



Sustainable bio-based functional additives for adhesives and coatings – A review

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ABSTRACT

The climate crisis, the depletion of fossil fuels and health concerns about petrochemical components in adhesives and coatings have led to a social and political paradigm shift. As a result, there has been an intense search for sustainable bio-based materials that are also comparable to petroleum-based products in terms of cost and performance. A promising approach is the development of materials with (multi)functional properties that can be achieved by adding functional additives. This review article first explains the requirements for adhesives and coating materials and then presents the current state of research on sustainable bio-based functional additives, with a focus on flame retardant, UV protective, antimicrobial and antioxidant, mechanical stability, superhydrophilic and superhydrophobic, as well as anti-icing properties. Great attention is paid to the use of readily available raw materials such as lignin, cellulose(derivatives) and tannic acid. However, many other substances such as dyes, essential oils or their components, proteins and biopolymers of plants and animals are also promising additives. Hence, there are still some challenges to overcome, such as higher susceptibility to microbial attack or lower durability, in order to obtain market-relevant products. The approaches taken so far are presented in this article, critically analysed and the remaining challenges discussed in detail.

1. Introduction

Adhesives and coatings generally belong to binder systems and have in common that they connect materials across phase interfaces. Depending on the specification, they are used for bonding objects, protecting against external influences, or fulfilling specific functions [1–3]. The global market for adhesives and coatings was valued at over 79 trillion USD in 2023. By 2032, a CAGR (Compound Annual Growth Rate) of 4.6 % is projected, corresponding to a value of around 118 trillion USD. This demonstrates the enormous market potential of adhesives and coatings. Although binder systems are used in almost every sector, such as the automotive industry, aerospace, or electronics, strong growth in the future will be primarily driven by the construction industry. There is also an increasing demand for innovative products [4]. Thus, the requirements for adhesive and coating systems have become more complex in recent years. In addition to adhesive and layer formation characteristics, other functionalities such as UV protection [5–9], antimicrobial and antioxidant properties [10–32], thermal and mechanical resistances [10,19,22,33–41], superhydrophilic or superhydrophobic attributes [6,31,36,42–51], ice-retardant properties [46,

48–57], electrical conductivity [58,59], plasticising abilities [60,61] or high flame retardant effects [37,62–65] are desired.

These features can be achieved by adding functional additives to the matrix material. And although the additives are usually only used in a few parts by weight, they play a decisive role in determining the performance of the entire material due to synergistic effects [66]. These synergies can be significantly enhanced by the use of nanoparticulate additives due to the very large surface-to-volume ratio of nanoparticles. But it may be necessary to functionalise the nanoparticles in advance to ensure a high affinity between the polymer matrix material and the additive [67,68].

However, in view of the current global climate crisis and the increasing depletion of fossil raw materials, the use of additives of petrochemical source is increasingly being disputed. Perfluorinated and polyfluorinated alkyl substances (PFAS) are a very well-known example in this context. Although they have long been criticised for their proven harmful effects on humans and the environment, they are still used in countless everyday products such as food packing materials, textiles, fire-fighting foam and artificial turf. Their highly hydrophobic properties in particular make them an extremely effective additive. Non-

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hazardous alternatives with similar performance do not currently exist, which makes it difficult to replace these substances [69,70].

The development of equivalent sustainable bio-based materials are therefore of great political and economic interest [71]. These materials can be obtained partially or completely from fungi, plants and animals [10,72–74] and can be used in almost every area of daily life, such as construction materials [33–35], medicines [17,28,75–79], textiles and packaging materials [18,25,27,80–84], food [14,15,18,20,25–27] or energy systems [85–87]. Although the term sustainability is not clearly defined, it primarily means reducing waste and greenhouse gases and reaching a high level of recycling. The goal is to protect the environment by conserving fossil resources and using more renewable resources [88].

The need for research in the field of sustainable bio-based additives is therefore correspondingly high. However, only a few materials have so far made it to market readiness, as the environmental factor alone is often not enough. The materials must also be competitive to petroleum-based products in terms of reproducibility, large-scale production, performance and costs [89]. The higher susceptibility of bio-based substances to weathering and to microorganisms as well as the mostly lower mechanical resistance remain a major challenge. In addition, the formulation technology can be very demanding, which is why there is still a need for research [71].

Adhesives and coatings in particular offer great market potential, as they have a very wide range of possible applications. This review article therefore focusses on these two areas. The aim of this paper is to present the current state of research in the field of bio-based sustainable additives for adhesives and coatings, in order to highlight the potential of bio-based alternatives. Firstly, the terms adhesives and coatings are defined and their requirements are explained. Afterward, the current literature using selected examples of bio-based sustainable additives is shown, including a critical review of previous solutions and a

presentation of the challenges still to be solved. An overview of this review article is shown in Fig. 1.

2. Adhesives and coatings – definitions and requirements

2.1. Adhesives

Adhesives are substances that are able to create a mostly permanent bond between two objects. The substrates are held together by close interfacial contact so that mechanical forces can be transferred across the surface. The interaction between the surface of the substrate and the adhesive is primarily responsible for the bonding effect. In order to ensure high adhesive forces, some important basic requirements must be met, such as the selection of a suitable adhesive and joint design with regard to the task, the wetting of the material, the correct bonding technique and the cleanliness of the surfaces [1,2]. In addition, the durability of adhesives is an important factor, as their performance can deteriorate over time, e.g., due to stresses in the material or thermal effects [90].

From antiquity to modern times, adhesives made from renewable raw materials such as birch pitch, beeswax, fish glue or gelatine. It was not until the beginning of the 20th century, with the discovery of phenol-formaldehyde resin, that petroleum-based adhesives were increasingly used. Polyurethane and multi-component epoxy resin systems were used in the 1930s, methacrylate-based adhesives in the 1940s, cyanoacrylates in the 1950s and UV and silane-crosslinking polymers from the 1970s to the 1990s. For some time now, the trend has been moving back towards sustainable binder systems such as methyl cellulose, casein, gluten glue, dextrans, starch, soya or colophonium [40,88,91,92]. Some of these adhesives are already used in a variety of applications, such as medical products, due to their excellent

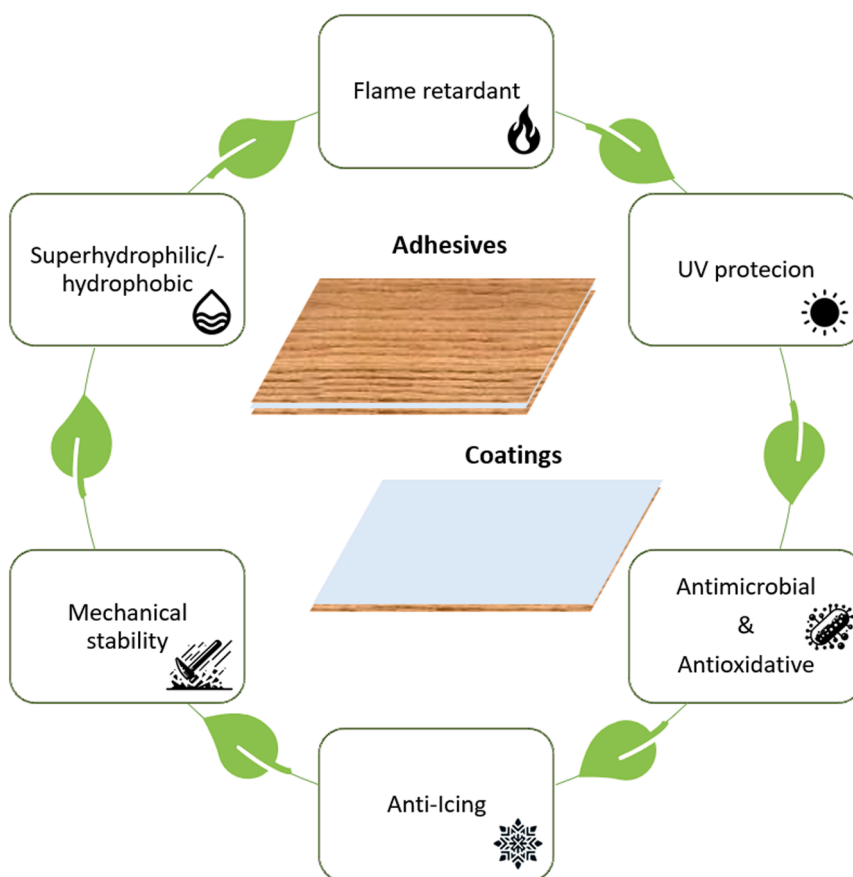


Fig. 1. Schematic overview of the functions of adhesives and coatings through the integration of sustainable bio-based additives shown in this article.

adhesive strength. However, incompatibilities and allergies can occur, as is sometimes the case with colophony [88,93].

2.2. Coatings

The surface of a material is more than just the spatial boundary of its cross-section. Rather, it is a point of attack for thermal, mechanical, chemical, electromagnetic and microbial forces. The resulting material damages cost the economy billions of dollars each year. Measures to protect surfaces are therefore of great interest. This can be achieved either by physical or chemical modification or by coating of the surfaces [94]. The choice of treatment depends on the task, which is essentially aimed at corrosion and wear protection, a defined tribological, optical or decorative feature [95].

In the past, polyurethanes and epoxy resins were mainly used as coating materials, as they have a high processability, mechanical, chemical and thermal resistance, high transparency and very good adhesion to surfaces [96–98]. However, some chemicals used in polyurethane or epoxy resins, such as isocyanates and bisphenol A (BPA), have been shown to be harmful to health [98–104].

Hence, there has been growing pressure on the coatings industry to replace or reduce critical agents. Bio-based polyhydroxy resins and polyurethane resins based on, e.g., castor oil, cashew nut shell liquid (cardanol) and tannins are considered to be very promising in this respect. These materials are therefore most commonly used not only because of their high availability, low costs and ecological benefits, but also due to their excellent properties such as good adhesion, high flexibility, good water and chemical resistance and high impact strength [47,105–107]. However, bio-based polyester resins, e.g., based on itaconic acid, are also frequently used to produce mechanically stable, transparent coatings [84,108–110]. Other examples of bio-based coatings are those based on eugenol [29], polylactide [111], chitosan [41, 111,112], alginate [112] and waxes [6,113,114]. However, some bio-based coating materials are quite expensive, such as chitosan, which is why their use is currently focussed on the biomedical sector [115].

The addition of additives to adhesive and coating systems allows certain functions to be set. The trend is towards multifunctional systems, which are achieved by adding several different additives or by the inherent multifunctionality of a single additive. By definition, functional additives are substances that fulfil a specific function in a product. Although the additives are usually used in concentrations of less than 5 wt%, they have a significant influence on the properties of the whole material [66].

3. Sustainable bio-based functional additives

3.1. Flame retardant additives

Flame protection has always been an essential need for people, as there is always an immediate danger to life and property. Even in the ancient Roman Empire, siege towers were coated with clay in which hair was incorporated as a fire protection additive [89]. Many years later, Gay-Lussac was involved in the development of a flame retardant for theatre curtains based on ammonium chloride and phosphate [116]. Subsequently, highly effective halogenated flame retardants such as chlorinated kerosene were developed [117]. Today, halogenated flame retardants are already banned in the European Union due to their toxicity in electronic components (Regulation (EU) 2019/2021), so research in the field of bio-based additives with flame retardant properties has been strongly promoted [89].

Polymers, which can be found in almost every product in our daily lives, such as houses, cars and furniture, are often good fuels and therefore require special protection. The fire mechanism is initially based on the melting of the polymer under releasing volatile components into the atmosphere. When these come into contact with heat and oxygen, the combustion process is initiated and the occurrence is kept

going by the flames and thus the heat. As long as heat, fuel and oxygen are present, this process continues. Depending on the type of material, this can lead to dripping of the polymer and thus to the spread of fire or to the release of toxic gases. As a result, various fire-retardant strategies have been developed. On the one hand, these consist of the heat reduction and the interception of reactive radicals from the atmosphere. On the other hand, endothermic reactions, e.g., through the decomposition of released water, can be initiated with the aim of cooling. In addition, targeted carbonisation can take place, which ensures that the substrate is thermally insulated. It effectively reduces the spread of flames and prevents the release of gases into the atmosphere [89]. Synergistic effects through the combinations of different measures are also particularly effective [37].

Lignin, for example, is a biobased, harmless flame retardant additive. Chemically, it is an amorphous macromolecule consisting of different phenylpropane units as p-coumaryl alcohol, coniferyl alcohol and sinapyl alcohol [89,118]. A schematic illustration of a section of the chemical structure of lignin and the units can be seen in Fig. 2.

At over 25 wt%, it is an essential component of plants and after cellulose the second most common polymer in the world. As it is a by-product of agriculture as well as pulp and paper production, it is available in large quantities and therefore very inexpensive [89,118]. Although several million tons of industrial lignin are produced every year, the vast majority of it is not used effectively, but incinerated or disposed of elsewhere [119]. This clearly shows that there is still great potential for the commercial use of lignin.

The flame retardant mechanism of lignin is based on its aromatic structure and thus its ability to scavenge free radicals, as well as its special thermal behaviour. The thermal decomposition of lignin occurs with the formation of charred residues ranging from 30 wt% to 50 wt%, significantly disrupting the combustion mechanism. However, this protective char layer degrades in the presence of oxygen, which is why lignin is often used in combination with other flame retardant additives or through targeted chemical modifications [120–124]. For example, Hu et al. [37] modified lignin with phosphorus elements and processed it with wood powder, polylactic acid (PLA) and triglycidyl isocyanurate (TGIC) in a melting process to create a biodegradable flame retardant composite coating material. With optimum mixing of the components, an LOI value of up to 29 % was achieved. The LOI value (limiting oxygen index) is the oxygen concentration at which the combustion of a material can just be maintained. The thermogravimetric analysis also showed a residue of up to 31 %, which indicates a high degree of charring. By comparison, pure PLA had an LOI value of 18 %, demonstrating that the material is highly flammable. The highlights of this study are shown in Fig. 3.

Other scientists have also confirmed the flame retardant properties of lignin [125–134]. In addition to lignin or lignin-based additives, other bio-based additives can also be used as flame retardants. For example, Wu et al. [63] produced a novel intumescent adhesive from a phosphorus-nitrogen flame retardant and a melamine-urea-formaldehyde resin. Fire retardants containing phosphorus and nitrogen compounds exhibit a synergistic effect in both the gas and condensed phases to enhance fire retardancy. In the gas phase, active radicals are generated to scavenge flammable components, while in the condensed phase, a carbonaceous layer is formed to limit contact with oxygen and combustible gases [135]. Intumescent flame retardants have gained increasing attention recently due to their affordability and high performance. One characteristic is that these materials expand upon contact with heat, creating a thermally insulating layer that inhibits heat transfer. The performance is significantly influenced by the matrix materials. Traditionally, melamine-urea-formaldehyde resins are used for their excellent flame retardant properties, low cost, and long-term stability [63]. For example, the flame retardant characteristics of melamine are primarily attributed to the high nitrogen content of the material, which leads to a reduction in smoke density on one hand and a dilution of flammable gases during combustion on the other.

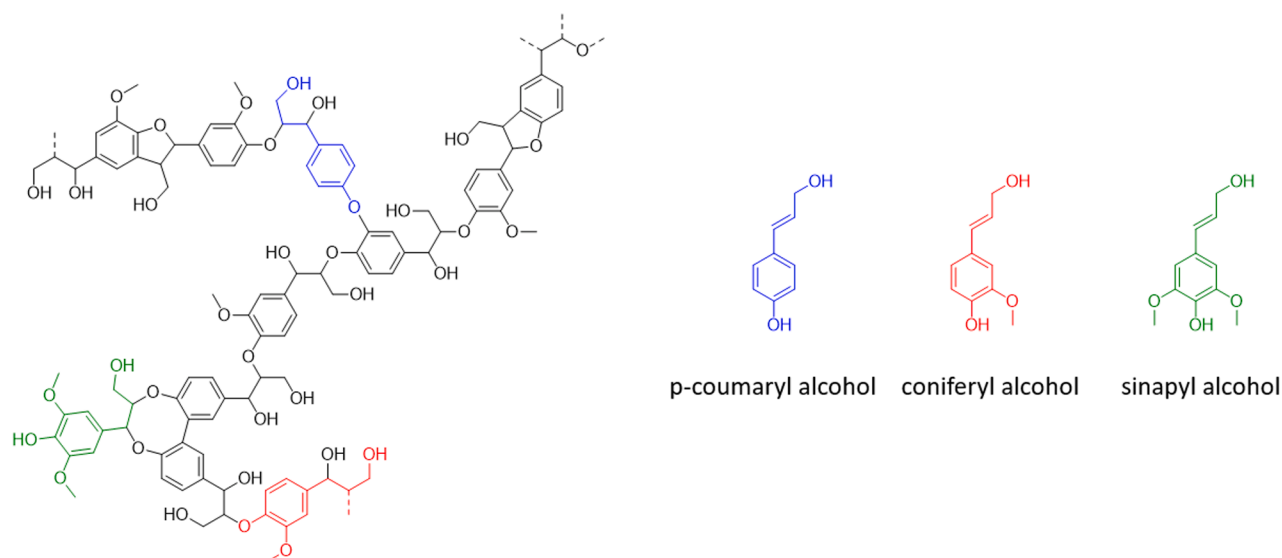


Fig. 2. Schematic representation of a section of the chemical structure of lignin (left) and the components it contains p-coumaryl alcohol, coniferyl alcohol and sinapyl alcohol (right).

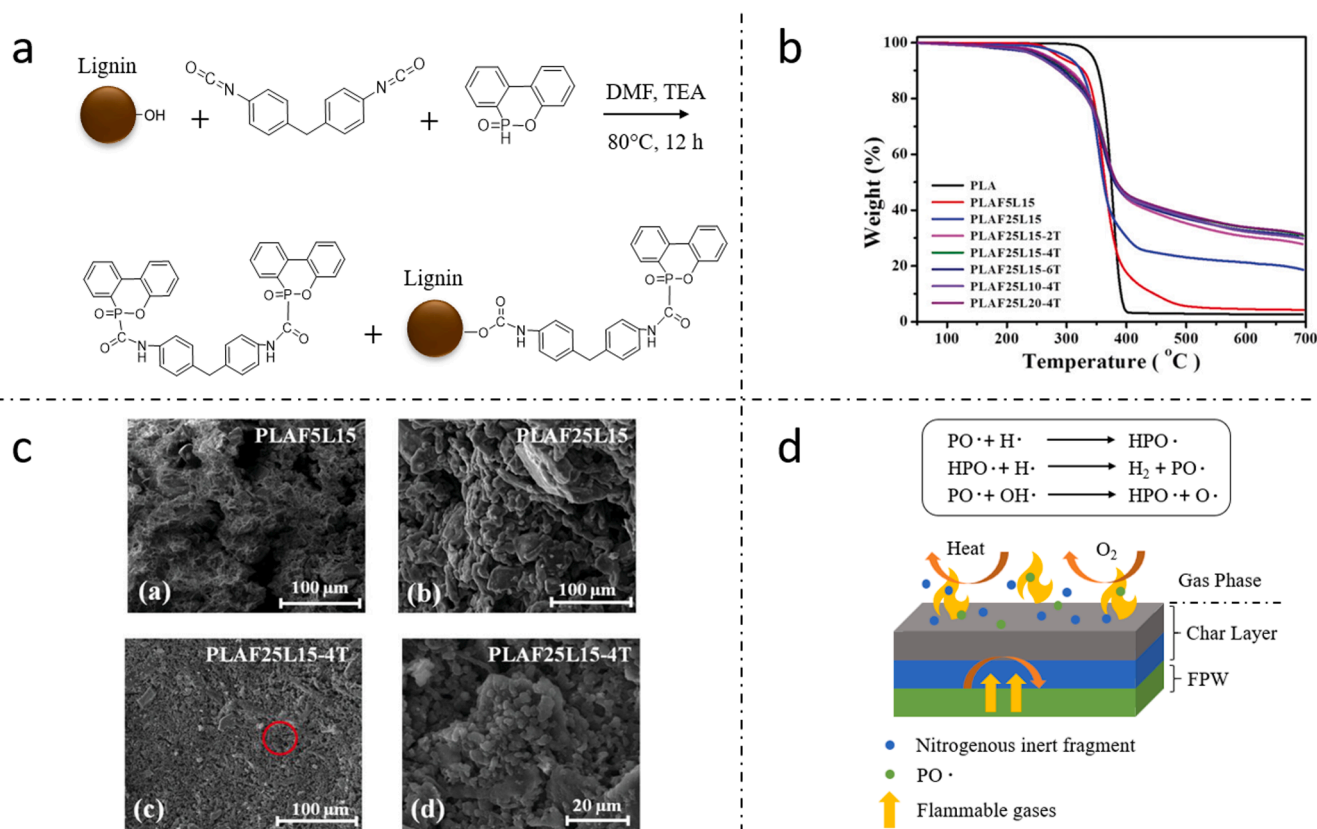


Fig. 3. Presentation of the main points of the study by Hu et al. [37] on a flame retardant bio-based composite coating material of wood powder, polylactic acid (PLA) and triglycidyl isocyanurate (TGIC). (a) Synthesis pathway of the phosphorus-containing lignin-based flame retardant. (b) Results of the thermogravimetric analysis of pure PLA, PLA and wood powder (PLAF) and PLA, wood powder and lignin-based flame retardant additive (PLAF) in various proportions. (c) SEM images of the composites. (d) Schematic representation of the applied flame retardant mechanism. Figure (a), (d) derived and (b), (c) taken from [37].

Additionally, melamine undergoes self-condensation, which in turn makes the protective char layer more compact [136]. However, melamine has recently been classified by the European Union as a category 2 carcinogen and as damaging to the urinary tract with prolonged and

repeated contact. This is likely to severely restrict its use in the future. It is therefore recommended to focus on non-hazardous materials [137].

Bio-based phytic acid is also commonly used as a flame retardant additive in coatings and adhesives [138–142]. It is obtained mainly from

plants such as grains, seeds or beans and consists of about 28 wt% phosphorus. When phytic acid is burned, a stable carbon layer forms, which provides an effective barrier between the combustible material and the flame [143]. Another plant-based additive with flame retardant features is tannic acid. It can be obtained, for example, from the bark or seeds of many tree species. Like lignin, it consists mainly of polyphenol units and also contains some other groups such as organic acids and sugars. As with lignin, the flame retardancy is primarily due to the aromatic structure, which traps free oxygen radicals during combustion. Furthermore, tannic acid also forms a charcoal layer that has a fire-retardant effect. An overview of the use of tannic acid as flame retardant additive is provided by Wang et al. [144].

In addition, inorganic particles such as silica are also increasingly used for their flame retardant properties [145]. An example in this context is given by Wang et al. [62], who developed an adhesive based on pi-pi stacking between siloxanes. The advantage of this adhesive is that, in addition to its good dry shear strength of up to 3.5 MPa on glass substrates and an LOI value of up to 34 %, it can be specifically activated and deactivated. Through targeted infiltration and elimination of ethanol, the pi-pi stacking of the siloxanes and thus the bonding effect could be influenced. This made it possible to reuse the adhesive up to 100 times without significant loss of performance. A combination of silica and phytic acid has proven to be particularly effective, as shown by Lin et al. [146].

3.2. UV protective additives

UV rays can accelerate the degradation of wood-based building materials, packaging materials and food, making the use of suitable sunscreens essential for many consumer goods and for health [7, 147–149].

Lignin also plays a major role as biobased additive in this case, as it has very good UV absorbing properties due to its numerous polyphenol groups. This is a very important characteristic in nature, as it protects the UV sensitive cellulose fibers of plants, preventing degradation and maintaining their strength [150]. Current literature therefore deals a lot with the topic of lignin-based additives as UV agent [8,151–154]. The use of lignin nanoparticles compared to the dissolved form is also interesting in this context, as the enormous increase in the surface-to-volume ratio means that a significant increase in UV absorbing characteristics can be achieved. In addition, some studies have shown that lignin nanoparticles exhibit better compatibility and dispersibility in aqueous solutions with a pH range between 3 and 9 [8, 155–159]. A stable lignin nanodispersion can also be produced in solutions with pH values greater than 10.0 or less than 2.5, provided the particles are chemically modified in advance. This can significantly expand the potential range of applications for lignin nanoparticles [155]. However, one disadvantage of lignin for some applications, is its inherent brownish colour. Initial research approaches aimed primarily at modifying the morphology, chemical structure or particle size of lignin already exist, but require even more intensive efforts in order to avoid a significant reduction in UV protective abilities as a result of the modification measures [8]. In addition to lignin, other wood components such as cellulose and hemicellulose are also suitable as UV filters, but they are also generally used in modified form as a UV protective additive in order to achieve a high effect [160]. Due to its chemical similarity to lignin, tannic acid can also be used as a UV protective additive, as demonstrated by Peng et al. [161]. Another promising UV protection material is biocarbon (BC), which is obtained by carbonizing organic materials, e.g., from by-products of forestry and agriculture, and whose UV protection is based on absorption mechanisms. The advantage of this material is, that its size, porosity and carbon content can be precisely adjusted through the production process [7,162]. For example, Marrot et al. [7] have developed wood coatings from tung oil and dispersed BC particles and analyzed their UV protection properties in terms of weather resistance. It was found that the wood substrate with

the coating without particles showed a greater color change and thus a greater loss of aesthetic appearance over a 6-month period than the coatings with particles. The overall color change ΔE was determined by spectrophotometric measurements of the brightness and the proportions of red/green and yellow/blue components. It was also shown that the higher the particle concentration, the better the reduction in water-repellent properties due to weathering could be prevented. However, it should be noted at this point that black carbon is, on the one hand, a black material and therefore not suitable for all applications and, on the other hand, it is classified by the WHO as a carcinogenic material. This requires special attention when handling and placing products with carbon black as an additive on the market [163].

Another example is given by Hayden et al. [149], who developed a bio-based coating with UV protection by encapsulating the UV filters quercetin, retinol and p-coumaric acid in nanocapsules made of ethyl-cellulose and zein. By specifically adjusting the thickness of the coating material, uniform broadband absorption spectra in the UV range could be achieved. However, there are still some challenges to be solved here. It was found that after 72 h, only 70 % quercetin and 64 % retinol remained in the capsules.

3.3. Antimicrobial and antioxidant additives

The growth of microorganisms on surfaces can result to undesirable losses in the durability and performance of a material [164]. Some nanoparticles, such as silver nanoparticles, have strong antimicrobial properties. Due to their high effectiveness, they are used in particular for medical products. But they are comparatively expensive, which is why they are not suitable for all life science areas [165]. The exact mechanism of the antimicrobial properties of silver particles is still not fully understood. Some of the mechanisms discussed in the literature are, among others, the release of silver nanoparticles and ions, damaging effects on cell membranes, interaction with DNA and the formation of reactive oxygen species (ROS) within the cells that can lead to cell damage and death. However, further studies in this context are still needed [166]. To achieve effective antimicrobial properties, the nanoparticles are either applied directly to the surface or integrated into the coating so that they are slowly released from the layer and kill the bacteria on the surface [167]. For example, Gogoi et al. [19] investigated the antimicrobial properties of a hyperbranched epoxy resin nanocomposite made of silver-decorated cellulose nanofibers and were able to show that effective inhibition of the growth of *Candida albicans* (minimum inhibitory concentration of 132 $\mu\text{g/mL}$) and *Staphylococcus aureus* (minimum inhibitory concentration of 120 $\mu\text{g/mL}$) could be achieved. In comparison, the pure epoxy resins had minimum inhibitory concentrations of at least 220 $\mu\text{g/mL}$. Hence, the use of epoxy resin in this context should be viewed critically, as it contains substances of concern, as mentioned earlier. A sustainable and biobased polymer material, such as polyester itaconate, is desirable [168,169].

Water-based adhesives in particular are very susceptible to microbial infestation. Antimicrobial properties are particularly important here, as they have a significant influence on the longevity of the adhesive. Silver can also be used to achieve very effective protection. In this context, Huang et al. [170] were able to show how excellent antimicrobial properties of a soy-based adhesive could be achieved. The bactericidal rate against *Staphylococcus aureus* and *E. coli* was almost 100 % and against *Candida albicans* almost 91 %. Other examples from the current literature on the use of silver in bio-based coatings and adhesives include [16,171,172]. As already mentioned, silver is an expensive material. To ensure that bio-based products are truly competitive, cheaper materials should be addressed.

Oxidative processes on surfaces can also lead to considerable product damage and, for example, shorten the shelf life of food [15]. Plant substances that are conventionally used as food or are processed in food are often used in this context because they are able to eliminate cell-damaging reactive oxygen species by means of various enzymatic

and non-enzymatic mechanisms [173]. Cranberry, sumac, turmeric, orange oil, cinnamon, cardamom, curry, garlic, thyme, sage, rosemary and oregano extracts are commonly applied [14,17,18,20,25–28,32,77–79,174]. Gonelimali et al. [175] examined the mechanism of the antimicrobial properties of some plant substances such as thyme on gram-negative and gram-positive bacteria. According to their results, the plant materials cause significant cell membrane impairment in the bacteria, which inhibits their growth. Some biopolymers also have antioxidant and antimicrobial abilities such as chitosan, which is obtained from fungi, plants or crustaceans. It is often used in combination with other plant substances to synergistically enhance the antioxidant and antimicrobial characteristics, as shown by Langroodi et al. [24] using a chitosan coating with oregano and grape seed oil. Despite the effective antimicrobial and antioxidative properties, it should be noted at this point that raw materials that compete with the food and feed industry are problematic. It can be assumed that there will inevitably be shortages and thus price increases.

Essential oils or their components are also known for their strong antimicrobial and antioxidant properties. Some examples from the literature are carvon and limonene [176], clove oil [177], oregano essential oil [178,179], orange essential oil [180], galangal essential oil [181], carum copticum essential oil [182], lavender essential oil [183–185], spearmint essential oil [186] and Citrus limonia essential oil [187,188]. A review article on the use of essential oils in coatings is also provided by Teixeira et al. [189]. But even substances that are usually used as colours in the food industry show antimicrobial and antioxidant properties. These include β -carotene [190] and curcumin [191–193], for example.

Furthermore, the combination of different antimicrobial and antioxidative substances can produce synergistic effects. Chang et al. [15] developed a biogenic coating of tannic acid (TA), chitosan (CS) and citric acid (CA) for the preservation of strawberries. They were able to show that the growth of *E. coli* and *S. aureus* could be inhibited most effectively with the TA/CS-CA films with up to around 90 %. A radical scavenging rate of around 90 % was measured for the TA and TA/CS-CA films. In contrast, the CS-CA films achieved around 5 %, indicating that the antioxidant properties are mainly determined by TA. Overall, the coated strawberries achieved up to twice as long freshness retention during a 7-day storage period at 19–25 °C. The highlights of the article are shown in Fig. 4.

Another example of the use of tannic acid as an antimicrobial and antioxidant additive is provided by Li et al. [194] with the development of an edible film made of sodium alginate.

Further to its use as a coating material, chitosan is likewise used as a bio-based adhesive, which in combination with bio-based functional additives can result in a product with multiple functions [20,21,25,115]. Currently, systems with chitosan and inorganic additives such as zirconium oxide and titanium dioxide are also of interest, as in addition to antimicrobial properties, high mechanical strength can be achieved and are therefore suitable for orthopaedic or dental applications [22]. This shows that chitosan is a well-researched and effective antimicrobial additive. However, chitosan has the disadvantage of being relatively expensive compared to other biopolymers, such as cellulose [115]. To be a real competitor to the petrochemical products, the price is very important. Therefore, more research should be done in the future on cheaper additives in this context. Furthermore, the use of titanium

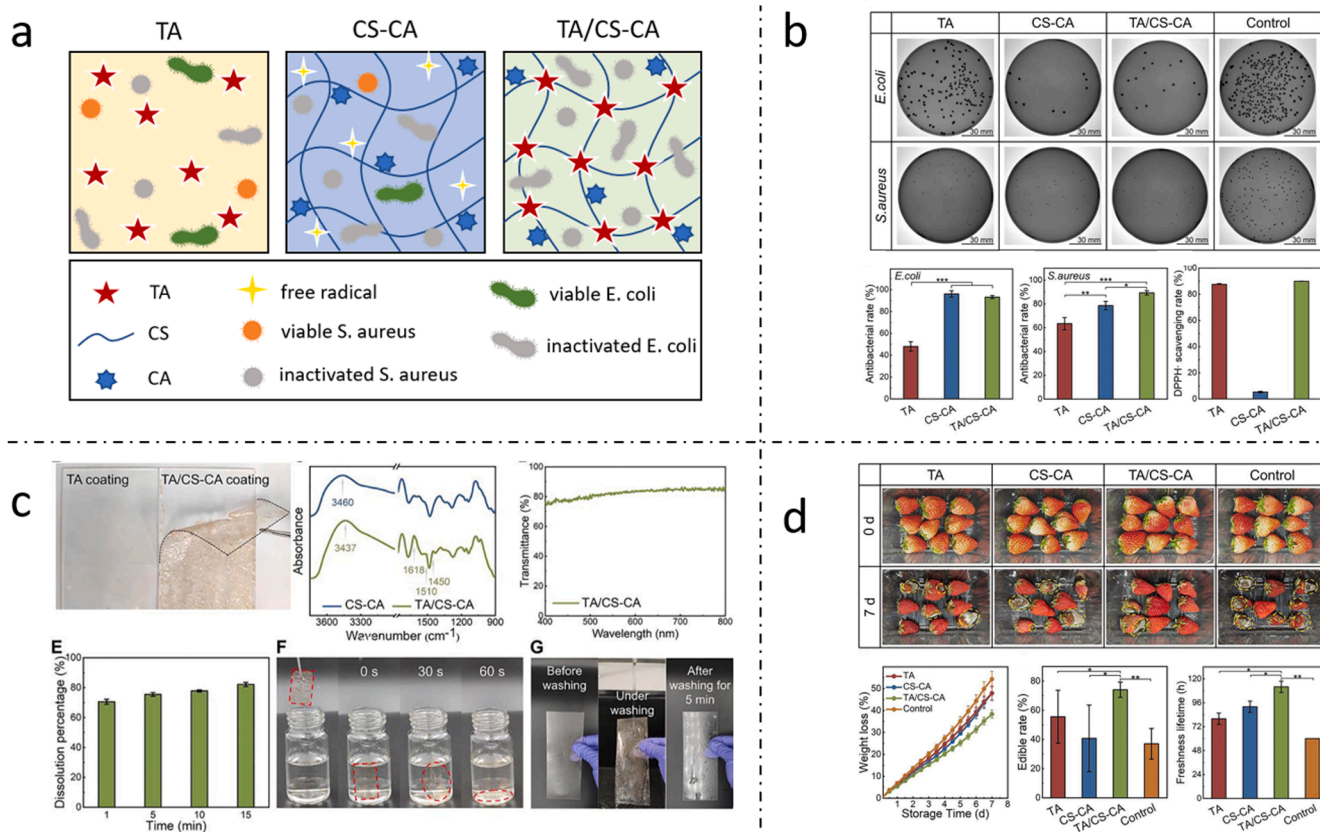


Fig. 4. Highlights of the study published by Chang et al. [15] on the production of a biogenic ternary coating made of tannic acid (TA)/chitosan (CS) and citric acid (CA) to increase the shelf life of strawberries. (a) Development design of the film that combines the strong antioxidant properties of TA with the strong antibacterial properties of CS-CA. (b) Investigation of the bacterial growth of *E. coli* and *S. aureus* as well as the radical scavenging rate of the coated and uncoated strawberries. (c) Analytical methods of the coatings such as photos of the coatings on acrylic plates, FT-IR measurements to investigate the hydrogen bonds, transmittance investigations, dissolution of the films in water over time and water washability. (d) Images, weight loss, consumption rate and freshness time over a storage period of 7 days at 19–25 °C of coated and uncoated strawberries. Figure (a) derived and figure (b), (c), (d) taken from [15].

dioxide nanoparticles is also being restricted due to its potential toxicity [195].

In addition, Lignin is of great interest in this context due to its antimicrobial and antioxidant characteristics. An example is provided by Yang et al. [30], who investigated the antimicrobial and antioxidant capabilities of polylactic acid (PLA) films with dispersed cellulose nanocrystals and lignin nanoparticles. A radical scavenging rate of up to 19.8 wt% was found, which the authors attributed mainly to the radical scavenging properties of the phenolic groups of the lignin. The films with lignin nanoparticles showed high antibacterial properties against the plant pathogens *X. axonopodis* pv. *vesicatoria* and *X. arboricola* pv. *pruni* compared to pure PLA films. The migration values for most of the films were also below the value permitted under EU law for contact with food. Lignin by itself or in combination of other substances are in general frequently used as an antimicrobial agent for coating materials [134, 196–198]. Bio-based adhesives in particular, e.g., those made from soya protein, are susceptible to microbial infestation, which severely limits their usability. Lignin can effectively contribute to the inhibition of microbial growth here [199,200].

Due to the increasing resistance to antibiotics, research has been focusing on the development of coatings with antimicrobial proteins (AMPs) for medical implants for some time. These are low-molecular-weight proteins with a wide range of antimicrobial and immunomodulatory activity [201]. Possible candidates in this case include lysozyme [202], hydramacin [202], cathelicidin [203] and histidine [204].

3.4. Additives to increase mechanical properties

The mechanical properties of adhesives and coatings such as strength, hardness, toughness, elastic modulus or residual stress are crucial for the reliability of the product, as the function can be severely impaired by material failure [39,205]. Significant improvements can be achieved by adding mechanically stable additives. For example, it has long been known that the addition of inorganic additives, such as aluminum oxide or silicon dioxide, results in improved mechanical characteristics because of their high material hardness, which they can transfer to the entire product. However, this can lead to a loss of transparency, an undesirable increase in the viscosity, an increase in brittleness and defects in the material. In this context, sol-gel methods can provide a remedy by adding a sol made of nanoparticles such as silica nanoparticles to the polymer solution to get transparent coatings with high mechanical resistance. This method is often carried out as an in-situ process, in which the inorganic phase is directly coupled with the organic phase by means of hydrolysis and condensation of alkoxysilanes or alkoxides via the formation of covalent bonds and thus creating an inorganic-organic hybrid material with close interfacial contact [66].

Silica is also often used in colloidal form to improve the properties of the adhesive or coating. In a study by Mamiński et al. [205], it was demonstrated that embedding silica nanoparticles in bio-based polyurethane adhesives resulted in a significant increase in adhesive strength. A series of tests showed an improvement from around 2 MPa to 7 MPa. Other examples of the enhancement the mechanical properties of a biobased coating with silica is given by Zhu et al. [206] and Saleh et al. [207]. A detailed overview article in this context is likewise provided by Yona et al. [208].

One potential application is opening up for the paper industry. For example, Chen et al. [36] showed a way to produce a high-strength and superhydrophobic paper from silica nanoparticles and microfibrillated cellulose. First, the microfibrillated cellulose was firmly bonded to the paper by a lamination process and then sprayed with silica nanoparticles, resulting in a multilayer coating material. This resulted in a significant increase in tensile strength by 56 % (dry) and 2277 % (wet) compared to pure cellulose paper. In addition, the coating had a contact angle of around 151.2°, which confirmed a high level of hydrophobicity. However, further processing into consumer goods such as straws or paper cups is questionable in this context, as the increased brittleness of

the films due to the silica particles could cause the coating to break when bent.

Wood components such as cellulose nanocrystals (CNCs) and nanofibrils (CNFs) are likewise be used as reinforcing materials for adhesives and coatings. Chaabouni et al. [209] investigated the shear strength of aqueous polyvinyl acetate (PVA) adhesives with different CNF contents from 1 wt% to 10 wt% and a value up to twice compared to the pure PVA. The improvement of the mechanical properties of coatings through the addition of CNCs or CNFs has also been observed by some other scientists [210–214].

In addition, Lignin nanoparticles show potential as additives to increase mechanical stability, as shown by Del Saz-Orozco et al. [215]. The incorporation of up to 8.5 wt% of lignin nanoparticles into phenolic foams led to an increase in compressive modulus of up to 128 % and an increase in compressive strength of up to 174 %. Strictly speaking, the term nanoparticle is not entirely correct here, since the particles were around 1.6 µm in diameter. Nevertheless, it shows that good properties can already be achieved with lignin particles in the lower micrometer range. It can be assumed that these can be improved by further reducing the particle size.

3.5. Superhydrophilic/superhydrophobic additives

The wetting properties of surfaces are a very important aspect, as they determine the adhesion of undesirable substances such as dirt or bacteria. The reprocessing of contaminated and/or damaged surfaces costs a lot of work, energy and money, which is why intensive research has been carried out in the field of superhydrophobic surfaces in recent decades [45].

Besides superhydrophobic coatings (contact angle > 150° and water droplets rolling off the surface at an inclination angle of < 5°), which target non-wetting of surfaces, there are opposite strategies with superwettability (contact angle < 5°) [43,216,217]. First of all, it is important to understand how the wetting of a surface takes place. This is fundamentally determined by the intermolecular interactions between the phases. Effective factors in this context are the geometric structure and the surface energy of the solid surface [218]. A classic example from nature are lotus leaves, which have strong water-repellent abilities due to their highly defined structuring at the nano and micro level. In contrast, the legs of water striders, moth eyes or butterfly wings show superwettability [216,219,220]. Superhydrophilic coatings are used primarily as antifouling and antifogging coatings that effectively prevent the adhesion of bacteria and fogging of glass or plastic surfaces, as demonstrated by Choi et al. [43]. Superhydrophilicity was achieved by building up a rough surface of chitosan and nanosilica in layers. In addition to very good antifouling and antifogging properties, these coatings also exhibited high transparency. They also proved to be resistant to gram-negative bacteria (*Pseudomonas aeruginosa*) and gram-positive bacteria (*Staphylococcus aureus*). Another example of a bio-based super wettability coating, in this case with a pH-responsive polyaniline coating, is provided by Li et al. [217].

In contrast, materials with very low chemical affinity to water are often used to produce superhydrophobic surfaces. Perfluorinated compounds (per- and polyfluoroalkyl substances (PFAS)) or organosilanes in particular have been widely used in the past because they are very cheap on the one hand and extremely water-repellent on the other. However, because they enter the environment via the products and are not biodegradable, they are increasingly putting a strain on our ecosystem and the health of humans and animals. Bio-based materials from plant or animal sources, which are hydrophobic either naturally or due to their surface structure, have particular potential here. However, there are still some challenges to be solved, e.g., with regard to adhesion strength, abrasion resistance or durability [69,221].

Lignin nanoparticles in particular are of increasing interest in this context. Henn et al. [222] succeeded in producing a bio-based multifunctional wood coating from lignin nanoparticles and glycerol

diglycidyl ether (GDE). The coating was resistant to abrasion, heat, water (contact angle of up to 120°), sunlight and dirt. The hydrophobicity of the coating was mainly caused by the spherical morphology of the lignin nanoparticles. It was found that the thin coatings in particular had a high degree of nanostructuring and surface roughness, which correlated with a high contact angle. However, it is not a superhydrophobic coating, as the water contact angle was $< 150^\circ$. Further tests would therefore have to be carried out here. Wei et al. [223] were even able to realise contact angles of up to 160° with the help of modified lignin nanoparticles.

Cellulose and its derivatives also suitable additives for superhydrophobic coatings. In addition to their high availability and sustainability, the fibres can be broken down into their building blocks at the nanometre level. These fibrillated celluloses give the material certain functions such as mechanical, thermal or optical properties, which is why it is used in a variety of ways. Nanocellulose in particular can greatly improve the properties of materials, for example as a reinforcing agent in composites. Moreover, the hydroxyl groups of the fibres can be used to specifically adapt the surface chemistry to improve the properties or to bind substances/particles to the surface. This demonstrates the potential of nanocellulose for the development of novel functional nanosystems [224]. Ye et al. [31] prepared a multifunctional coating made of nanofibrillated cellulose, tetraethyl orthosilicate and cetyltrimethoxysilane, which, in addition to its superhydrophobic nature, achieved antibacterial, self-cleaning and antiseptic effects. Huang et al. [44] were able to produce a superhydrophobic coating based on nanofibrillated cellulose with good abrasion resistance and durability. Specifically, resistance to sandpaper penetration, knife scratches, fingerprints, long-term storage in water, low temperatures and UV rays were investigated and fulfills the requirements. Current research in this context is on hybrid materials made from wood components such as cellulose and bio-based sustainable inorganic additives such as silica, as

they are suitable for the production of multifunctional products. Other examples from the current literature show the continued great interest in hydrophobic cellulose(derivatives) for adhesives and coatings [225–230].

Chen et al. [42] were able to show that a superhydrophobic coating can be produced from hydrophobic silica nanoparticles and cellulose nanofibers, which exhibited both a high contact angle of 162° and a moisture absorption reduced by up to 34 %. Another example of superhydrophobic coatings with hydrophobic silica nanoparticles is given by Latthe et al. [45]. Various substrates such as window glass or solar cells were coated with the nanoparticles using a dipping and/or spraying process. Each treated material had a water contact angle of almost 160° and a sliding angle of $< 6^\circ$. In addition, the authors investigated the self-cleaning properties of the coatings with soot powder (hydrophobic) and fine dust (hydrophilic) as well as the possibility of oil-water separation. Fig. 5 shows the focal points presented in the study.

Further examples of the use of silica nanoparticles for biobased superhydrophobic coatings and adhesives are [231–233].

Wetting also plays an important role in the formation of ice on surfaces, which is why the use of hydrophobic additives in coatings can also be used to realise anti-icing coatings [50,55,56,234–237]. As the anti-ice effect is a very specialised application and other additives are also used for anti-ice coatings, it is treated as a separate chapter.

3.6. Anti-icing additives

Ice formation on surfaces can lead to a loss of performance (e.g., icing of facilities) but can also pose a safety risk (e.g., in aviation). The anti-ice mechanism is based on the targeted non-wetting of surfaces with liquid droplets or ice, which is achieved by high water contact angles and/or high surface roughness [238].

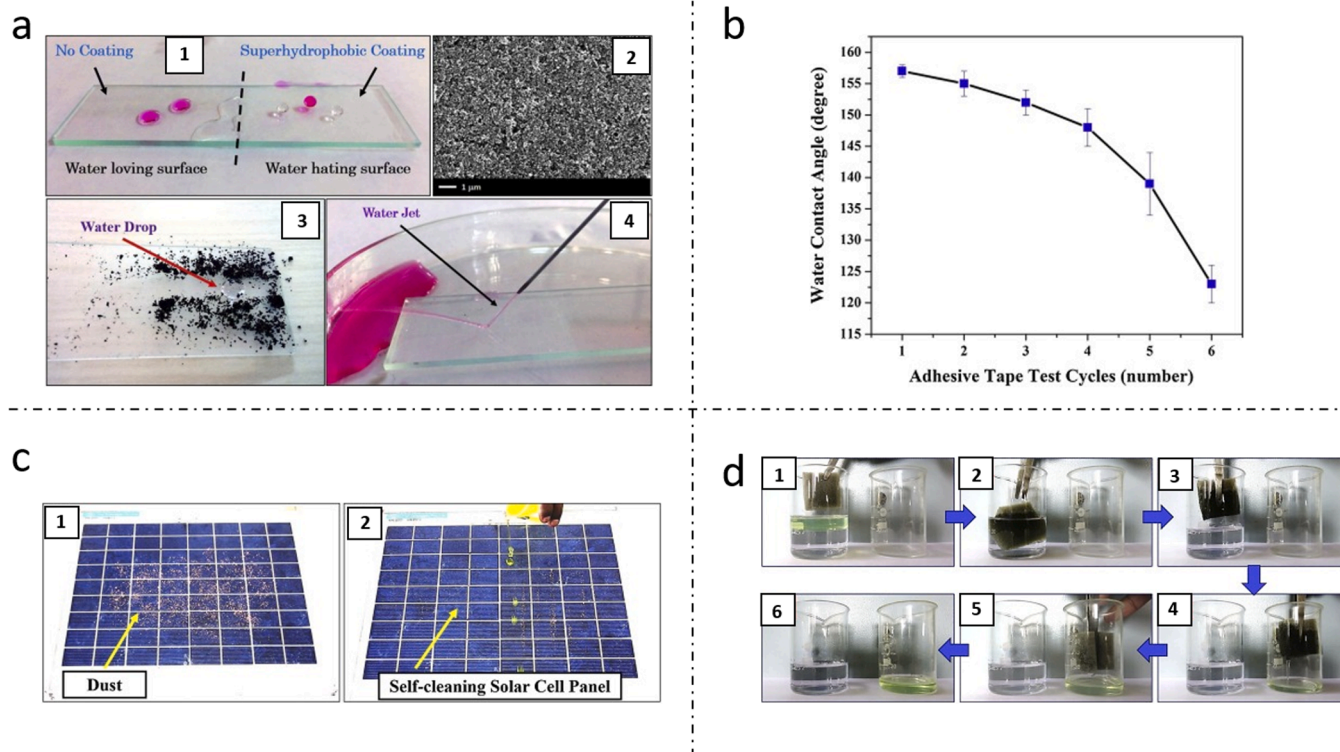


Fig. 5. Presentation of the main results published by Latthe et al. [45] about coatings made of hydrophobic silica nanoparticles with superhydrophobic and self-cleaning properties. (a) Optical comparison of the wettability of water drops on glass substrates with and without coating (1), SEM image of a coating (2) and the resistance to dirt particles from soot (3) and a water jet (4). (b) Results of the adhesive tape test. (c) Presentation of the self-cleaning properties of uncoated (1) and coated solar cells by contamination with fine dust (2). (d) Results of the oil-water separation process, in which a coated sponge absorbs the oil phase in a mixture of diesel oil and water (1–3) and releases it again in a separate beaker (4–6). Figures taken from [45].

In the past, either electrothermal de-icing methods with high energy consumption were used or the materials were coated with epoxy resins, which, contain critical ingredients [51,56]. An alternative is shown by Wu et al. [56]. They developed coatings from bio-based epoxy resins with high transparency and anti-ice properties by using hydrophobic additives such as a fluoroalkyl-functional oligosiloxane, aminosiloxane emulsion and aminofunctional polysiloxane. The coatings showed a reduction in moisture absorption, ice adhesion strength and ice formation time. The use of (3-glycidyloxypropyl)trimethoxysilane (GLYMO) showed a further improvement compared to ethanol in the formulation. The risk to health and the environment posed by fluoroalkyl-functional oligosiloxanes must, nevertheless, be viewed critically. The development of a completely bio-based material is desirable in this context. An overview of the highlights from the study is shown in Fig. 6.

Another example is provided by Zhu et al. [49] through the development of an anti-icing coating of plant origin. The coating was obtained by a hydrosilylation reaction of long alkyl diene monomers with 1,1,3,3-tetramethyldisiloxane and showed an ice adhesion strength of 85 kPa, which the authors attributed mainly to a lower crystallinity and an increased chain mobility compared to conventional polysiloxane coatings.

A promising approach was also shown by Wang et al. [55] with an anti-icing coating made of a bio-based epoxy resin, hydrophobic polydimethylsiloxane and 4,4'-diaminodiphenyl disulfide, which form reversible disulfide bonds, whereby self-healing properties could be integrated in addition to the anti-icing properties (measured ice adhesion strength of 33 kPa). Liu et al. [50] developed an anti-icing coating with very high water contact angles of up to 162° and very low ice adhesion strengths of up to 14 kPa, which also exhibited self-healing properties.

However, the durability of the coatings has so far received little

attention in the literature. As Farhadi et al. [238] were able to show, surface roughness can decrease as a result of multiple icing/de-icing cycles, which also significantly worsens the anti-ice properties. Humidity also plays a crucial role, because they also found that at very high humidity, anti-ice efficiency decreased as condensation formed on the surface and between the bumps. Therefore, more emphasis should be placed on durability.

Anti-icing properties can likewise be achieved by using cryoprotectants based on antifreeze proteins. Gwak et al. [54] developed a coating for aluminium plates with an antifreeze protein from the marine diatom *Chaetoceros neogracile*, which originates from the antarctic and is therefore exposed to particularly cold environmental conditions. To ensure good adhesion, a peptide was used as a conjugator. The protein-conjugator complex was shown to effectively reduce the supercooling point from around −6 °C to −21 °C compared to pure water. If the complex was coated on an aluminium plate, on the other hand, the subcooling point rose again to −9 °C. A few optimisation measures are therefore still necessary. The authors also examined various aluminium plates in the cold chamber. Ice formation was observed after 3 h on the aluminium plate without coating and with a hydrophilic coating of zirconium dioxide, while the plate coated with the protein and the conjugator showed no ice formation. Other examples of anti-icing proteins are also subject of current research [239–242]. The cost-effectiveness of the material is questionable in this context, since it can be assumed that the availability of these special proteins is not high. This inevitably leads to a higher price, making it less competitive than other (petrochemical) products.

4. Conclusions

The focus of this article is on the use of bio-based additives that

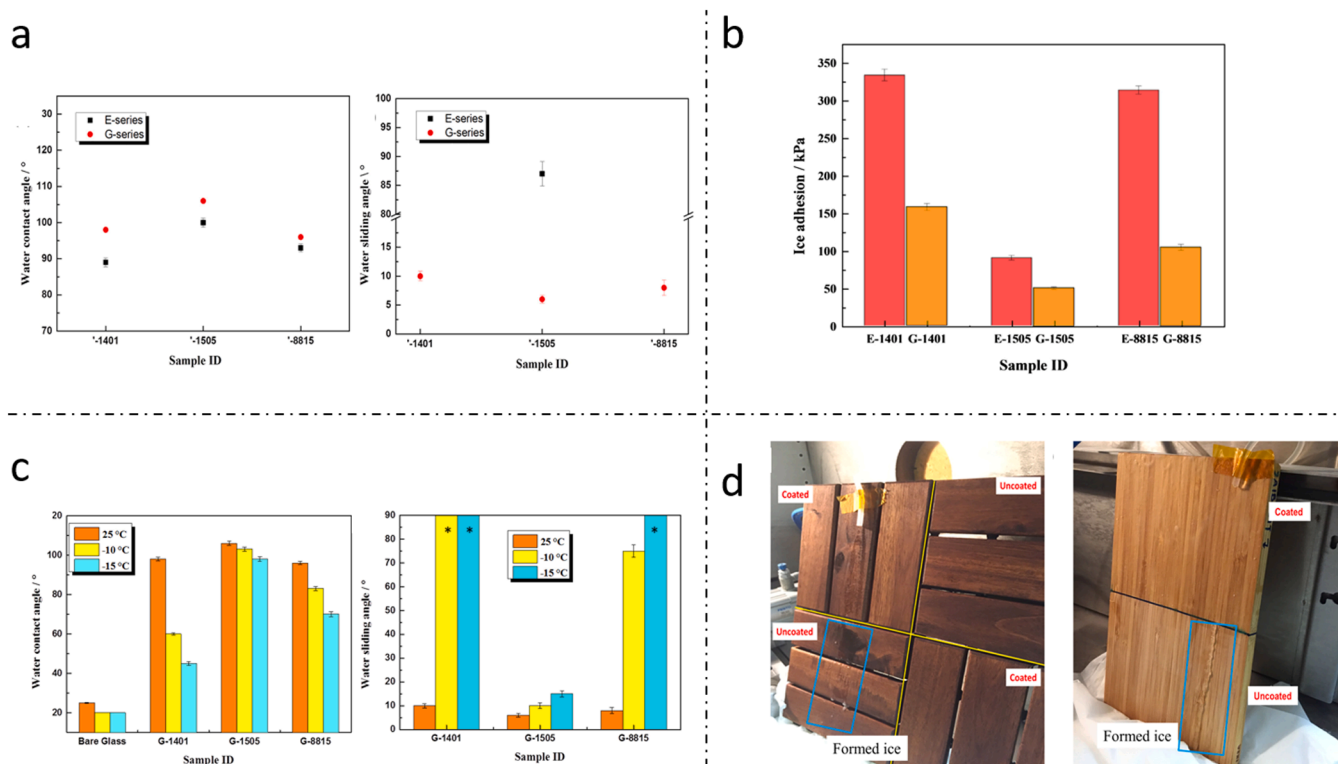


Fig. 6. Key results of the study by Wu et al. [56] by investigating the anti-icing properties of bio-based epoxy resin coatings with hydrophobic additives. (a) Measured water contact angles and sliding angles of the coatings using ethanol (E-series) and (3-glycidyloxypropyl)trimethoxysilane (GLYMO) (G-series) for the formulations as well as using various hydrophobic additives such as fluoroalkyl-functional oligosiloxane (Dynasylan F8815), aminosiloxane emulsion (Tego 1401) and amino-functional polysiloxane (Tego Phobe 1501). (b) Determined adhesion strengths of the coatings on ice at −20 °C. (c) Investigation of the water contact angles and sliding angles of the hydrophobic coatings with GLYMO at different temperatures. (d) Test results of the anti-icing properties of coated and uncoated wooden boards when in contact with water in the climate chamber at −15 °C. Figures taken from [56].

introduce one or more functionalities into adhesives or coatings. The current state of research for additives with flame retardant, UV protective, antimicrobial and antioxidant, high mechanical, superhydrophilic and superhydrophobic as well as ice-inhibiting properties was presented.

Wood components such as lignin, cellulose(derivatives) and tannic acid are used in many areas because they have several characteristics such as flame and UV protection as well as antimicrobial and antioxidant properties. Another advantage is that they are generally readily available. However, they usually need to be used in combination with other additives or their surface chemistry needs to be specifically adapted to increase their effectiveness.

Other sustainable and bio-based additives such as dyes from plants, essential oils or their components, proteins and biopolymers of plant and animal origin have also been used as promising additives for coatings and adhesives due to their (multi)functional properties.

A trend can also be observed in the development of inorganic-organic hybrid materials, as different material features, such as high mechanical resistance on the one hand and antimicrobial characteristics on the other, can be combined in a targeted manner. Silicon dioxide is a frequently used material in this context as an inorganic component due to its safety and high availability. Hydrophobic silicic acid and siloxanes can be used to produce superhydrophobic coatings and represent a promising alternative to PFAS compounds. However, the wetting or non-wetting of surfaces can also be achieved through controlled nano- and microstructuring. The current literature clearly shows the advantages of nanoparticulate additives over macroscopic additives in coatings and adhesives, as the synergistic effects can be significantly enhanced.

The current literature show that a great deal of research has already been conducted in recent years in the field of sustainable bio-based additives for adhesives and coatings. There is huge potential for these products to become commercially relevant in the near future. Due to their diverse and often multifunctional properties, these products are suitable for a wide range of applications, such as flame retardant coatings for building materials, protective coatings for fruit and vegetables, abrasion and impact resistant coatings for outdoor use or resistant coatings against dirt and ice formation for the automotive industry.

However, there are still a number of challenges to be overcome. On the one hand, the switch to harmless water-based products leads to greater susceptibility to microbial attack, which must be taken into account. On the other hand, petrochemical and potentially harmful substances should be avoided altogether wherever possible. Bio-based additives are increasingly being used, but often in combination with other controversial substances to enhance performance. In addition, there should be a stronger focus on durability and availability. These are the decisive factors in order to represent a market-relevant alternative to its petrochemical counterpart. It is also important to ensure that no raw materials that compete with the feed and food industry are used, as this would also have a negative impact on price.

Author contribution

Conceptualization, J.M. and S.E.; writing- original draft preparation, J.M.; writing- review and editing, J.M. and S.E.; supervision, S.E.; funding acquisition, S.E.. All authors have read and agreed to the published version of the manuscript.

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.

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