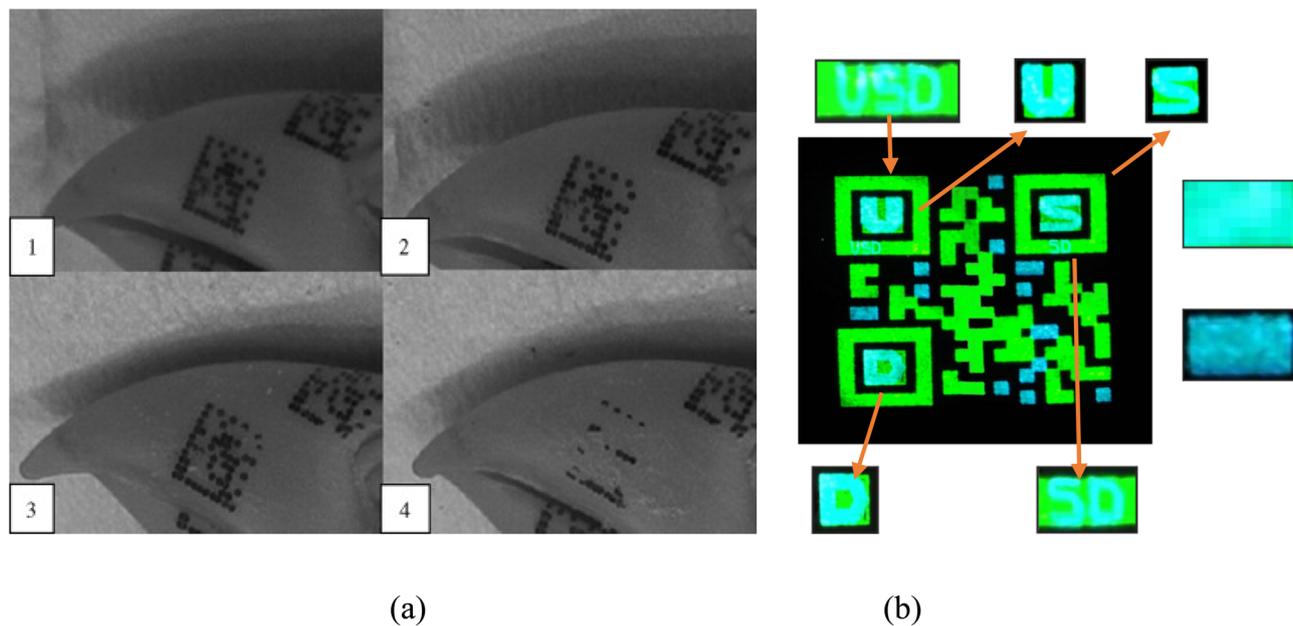
(QR) codes, text, and pictograms were overprinted using inkjet printing.<sup>101</sup> The results demonstrated that both types of films exhibited good print quality, with the overprinted QR codes being readable on smartphones. However, while potato starch films displayed higher optical density, the quality of lettering and sharpness was observed to degrade compared to corn starch films. Print quality was evaluated using image analysis, colorimetric parameters, and optical density measurements. The optical densities of potato starch films ( $1.61 \pm 0.06$ ) and corn starch films ( $1.46 \pm 0.06$ ) were significantly higher than those of conventional paper. These findings highlight the potential of starch-based films as modern, environmentally friendly packaging materials, capable of achieving high-quality printing. Further investigations could explore their applications in the food industry.

The application of barcoding technology can also serve to mitigate counterfeiting, which incurs significant financial losses for governments and private industries annually. As a potential solution, a study explored the use of lanthanide-doped  $\beta$ -NaYF<sub>4</sub> nanoparticles for security printing applications.<sup>98</sup> The ink formulation incorporated  $\beta$ -NaYF<sub>4</sub> nanoparticles doped with Yb<sup>3+</sup>/Er<sup>3+</sup> and Yb<sup>3+</sup>/Tm<sup>3+</sup>, with oleic acid serving as a capping agent, dispersed in toluene and methyl benzoate, and stabilized using poly(methyl methacrylate) (PMMA) as a binding agent. This ink was utilized to print Quick Response (QR) codes *via* Optomec's direct-write aerosol jetting technique. Enhanced security was achieved by combining green and blue upconverting inks, derived from Er<sup>3+</sup>/Yb<sup>3+</sup> and Tm<sup>3+</sup>/Yb<sup>3+</sup> pairs, respectively, within a single QR code, as shown in Fig. 6(b). The printed codes were invisible under ambient lighting conditions but could be activated using a near-infrared (IR) laser and scanned with a smartphone. This research highlights the development of security inks for QR code printing, offering potential applications in secure information sharing and anti-counterfeiting measures.

A self-healing, photo-cross-linkable polymeric film was developed using poly(ethyleneimine) (PEI) and photoreactive poly(acrylic acid) (PAA) assembled through a layer-by-layer (LbL) interaction, forming (PEI/PAA-N<sub>3</sub>)<sub>10</sub> structures.<sup>99</sup> This material exhibits a dynamic surface that allows smart labeling patterns of 2D barcodes with distinct color contrast, which can be reliably scanned using a CortexScan barcode reading application on a smartphone. The films remain stable for at least one year under standard temperature and humidity conditions. When subjected to abrasion with sandpaper, the barcode patterns became fuzzy and unreadable, as shown in Fig. 6(c). However, in a 100% relative humidity (RH) environment, the film's macroscopic appearance and machine readability were effectively restored. This recovery was attributed to the stable covalent network developed within the cross-linked regions, which restricted the mobility of most polymer chains. These embedded symbology techniques show promising potential for use in food traceability systems. A summary of self-healing-based embedded symbology is presented in Table 3.

The integration of self-healing embedded symbology into food traceability systems offers substantial sustainability benefits across economic, environmental, and social





**Fig. 6** (a) Sequence of deterioration of an inkjet-imprinted barcode by abrasion. Reprinted (reproduced) with permission from ref. 47. Copyright © 2010 Elsevier. (b) Upconverting image of QR code which has the literal text 'USD'; 'U'; 'S'; and 'D'; and 'SD' inserted in the code image with blue upconverting ink. Reprinted (reproduced) with permission from ref. 98. Copyright © 2012 IOP Publishing Ltd. (c) Optical images showing the recovery process of a (PEI/PAA- $N_3$ )<sub>10</sub> film that was encoded with a QR code. (1) Film without any treatment. (2) Damaged film. (3) Healed film after being exposed to 100% RH for 12 h. Reprinted (reproduced) with permission from ref. 99. Copyright © 2018 American Chemical Society.

dimensions. As reported in the research, self-healing systems frequently employ highly corrosive reagents (such as PEI, acrylamide derivatives), harsh organic solvents, and photo-reactive initiators that exhibit significant ecotoxicological risks. Therefore, future research must focus on the development of biodegradable materials with intrinsic self-healing polymeric coatings. This lowers the overall ecological footprint of supply chains. Self-healing mechanisms extend the functional lifespan of identification tags, decreasing the frequency of reprinting

and material consumption, which, in turn, reduces the energy use and carbon emissions associated with label production and replacement. Collectively, self-healing technology aligns with global sustainability goals by employing biopolymers in agro-food systems.

**4.4.2 Wireless-based detection technologies.** Wireless-based food safety detection systems utilize devices that collect data on food properties and wirelessly transmit this information for further analysis. This technology eliminates the need



Table 3 A summary of self-healing-based polymers and their interaction in food-related studies

Technology	Bio-polymer	Interaction	SHE	Purpose	References
Fabrication of polyelectrolyte multilayer films (PEMs) with self-healing properties	Chitosan (CS) and carboxymethyl cellulose (CMC)	Ionic bonds ( $\text{COO}^-$ and $\text{NH}_3^+$ ) and hydrogen bonding	87.4%	Preserving fresh-cut apples	63
	Carbon nitride ( $\text{C}_3\text{N}_4$ ), poly(ethylenimine)- $\beta$ -cyclodextrin (PEI- $\beta$ -CD), and poly(acrylic acid)-adamantanamine (PAA-AD)	Host-guest interactions through LbL assembly		Fresh-keeping packaging film for bananas	79
Enhancement of self-healing efficiency	LbL assembly of chitosan (CS) and sodium alginate (SA)	Hydrogen bond and ionic bonds	97%	Fresh-keeping packaging film for strawberries	80
	Polymeric blend of poly(ethylene imine) (PEI) and cetyltrimethylammonium bromide (CTAB), into poly(ether-thioureas) (PETU)	SHE was enhanced using poly(ethylene imine) (PEI) and cetyltrimethylammonium bromide (CTAB)		High bactericidal rate (>99.9%) against <i>E. coli</i> and <i>S. aureus</i> and has potential application in food industry	82
	Soy protein isolate (SPI)-based films combined with PEI and metal ions ( $\text{Cu}^{2+}$ or $\text{Zn}^{2+}$ )	Amine groups in poly(ethylene imine) (PEI) enhanced the mechanical strength and formed extensive hydrogen bonds with SPI	SHE was recovered to 90.17%	Superior self-healing and antibacterial properties	85
	Corn and starch films were developed with 50% glycerol	Self-healing mechanisms were not reported. Quick response (QR) codes, text, and pictograms were overprinted using inkjet printing		Potato starch films showed superior printing properties	101
Embedded symbology techniques and the assessment of self-healing and printing properties	Photo-cross-linkable polymeric film was developed using (PEI/PAA- $\text{N}_3$ ) <sub>10</sub>	Stable covalent network developed within the cross-linked regions, which was attributed to recovery		Label material	99
	Biodegradable film fabricated from banana peel and by incorporating beeswax	Aminolysis treatment was provided by introducing $\text{NH}_2$ groups using 1,6-hexanediamine	SHE was enhanced to 77%	Better printability properties and has potential application in food traceability	81



for additional circuits, enabling the wireless transfer of data in both active and passive modes.<sup>102</sup> Wireless Sensor Networks (WSNs) possess key features such as self-configuring, self-organizing, self-diagnosing, and self-healing capabilities,<sup>103</sup> making them advantageous for mobile and user-friendly food safety detection.

Wireless sensor networks are interconnected through gateways and coordinating devices, which monitor the physical and environmental conditions of the system. Each node in the network contains one or more passive or active sensors that communicate with each other to transmit data to a server that manages the entire network. Common technologies and communication protocols employed in WSNs include Bluetooth and Wi-Fi for physical and Media Access Control (MAC) layers, and ZigBee and 6LoWPan for network, security, and application layers.<sup>104</sup> A study involving a farm-to-fork project combined RFID technology with WSNs for wine traceability, tracking the product from vineyard to consumer glass.<sup>105</sup> In the pilot study, the ZigBee protocol was deployed in the vineyard to monitor environmental parameters. The data were collected in databases, and consumers will be able to retrieve information using the RFID/NFC/QR code. By integrating RFID and sensor network technologies, this system enables the precise recording of a food product's history, allowing Small and Medium-sized Enterprises (SMEs) to optimize business processes and maximize returns. Additionally, the system enhances consumer confidence and producer margins by demonstrating the quality and freshness of the product.

Self-healing technology has the potential to revolutionize electronics and wireless-based sensors by developing a system with soft, elastic and ultra-comfortable structures.<sup>106</sup> Recently, the development of flexible and self-healing electrodes has enabled wireless-based detection to monitor food and increase safety.<sup>6</sup> Pournoori *et al.* (2024) reported the design of a multi-phase-heterogeneous blend for a UHF RFID tag, which has the ability to autonomously repair micro-scale damage.<sup>106</sup> It consists of a conductive network (PEDOT (poly(3,4-ethylenedioxythiophene)-rich nanofibrils)) that is a multi-phase, self-healing elastomer matrix, achieving conductivity of approximately  $150 \text{ S m}^{-1}$ . The PEDOT polymer can reorganize itself to restore both conductivity and mechanical integrity when damage has occurred. This polymer aims to fabricate a proposed self-healing RFID antenna, which has huge potential in the industry.

## 5. Future scope and challenges

Conventional techniques in food traceability have become outdated in this current digital era, and further advancements will require the use of blockchain, AI, IoT, or big data. These advanced technologies can enable digital, intelligent, and real-time food traceability systems to improve food authenticity, quality, and safety, and prevent food fraud, and reduce food waste. For instance, numerous studies have highlighted the advantages of combining IoT and blockchain to improve traceability.<sup>107</sup> The research was mainly conducted on aquatic animals in cold chain logistics. The results ensured the tamper-

proof integrity and accuracy of the data and improved customers' trust. Therefore, the IoT can be combined with other smart traceability tools such as RFID, WSN, and barcodes to improve the efficiency of the traceability system. Blockchain has also received research attention as a promising technique to ensure safety and prevent fraud. However, consumption of large amounts of energy, high implementation costs, latency, and scalability compromise the functioning of blockchain.<sup>4</sup> AI has the potential to enhance digitalization and automation, detect adulteration and food fraud, while big data helps enhance food quality and safety, and improve decision-making. Thus, integration of AI-based technologies has great potential to enhance food traceability in real-world scenarios.

Integrating self-healing capabilities into digitized food traceability systems promises to reduce maintenance costs, extend operational lifespans, and bolster reliability across diverse supply-chain applications. As regulatory frameworks increasingly emphasize transparency and sustainability, self-healing materials are capable of autonomously repairing sensors, labels, and packaging films, which has the ability to enhance the resilience of next-generation traceability platforms. Nevertheless, widespread adoption of fully digitized traceability faces persistent barriers such as high implementation costs, poor compatibility with legacy industrial processes, and a shortage of technical expertise to manage advanced digital tools.<sup>4</sup> Overcoming these challenges will require sustained technical innovation, cross-sector collaboration among industry, regulators, and academia, and targeted training programs to build the skills needed for deploying AI-driven traceability and self-healing technologies on a large scale.

## 6. Conclusion

Food traceability systems are inherently data-intensive, as they involve collecting large volumes of data from various critical control points throughout the food supply chain, including harvesting, transportation, storage, processing, and distribution. There is a pressing need to develop advanced smart food traceability systems to enhance food safety, mitigate foodborne outbreaks, and ensure food security. Recent studies have focused on the development of self-healing materials through dynamic non-covalent and covalent interactions, or their synergistic effects. This review highlights recent advancements that have the potential to incorporate self-healing capabilities into food traceability systems.

Although literature on integrating self-healing with technologies such as RFID, WSN, and barcodes into food traceability systems is limited, substantial progress has been made in applying self-healing materials to food packaging, where films and coatings restore mechanical and barrier functions to enhance product protection. Efforts have explored self-healable UHF RFID tags and barcode-encoded photo-crosslinkable films; however, industrial adoption remains hindered by high implementation costs and the need for reliable long-term stability. This review highlights innovative perspectives for developing industry-oriented, self-healable, digitized food traceability tools, aiming to inspire future advancements in this interdisciplinary field.



## Author contributions

Krishna Gopalakrishnan: conceptualization, methodology and writing-original draft preparation. Arnav Bayan: writing-original draft preparation. Prof. Poonam Mishra (corresponding author): conceptualization, validation, supervision, writing-reviewing and editing.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

No data are generated for the present study.

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