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Biodegradable Polymer Coatings: Current State-of-the-Art, Recent Advances, and Still Open Challenges

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ABSTRACT

Polymer coatings represent both a key academic research topic and a viable and well-established industry market. Their characteristics are unique and allow for their application on different types of substrates (such as metal alloys, ceramic materials, polymers, paper, and paperboard), even for uses characterized by high durability. The latter property is perhaps the one behind their extensive diffusion since it guarantees performance over a wide range of time, often comparable with the lifetime of the coated substrate. However, the durability of a coating is not always a desired and sought-after feature: in fact, there are applications for which the possibility to remove the coating on demand from the coated substrate becomes an extremely important goal, even in the logic of the circular economy. In this context, academic research is trying to develop and implement biodegradable polymer coatings, i.e., thin layers of material that may start interacting with the environment in specific conditions, breaking down into simple substances that do not exhibit toxicity or hazard. This work aims to review the current state-of-the-art related to biodegradable polymer coatings, providing the reader with an overview of the progress made so far in this research field and some perspectives for the coming years.

1 | Introduction

Polymer coatings are thin polymer layers applied to flat substrates or objects showing irregular geometries/shapes [1]. Polymeric coatings can be functional (adhesives, photographic films), protective (anti-corrosion, barrier systems, etc.), or decorative (paint and varnishes) [2–6]. They are also used to modify the surface properties of different types of substrates (paper coatings, hydrophobic/oleophobic coatings) [7].

Although polymeric coatings are mostly organic, they can also include ceramic or metal particles to increase durability, functionality, or aesthetics. Besides, they can be synthesized through sol-gel methods, usually starting from alkoxy precursors. In this case, they are classified as hybrid organic/inorganic (O/I) coatings and employed mainly as protective systems for different substrates [8, 9].

It is worth noticing that polymer coatings have been and are still mainly employed for durable uses: this means that these coatings should be able to tolerate the operational environmental conditions without showing any degradation phenomena, at least before the end-of-life of the substrate material to which they

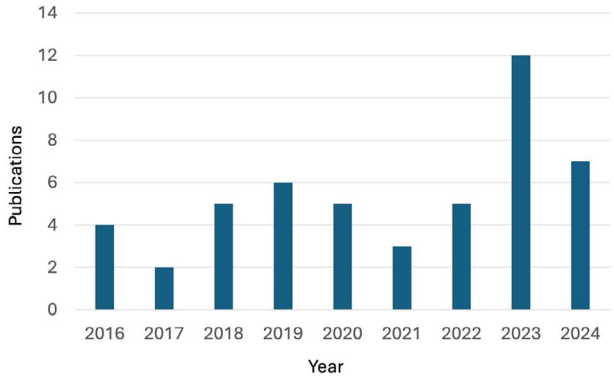

FIGURE 1 | Number of publications (from 2016 to 2024) in peer-reviewed journals, dealing with biodegradable polymer coatings (data collected from the Scopus database, first using the Boolean combination of "biodegradat*" OR "biodegradab*," "poly*," and "coating*," then, carefully reading the titles and abstracts of each article and eliminating those that contained the three words but not referring to biodegradable polymer coatings; accessed on March 07, 2025).

are applied. This way, the polymer coatings can guarantee their function(s) (e.g., aesthetic/decorative, protective, etc.) with high effectiveness.

Despite the reasonable need to design durable polymer coatings, nowadays there is an emerging need to design the so-called biodegradable coatings. These coatings can ensure their functionality but can also be removed from substrates and easily degraded on demand. Their degradation, of course, must not give rise to toxic, hazardous, or polluting byproducts, either to humans or to the environment.

The academic investigation of biodegradable polymer coatings is quite recent and somehow still in its infancy, as witnessed by the publications appearing in the scientific literature so far (Figure 1).

In particular, the research on biodegradable coatings has mainly focused on polyurethane-based and epoxy-based systems, despite a few examples referring to other types of coatings (such as acrylic coatings). The next paragraphs will describe these coatings, providing the reader with an overview of the progress achieved so far and highlighting the most interesting research outcomes. Before investigating the structure-biodegradability relationships in biodegradable coatings, the methods employed for assessing their biodegradability will be thoroughly elucidated.

2 | Assessing the Biodegradability of Polymer Coatings

Since 1992, with the celebration of an international workshop on biodegradable materials, experts worldwide have reached a broad agreement on definitions, standards, and testing methodologies [10]. Despite this established consensus, the following basic points are worth remembering. The term biodegradability refers to the ability of a material, herewith the coatings that are the objective of this critical review, to be decomposed by microorganisms under aerobic natural conditions into water, CO_2, and biomass, and also CH_4, for anaerobic processes, always through the involvement of enzymes. In case, also the effect of metabolic acids released by, e.g., bacteria and fungi, has to be considered [11]. On the other hand, a material truly biodegrades only when specific environmental conditions are present, either through naturally occurring processes or artificially forced during specific assays.

To assess biodegradability, it is necessary to induce and follow the biodegradation pathways, which may be done either using standardized methods or more complex and specific methodologies. This also depends on the final aim, e.g., the simple commercial need to define a coating as partially or completely biodegradable through standardization and certification systems or a deeper academic study of the microbiological processes and chemical intermediates involved in the degradation routes found in natural environments. Biodegradation may be studied by monitoring the coating's property changes (from structural to mechanical, through mass changes), the microbial growth, or the released degradation products. In addition, biodegradability is evaluated by estimating indices related to such changes either from standard tests or using other specific quantifiers. More detailed information on the methodologies and standard tests (American Society for Testing and Materials, ASTM, and International Organization for Standardization, ISO, being the most used worldwide) for evaluating the biodegradability of polymers may be found elsewhere [12, 13].

Among the simulated environments in which polymeric material biodegradation may occur, those that better reflect the natural degradation of protective and decorative coatings during their service life essentially refer to the so-called high solids conditions, as in solid waste treatment or littering, with some interest also for aquatic environments found in wastewater treatment plants or surface waters and marine environments [11].

Most valuable assays for coating, which simulate such conditions at different levels, i.e., enzymatic assays, plate tests, respirometry and gas analysis, and soil burial, are shortly introduced and discussed below, also through some examples. This highlights advantages and limits while considering the ease of application and indexing.

2.1 | Enzyme Assays

Enzymes are biological catalysts with an absolute specificity for a given substrate or a family of linkages. They can be selected to induce the cleavage of specific chemical groups in polymer chains. Of special interest for biobased coatings are the tests with esterases, particularly lipases (a group of carboxylic acid hydrolases), which catalyze the hydrolysis of triglycerides and other polyesters. As representative examples of the procedure, coating specimens of epoxy resins with epoxidized oleic acids [14], waterborne polyurethane (PU) with castor oil and polycaprolactone diol as the soft segments [15, 16] or modified with b-cyclodextrin and poly(ethylene glycol) [17] have been immersed in a buffered lipase solution and soaked at the enzyme optimal temperature for several weeks. The biodegradation rate, controllable by adjusting the enzyme dose, could be quantified by following the mass loss rate. In addition, optical microscopy

or scanning electron microscopy (SEM) has also been used to visualize the degradation processes [15].

Enzyme tests are an excellent screening tool to evaluate the biodegradability of a coating. However, they do not allow a direct comparison between polymers of different families, as the type of enzyme or the mixture of selected enzymes is paired to only one polymer functional group family. In the end, enzymic methods require precise enzyme selection and a relatively simple experimental design, easily generating a biodegradation index. On the contrary, enzymes can be inhibited or partially inactivated due to inappropriate storage or purification.

2.2 | Plate Tests

Simple and traditional methods to verify qualitatively if a material can support the growth of some microorganisms are the so-called plate tests, which may also be performed following standard protocols (as defined by ISO 20645) [18]. After a nutrient agar gel that does not contain any carbon source is poured into Petri dishes and inoculated with known bacteria or fungi, coating fragments are placed onto the surface and then incubated at a constant temperature. Even after a short time, in the order of a few hours, it is possible to appreciate microorganism growth. In any case, a positive response does not automatically indicate the biodegradability of the polymer component of the coating, as microorganisms may be using oligomers, fillers, or plasticizers as carbon sources. Conversely, a negative result may also be ascribed to the inhibition effect of secondary components or employed to verify the coating's antimicrobial activity [19].

2.3 | Respirometry and Gas Release

The extension and rate of biodegradation can also be quantified following the amount of oxygen utilized, or the CO_2 emitted, during aerobic microbial activity or the evolution of CH_4, CO_2, or, more in general, a biogas mixture, under anaerobic conditions. Biodegradation oxygen demand (BOD) is determined in a closed system, i.e., a respirometer consisting of a hermetic bottle equipped with a manometer, where a known amount of specimen is dispersed in a saline solution. To mimic real environmental conditions, the microbial population is introduced as an inoculum of, e.g., activated sludge from wastewater treatment plants [20] or directly using seawater [21]. It naturally contains bacterial consortia and the saline components needed for their development. As the emitted CO_2 is trapped inside the device, a decrease in pressure or an electrochemical measurement allows the quantification of the BOD, expressed as the oxygen consumed per weight unit of material over time [20, 21].

Similarly, it is possible to quantify the gaseous components released during anaerobic biodegradation of specimens incubated in an opportunely inoculated saline solution. Alternatively, a direct monitoring of CO_2 production over time by gas chromatographic methods [22] or by titration of a trapping solution [23] in both aerobic and anaerobic experiments revealed the degradation capacity of the bacterial consortia.

2.4 | Other Lab-Scale and Natural Biodegradation Tests

Other methodologies, easy to develop, consist of exposing the specimen in the form of films in natural or synthetic aquatic media or soil samples. In recent studies, sterilized bio-derived PU films have been immersed in an activated water solution containing bacteria from a natural source, e.g., from active sewage sludge [24], or appositely added as a co-culture of *Bacillus subtilis* and *Pseudomonas aeruginosa* [25], well known for their bioremediation potential to induce decomposition. For transparent films, the bacteria growth could be checked by optical density (using the absorbance value at 600 nm). More in general, SEM images reveal surface erosion [24, 25].

Also, biodegradation in soil can be studied using standard tests, such as the ISO 17556 [26] or the corresponding ASTM D5988 [27], focusing on assignment rules of certification. At the research level, assays are carried out either at lab-scale (in a closed jar, as in standard tests) or in a natural field to obtain some indexes from mass loss or disintegration, assessing the biodegradability under specific conditions, but also for following other parameters that offer information on the biodegradation mechanisms. For instance, burial of cardanol-based acrylates [28] and waterborne PU [15–17] for up to 1–3 months in natural soils under environmental conditions induced an extensive fragmentation, indexed by mass loss. SEM characterization of surfaces at increasing time intervals allows for monitoring cracking, detachments, erosion phenomena, and an increase in surface roughness. FTIR spectroscopy measurements show polymer structural changes, elucidating degradation pathways [16]. In addition, XRD of semicrystalline coatings could indicate the disruption of crystalline structure during degradation [15].

Concerning the assessment of biodegradation by fungi, treatments are performed by spreading the specimen with a spore suspension of the selected fungi strains. After incubation in a chamber at optimal temperature and humidity, the fungi's morphology, their ultrastructure, and the coatings' surface can be shown by SEM, FTIR, and X-ray photoelectron spectroscopy [29, 30]. In this regard, SEM can provide valuable information on fungi morphology and ultrastructure. Meanwhile, FTIR and X-ray photoelectron spectroscopy can shed light on the chemical changes that occur on the surface of coatings as a result of biodegradation. As a fact, fungi can induce porous corrosion and cracks associated with the growth of mycelium and hydrolysis, e.g., of ester and urethane bonds induced by the secreted carbonyl acids [30].

As a final remark, the methods presented in this section permit to compare the behavior of a series of coatings through routine measurements. Moreover, the implementation of mechanistic studies using spectroscopic techniques is always possible. On the other hand, the main drawback is the need for accurate cleaning of specimens incubated in aquatic media or soil. Protocols to remove residues and bacteria strongly adhered to the coating surfaces must be fine-tuned for carrying out kinetic studies based on mass loss measurements and before drawing conclusions from FTIR spectroscopy changes. The interference of residual biofilm can lead to the misattribution of degradation products [22].

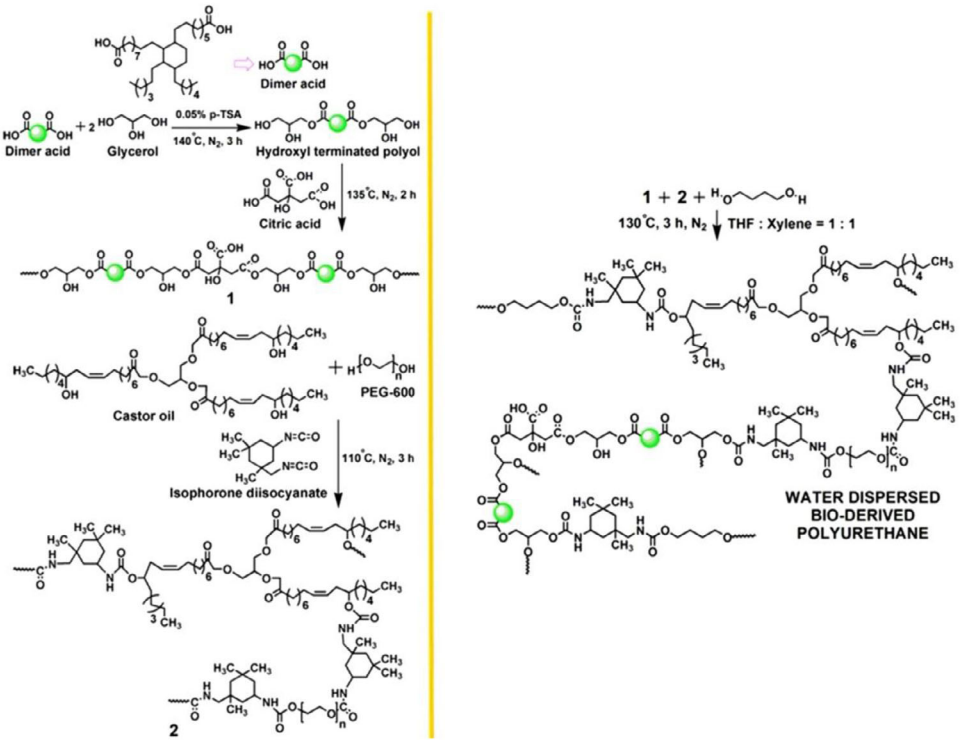

FIGURE 2 | Scheme of the synthetic path of water-dispersed bio-derived PU coatings: preparation of bio-derived polyol (1); fabrication of chain extender (2). Adapted with permission from ref. [25] Copyright 2020, Elsevier.

3 | Recent Advances in Biodegradable Coatings

3.1 | Biodegradable Polyurethane-Based Coatings

Among polymer coatings, PU systems show interesting features (such as toughness, mechanical strength, abrasion resistance, chemical and weathering resistance, and tunable flexibility – even at low temperatures), which justify their use in a wide variety of application sectors, including automotive and chemical industry, among others [31, 32]. The possibility of formulating biodegradable PU coatings through the use of bio-sourced components (e.g., vegetable oils, cashew nutshell liquid, terpenes, Eucalyptus tar, and other bio-renewable sources [33]) for synthesizing polyols or isocyanates has paved the way toward sustainability in materials science and technology. Further, it is worth noticing that the biodegradation of PU coatings is controlled by several parameters, such as the ratio of hydrophilic to hydrophobic moieties in the PU structure, the degree of crosslinking, and the bio-based fragment content [34–36]. The progress achieved so far will be described in the following paragraphs.

Chaudhuri and Karak [25] synthesized water-dispersed bio-derived transparent PU coatings starting from various bio-derived precursors comprising poly(ethylene glycol)-600, dimer acid, castor oil, and glycerol (Figure 2). The biodegradation of the coatings was assessed utilizing *B. subtilis* (gram-positive) and *P. aeruginosa* (gram-negative) bacterial strains: in particular, the bacterial growth on the PU coating concerning the initial condition was tested by measuring the absorbance at 600 nm as the optical density of the two bacterial strains and the weight loss% of the degraded coating was evaluated. As shown in Figure 3a, the optical density values of the two bacteria strains increased with increasing test time. Further, because of the differences in wall structures of the *B. subtilis* and *P. aeruginosa* bacteria cells, the latter bacteria showed higher binding capabilities and degradation efficiency of the PU coating compared with *B. subtilis* bacteria. Also, the higher biosurfactant activity and surface hydrophobicity of bacteria cells of *P. aeruginosa* than *B. subtilis* accounted for a greater degradation capability of the former bacteria. Finally, the biodegradation was confirmed by SEM analysis carried out on the PU coating before (Figure 3c) and after (Figure 3d) the tests.

Fan and co-workers [17] investigated the biodegradability of waterborne PU coatings containing β-cyclodextrins modified with poly(ethylene glycol) (with different average molecular weights, namely, 1000, 2000, and 4000) and synthesized through the reaction of poly(caprolactone) diol and isophorone diisocyanate. In particular, the biodegradation tests were carried out in soil (buried samples) and in phosphate-buffered saline solution (pH 7) containing lipase (200 μ/mL, 0.6 wt.%). The temperature was kept at 50°C, and the tests were conducted for up to 60 days. The biodegradation rate was calculated based on the sample weight loss: in particular, it was found that the incorporation of β-cyclodextrins into the PU accounted for an increase in biodegradation due to the hydrophilic character of the β-cyclodextrins. Furthermore, the modification of these latter with poly(ethylene glycol)s having increased molecular weights further enhanced the biodegradation rates that achieved 31.2% in lipase phosphate-buffered saline solution and 14.8% in soil (respectively with 39.8% and 24.3% increase compared to the unfilled PU coatings). These findings were ascribed to the higher hydrophilicity provided by poly(ethylene glycol)s to the β-cyclodextrins, which made the PU more prone to biodegrade.

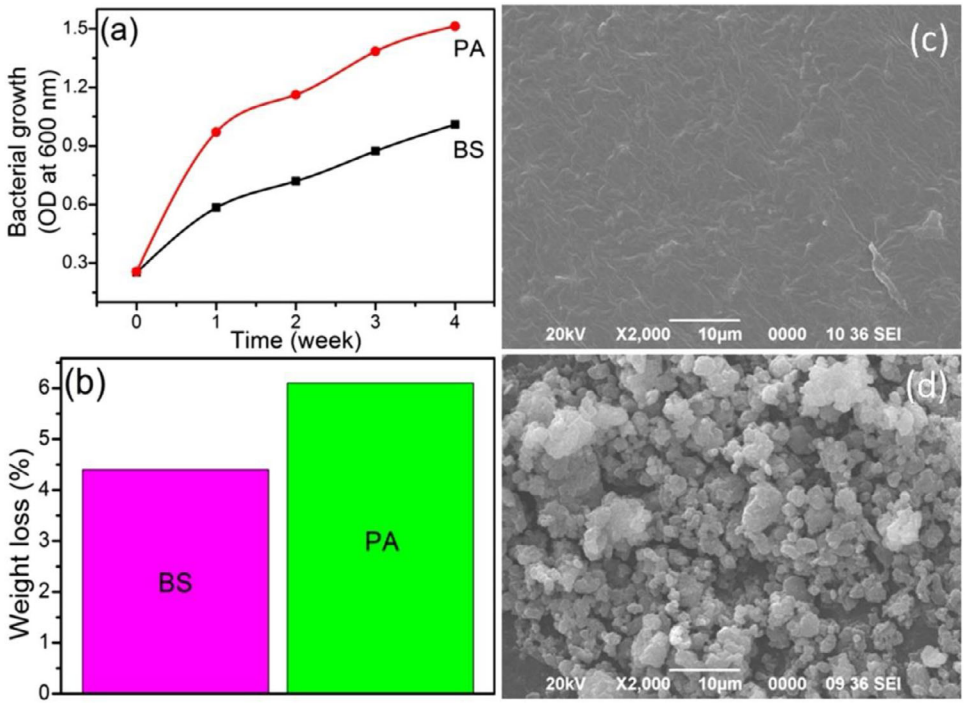

FIGURE 3 | (a) Growth curves of *B. subtilis* (BS) and *P. aeruginosa* (PA) on water-dispersed bio-derived PU coatings containing 5 wt.% of 1,4-butanediol and (b) weight loss diagram of water-dispersed bio-derived PU coatings due to bacterial degradation. Typical SEM images of control (c) and degraded surface (d) of water-dispersed bio-derived PU coatings. Reprinted with permission from ref. [25] Copyright 2020, Elsevier.

Hao et al. [30] incubated two different fungi, namely, *Talaromyces funiculosus* and *Phanerochaete chrysosporium*, on a PU coating for 2 weeks at 30°C and 95% relative humidity. Although both fungi promoted the biodegradation of the PU coatings, the contribution of *T. funiculosus* was prevalent because of two main reasons: first, its mycelium weight was almost three times more than that of *P. chrysosporium*, hence this mycelium found more substrate deep in the coating to obtain more nutrients in the medium during colonization. Then, *T. funiculosus* produced such carboxylic acids as tartaric acid, succinic acid, citric acid, and propanoic acid, which enhanced the hydrolysis of the urethane and ester bonds of the PU coating: this way, its mycelium could rapidly proliferate (Figure 4).

Cao and co-workers [37] synthesized a biodegradable dihydroxyl-terminated poly[(ethylene oxide)-*co*-(ethylene carbonate)] and subsequently polycondensated it with isophorone diisocyanate. The presence of ethylene carbonate units as soft segments in the PU macromolecular chains (which also showed protein resistance and antibacterial adhesion features) accounted for a high biodegradability, as assessed by treating the PU coatings in lipase phosphate-buffered saline solution (lipase concentration: 0.8 g/l; pH 7.4). In particular, during the first 2 weeks of biodegradation, the remaining mass of the film almost linearly decreased, achieving around 30% and highlighting a homogeneous degradation of the PU coating.

Li and co-workers [38] exploited ultrasound-assisted inject printing for depositing self-healing and biodegradable PU coatings containing carbon nanotubes onto knitted cotton fabrics for the design of flexible and high-performing strain sensors. As shown in Figure 5, poly(lactic acid), synthesized on purpose, together with poly(tetramethylene ether glycol), was reacted with isophorone diisocyanate to obtain a PU prepolymer that further reacted with 1,4-butanediol, hence obtaining the biodegradable PU coating.

The PU coatings obtained were cut into 10 × 10 mm^2 specimens and dipped into lipase phosphate-buffered saline solution (pH 7.4, T = 37°C) for several weeks (up to 12) to assess their biodegradability.

Figure 6a–d shows the morphology of the coatings during the biodegradation tests: in particular, it is worth noticing that, after 4 weeks of the degradation test, the water absorption and the subsequent swelling of the coating accounted for the appearance of a certain roughness. This latter increased after 8 weeks; after 12 weeks, several gullies and cracks appeared. These phenomena were accompanied by a progressive decrease in the average molecular weight of the PU (Figure 6e, assessed through Gel Permeation Chromatography analyses), as well as in the remaining mass (which approached around 25%—Figure 6f—at the end of the 12th week).

The degradation mechanism of the PU coatings (Figure 6g) was mainly ascribed to the conversion of the ester (R-COO-R') and carbamate (R-COO-NH-R') bonds into R-COOH and R-NH-COOH species in the presence of the phosphate-buffered saline solution. Besides, the formation of carboxyl groups further accelerated the degradation of the coatings.

De Smet et al. [39] designed and developed a PU coating for polyester textiles, reacting a polycaprolactone polyol with isophorone diisocyanate; the resulting PU was subsequently

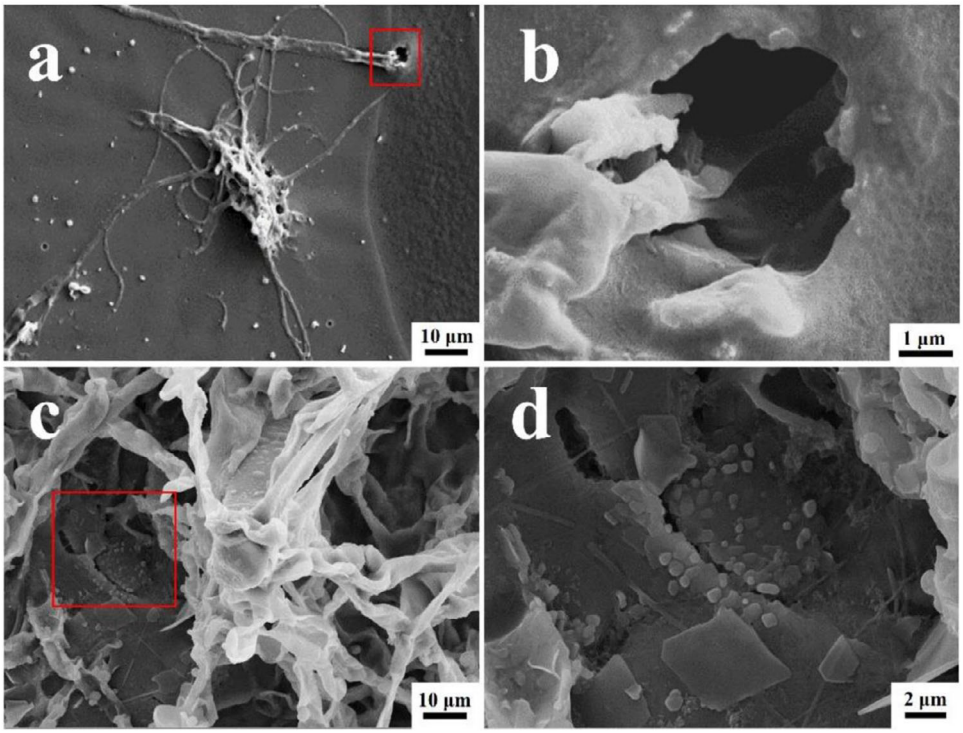

FIGURE 4 | SEM images of the surface morphology of PU coatings after colonization by *T. funiculosus* (a,b) and *P. chrysosporium* (c,d) for 7 days (at 35°C and 95% relative humidity). Reprinted from ref. [30] under CC-BY License.

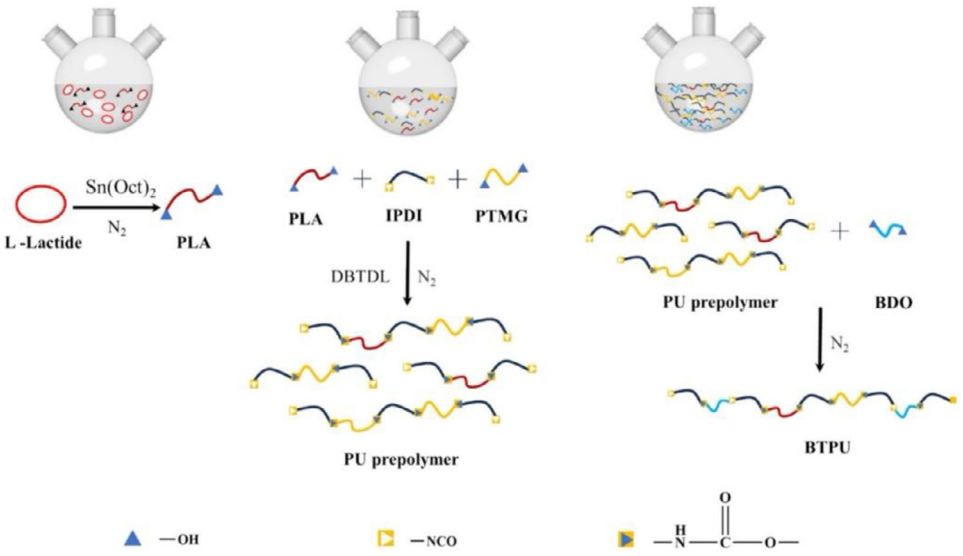

FIGURE 5 | Synthesis of the biodegradable PU. Legend: PLA = poly (lactic acid); IPDI = isophorone diisocyanate; PTMG = poly(tetramethylene ether glycol); DBDTDL = dibutyl tin di lauric acid; BDO = 1,4-butanediol. Reprinted with permission from ref. [38], Copyright 2024, Elsevier.

crosslinked with a polyisocyanate-based crosslinker. The biodegradability of the PU coatings was assessed through disintegration tests (up to 8 weeks), according to a soil burial test, and biodegradation tests in soil (up to 180 days), following ISO 17556 [26]. For this latter, natural sieved soil was employed as the inoculum (water content: 40%–60%; pH: 6–8; C:N ratio ≥40:1).

Figure 7 compares the disintegration tests performed on the PU coatings before and after crosslinking with a 1% polyisocyanate-based crosslinker. Unlike the non-crosslinked coatings, which disintegrated quickly and already exhibited significant disintegration after 2 weeks, the crosslinked counterparts did not show disintegration in the first 4 weeks; however, after 8 weeks, a remarkable disintegration was observed.

Figure 8 shows the results of biodegradation tests in soil for the non-crosslinked PU, using cellulose as a reference. The biodegradation rate of cellulose during the first month of the test

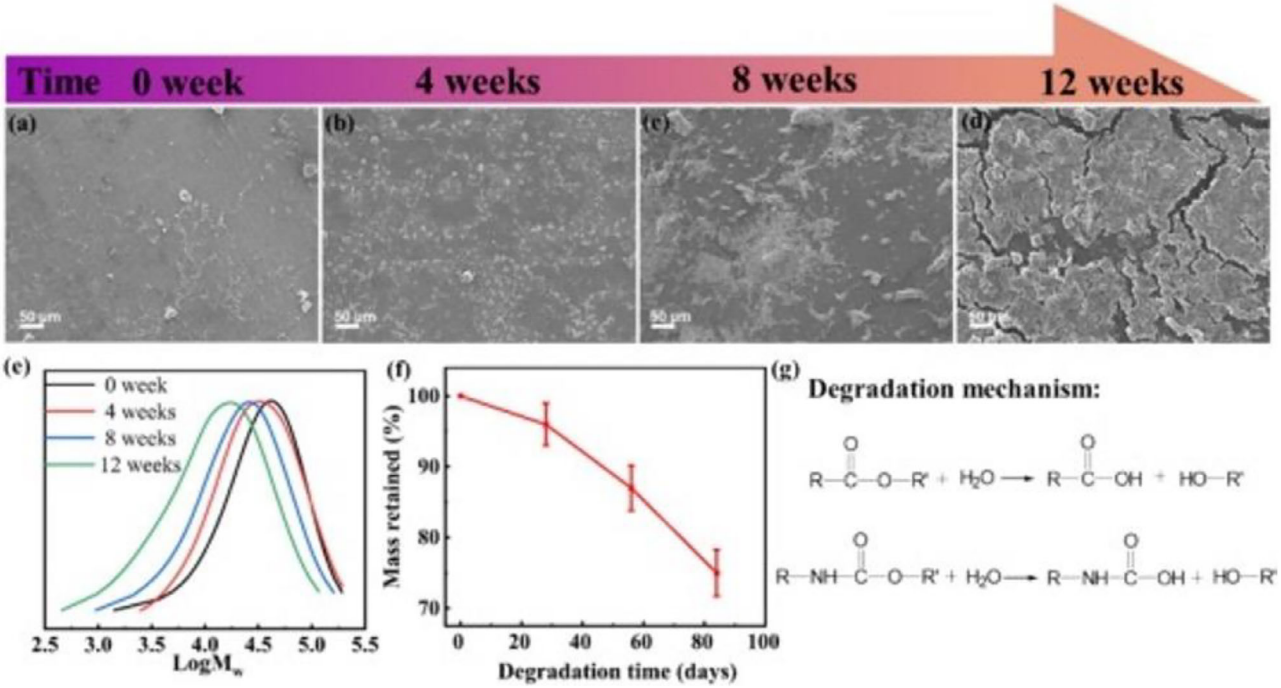

FIGURE 6 | (a–d) typical SEM images of the PU coatings during the biodegradation tests, (e) Gel Permeation Chromatography curves of the coatings at different biodegradation times; (f) weight loss versus degradation time, and (g) degradation mechanism of the PU coatings. Adapted with permission from ref. [38], Copyright 2024, Elsevier.

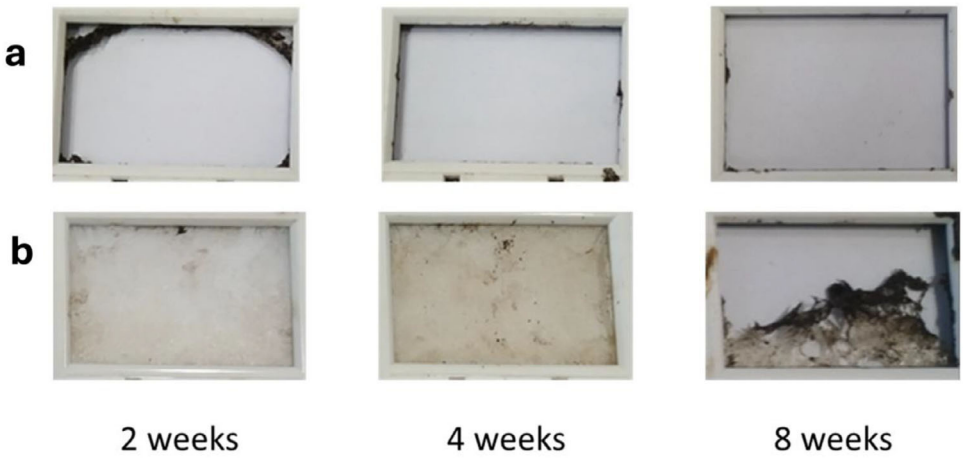

FIGURE 7 | Results from disintegration tests carried out on non-crosslinked (a) and crosslinked (with 1% crosslinker) PU coatings as a function of time. Adapted from ref. [39], under CC-BY License.

was higher than that of the PU. After 180 days, the reference cellulose was almost 75% degraded. The PU coating started to degrade after 10 days and was already degraded by about 50% after 60 days; then, degradation gradually increased, achieving about 60% after 180 days.

Esfahani and Rafiemanzelat [24] investigated the role of natural reactive deep eutectic solvents on the biodegradability of crosslinked PU coatings cured with isophorone diisocyanate in the presence of glycerol. The coatings were dipped into a sewage sludge vessel to assess their biodegradability, measuring the weight changes over 28 days. The presence of increasing amounts of the reactive deep eutectic solvents accounted for an increased biodegradability of the PU coatings, as the former augmented the free volume within the polymer network, hence favoring the water uptake, the weight loss (up to around 40% after 4 weeks), and the overall biodegradation of the coatings.

Zhang and co-workers [40] synthesized covalently crosslinked polyester polyols (namely, poly(glycerol adipate), poly(glycerol sebacate), and poly(glycerol dodecanoate)) through the polycondensation of glycerol with the corresponding bioderived aliphatic dicarboxylic acids. Then, these polyester polyols were reacted with dicyclohexylmethane diisocyanate to obtain PU coatings.

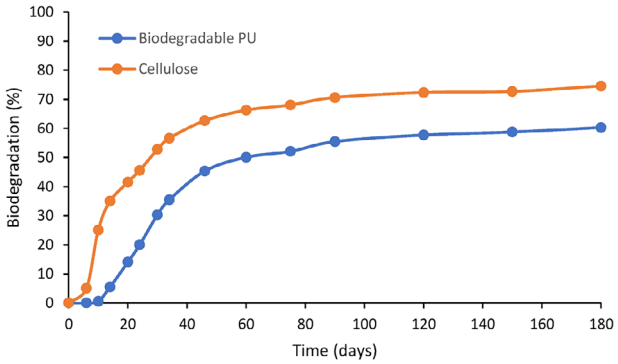

FIGURE 8 | Biodegradation in the soil of the non-crosslinked PU coating, compared with cellulose, used as a reference. Reprinted from ref. [39], under CC-BY License.

The biodegradability of the latter was assessed employing lipase phosphate-buffered saline solution (concentration = 20 U/ml, which corresponds to 0.6 wt.%; pH 7.4; T = 37°C). In particular, it was observed that the biodegradation was strictly correlated with the water absorption capability of the PU coatings. More specifically, the systems containing poly(glycerol dodecanoate) highlighted the highest biodegradation rate, achieving around 18% weight loss after 42 days of biodegradation test.

Interestingly, Liu and co-workers [29] investigated the effect of the space microgravity environment on the biodegradation of a commercially available PU coating (not specified) by *Aspergillus brasiliensis* ATCC 16404TM, a spore-producing fungus. The biodegradation results obtained in microgravity conditions were compared with those gathered on Earth using the same fungus (Figure 9). In particular, it was found that the fungus could produce mature biofilms in space, remarkably affecting the degradation of the PU coatings. Unlike earth samples, which showed a porous degraded layer, for the space counterparts, degradation was associated with the aggressive growth of mycelium, and no porous degraded layers were observed. Besides, an increased oxalic acid accumulation was observed in the microgravity environment.

Yu et al. [15] synthesized poly(L-malic acid ethylene glycol diester) and reacted with isophorone diisocyanate in the presence of poly(caprolactone), hence obtaining crosslinked PU coatings. Their biodegradability was thoroughly assessed through tests in soil and in a lipase phosphate-buffered saline solution (lipase concentration = 20 U/mL, corresponding to 0.6 wt.%), taking out the degraded specimens after 1, 2, and 4 weeks. From an overall point of view, the soft segments in the PU network (i.e., those based on poly(L-malic acid ethylene glycol diester) underwent cleavage: 40 mol.% of the diester accounted for about 44% and 54% weight loss in soil and lipase phosphate-buffered saline solution, respectively.

Very recently, Wu et al. [41] succeeded in preparing waterborne PUs (derived from poly(tetramethylene ether glycol) and isophorone diisocyanate) containing lignin at different loadings (namely, 5, 10, and 20 wt.%) through an addition reaction and the formation of H-bonds between the hydroxyl groups of lignin and the isocyanate and hydroxyl groups of PU. Then, this composite system was applied as a coating on the surface of a paper-based substrate to prepare grass paper mulch films. The biodegradability of these latter was assessed by burying them in natural soil (10 cm depth; temperature range: 10°C–35°C; relative humidity: 20%–70%) and evaluating their morphology changes after 6 months (Figure 10). As shown in Figure 11, 20 wt.% lignin accounted for about 61% degradation rate (much higher than that measured for straw paper films not containing lignin—around 24%).

Dong et al. [42] exploited a one-pot method for preparing PU coatings, obtained from castor oil and ethyl ester L-lysine triisocyanate, to be used for controlled release of fertilizers. The biodegradation of the coatings was evaluated by burying them in the soil in nylon mesh bags and taking out the degraded specimens after 1, 3, 5, 9, and 12 months. A significant degradation rate of about 27% was observed after 12 months (Figure 12(a)); furthermore, after 3 months of degradation, the surface morphology of the coatings started to change, highlighting the formation of pores and fragments because of the erosion induced by the microorganisms (Figure 12(b)).

Yang et al. [43] incorporated sorbitan monooleate and quercetin (added as co-reactants) into the formulation of waterborne PU coatings derived from poly(caprolactone) diol and isophorone diisocyanate. Their biodegradability was assessed through tests both in moist soil enriched with microorganisms and maintained at 25°C and in a lipase phosphate-buffered saline solution (lipase concentration = 20 U/mL, which corresponds to 0.6 wt.%), taking out the degraded specimens after 1, 2, and 4 weeks. At the end of the degradation period in soil and in lipase phosphate-buffered saline solution, the weight of the PU coatings decreased to 53% and 48%, respectively, hence demonstrating a high biodegradability.

Table 1 summarizes the main research outcomes for biodegradable polyurethane-based coatings.

3.2 | Biodegradable Epoxy-Based Coatings

Wang and co-workers [44] studied the impact of *P. putida* on the corrosion resistance of marine Bisphenol A-derived epoxy coatings. To this aim, some steel substrates, coated with the epoxy resin, were incubated with the bacterium in both seawater and sterile seawater from 48 h to 25 days. The presence of the *P. putida* accounted for the formation of an extended biofilm on the epoxy coating and a significant decrease in the corrosion resistance (as revealed by electrochemical impedance spectroscopy measurements), hence indicating the occurrence of biodegradation. Furthermore, as shown in Figure 13, some pulverization and tiny holes developed on the epoxy coating surface after the biofilm formation.

Deng and co-workers [45] coated carbon steel coupons with an epoxy resin (chemical structure not specified) and then dipped the coated specimens into sterile seawater containing *B. flexus* for up to 30 days. As assessed by electrochemical impedance spectroscopy measurements, the corrosion resistance significantly worsened during the first 19 days of incubation due to the degradation of the epoxy coating. The increase of surface roughness, as well as the appearance of holes and pores

TABLE 1 | Main research outcomes for biodegradable PU-based coatings.

PU coating formulation	Biodegradation assessment	Main outcomes	Refs.
Poly(ethylene glycol) 600, dimer acid, castor oil, glycerol	With *B. subtilis* and *P. aeruginosa* strains	Higher biosurfactant activity and degradation ability of *P. aeruginosa* than *B. subtilis*.	[25]
Poly(ethylene glycol) 1000, 2000 and 4000-modidied β-cyclodextrins, poly(caprolactone) diol, isophorone diisocyanate	In soil and in lipase phosphate-buffered saline solution (pH 7, T = 50°C)	Higher biodegradation in the presence of β-cyclodextrins. 31.2% biodegradation rate in lipase phosphate-buffered saline solution. 14.8% biodegradation rate in soil.	[17]
PU coating (not specified)	With *T. funiculosus* and *P. chrysosporium* (T = 30°C; 95% R.H.)	Higher biodegradation induced by *T. funiculosus* than *P. chrysosporium*. Production of carboxylic acids enhancing hydrolysis reaction in the PU coating.	[30]
Poly[(ethylene oxide)-co-(ethylene carbonate)], isophorone diisocyanate	In lipase phosphate-buffered saline solution (pH 7.4)	About 70% mass loss of the PU coating after 2 weeks.	[37]
PU coatings containing carbon nanotubes onto knitted cotton fabrics	In lipase phosphate-buffered saline solution (pH 7.4, T = 37°C)	About 75% mass loss of the PU coating after 12 weeks.	[38]
Polycaprolactone polyol, isophorone diisocyanate	Soil burial test (pH 6–8; H_2O content = 40%–60%)	Remarkable disintegration after 2 weeks. About 60% degradation after 180 days	[39]
Glycerol, isophorone diisocyanate, deep eutectic solvents	In a sewage sludge vessel	Increase in the biodegradation with increasing the amount of deep eutectic solvents; about 40% mass loss after 4 weeks	[24]
Glycerol, bioderived aliphatic carboxylic acids, dicyclohexylmethane diisocyanate	In lipase phosphate-buffered saline solution (pH 7.4, T = 37°C)	Highest biodegradation rate observed for the coatings containing poly(glycerol dodecanoate) with 18% mass loss after 42 days.	[40]
PU coating (not specified)	Space microgravity environment in the presence of *A. brasiliensis*	No porous degraded layers produced by *A. brasiliensis* in the microgravity environment. Increased oxalic acid accumulation in the microgravity environment	[29]
Poly(L-malic acid ethylene glycol diester), poly(caprolactone), isophorone diisocyanate	In soil and in lipase phosphate-buffered saline solution	About 44% and 54% mass loss, respectively in soil and lipase phosphate-buffered saline solution in the presence of 40 mol.% of the diester.	[15]
Poly(tetramethylene ether glycol), isophorone diisocyanate, lignin	In soil (T= 10°C–35°C, 20%–70% R.H.)	About 61% degradation in the presence of 20 wt.% lignin.	[41]
Castor oil, ethyl ester L-lysine triisocyanate	In soil on nylon mesh bags	Occurrence of bioerosion phenomena after 3 months	[42]
Poly(caprolactone) diol, isophorone diisocyanate, sorbitan monooleate, quercetin	In moist soil and in lipase phosphate-buffered saline solution (T = 25°C)	About 53% and 48% mass loss, respectively in soil and lipase phosphate-buffered saline solution, after 4 weeks.	[43]

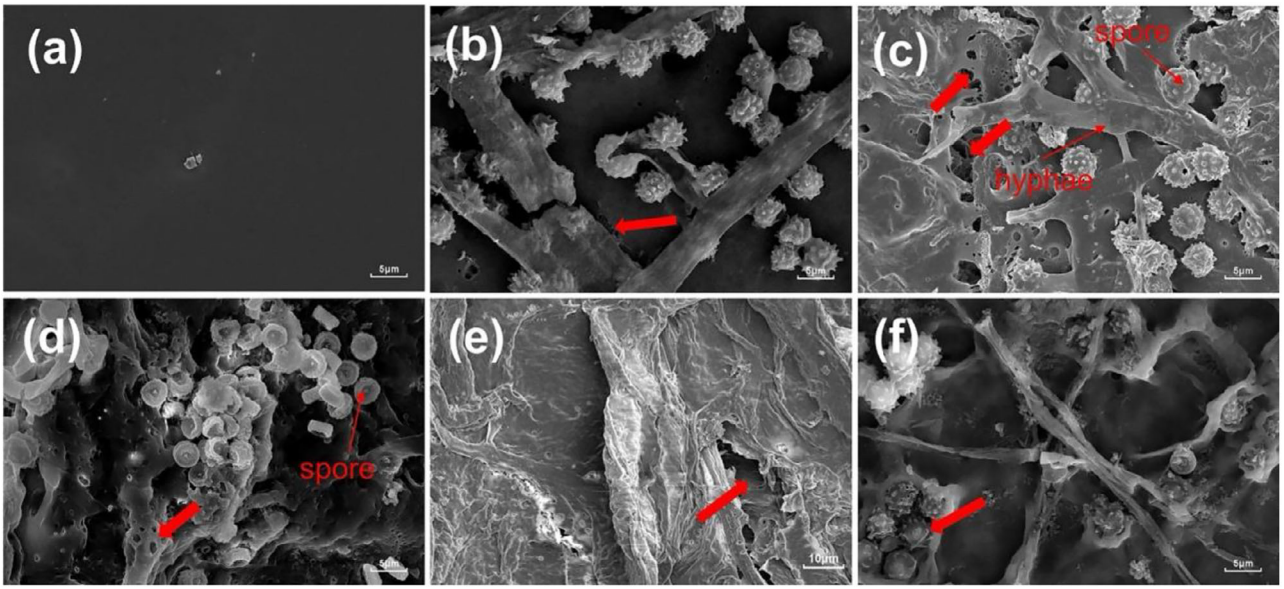

FIGURE 9 | SEM images of PU coating after a 90-day fungal degradation test. (a) Abiotic control; (b,c) Earth sample; (d–f) Space sample. Reprinted with permission from ref. [29], Copyright Elsevier, 2024.

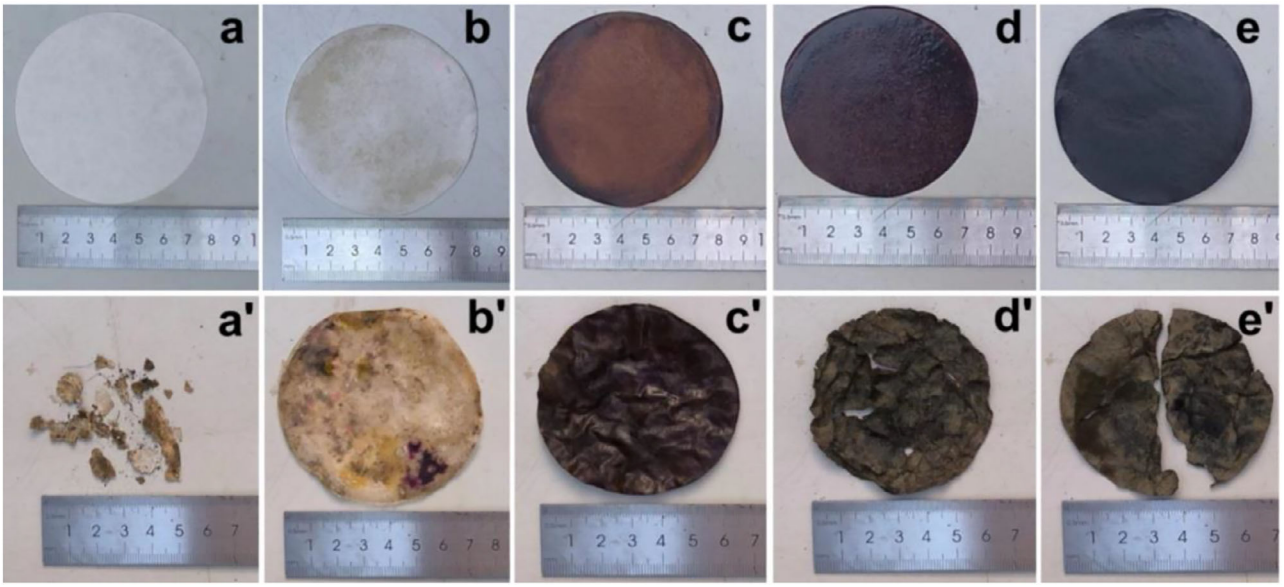

FIGURE 10 | Biodegradability tests of the grass paper mulch films in a natural soil environment: (a) original paper; (b) paper coated with PU; (c) paper coated with PU containing 5 wt.% lignin; (d) paper coated with PU containing 10 wt.% lignin; (e) paper coated with PU containing 20 wt.% lignin. (a'–e') Refer to the same systems after 6 months of biodegradation test. Reprinted from ref. [41], Copyright Elsevier, 2025.

in the surface, witnessed the activity of the microorganism in biodegrading the coating.

Similarly, Zhang and co-workers [46] investigated the biodegradation of epoxy coatings (obtained from the curing of diglycidyl ether of Bisphenol A with Jeffamine D230) induced by *P. aeruginosa*. For this purpose, the epoxy coating was applied on carbon steel plates that, in turn, were dipped into sterile and inoculated culture media, using various nutrient loadings (0%, 10%, and 100%) for up to 28 days. The biodegradation of the coatings, confirmed through morphological (SEM) and electrochemical tests, was ascribed to the break of C—O and C—O—C groups in the epoxy network by the marine microorganism.

Negi et al. [47] developed six bacteria isolates into two consortia (namely, consortium-1, comprising *Microbacterium sp*., *P. putida*, and *Bacterium Te 68R*, and consortium-2, consisting of *P. putida* and *P. aeruginosa*), which were utilized for investigating the in-vitro biodegradation of epoxy and epoxy-silicone blend coatings. Consortium-1 was more effective than consortium-2 in biodegrading epoxy and epoxy-silicone blend coatings, highlighting weight losses of around 34% and 37%, respectively, when incubation was carried out in aerobic conditions for 15 days.

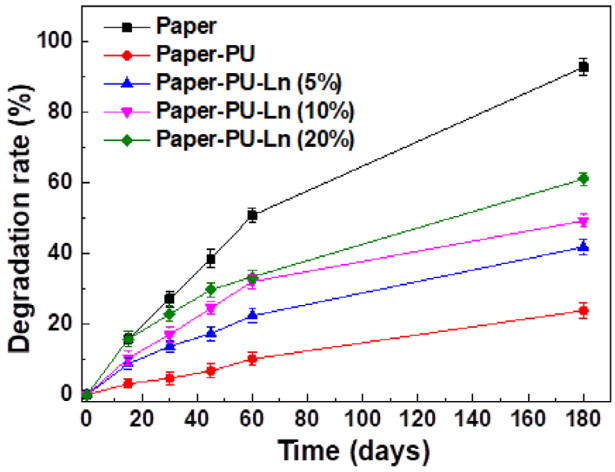

FIGURE 11 | Biodegradability tests of the grass paper mulch films in a natural soil environment: degradation rate vs. time for the original paper (Paper) and the paper coated with PU (paper-PU) and with PU containing lignin (Paper-PU-Ln) at 5, 10, and 20 wt.% loading. Reprinted from ref. [41], Copyright Elsevier, 2025.

Borah and Karak [48] synthesized bio-based tannic acid Bisphenol A epoxy coatings exploiting a surface modification of tannic acid; the epoxy resin was cured with poly(amido amine), a bio-based hardener. The biodegradability of the coatings was investigated by the McFarland turbidity method using *P. aeruginosa* (gram-negative) and *B. subtilis* (gram-positive) bacterial strains. After 30 days of exposure time to the bacteria, a considerable amount of surface erosion occurred in the coatings containing tannic acid; however, the biodegradation was somewhat quite limited, also considering that tannic acid exhibits antimicrobial features against different positive and negative bacterial strains, which slow down the biodegradation processes.

Pardi-Comensoli and co-workers [49] incubated epoxy coatings (derived from the curing of a Bisphenol A/Bisphenol F commercial epoxy resin with an aromatic amine) in soil microcosms inoculated with *Ganoderma adspersum* (a fungus that produces laccase and lignin peroxidase) for 1, 3, 6, and 9 months. A sterile soil was used as a reference. However, no clear weight loss data were observed, except for a decrease in the hydrophobicity of the coating surface, which was an indirect indication of oxidation by the fungus.

Table 2 summarizes the main research outcomes for biodegradable epoxy-based coatings.

3.3 | Other Types of Biodegradable Coatings

Pramanik and co-workers [50] synthesized biodegradable poly(ester amide) coatings through the reaction of *N,N*-bis(2-hydroxy ethyl) fatty amide of castor oil with maleic anhydride, phthalic anhydride, and isophthalic acid (the mole ratio was set at 100:30:35:35). The biodegradation of the coatings was evaluated by incubating them in a sterilized nutrient salt broth medium containing either *P. aeruginosa* or *B. subtilis* bacterial strains up to 6 weeks (working temperature: 37°C); the specimens were taken out at different biodegradation times and weighed. As shown in Figure 14, the *P. aeruginosa* bacterial strain exhibited a comparatively higher degradation rate (achieving about 14% weight loss after 6 weeks of incubation) compared to the *B. subtilus* counterpart (about 3.2% weight loss). This finding was attributed to the higher cell surface hydrophobicity and biosurfactant activity of *P. aeruginosa* compared to *B. subtilis*.

Liu and co-workers [28] synthesized a series of multi-arms (three-, four-, and five-armed) cardanol-based acrylates starting from renewable cardanols and bio-based polyols (namely, glycerol, diglycerol, tripropylglycerol) as precursors. After UV-curing, the so-obtained coatings were tested for biodegradation. To this aim, the specimens were buried in soil for different times, taking them out after 30, 60, and 90 days. As shown in Figure 15, increasing the test duration accounted for changes in the surface morphology of the coatings, with the appearance of a certain roughness, which proved the occurrence of the degradation (after 60 and 90 days, the degradation rates were beyond 6% and 8%, respectively, regardless of the employed polyol). These findings were ascribed to the presence of cardanol and polyhydric alcohols in the polymer network, which behave as weak biodegradable segments.

Piroonpan et al. [51] exploited ring-opening polymerization for obtaining a sugar-based multibranched poly(lactic acid) that, in turn, was blended with tripropylene glycol diacrylate (from 10 to 90 wt.%), added a suitable photoinitiator, and finally UV-LED cured (Figure 16). The biodegradability of the coatings was assessed by continuously measuring the CO_2 production (measured automatically and in real-time every 10 s) after burying them in soil for 17 days at 30°C.

As shown in Figure 17A, the CO_2 production of the UV LED-cured coating in soil showed a remarkable increase (about 2.7-fold compared to the control soil) at the initial measurement step. Then, the carbon dioxide production leveled at about 1.8–2.8-fold until the twelfth day and subsequently lowered (1.5–2.1-fold), probably because of the loss of moisture content in the soil. Besides, at the end of the biodegradation tests, the coatings became brittle, showing the appearance of white spots on their surfaces (Figure 17B).

Another family of interesting polymers suitable for the design of biodegradable coatings refers to polyhydroxyalkanoates, which embrace a group of natural biodegradable polyesters synthesized by microorganisms [52–55]. In this context, Mousavion and co-workers [56] exploited melt extrusion for obtaining blends of poly(hydroxybutyrate) (PHB) and lignin (extracted from bagasse) at different loading of the latter (i.e., from 10 to 90 wt.%). In situ biodegradation was performed in a garden soil, burying the polymer films (according to the ASTM D 5988 standard) for up to 1 year. The soil pH was maintained at 6.7, and the temperature was between 12°C and 27°C; water content was around 20%. As shown in Figure 18, unlike the unfilled PHB film that disintegrated and lost around 45 wt.% of mass within one 1-year test, increasing amounts of lignin in the film formulation significantly inhibited the biodegradation. This finding was attributed to (i) the biochemical protection effect exerted by lignin against the attack by bio-organisms on PHB, or (ii) the segregation of lignin on the film surface, which inhibited the biofilm formation and the access to the underlying polymer.

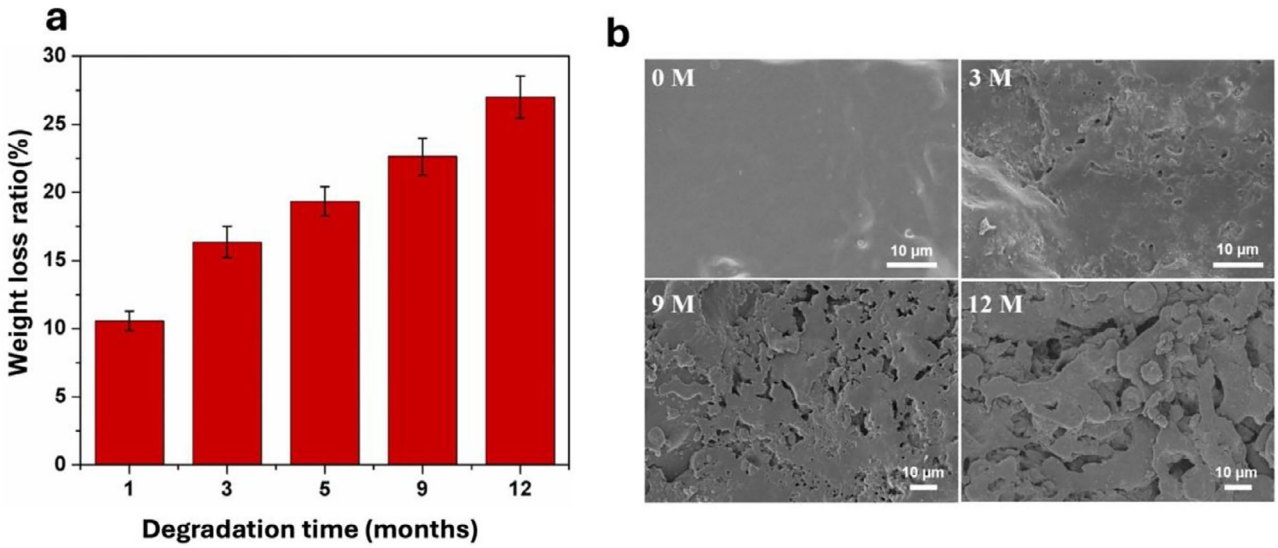

FIGURE 12 | (a) Ratio of weight loss for the PU coatings buried for various periods in soil; (b) typical SEM images of the PU coatings buried in the soil at different intervals (0, 3, 9, and 12 months). Reprinted from ref. [42], under CC-BY license.

TABLE 2 | Main research outcomes for biodegradable epoxy-based coatings.

Epoxy coating formulation	Biodegradation assessment	Main outcomes	Refs.
Bisphenol A-derived epoxy coating	With *P. putida* in seawater	Extended biofilm formation induced by the microorganism.	[44]
Epoxy coating (not specified)	With *B. flexus* in seawater	Worsening of the corrosion resistance during the first 19 days of incubation. Appearance of holes and pores on the coating surface.	[45]
Bisphenol A-derived epoxy coating	Dipping in inoculated culture media for up to 28 days	Biodegradation confirmed by SEM and electrochemical tests.	[46]
Epoxy and epoxy-silicone blend coatings	In two bacterial consortia	About 34% and 37% mass loss for the consortium comprising *Microbacterium sp.*, *P. putida*, and *Bacterium Te* 68R, after 15 days of incubation	[47]
Epoxy coatings from Bisphenol A, tannic acid, poly(amido amine)	In *P. aeruginosa* and *B. subtilis*	Occurrence of surface erosion due to the presence of tannic acid. Limited biodegradation.	[48]
Epoxy coatings from Bisphenol A, Bisphenol F, aromatic amine	In soil microcosms inoculated with *G. adspersum*	No clear mass loss even after 9 months. Decrease in the hydrophobicity of the coating.	[49]

Etxeberria et al. [57] formulated bio-alkyd coatings by reacting pentaerythritol, azelaic acid, dehydrated castor oil fatty acids, and glycerol. Besides, cellulose microcrystalline powder was incorporated in an additional coating formulation at 0.8 wt.% loading. To assess the biodegradability of the coatings, respirometric tests were performed, following the ISO 14855-1 standard. Under composting conditions, irrespective of the presence of microcrystalline cellulose, a significant biodegradability (about 70% in less than 90 days) was observed, which was noticeably higher than that of a commercial reference alkyd coating (34.7%) under the same conditions, as shown in Figure 19.

Table 3 summarizes the main research outcomes for the biodegradable coatings discussed in the present paragraph.

3.4 | Key Design Principles to get Efficient Biodegradable Coatings

Several design principles can be followed to create efficient biodegradable coatings. They will be discussed in the following.

First, it is recommended that labile bonds be incorporated into the polymer coating. It is crucial to choose polymer backbones with bonds susceptible to hydrolysis (breaking down in water), enzymatic degradation, or photodegradation (degradation by light). Polyesters, polyanhydrides, and poly(ester amides), for example, contain hydrolytically unstable bonds that can be incorporated into the coating material [58–60].

TABLE 3 | Main research outcomes for other biodegradable coatings.

Coating type	Biodegradation assessment	Main outcomes	Refs.
Poly(ester amide) coatings	With *P. aeruginosa* and *B. subtilis* in salt broth medium (T = 37°C)	Higher degradation rates observed with *P. aeruginosa* (about 14% mass loss after 6 weeks of incubation) than *B. subtilis* (around 3.2% mass loss after 6 weeks of incubation)	[50]
UV-curable multi-arms cardanol-based acrylates coatings	In soil	Degradation rates beyond 8% after 90 days of test.	[28]
UV-LED curable sugar-based multibranched poly(lactic acid)/tripropylene glycol diacrylate coatings	In soil (T = 30°C)	2.7-fold carbon dioxide production at the initial measuring step. Appearance of white spots on the coating surface after 17 days of incubation.	[51]
Poly(hydroxybutyrate) coatings containing lignin	In garden soil (pH 6.7, T = 12°C–27°C)	Inhibition of the biodegradation due to the incorporation of increasing amounts of lignin	[56]
Bio-alkyd coatings derived from pentaerythritol, azelaic acid, dehydrated castor oil fatty acids, glycerol	With respirometric tests	About 70% degradation achieved in less than 90 days of incubation. Negligible effect on the biodegradation rate provided by the incorporation of microcrystalline cellulose.	[57]

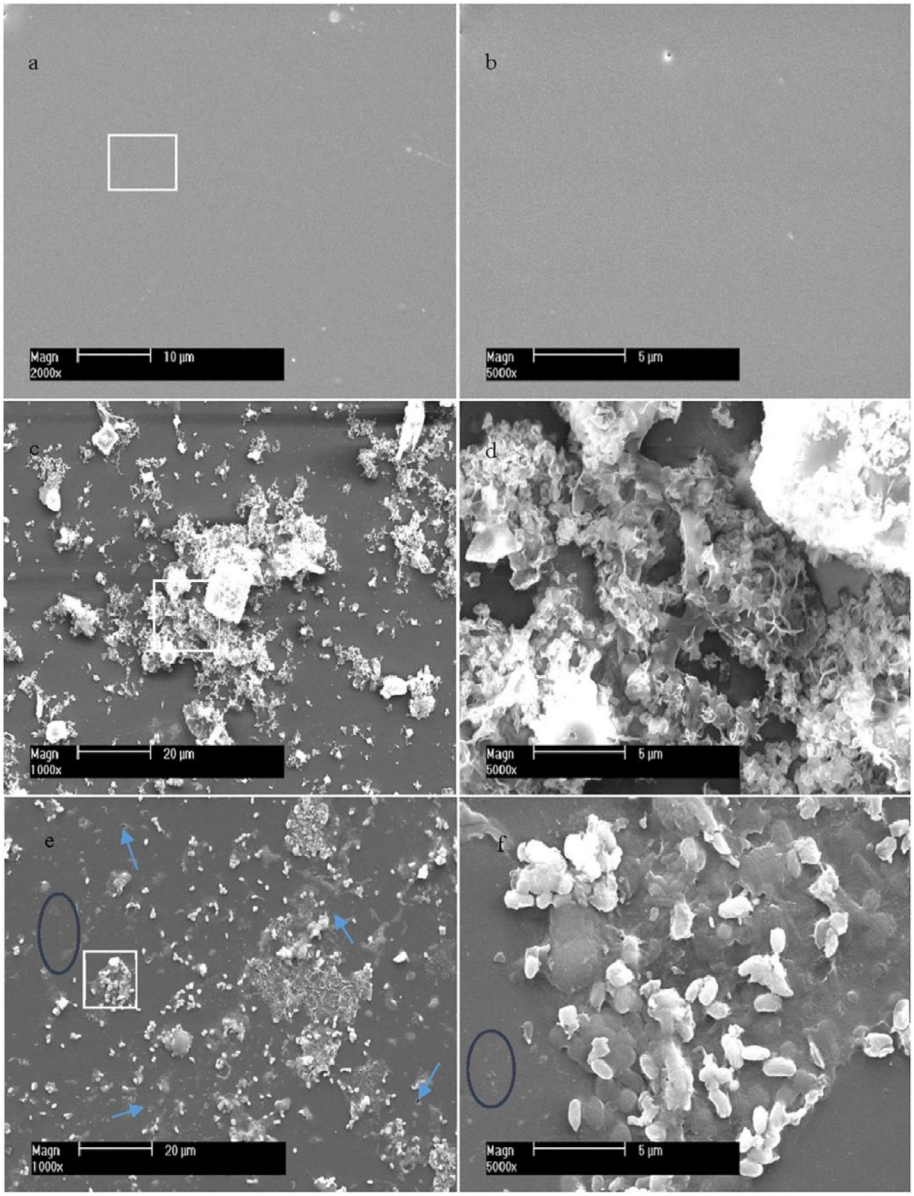

FIGURE 13 | Typical SEM micrographs of the surfaces of the epoxy coatings before immersion (a,b), after 30 days immersion in sterile seawater (c,d), and after 30 days immersion in seawater inoculated with *P. putida* (e,f). Reprinted with permission ref. [44], Copyright Elsevier, 2016.

Another strategy involves the use of biodegradable functional groups, and especially those from bio-sourced components. Indeed, some functional groups are more susceptible to degradation than others. Carboxylic acids, alcohols, and amines, for instance, are generally more biodegradable than aromatic or highly branched structures [61–66].

Furthermore, the molecular complexity of the coatings' structures can have a significant impact on their biodegradability. Simpler structures with fewer substituents and less steric hindrance tend to degrade more readily than highly complex structures. This is because simpler macromolecules offer less resistance to enzymatic or hydrolytic attack [67–70].

The degradation rate of the coatings can also be affected by their degree of crystallinity and their overall morphology. In particular, highly crystalline regions tend to be less susceptible to degradation than amorphous regions [71, 72].

Due to increased exposure to degrading agents, coatings with a higher surface area to volume ratio, such as thin films, will degrade faster [73, 74].

3.5 | Tuning Coatings' Biodegradation: The Role of Chemical Handling

The biodegradation of coatings can be controlled by adjusting various chemical parameters.

Many biodegradable polymers exhibit pH-dependent degradation, meaning their degradation rate is affected by the surrounding pH level [75–77].

Temperature affects the rates of hydrolysis and enzymatic degradation. Generally, increasing the temperature accelerates the degradation process [58, 78].

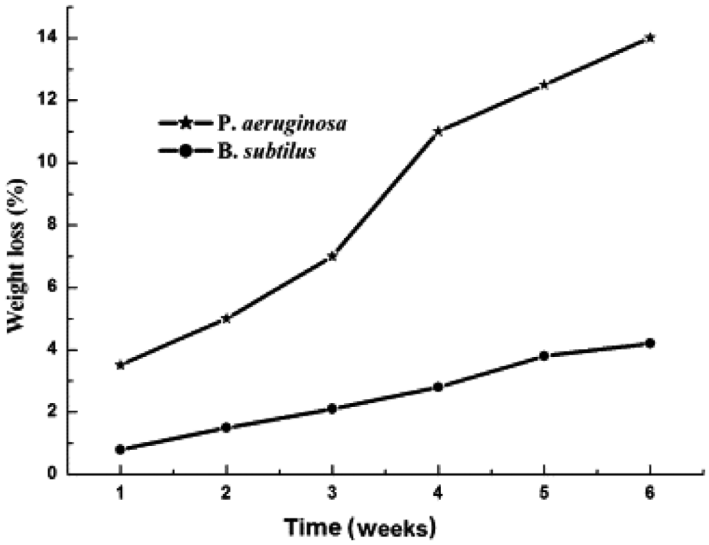

FIGURE 14 | Biodegradation of poly(ester amide) coatings using *Pseudomonas aeruginosa* and *Bacillus subtilis* bacterial strains: weight loss (%) vs. time curves. Reprinted with permission from ref. [50], Copyright Elsevier, 2012.

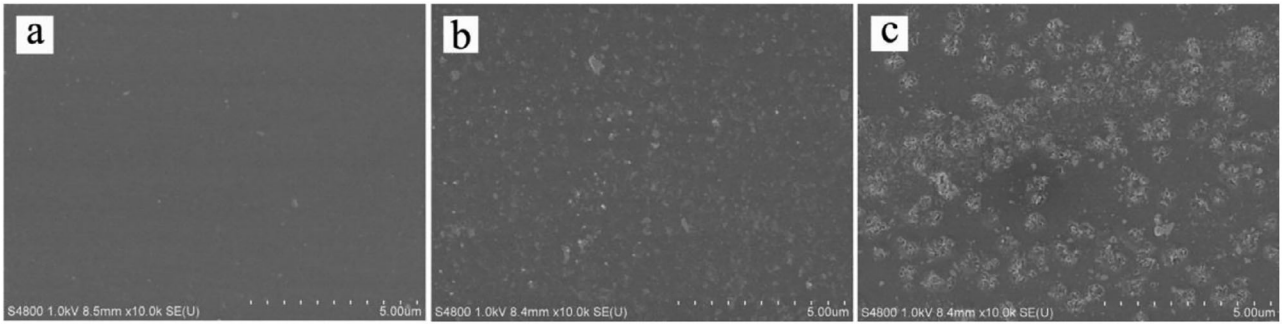

FIGURE 15 | SEM micrographs of UV-cured three-armed cardanol-based coatings buried in soil for different times: 0 (a), 30 (b), and 60 days (c). Reprinted with permission from ref. [28], Copyright Elsevier, 2016.

Furthermore, the presence of specific enzymes in the environment can trigger the degradation of certain biodegradable coatings. For instance, lipases can break down polyesters [79, 80], and cellulases can degrade cellulose-based coatings [81, 82].

Hydrolytic degradation depends heavily on the presence of water. Higher humidity and moisture levels can speed up the breakdown of hydrolytically unstable polymer coatings [83, 84].

Finally, the degree of crosslinking in a polymer network affects its degradation rate. Crosslinking can increase a coating's resistance to biodegradation, while reducing crosslinking can speed up the degradation process [85–87].

4 | Conclusions and Perspectives

The world of polymer coatings has based its development on the need to design, synthesize, and apply systems with high mechanical, thermal, functional, and even aesthetic performance. All this research activity has ensured the achievement of very attractive characteristics in terms of durability, which has been, until not too many years ago, the driving force behind the development of these peculiar polymeric systems.

Nowadays, the need to be able to employ polymeric coatings that can interact on demand with the environment (and in particular with the microorganisms present, under certain conditions of relative humidity, temperature, pH, etc.) and therefore biodegrade, has become one of the most important objectives, also in compliance with the very current concept of circular economy.

Different chemistries in biodegradable coatings have, therefore, been developed, with a particular focus on PU and epoxy systems, for which there has been a strong interest, adequately demonstrated by the nice works reported in the scientific literature.

Despite the strong interest in the formulation, synthesis, and application of biodegradable polymeric coatings, the topic is still under development and implementation. It will require, for years to come, further research efforts to overcome the current limitations. In particular, it is first of all necessary to expand the chemistries behind biodegradable polymer coat-

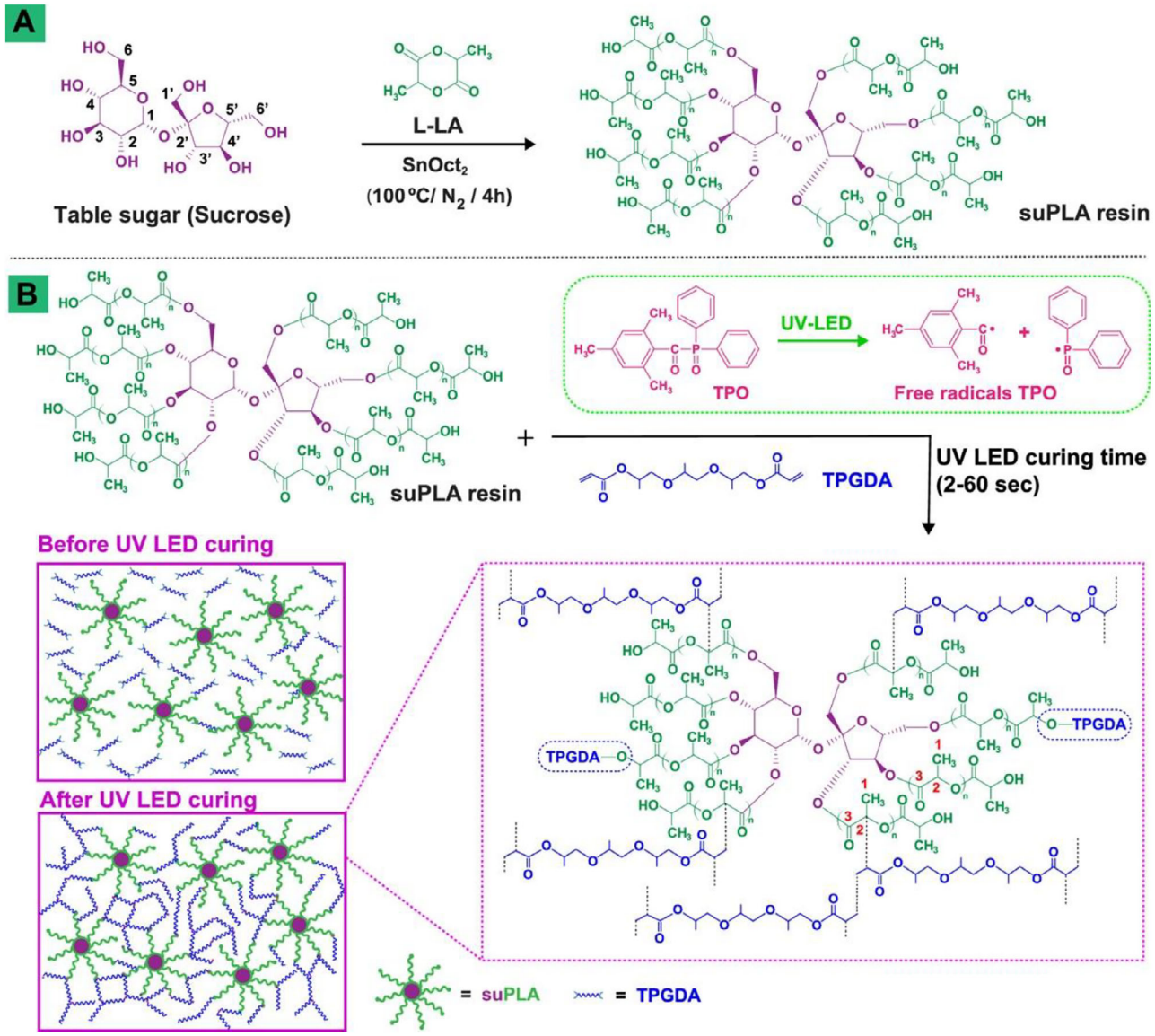

FIGURE 16 | (A) Synthesis of the sugar-based multibranched poly(lactic acid) (suPLA resin) reacting L-lactic acid (L-LA) with sucrose; (B) UV-LED curing of the blends of suPLA resin with tripropylene glycol diacrylate (TPGDA) and possible structure of the network after UV-LED curing (TPO, diphenyl(2,4,6-trimethylbenzoyl) phosphine oxide, is the photoinitiator). Reprinted with permission from ref. [51], Copyright the American Chemical Society, 2022.

ings, which at present are still quite limited: efforts made in this direction can certainly increase the types of systems that can be used, provided that their thermal, mechanical, functional, and aesthetic performance is safeguarded and the on-demand biodegradation phenomena can be easily triggered and carried out. An innovative approach that is worth exploring to enable biodegradation at the coatings end of service life concerns the use of nanotechnology, e.g., by adding intrinsically degradable nanofillers, as proposed for food packaging applications [53].

In addition, it is also necessary to work on the microorganisms used in the biodegradation of coatings, identifying the best performing ones and also choosing them based on the non-toxicity or limited toxicity of the biodegradation products. Further, there is a need to improve the biodegradability performance of the coatings through increased biodegradation rates and decreased biodegradation times.

Notwithstanding the obvious need to face scalability issues, to fulfill the circular economy requirements, and to establish the environmental impact of biodegradation, the investigations available in these subjects are scarce. A particularly challenging aspect of current limitations in biodegradable coatings is that biodegradation has historically been investigated primarily within laboratory settings. The successful implementation of biodegradable coatings on a pre-industrial scale necessitates a substantial scale-up process, a notoriously arduous task. That is also addressing the potential long-term environmental implications of biodegradation products.

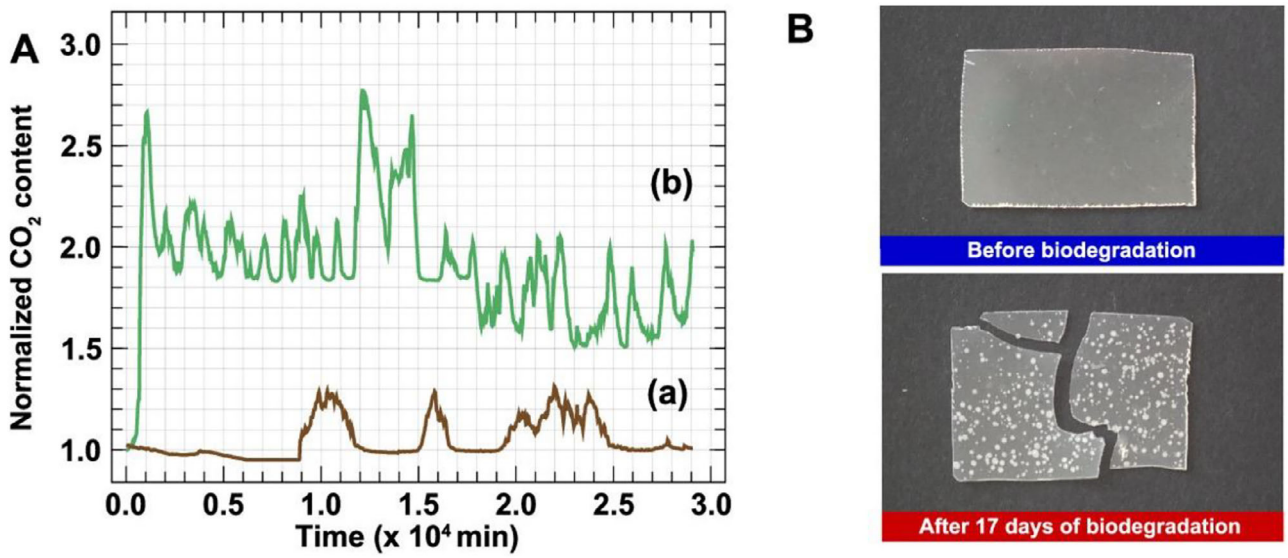

FIGURE 17 | (A) Biodegradation profiles measured from CO_2 production: control soil (a) and soil containing the UV-LED cured coating (b). (B) Photographs of the UV-LED cured coating before (a) and after (b) disposal for 17 days. Adapted with permission from ref. [51], Copyright the American Chemical Society, 2022.

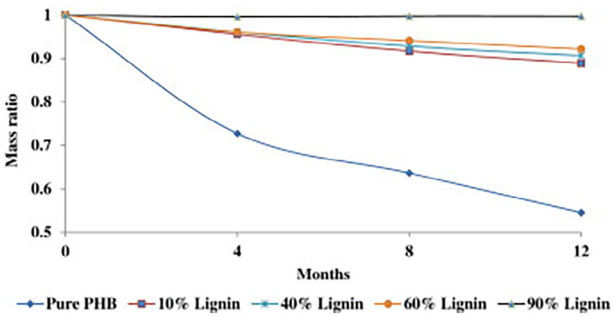

FIGURE 18 | Variation of mass ratio of PHB and PHB-lignin films (from 10 to 90 wt.% of lignin loading) over 12 months of soil burial. Reprinted with permission from ref. [56], Copyright Elsevier, 2012.

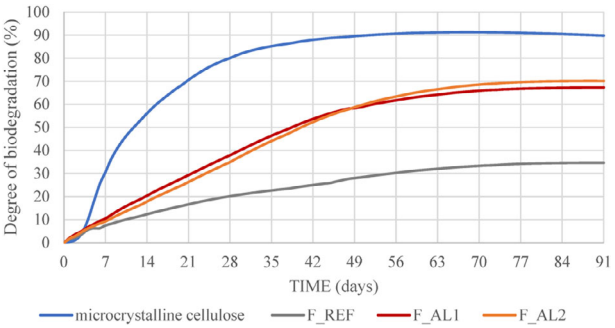

FIGURE 19 | Biodegradation of alkyd coating samples: Legend: F_REF= commercial reference alkyd coating; F_AL1: bio-alkyd coating without cellulose microcrystalline powder; F_AL2: bio-alkyd coating containing 0.8 wt.% of cellulose microcrystalline powder. Reprinted from ref. [57], under CC-BY License.

However, research can be expected to be implemented in the coming years, leading to solutions in terms of better-performing materials and (bio)technologies. Thus, it will contribute to the development of biodegradable polymer coatings, and possibly to their introduction to the market.

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Conflicts of Interest

The authors declare no conflicts of interest.

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