



Review and Perspectives of sustainable, biodegradable, eco-friendly and flexible electronic devices and (Bio)sensors

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ABSTRACT

The demand for increasingly advanced bioelectronic devices with smart functions, long-term stability, and the ability to operate outdoors for extended periods of time has been growing rapidly in modern society. In addition, excellent flexibility, biocompatibility, lightweight design, sustainability, biodegradability and environmental friendliness are equally important considerations when choosing mobile electronics. To meet these requirements, flexible bioelectronic devices need to be equipped with efficient portable tasks. However, most of these devices are currently manufactured using plastic substrates/supports, which is not environmentally sustainable. This review is focused on research attempts to shift biodegradable polymeric materials toward flexible and sustainable components. We present recent scientific achievements in the strategies to obtain eco-friendly polymers for flexible substrates/support to fabrication of bioelectronic devices. We summarize the introduction of biodegradable polymers and techniques to fabrication of portable bioelectronic devices as components to provide flexible and sustainable substrates/support. Lastly, we summarized the few applications reported on biodegradable polymeric films toward flexible bioelectronic devices and commented on the challenges and future prospects for unexplored sensing and biosensing.

1. Introduction

For many years, petroleum-based polymers have been produced and used on a large scale due to their attractive properties, such as lightweight, cheap, and easy to process, or having high strength and stiffness (Kasprzak et al., 2022). These materials can be tailored to suit various applications, making them ideal for flexible and wearable energy storage and (bio)sensing systems used to mobile phones, tablets, laptops and smartwatches (Kasprzak et al., 2022; Siddiqa et al., 2023). However, conventional non-biodegradable polymers pose a serious environmental problem since they are resistant to aging and biological degradation, which could result in pollution after disposal (Kasprzak et al., 2022; Siddiqa et al., 2023). Furthermore, the most of monomers used to create plastics are from fossil hydrocarbons, accelerating climate change (Kasprzak et al., 2022; Siddiqa et al., 2023). The synthesis process of petroleum-based polymers also involves harmful compounds or toxic

byproducts (Kasprzak et al., 2022; Siddiqa et al., 2023). To address these concerns, researchers have been intensifying their efforts to find eco-friendly alternatives to petrochemical-based materials by exploring bioresources (Kasprzak et al., 2022).

Biodegradable polymers are molecularly chain containing repeated units as such as proteins (silks and gelatin) and polysaccharides (cellulose, sodium alginate, chitin and chitosan) (Cui et al., 2021). They can be classified in agreement with the origin source being i) plant-based: cellulose, hemicellulose, glucomannan, agar, starch, acacia gum; or ii) animal: chitin, alginates, carrageenans and xanthan gum. In cellulose-based materials, chemical modification by esterification processes adding functional groups lead to the new materials as such as hydroxypropyl methyl cellulose, carboxymethyl cellulose and methyl cellulose while etherification processes produce cellulose nitrate and cellulose acetate (Aziz et al., 2022). Chitin is a classical example that can be considered a cellulose derivative in which the hydroxyl groups on the

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second carbon of each glucose unit have been replaced by the acetamide group ($-\text{NH}(\text{C}=\text{O})\text{CH}_3$) (Kulkarni et al., 2012), while deacetylation of chitin results in chitosan production (Wang et al., 2020). Another possibility is the obtaining of biocomposites which consist of the mixture of two or more components being a matrix and a reinforcement to produce a hybrid material with unique features compared to their mono-constituted counterpart (Boonsiriwit et al., 2021). Flexible devices have been integrated into different materials such as paper, thin films, elastic structures and fabrics (Lou et al., 2020; Teymourian et al., 2020; Yang and Cheng, 2020). Due to the great adaptability to the different substrates, structures, surfaces and matrices, flexible devices can be manufactured based on biodegradable polymers (Huang et al., 2019). Besides coming from renewable resources, biodegradable polymers increase their versatility by being non-toxic, low cost, biodegradable, biocompatible and resistant to mechanical stress (Cui et al., 2021; da Silva et al., 2020; Otoni et al., 2020). Not much emphasis on earlier work on biosensor development using ecofriendly-biodegradable materials and their comparison with biosensors made of non-biodegradable in terms of sensitivity, usability and cost was given here because most of the published papers on biosensor development using ecofriendly-biodegradable materials are focused on the use as modifier, part of device composition (composites), sensing elements or matrix. Differently, our review is focused on the use of flexible and sustainable substrates/support substituting plastics from petrochemical sources in which this topic has been few explored, however the researchers have directed the attention due to economic, social and environmental importance of this hot topic. In addition, we could see clearly that the most of published manuscripts did not contain comparison with devices made of non-biodegradable substrates in terms of sensitivity, usability and cost may be because non-biodegradable materials had a superior performance.

The aim of this review is to highlight biodegradable polymers that can be used as solid support for the development of flexible bioelectronic

devices and sensing. As the application of biodegradable polymers is innovative and the most of them have hydrophilic features, it is crucial understand the production process and the main features of biodegradable polymers to assemble the sensors and bioelectronic devices. Fig. 1 provides a schematic overview of the potential applications of biodegradable polymeric materials in flexible bioelectronic devices. More detailed information on these applications will be provided later. Our review contain the main biodegradable polymeric materials as such as cellulose acetate (CA), methyl cellulose (MC), hydroxypropyl methyl cellulose (HPMC), polyvinyl alcohol (PVA), chitosan (QT), carboxymethyl cellulose (CMC), starch (AM) and poly (lactic acid) (PLA) used to assemble devices allowing a more complete understanding of how biopolymers can be modulated for the development of solid supports. These biopolymers were chosen due to the biocompatibility, biodegradability, bioactivity, excellent flexibility, light weight, mechanical, thermal and chemical stability that are desirable features to use as substrate/support for flexible electronics devices (Alexandra Rodriguez Gaviria et al., 2021; Kasprzak et al., 2022; Liu et al., 2020, 2022; Miao et al., 2018). Further, they are found to be an inexpensive source of polymeric material offering a renewability, sustainability and scalability (Kasprzak et al., 2022). Moreover, biodegradable and sustainable polymeric materials have attracted growing attention in the development of ecofriendly, biocompatible and flexible bioelectronic devices, in especial supercapacitors, batteries and sensing of visualized UV, temperature, pH indicator, strain, pressure, temperature and wearable electronics (Alexandra Rodriguez Gaviria et al., 2021; Liu et al., 2020, 2022; Miao et al., 2018) indicating to be an efficient and green alternative for the design of electronic devices.

2. Fabrication methods of ecofriendly substrates

Solvent casting (Teixeira et al., 2021) and extrusion (Karkhanis et al., 2017) are the most common methods used to fabrication of substrates.

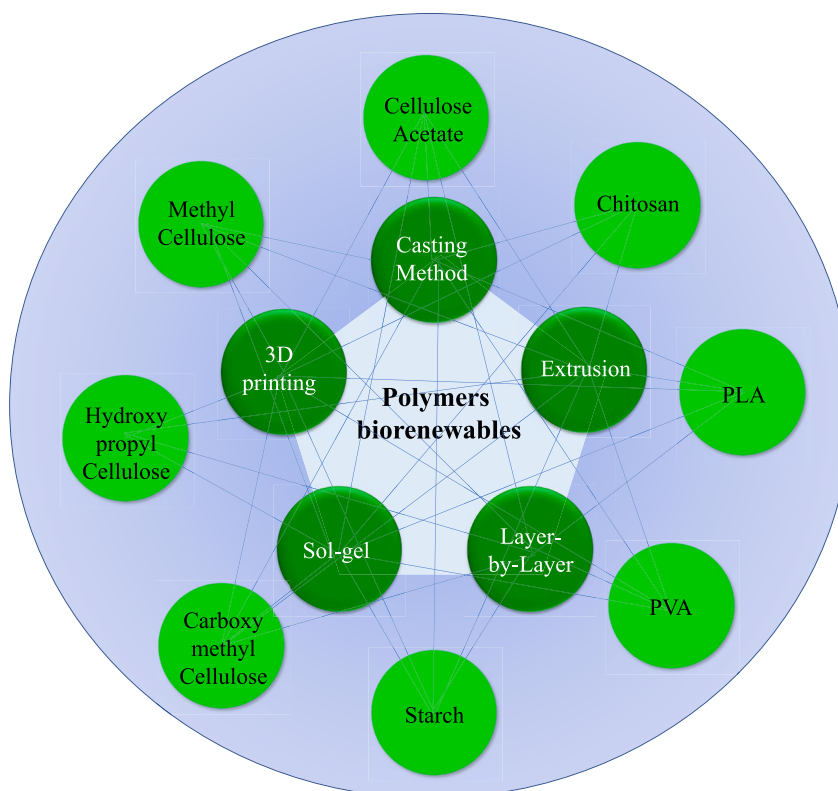


Fig. 1. Potential routes for applications of biodegradable polymeric films in flexible bioelectronic devices.

However, several new advancements and novel procedures have been developed in recent years, e.g., layer-by-layer (Moon et al., 2020), solution blow spinning (O Gomes et al., 2023; Paschoalin et al., 2022), bacterial cellulose films (Silva et al., 2020), sol-gel (Owens et al., 2016) and 3D printing (Li et al., 2022).

2.1. Solvent casting

At laboratory and pilot scales, the casting method (also known as solvent casting) is commonly employed to preparation of polymeric substrates in which it is necessary to know the chemical and physical properties of the components used to film formulation (Fig. 2) (Suhag et al., 2020). The substances must be dispersed in compatible solvents, e.g. cellulose acetate is dispersed in acetone, while HPMC is dispersed in water. Some polymers require a specific time in contact with the solvent before the complete dispersion followed by stirrer (magnetically) to achieve uniformity of the particles. The dispersion is then spread on glass plates (Petri plates are commonly used), and plates are placed in ovens for solvent evaporation and drying. The films are peeled off and cut to the desired and stored in packaging (Hay et al., 2018; Salawi, 2022; Teixeira et al., 2021).

The key benefit of this processing approach is the ease and simplicity of preparation without the use of sophisticated equipments. The major disadvantages of solvent casting are the shape limitations usually flat; sheets are the only geometries that may be made; the possibility of solvent retention in the films; the denaturation of the proteins and other molecules due to the use of organic solvents to dissolve the reagents; and the toxicity of organic solvents producing waste and environmental issues (Chen et al., 2008).

2.2. Extrusion

Fig. 3 shows the extrusion method that was developed to meet the limitations of solvent casting. Extrusion technique is used to polymer processing industries being a continuous high-speed manufacturing method producing thin, highly aligned monolayer or multilayer films (Barborik and Zatloukal, 2020). The technique uses an extruder to spread, homogenize, compact and melt polymer pellets being subsequently pushed through the uniform die. The thick sheet formed is then intensely stretched toward the machine employing a constant rotation with a circumferential speed, larger than the average polymer melt speed. The macromolecules are oriented and decrease the film thickness, and the film dimensions are set owing to the sufficiently high cooling rate (Panzer and Petoussi, 1993).

Manufacturers process a wide range of materials employing film casting technology in according with the current industry practice, where a wide variety of films are created with a necessity for usage in several applications. Low-density polyethylene (LDPE), high-density polyethylene (HDPE), linear low-density polyethylene (LLDPE), polypropylene (PP), polyethylene terephthalate (PET) and polystyrene (PS) are non-biodegradable polymers obtained by extrusion (Barborik and Zatloukal, 2020). The extrusion also is employed for the production of biodegradable polymeric substrates with hydrophobic features such as cellulose acetate and PLA (Erdmann et al., 2021; Karkhanis et al., 2017; Mallegni et al., 2018; Mohanty et al., 2003).

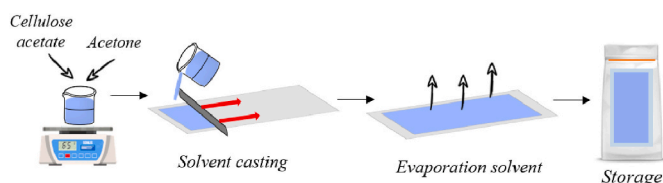


Fig. 2. Casting method of film formation.

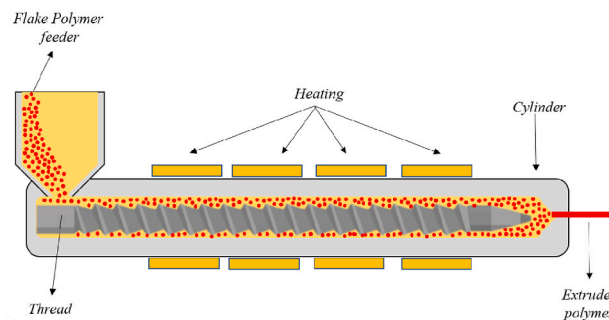


Fig. 3. Schematic representation of film production by extrusion method.

2.3. Layer-by-layer

Layer-by-Layer (LbL) is a process for assembling multilayer films in which the layers are deposited on a substrate surface through alternative adsorption of interacting materials (Fig. 4). The materials interact with each other through driving forces such as electrostatic interactions, hydrogen bonds, covalent bonds, or biospecies interactions. These properties enable the controlled release of added additives, such as antioxidants, with great applicability in the biomedical field (Salari et al., 2018). The LbL process is a versatile and flexible technique for building multilayer thin films with precise control over film thickness and composition. The films can be made from a wide range of materials, including synthetic and natural polymers, nanoparticles, and biomolecules. The LbL process can be performed in solution or on solid surfaces, and the resulting films can be patterned and shaped to create complex three-dimensional structures. The controlled release of additives from LbL films is of particular interest in the biomedical field, where it can be used to deliver drugs or growth factors to specific tissues or cells. The release rate of the additives can be controlled by varying the thickness and composition of the film, allowing for precise control over the timing and dosage of the released substances. LbL films have also been used for the immobilization of enzymes and other biomolecules, creating biocompatible and biologically active surfaces for a wide range of applications. The LbL technology is a powerful tool for creating functional thin films with a wide range of applications, including drug delivery, biosensing, and tissue engineering. Its flexibility and precise control over film composition and thickness make it an attractive option for the development of complex and functional materials (Moon et al., 2020).

The fabrication of multilayers on a substrate by consecutive adsorption of typically polymeric polyanions and polycations leads to the formation of one-dimensional structures along the normal layer (Decher et al., 1998). The steps in the production process of Layer-by-Layer films can be understood as shown in the work of J. Liu et al. (2019) (Liu et al., 2019). The mixture prepared by the authors became a gel and was dried in a vacuum oven. Then the gel was immersed in an aqueous solution of copper sulfate and rinsed with

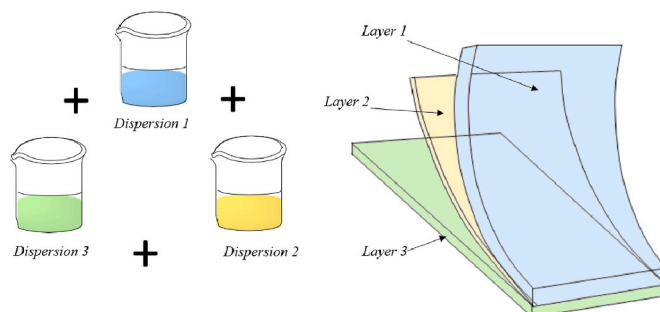


Fig. 4. Layer-by-Layer film production.

distilled water repeated by three times. The obtained multilayer films were dried in a vacuum oven at 40 °C for 24 h. LbL films are advantageous for industrial applications due to the mean three reasons: i) simplicity in the fabrication procedures; ii) variety of materials that may be employed; and iii) film robustness (Oliveira Jr. et al., 2022).

2.4. Sol-gel

The sol-gel technique is a versatile method for creating a wide variety of solid materials with diverse morphologies, including films, coatings, powders, and fibers. The sol phase is a suspension of solid particles, typically nanoparticles, in a liquid medium, while the gel phase is a continuous solid network that is formed when the sol is allowed to undergo gelation and finally film formation, as shown in Fig. 5. The sol phase can be modified by changing the chemical composition or concentration of the sol, or by introducing additives to the sol. Similarly, the gel phase can be manipulated by varying the gelation conditions, such as the temperature, pH, and drying method. These variations in the sol and gel phases allow the creation of materials with tailored properties and structures. Sol-gel technology has numerous applications in fields such as materials science, optics, electronics, catalysis, and biomedicine. For example, sol-gel-derived coatings can improve the durability and functionality of metal and glass surfaces, while sol-gel-derived nanoparticles can serve as catalysts for a range of chemical reactions (Owens et al., 2016).

The sol-gel transition is the transformation of a sol, or colloidal solution, into a gel through the growth of links between particles, or molecular species, resulting in the development of a three-dimensional solid network. As a result, the initially viscous system takes on an elastic character. Nonetheless, this transition differs significantly from the traditional solidification of a liquid (Brinker and Scherer, 1990). Li et al. (2007) (Li et al., 2007) proposed that ϵ -caprolactone, an important polyester, can be hybridized with inorganic titanium dioxide via in situ sol-gel process to suppress the crystallization of the polyester chains and thus increase the biodegradation rate by controlling the formation of nanophase structure of the hybrid materials.

2.5. 3D printing

3D printing has emerged as a powerful tool in materials engineering, allowing the rapid production of complex structures with precise control over shape, size, and composition (Fig. 6). This technology has enabled the development of a wide range of materials, including polymeric films with specific shapes and composite structures. Compared to traditional manufacturing methods, such as tape casting, 3D printing offers several advantages. One key advantage is the ability to create complex geometries and shapes that are difficult or impossible to achieve with conventional methods. 3D printing also allows a precise control of the material properties, such as porosity, pore size, and pore structure, which is critical for many applications. In the context of polymeric films, 3D printing can be used to produce films with specific shapes and composite structures, which can be challenging to achieve with traditional tape casting methods. With 3D printing, materials engineers can quickly prototype and test different film designs and composites, accelerating the research and development process. Overall, 3D printing presents a new platform for materials engineering research and has the potential to revolutionize the way materials are designed,

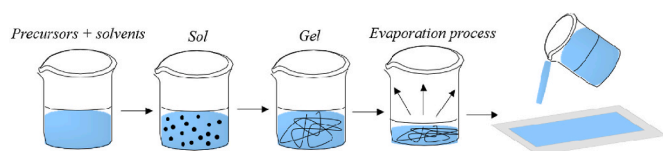


Fig. 5. A schematic view of sol-gel process.

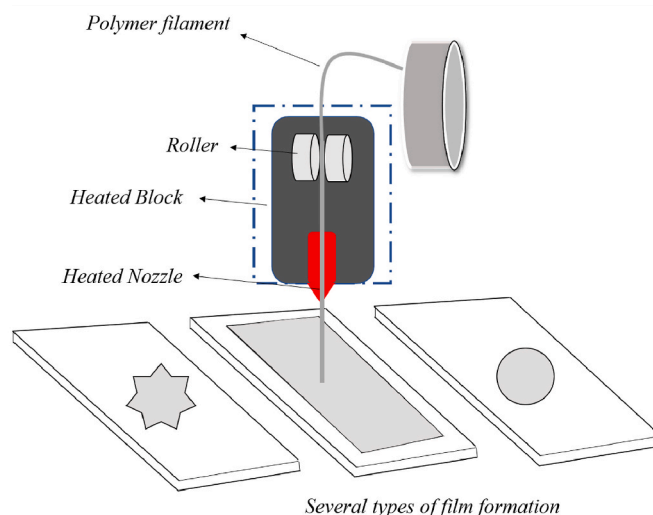


Fig. 6. 3D printing of polymeric films.

manufactured, and used in a wide range of applications (Li et al., 2022). In order to develop a cassava starch film with antibacterial and antioxidant properties, Li et al. (2022) (Liu et al., 2022) used 3D printing in which the starch was dissolved in acetic acid and other components to generate a polymeric dispersion to be placed in a cylinder to be printed using a 3D printer and the film was formed.

3. Biomaterials for substrates production

3.1. Cellulose acetate (CA)

Cellulose acetate is a modified natural polymer, biodegradable, obtained through the esterification of cellulose with acetic acid, responsible for the substitution of cellulose hydroxyl groups by acetyl groups (Watabe et al., 2018). The degree of substitution (DS) significantly changes material properties, particularly thermal properties, hydrophobicity, transparency, processability, and solubility. Among the altered material properties, hydrophobicity and processability differ greatly from natural cellulose in which the CA can not be dissolved in common solvents (Zhou et al., 2016). Furthermore, the glass transition temperature of cellulose acetate tends to decrease with increasing DS (Teramoto, 2015).

Cellulose acetate is a versatile and cost-effective material with a range of potential applications including an attractive choice for use in textiles, filters, photographic films, plastic materials, and packaging (Teixeira et al., 2021, 2022). Recent research has explored new uses for cellulose acetate, including impregnating films with carvacrol for antimicrobial properties (Adamovic et al., 2018; Rodriguez-Amaya, 2019); bio-based cellulose acetate films plasticized with glycerol or triethyl citrate for food packing (Teixeira et al., 2022); highly flexible and stable carbon nitride/cellulose acetate porous films with enhanced photocatalytic activity for contaminant removal from wastewater (S. Wang et al., 2020). The applications are underpinned due to the better processability than pure cellulose; biocompatibility; excellent optical clarity and high toughness; dispersed in various solvents; versatility in processing method, including extrusion, injection and compression, as well as its comparatively low cost to performance ratio compared to other polymers.

Considering the properties of cellulose acetate, its use as a support material for printing bioelectronics devices is promising. Some methodologies employed for the fabrication of cellulose acetate substrates including the dispersion of 10% CA in acetone, sealed and kept at rest for 24 h, with subsequent manual homogenization for 2 min, poured onto glass plates (25 × 10 cm²) for solvent evaporation (25 °C + 2 °C)

(Teixeira et al., 2021).

Another methodology involved preparing films from cellulose acetate by dissolving 1 g of CA in 10 mL of acetone and stirring the mixture for 24 h at room temperature. The resulting solution was homogenized and poured onto glass plates measuring $25 \times 10 \text{ cm}^2$. The solvent was then evaporated using forced air circulation for 30 min at room temperature. The films were stored in polyethylene/nylon packaging under vacuum. Prior to analysis, the substrates were conditioned at $23 \pm 2 \text{ }^\circ\text{C}$ and $50 \pm 2\%$ relative humidity for 16 h (Freitas et al., 2020). Laroque et al. (2021) (Laroque et al., 2021) prepared the substrates by mixing 50 g of CA per liter of acetone, subsequently 10 mL of the dispersion was poured into a Petri dish (9 cm diameter) and drying was performed in a closed plate. The films were stored at $23 \pm 1 \text{ }^\circ\text{C}$ and $67 \pm 3\%$ relative humidity (RH). Teixeira and co-workers (Teixeira et al., 2022) used the casting method to prepare cellulose acetate films with the plasticizers glycerol (GLY) or triethyl citrate (TEC) combined with natural anthocyanins extracted from açai producing a colorimetric sensor for microbial spoilage in shrimp samples (Teixeira et al., 2022).

The use of cellulose acetate as a substrate for flexible bioelectronics has not been extensively reported in the literature, as it is a relatively new and innovative application of this material. However, as mentioned, there are other studies that have used cellulose acetate for different applications, such as immobilization of immunoreagents, synthesis and functionalization with zinc oxide nanocomposites (Ahmadi et al., 2021; Florou, 2000; Padmalaya et al., 2019; Wu et al., 2006), and as a matrix or sensing layer for enzymatic electrochemical (bio)sensors (Ren et al., 2009) and immunosensors (Chauhan and R. Solanki, 2019). We have summarized in Table S1 the current studies focused on applying and adapting of CA with conductive materials as composite materials for devices dedicated batteries, triboelectric nanogenerator, self-powered active vibration sensor, optical biosensors, biofuel cell, and piezoresistive, fluorescent and strain sensor (Fu et al., 2018; Han et al., 2021; Jia et al., 2019; Monisha et al., 2017; Paixão et al., 2021; Qin et al., 2022; Varghese et al., 2022; Yu et al., 2019). These studies highlight the versatility of cellulose acetate as a biodegradable and sustainable material with potential applications in a wide range of fields.

3.2. Methylcellulose (MC)

Methylcellulose (MC) is a cellulose ether that finds widespread use in various industrial applications, including the development of polymeric films (de Carvalho Oliveira et al., 2010; Nasatto et al., 2015; Silva et al., 2022) in which it is synthesized by methylation at positions 2, 3, and/or 6 of the glycosidic units of cellulose. MC is a biopolymer with desirable properties such as non-toxicity (GRAS), renewability, easy synthesizability, industrial viability, and water solubility (in contrast to cellulose) (FDA, 2016). The methylation of methyl groups along the polymer chain can be either heterogeneous or homogeneous when the chains are dissolved before the methylation reaction (Ruda et al., 2021).

The versatility and applicability of MC make a promising substrate for producing flexible bioelectronic devices. However, its high hydrophilicity is a major drawback because sometimes its use requires contact of the substrate with water in one of the steps. To overcome this challenge, some alternatives can be employed, such as developing polymer blends with starch or adding other compounds such as hydrophobic plasticizers, cellulose nanocrystals, or other chemicals to minimize its high hydrophilicity.

The production of MC films is based on the dispersion of the polymer in heated water. Some methodologies can be highlighted as the developed film by Silva et al. (2020) (da Silva et al., 2020) where 1% (w/v) of MC was dispersed in distilled water at pH 7 and maintained at mechanically stirred, with a gradual increase in temperature. In another study, a dispersion was prepared by mixing of MC powder with 1/3 of the total required volume of distilled water heated at $70 \text{ }^\circ\text{C}$ and using magnetic stirring to complete dispersion of particles (Braham et al.,

2022). The dispersion was then stored overnight at $4 \text{ }^\circ\text{C}$ for removal of bubbles. Films were made by the casting technique where 70 g of the material was spread on acrylic plates using a film applicator. The films were dried in a climatic chamber for 24 h ($40 \text{ }^\circ\text{C}$ and 60% R.H.), removed from the acrylic plates and then conditioned at $25 \text{ }^\circ\text{C}$ and 53% R.H.

Hashim and co-workers (Hashim et al., 2023) reported the preparation of a film of methyl cellulose and agar by the solvent evaporation method to which sunflower wax (SFW) and Chinese cabbage (CPC) anthocyanins were incorporated. The MC/Agar/SFW/CPC film showed an acceptable response level with good color variation for acidic media between pH from 2.0 to 6.0 and in basic pH range between 7.0 and 14 (Hashim et al., 2023). The biodegradable film was applied to monitor quality of Fresh food (chicken breast) aiming at the design of intelligent packaging for food quality control (Hashim et al., 2023). MC films can be prepared with soy wax and anthocyanins from red cauliflower (Hashim et al., 2022), and chitosan nanofiber and anthocyanins from barberry (Alizadeh-Sani et al., 2021) to monitor pH levels in food samples. In Table S2, we present a summary of the MC applications in triboelectric nanogenerator for human physiological monitoring (Wang et al., 2022), supercapacitor (Nadirah et al., 2020) and capacitor (Dannoun et al., 2022) in which MC was combined with cellulose nanocrystal (Wang et al., 2022), polyacrylonitrile/LiI (Nadirah et al., 2020) and dextran (Dannoun et al., 2022). In flexible bioelectronics devices, the MC has been used to only modify transducers (Sohouli et al., 2020), however, this kind of application is not the focus of this review. A transparent and flexible methyl cellulose-based substrate was prepared using a simple solvent evaporation process. The interconnect electrodes were made with a pattern of silver/carbon nanotube nanocomposite by using a stencil mask for wearable touch sensor applications in a light-emitting diode display (Kim et al., 2022). Due to the excellent features, the use of MC as substrate for producing flexible bioelectronics is promising specially for sensors, supercapacitor, biofuel cell and batteries applications.

3.3. Hydroxypropylmethylcellulose (HPMC)

Hydroxypropylmethylcellulose (HPMC), also known as hypromellose, has interesting properties such as water solubility, superior film forming quality, good biocompatibility and biodegradability (Ding et al., 2015). HPMC, a cellulose derivative, is produced by treating natural cellulose with sodium hydroxide toward directed alkylation of hydroxyl groups from natural cellulose (Mašková et al., 2020). Various strategies have been investigated to manufacture films for the use in tissue engineering, in pharmaceuticals and in the food industry (Burdock, 2007; Ghadermazi et al., 2019; Ghorbani et al., 2018; Mašková et al., 2020).

HPMC types differ by their chemical structure (degrees and ratios of substitution), resulting in different molecular weights, viscosities and particle sizes. Regarding their mode of preparation, aqueous solutions are most commonly used, although hypromellose can also be dispersed in aqueous alcohols such as ethanol and 2-propanol as long as the alcohol content is less than 50% w/w. Mixtures of dichloromethane and ethanol can also be utilized as solvents. However, solutions prepared with organic solvents tend to be more viscous (Shah et al., 2020). The gel formation process for film preparation is highly influenced by temperature, and complete polymer hydration takes place at low temperatures (Haque and Morris, 1993; Mitchell et al., 1990).

We highlighted two studies that proposed the preparation of films based on HPMC. Akhtar et al. (2012) (Akhtar et al., 2012) dissolved 6 g of HPMC in a 35% ethanol solution for 40 min at $65 \text{ }^\circ\text{C}$ using a magnetic stirrer with heating. Once the stirring was finished, the polymeric dispersion was degassed at room temperature under vacuum for 30 min. Films were made by pouring 6 g of the dispersion into Petri dishes and left in a dark room (pre-equilibrated at $20 \text{ }^\circ\text{C}$, 50% RH) to dry on a level surface for 48 h. Atarés et al. (2011) (Atarés et al., 2011) developed films

with 5% (w/w) HPMC in deionized water at 80 °C keeping the stirred overnight. Homogenization was performed using a vacuum mixer at 13,500 rpm for 3 min, and the dispersions were degassed at room temperature. The material was poured onto plates and left to dry on a flat surface for 24 h at 45% RH and 20 °C. The films were removed from the surface of the plates and stored at 25 °C in controlled relative humidity.

However, like MC, HPMC polymer has high hydrophilicity representing a challenge for use in electrochemical detection. Various strategies have been investigated to minimize the hydrophilicity, including the addition of hydrophobic plasticizers and the incorporation of other compounds such as cellulose nanocrystals or other chemicals. Hay et al. (2018) (Hay et al., 2018) aiming to improve the properties of HPCM films, suggested the ability of amylose to form helical complexes with hydrophobic ligands to incorporate lipids into a polymer film. Incorporating the hydrophobic sodium palmitate ligand via the amylose complex increases the crystallinity and reduces the hydrophilicity of the films resulting in improvements in several properties, such as gas barrier. The incorporation of a hydrophobic agent into a polymeric material is a more effective way to obtain a product with enhanced physical properties.

The HPMC films are essentially applied to construct colorimetric sensors for pH and ammonia monitoring in foods using smart packaging (Boonsiriwit et al., 2021; Huang et al., 2021; Yao et al., 2021; Zhou et al., 2021). We have summarized in Table S3 the other current studies focused on applying and adapting of HPMC with silver nanowires (Zhang et al., 2022), silver nanofibers (Li et al., 2021), MgTf2/BMIMTf (Chong et al., 2016) and PVA/CuO (Sandhya Rani et al., 2022) conductive nanomaterials for devices devoted to wearable strain sensor (Zhang et al., 2022), electrochromic devices (Li et al., 2021), biodegradable solid polymer electrolyte (Chong et al., 2016) and ion conducting thin films (Sandhya Rani et al., 2022). Similarly, to the MC films, there were no reports on the use of HPCM as a substrate for flexible bioelectronic devices.

3.4. Carboxymethylcellulose (CMC)

Carboxymethyl cellulose (CMC) is a commonly used polysaccharide produced through a chemical modification of cellulose, whereby carboxymethyl groups are attached to the hydroxyl groups of the cellulose chain (Kanikireddy et al., 2020). The degree of substitution of CMC can affect its properties including the molar weight (Kanikireddy et al., 2020). CMC has a wide range of applications including in materials engineering and pharmaceutical/biomedical areas (Kanikireddy et al., 2020; Liyun et al., 2009; Teleszko et al., 2019). One notable application of CMC is in the food industry as a thickening and stabilizing agent due to its water-solubility and edible properties. CMC is also used to produce biodegradable, flexible films with good mechanical properties, which can have potential applications in a range of fields, such as packaging and biomedical engineering (Su et al., 2010).

The production methods of CMC substrates are based on the dispersion of the polymer in water. Abdollahi et al. (2019) (Abdollahi et al., 2019) produced films dispersing 2 g of CMC in 200 mL of distilled water by continuous stirring at 1200 rpm for 30 min in a heated bath at 70 °C. The polymeric dispersion was degassed under vacuum for 5 min to remove air bubbles. Then 15 g of the material was poured into Petri dishes and the solvent was evaporated in a ventilated oven at 45 °C for about 24 h. Finally, the film was removed from the plate and stored in a desiccator at $50 \pm 5\%$ relative humidity (saturated magnesium nitrate solution) and 25 ± 2 °C for 48 h. Similarly, Trevisol et al. (2019) (Trevisol et al., 2019) prepared CMC films dispersing 1.5 g of the feedstock in 100 mL of distilled water under stirring at 900 rpm at 50 °C. After complete dispersion, the mixture was deaerated with a vacuum pump. Amounts of 100 g of the dispersion was poured into Petri dishes and dried by a casting method in a convection oven at 40 °C for 20 h. The dried film was removed from the Petri dish and conditioned until use.

The large number of hydroxyl and carboxyl groups in the CMC

structure results in highly water-sensitive, which can limit the use in certain applications where water stability is important, such as in the development of solid supports or other materials that require resistance to moisture.

An interesting alternative was developed by Abdollahi et al. (2019) (Abdollahi et al., 2019) in which the authors proposed the addition of agar that presents highest water resistance and essential oil with hydrophobic characteristic already known. The contact angle analysis indicated that the incorporation of oil into the CMC resulted in a decrease in water affinity of the films and a lower interaction rate between film components and water molecules. This suggests that the addition of oil can reduce the water sensitivity of the films and improve their water resistance, which can be beneficial for a range of applications where water stability is important (Abdollahi et al., 2019).

Table S4 summarize applications of flexible and biodegradable CMC films. While CMC has been widely studied and utilized in various fields, including as a functionalizer, stabilizer, synthesizer and modifier for electrochemical sensing (Araque et al., 2014; Chu et al., 2021; Medřiková et al., 2019; Piovesan et al., 2017), and also colorimetric sensing of melanine in environmental and clinical samples (Kong et al., 2022), of silver and iron ions (Da Silva Júnior et al., 2021) and of pH (Yang et al., 2022), the use as a substrate has not been widely reported in the literature. The unique properties of CMC, including the water solubility and biocompatibility, represent a promising material for a range of applications, including in the field of flexible bioelectronic devices. In biosensing applications, nanofibrous mat of CMC is properly used to immobilize polyaniline nanorods and laccase (Lac) for the detection of catechol (Fu et al., 2015) or for fabrication of microarrays strips to detect three bacterial toxins: cholera toxin, staphylococcal enterotoxin A and toxic shock syndrome toxin in an immunoassay with high storage stability (Shlyapnikov et al., 2014). However, the use as a substrate for this purpose may require further investigation and optimization, as the hydrophilicity and water sensitivity of CMC can present challenges in certain environments. Nonetheless, researchers continue to explore the potential of CMC and other cellulose-derived polymers in a range of applications and it is possible that new advances and developments in this area may emerge in the future.

3.5. Polyvinyl alcohol (PVA)

Polyvinyl alcohol (PVA) is a synthetic polymer made by the polymerization of vinyl acetate to form a water-soluble polymer by solution casting technique which is an excellent property to obtain flexible bioelectronic devices. PVA is commonly used in a variety of applications due to its excellent film-forming properties, high tensile strength, and good adhesion to various substrates. PVA is non-toxic, biocompatible and biodegradable with easy processability and high hydrophilicity making it useful in many different industries, including textiles, paper, food packaging, and pharmaceuticals (Saini et al., 2017; Shah et al., 2020). PVA can also be used as a binder, thickener, emulsifier, and dispersant, and it has a wide range of molecular weights and degrees of hydrolysis that can be tailored to specific applications (Agunos et al., 2020; Liu et al., 2021; Ma et al., 2018).

The solubility of PVA depends on some factors: i) the size of the macromolecules; ii) the number of hydroxyls that remain free in the molecule; iii) the amount of acetate groups and aldehyde radicals are attached to it; iv) and the etheric bonds present in the molecule. Generally, PVA with higher molecular weight is less soluble than derivatives at a lower stage of polymerization (Julinová et al., 2018).

The production methods for PVA films basically encompass the casting technique, which is widely used for the production of biodegradable materials. In this technique, the polymer is dispersed in a solvent to obtain the film-forming dispersion, which is then placed on a support allowing the solvent evaporation. Ogunsona and Mekonnen (2020) (Ogunsona and Mekonnen, 2020) produced 10% (w/v) PVA films by dissolving the powder in deionized water at 90 °C under

stirring. A portion of 50 g of dispersion was poured in a 165 × 115 mm rectangular glass template. The dispersion was left to dry at room temperature overnight followed by removal of the film from the plate, they were dried in an oven at 80 °C for 2 h. In another work, 4 g of PVA powder were dispersed in 250 mL of deionized water under stirred for 10 min. The mixture was kept overnight to promote better polymer dispersion. The next day, the PVA dispersion was magnetically stirred up to 65 °C for 4 h and poured onto glass plates forming the film by drying in a hot oven for 1 h (Jha and Shimpi, 2022).

PVA is highly hygroscopic, susceptible to water vapor transmission, and exhibits relatively poor mechanical performance compared to other polymers commonly used for substrate applications. Therefore, an alternative approach has been proposed to mitigate these issues is developing multi-layer films consisting of different polymer layers that possess specific properties such as excellent water vapor and oxygen barrier performance. Ogunsona and Mekonnen (2020) (Ogunsona and Mekonnen, 2020) developed films with the addition of layers of cellulose nanocrystal (CNC) to PVA films. The presence of multiple CNC layers suggests that moisture penetration is impeded as the transport path for water molecules through the films becomes more tortuous. Despite the hydrophilic nature of CNCs, the authors expected the water resistance to decrease further. However, the results were promising, as water permeability actually decreased possibly due to the restrictive effect of the CNC layers, which block and prevent water molecules from permeating through the PVA polymeric networks (Ogunsona and Mekonnen, 2020). Another alternative is to use the mixing of PVA with less hydrophilic polymers to avoid the desintegration of the films in the presence of high humidity. Singha and Kapoor (2014) (Singha and Kapoor, 2014) synthesized PVA and starch using citric acid as the plasticizer and glutaraldehyde as the crosslinking agent. The hydrophilicity of PVA decreased because the free volume between the polymer chains was filled due to hydrogen bonding between PVA/starch and the interactions promoted between citric acid and glutaraldehyde.

In Table S5 we summarized preparations method and use of flexible and biodegradable PVA films. The literature on sensing shows that PVA is used for gel-electrolyte formation, electrode modification and as a functionalizing agent for developing immunosensors (Buledi et al., 2022; Charoenkitamorn et al., 2020; Nguyen et al., 2021; Wen et al., 2022). Water-soluble PVA substrate was used with carbon nanotube (CNT) to develop field-effect transistors (Yoon et al., 2018). However, there are few reports of its use in biosensing applications restricts for matrix preparation toward biomolecule immobilization (Rana et al., 2022; Sanaeifar et al., 2017; Schyrr et al., 2014).

3.6. Chitosan (QT)

Chitosan (QT) is a partially deacetylated product of chitin, which is the second most abundant natural biopolymer, after cellulose, consisting predominantly of β -(1 → 4)-2-amino-2-deoxy-D-glucose (deacetylated unit) and a smaller percentage (usually less than 20%) of β -(1 → 4)-2-acetamido-D-glucose (acetylated unit) (Shahidi et al., 1999). QT is a non-toxic, biodegradable and biocompatible cationic biopolymer commercially prepared from processing of seafood waste (Tharanathan and Kittur, 2003).

In recent years, the application of chitosan in food, pharmaceuticals, and the development of polymeric films has received considerable attention due to its functional physical and chemical properties (Afshar et al., 2020; Agunos et al., 2020; Ebrahimi Tirtashi et al., 2019; Kanatt et al., 2008; Lozano-Navarro et al., 2018; Ma et al., 2018; Wu et al., 2019). The solubility of chitosan is the most interesting feature considering its use in substrate development. QT is sparingly soluble in water, practically insoluble in ethanol (95%) and other organic solvents, and neutral or alkaline solutions with pH above about 6.5. QT dissolves easily in dilute and concentrated solutions of most acidic organic products and to some extent in inorganic mineral acids (except phosphoric and sulfuric acids). Solubility is also greatly influenced by the

addition of salts to the solution. The increase of the ionic strength leads to the decreasing of solubility (Mulla et al., 2020). The functional properties of QT depend on the chain length (molecular weight), charge density, and charge distribution. Considering QT features, substrates based on QT are highly promising for use in solid supports for flexible bioelectronic devices and sensing.

The methodologies employed for chitosan films preparation generally involve the dispersion in acidic solutions. The methodology described by Liu et al. (2013) (Liu et al., 2013) involved the dispersion of QT (2%, m/m) in an aqueous solution containing 0.5% of acetic acid. The chitosan (2 g) is added slowly to the solution under stirring, and the resulting dispersion is left to stir overnight to ensure complete dispersion of the polymer. The dispersion is then poured onto polystyrene plates and dried overnight at 65 ± 1 °C. Finally, the films are conditioned in a desiccator containing lithium chloride to control the humidity level (11.3% RH at 25 ± 1 °C). This methodology is commonly used for the preparation of chitosan films and allows the production of high-quality films with desirable properties for various applications. Due to its moderate solubility in water, QT can be directly used as a solid support for electrochemical sensing without the need to add other chemical components to reduce its hydrophilicity. Kalpana et al. (2019) prepared QT films by dispersing a 2% amount of the polymer in an aqueous acetic acid solution (1% v/v) and magnetically stirring the dispersion overnight at 70 °C. The dispersion was then filtered to remove impurities and poured onto flat glass plates to dry at room temperature for 72 h. The finished films were stored in a desiccator at 0% relative humidity and 25 °C temperature.

Table S6 summarize preparations method and use of flexible and biodegradable QT films. Differently of cellulose-based films, we found some applications of biodegradable QT films as substrate for flexible electric circuit (Bonardd et al., 2020), flexible piezoelectric Gly/CS-based pressure sensor (Hosseini et al., 2020) and colorimetric sensor for pH sensing (Zheng et al., 2023). Chitosan combined with CaCl₂ (Charoonsuk et al., 2022), NH₄SCN/Potato starch (Abdulwahid et al., 2022), MXene (Ding et al., 2022), potato starch (Zheng et al., 2022), carbon films (Xu et al., 2022), TiO₂ (Rajesh Banu et al., 2020) and Cu-doped carbon dot (Tummala et al., 2022) are an excellent strategy for nanocomposite production with distinct characteristics that can be applied to energy storage devices (Abdulwahid et al., 2022; Ding et al., 2022), triboelectric nanogenerators (Zheng et al., 2022), bioenergy production (Rajesh Banu et al., 2020) and colorimetric sensor (Tummala et al., 2022).

3.7. Starch (AM)

Starch is a type of carbohydrate commonly found in plants, which serves as a source of energy for the plant, composed of long chains of glucose molecules that are linked together. Starch is produced by many different plant species and can be found in a variety of plant-based foods, including grains, vegetables and legumes. Starch is commonly used in food products as a thickener or stabilizer, and is also used in industrial applications, such as the production of paper and textiles. Starch is a complex carbohydrate, which means it takes longer to break down in the body and provides sustained energy.

Starch is a polysaccharide consisting of two components: linear amylose and branched amylopectin. Both amylose and amylopectin are made up of alpha-D-glucose units and have a semicrystalline structure. In the starch granule, amylopectin forms the crystalline portion (Mulla et al., 2020). Starch is commonly used in various industries, including pharmaceuticals and food, due to its biodegradable, biocompatible, and non-toxic properties. Starch is often used as a binder, diluent, disintegrating agent, and in the development of polymeric films (Diyana et al., 2021; Ezati et al., 2019; Glover, 1993; Kulkarni et al., 2012).

The solubility of polysaccharides, such as starch, is a critical characteristic for the development of solid supports like films. Polysaccharides have a high affinity for water molecules, but this also results

in strong hydrogen bonding between molecules, affecting solubility. The polysaccharides have a strong affinity for water molecules due to the presence of OH groups. However, this also leads to a strong interaction between polysaccharide molecules via hydrogen bonding. Therefore, the balance between molecule-molecule interaction and molecule-water interaction is the key to understanding the solubility of polysaccharides (Guo et al., 2017). The molecular weight of the polymer is also an important factor to consider, with high molecular weight polysaccharides being less soluble, for example, amylose and amylopectin in starch are reluctant to dissolve in cold water due to high molecular weight, while maltodextrin (starch after chain cleavage by acid or enzyme) with low molecular weight shows very good solubility in cold water. Other characteristics like molecular weight distribution, charge, and the presence of hydrophobic groups can also affect solubility and should be taken into account (Guo et al., 2017).

Complete dispersion of starch only occurs when heat is employed, and there are already several methodologies widely documented in the literature for the production of starch substrates (BeMiller and Whistler, 1984; Shi et al., 2013; Teixeira et al., 2015). Shi et al. (2013) (Shi et al., 2013) dispersed 7.0 g of previously dried corn starch (dried at 40 °C for 6 h) and 3.0 g of plasticizers (glycerol and xylitol in a 1:1 ratio) in 200 mL of deionized water. The dispersions were stirred at 300 rpm for 1 h under boiling conditions. After gelatinizing the starch at 100 °C for 1 h and cooling it down to 70 °C, the films were poured into Petri dishes and dried for more than 8 h at 45 °C.

One of the major problems encountered in the production of starch films for use as substrates in flexible bioelectronic devices is cracking that can occur after drying. However, this problem can be minimized by controlling the time and temperature during the final drying step. Another effective approach is to combine AM with other polymers, such as the addition of PVA. The study by Frone et al. (2015) (Frone et al., 2015) aimed to improve the water resistance of AM films by incorporating PVA and cellulose nanofibers (CN). To create the films, the researchers mixed 16 g of starch, 4 g of PVA, 6.6 g of glycerol, 2.0 mL of formaldehyde solution (37 wt%), and 0.2 g of ammonium chloride in distilled water (1/10 w/v) at room temperature, with constant stirring for 15 min at 2000 rpm. They then added different amounts of CN suspension to achieve final concentrations ranging from 0 to 10 wt%, based on starch + PVA. The mixtures were heated to 90 °C for 15 min with constant stirring to gelatinize the starch and then for another 45 min at 98 °C to complete gelatinization. Finally, the films were poured onto PET plates and left to dry overnight at room temperature. The incorporation of PVA and CN in the AM films improved their water resistance, which is a desirable for substrates used in flexible bioelectronic devices and sensing.

Films based on cassava starch and anthocyanins from jambolan (*Syzygium cumini*) fruit can be used to classical pH-indicator to monitor the meat freshness (Alexandra Rodriguez Gaviria et al., 2021). Table S7 summarize the materials, preparation method and use of starch films. We highlight the substrate of starch film to fabrication of wearable device resulting in a multi-functional, recyclable, and flexible sensing device with high reproducibility for visualized UV, temperature and sweat pH sensing (Liu et al., 2022). Due to molecular miscibility, a blend of starch and chitosan from abundant and inexpensive potato and crab shells opened great opportunities to develop a natural and biodegradable substrate for flexible and wearable electronics in optoelectronic device applications (Miao et al., 2018). Potato starch and chitosan films were made by the solvent evaporation method, serving as substrate for the projection of electronic devices (Miao et al., 2018). Pure potato starch films produced from solvent evaporation method can also be used as substrate for fabrication of flexible sensors (Liu et al., 2020). These devices can be prepared by transferring a carbon film produced by laser carbonization of polyimide to the surface of the potato starch film. Flexible devices made with potato starch films indicated efficiency in monitoring multiple stimuli such as strain, pressure and temperature proving to be an efficient and green alternative for the design of

electronic devices (Liu et al., 2020). The high-performance of biodegradable, flexible and transparent substrates based on starch are a promising material for next-generation of green optoelectronics, transient electronics and edible electronics devices (Alexandra Rodriguez Gaviria et al., 2021; Liu et al., 2020, 2022; Miao et al., 2018).

3.8. Poly (lactic acid) (PLA)

Poly (lactic acid) (PLA) is a biodegradable and bio-based polyester family made from α -hydroxy acids of renewable resources such as corn starch or sugarcane. The building block for the production of PLA is lactic acid (2-hydroxy-propanoic acid), which can exist in two optically active forms (D- or L-enantiomers). PLA is a type of thermoplastic that can be processed using conventional polymer processing techniques like injection molding, extrusion, or thermoforming. PLA has gained increasing attention as a sustainable alternative to conventional petroleum-based plastics because it can biodegrade under certain conditions, reducing environmental impact. Additionally, PLA has several desirable properties, including good physical, mechanical and barrier properties, high strength and elasticity and stiffness. PLA has various applications and can be produced in a range of colors and finishes, such as in packaging, disposable tableware, biocompatible devices, textiles, medical implants, and 3D printing (Auras et al., 2003; Garlotta, 2001; Waris et al., 2004).

PLA is soluble in several organic solvents including dichloromethane, tetrahydrofuran, ethyl acetate, chloroform, hexafluoro-2-propanol, and acetone. However, it is insoluble in water, which makes it useful for electrochemical sensing applications (Shah et al., 2020). There are various methodologies for developing PLA substrates, including the solution-blow spinning method used by (O Gomes et al., 2023; Paschoalin et al., 2022) to produce mats of PLA fibers. However, the literature widely reports the use of the casting method, as described in the works of González and Alvarez Igarzabal (2013) and Gwon et al. (2016) (González and Alvarez Igarzabal, 2013; Gwon et al., 2016). In summary, the authors proposed dispersing PLA in chloroform (which can be replaced by dichloromethane or a combination of the two solvents) at a specific time and temperature, followed by drying the film on plates in a controlled environment, removing the substrates from the plate, and storing them.

The literature has already reported the use of PLA as substrate for electrochemical sensing, as shown in the work of (O Gomes et al., 2023; Paschoalin et al., 2022) using solution blow spinning to produce mats of PLA nanofibers. Additionally, PLA has been utilized as a support for producing microneedle arrays for dermal biosensing, and was printed using 3D techniques to detect dopamine and glucose (Fontana-Escartín et al., 2022; Skaria et al., 2019). Biodegradable bioplastics of PLA blended with polyhydroxybutyrate (PHB) were produced to be used as support for flexible electronic devices with applications in capacitor, printed conductors and touch sensor (Bozó et al., 2021). Biodegradable PLA substrates we prepared using spin-coating technique to printing of resistive and interdigitated (IDE) capacitive devices for temperature and humidity sensing applications (Quintero et al., 2014). The opportunities for the practical application of PLA films as substrates in flexible bioelectronics devices are opened to be explored.

4. Limitations and future perspectives

Sustainable and flexible electronic devices are arising to meet the growing demand for green technology solutions in biosensing applications. Renewable natural resources-derived biopolymers are standing out owing to intrinsic physico-chemical properties including mechanical, thermal, and chemical stability combined with flexibility, durability, biocompatibility, biodegradability and bioaccessibility to assemble green devices (Kasprzak et al., 2022; Liu et al., 2020). Although extensive efforts are still being devoted to the preparation and development of flexible ecofriendly biopolymers as support/substrate

for sustainable, modern and green devices, they still are in an initial technological degree. However, the development of sustainable support/substrate based on renewable natural bioresources-derived polymers have limitations, challenges and opportunities to address real practical scenario including sources of natural bioresources, scalability of preparation, processability, full degradability and competitive costs with petroleum-based support/substrates. The origin of raw materials of the bio-based polymer are obtained from biological environment such as plants, algae, food and crustaceans in which will influence the properties, features and potential applications. A standardization of the biological materials used for the production of biopolymers will be imperative, specifically on the steps involved for extraction and purification of these bio-based feedstocks. The cost effectiveness, large scale production, and well-controlled production steps of bio-based support/substrates as well as whole flexible electronic devices and biosensors should be optimized, explored and developed for the practical application on an industrial scale identifying processability parameters owing huge variety of materials offered by renewable natural bioresources. Although significant efforts are being made to develop whole biodegradable biosensing devices, the use of several non-biodegradable elements remains necessary for high performance biosensing applications. The concept of flexible electronic devices manufactured on bioplastics is in the preliminary development stage. Despite its challenges, the polymeric substrates from renewable natural bioresources field has tremendous opportunity with promising results and impact that can be extensively explored by industry and scientific community. Although the use of materials from renewable natural bioresources in flexible electronic devices is still in their infancy, we believed that these polymers derived from biomaterials have promising applicability for industrialization in the future.

Considering the vast developments in the field of biopolymeric and biodegradable substrates, it is clear that the processing of polymeric films of petrochemical origin should be minimized due to the pollution that they cause to the environment and other associated issues. The use of alternative, suggested polymers can lead to improved mechanical characteristics for the development of flexible electrochemical substrates. Although some polymers have high hydrophilicity, the addition of other polymers such as starch and cellulose nanocrystals can improve not only this drawback, but also other material properties. Moreover, the composition of different polymers can also impact properties such as surface morphology and thermal characteristics. Despite the fact that significant economic, technological, and political efforts are still needed to achieve really sustainable and flexible bioelectronic devices, recent scientific advancements provide us with confidence that biodegradable-based flexible bioelectronics will soon be industrialized and integrated into our daily lives.

5. Conclusions

We hope that the fabrication techniques of eco-friendly substrates, fundamental concepts, adaptation of polymeric materials and applications shown here has highlighted the importance of biopolymeric and biodegradable films for sustainable and flexible devices dedicated to several frontier areas in science and technology. In this review, we have outlined recent research that focused on the use of biodegradable-based biosustainable films in flexible and bioelectronic devices. The incorporation of chemical active additives or different polymers (blends) in the production of biopolymer-based films represents a promising approach to develop green, flexible bioelectronic devices. The biopolymers introduced here for production of substrates can provide flexibility and sustainability making a valuable and promising alternative to conventional materials from nonrenewable polymers.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships 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

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