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## Trash to treasure: advancing resource efficiency using waste-derived fillers as sustainable reinforcing agents in bioplastics

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The escalating environmental challenges posed by different waste sources, including agricultural residues and industrial byproducts, necessitate innovative solutions for waste utilization. Converting waste into valuable resources offers a sustainable approach to mitigating pollution and conserving natural resources. Driven by the urgent need for eco-friendly packaging solutions, this review explores the potential of waste-generated fillers to enhance bioplastic performance. The integration of waste-derived fillers, including nanofillers, into bioplastic matrices significantly improves the mechanical, thermal, and barrier properties, promoting the principles of circular economy and industrial symbiosis. This approach also contributes significantly to reducing carbon footprints by minimizing waste and promoting the reuse of byproducts for sustainable bioplastic production. Addressing the growing concern over the potential toxicity of commercial fillers, specifically metal and metal oxide-based nanofillers, bio-based fillers have emerged as a promising alternative, offering a safer and more eco-friendly solution. An in-depth analysis of recent advancements in processing, production, utilization, challenges, and future prospects would serve as a valuable guide for researchers, industry professionals, and policymakers. The key findings of this review emphasize the necessity of modifying or pre-treating waste fillers to optimize the properties of bioplastic composites. According to the literature, corn processing residues, coffee waste, eggshell waste, and sugarcane bagasse-based fillers are particularly notable among the most studied materials for green composites. Polylactic acid is the most commonly used biopolymer for experimentation with waste-derived fillers. This review underscores the transformative potential of waste valorization in enhancing bioplastic performance, stressing the need for continued research, innovation, and supportive policies to drive sustainable development in this field.

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## Introduction

Agricultural waste residues, industrial byproducts, and agro-industrial waste present substantial environmental challenges, contributing to soil and water contamination, and greenhouse gas (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, etc.) emissions. The two most popular techniques for managing these wastes are landfilling and

incineration, which are environmentally detrimental.<sup>1</sup> Nevertheless, since commercialization to date, approximately 8.3 billion metric tonnes of plastic have been produced, among which only 9% is recycled, 12% is burned, and the remaining 79% is accumulated in sanitary landfills or the surrounding environment. By 2050, the amount of plastic waste in water is expected to exceed the volume of fish. This alarming prediction stems from increased plastic production, inefficient disposal practices, and low recycling rates.<sup>2</sup> The generation of non-degradable solid waste and the depletion of finite resources highlight the urgent need for alternative materials. Bioplastics offer a compelling alternative to conventional plastics owing to their biodegradability and renewable origins. By reducing reliance on finite resources, lowering carbon footprints, and helping mitigate plastic pollution, they represent a significant advancement in sustainable materials.<sup>3–12</sup> Fig. 1 presents the current research trends on sustainable packaging around the world.

The current performance of bioplastics is often inferior to that of traditional plastics, highlighting the need for significant

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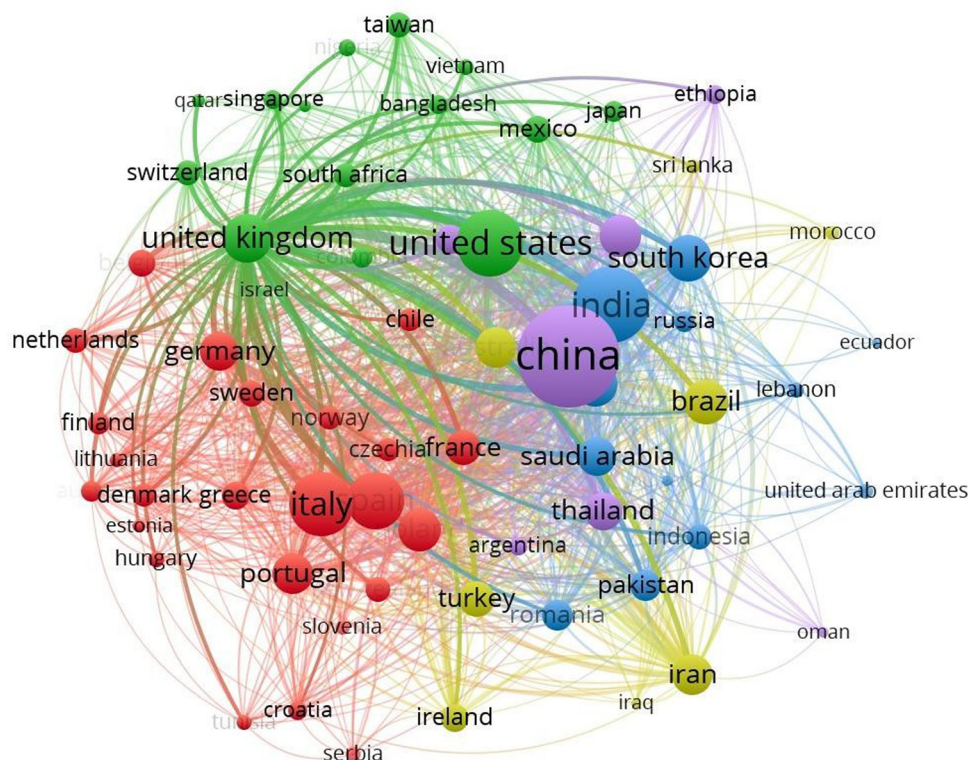


Fig. 1 Sustainable packaging research trend around the world (source: Dimensions AI).

advancements. Recent developments in nanotechnology and polymer engineering have revolutionized applications across various fields. Hence, a key strategy for enhancing bioplastic performance is bioplastic reinforcement, which involves the incorporation of natural or synthetic materials, such as fibers, fillers, or nanomaterials, to improve their mechanical, thermal, and functional properties while preserving their environmental sustainability. Nanofillers, which have at least one dimension in the nanometer range (1–100 nm), are integrated into a matrix to significantly enhance their overall properties.<sup>10,12–16</sup>

Nanofillers have the potential to enhance bioplastic properties, such as mechanical strength, durability, and performance. However, commercially available fillers such as metal and metal oxide nanoparticles raise concerns about their potential toxicity. Consequently, researchers are increasingly focusing on bio-based fillers, such as ZnO, TiO<sub>2</sub>, aluminum, silver, iron oxide, fullerene, and silica, which have raised concerns due to their potential to leach toxic ions, leading to environmental persistence and health risks, including cytotoxicity and bioaccumulation in ecosystems.<sup>17</sup> Their non-biodegradable nature exacerbates these issues and contributes to long-term ecological damage. Studies have also highlighted the risks associated with nanoparticle size, such as increased reactivity and penetration into biological systems, which can cause oxidative stress and inflammation. In contrast, bio-based fillers derived from agricultural or industrial waste offer a safer, renewable, and biodegradable alternative, aligning with sustainability goals while providing comparable or enhanced material properties. Various fillers for bioplastic reinforcement and their

characteristics are depicted in Fig. 2 and 3 highlights the high performance of these types of eco-friendly bioplastics.

Adopting a circular economy paradigm, as opposed to the traditional linear economy, offers significant advantages by promoting resource efficiency and sustainability through the continuous reuse and recycling of materials (Fig. 4). Industrial symbiosis further enhances this approach by fostering inter-industry collaboration, wherein waste from one process becomes a valuable input for another, thus optimizing both economic and environmental outcomes. This strategy not only addresses waste management issues but also supports broader goals of sustainable development and ecological preservation.<sup>12,20–22</sup>

This review article summarizes recent developments in sustainable packaging by utilizing fillers derived from waste sources and addresses the growing concern over the potential toxicity of commercial fillers. Bio-based fillers have emerged as promising alternatives, offering safer and more eco-friendly solutions. The current work comprehensively discusses various aspects of waste filler-reinforced bioplastics for food packaging, including their processing methods and the enhancement of their properties for improved applicability.

## Agricultural waste

The world's population, which reached 7.9 billion by 2021, is projected to grow to 9 billion by 2050 and further to 11 billion by 2100. To meet the increasing demands of the burgeoning global population, there has been a significant increase in both





Fig. 2 Improved characteristics of waste-derived fillers incorporated bioplastic [the image is created by the 1st author Zeba Tabassum; information is obtained from ref. 18 and 19].

livestock and crop production. This increase has, in turn, contributed to the substantial generation of agricultural waste. Over the last century, regions such as China, India, and Africa have not only witnessed rapid population and economic growth but also a significant increase in agricultural waste production. Each year, India generates a large amount of solid waste, with agricultural waste being the most significant, ranging from approximately 350 to 990 million tonnes per year.<sup>23</sup>

Agricultural processing residues have significant potential for repurposing value-added products, contributing to waste reduction and sustainability. Crop residues, process residues, and fruit and vegetable waste are agricultural byproducts that are often wasted, with only a small portion used for animal feed, while the rest are incinerated, harming the environment. Various types of agricultural waste, such as straw, husk, seed, seed pod, peel, shell, bagasse, de-oiled seed cake, stem, leaves,

and bran are the types of agricultural residues that are often underutilized, as presented in Fig. 5. The current work discusses the utilization of rice husk, wheat husk, corn husk/cob/stalks/stover, and oat hull as sustainable resources for filler production and utilization in bio-based polymer matrices. As depicted in Fig. 6, lignocellulosic fibres such as sugar cane bagasse, rice and wheat straw have been widely used as reinforcements in bioplastics.<sup>24</sup> Table 1 summarizes the recently published research on the incorporation of such fillers into different polymer matrices, which has shown the potential for creating more eco-friendly packaging materials by reducing reliance on fossil fuel-based polymers and minimizing agricultural waste disposal issues.

As the world's second-largest producer of agricultural waste after China, India produces over 130 million tonnes of paddy straw annually. Of these, about half is used as fodder, while the



Fig. 3 Properties and types of fillers [image created by the first author, Dr Zeba Tabassum, based on information from ref. 18].





Fig. 4 Linear economy vs. circular economy [the image is created by the 1st author, Zeba Tabassum].

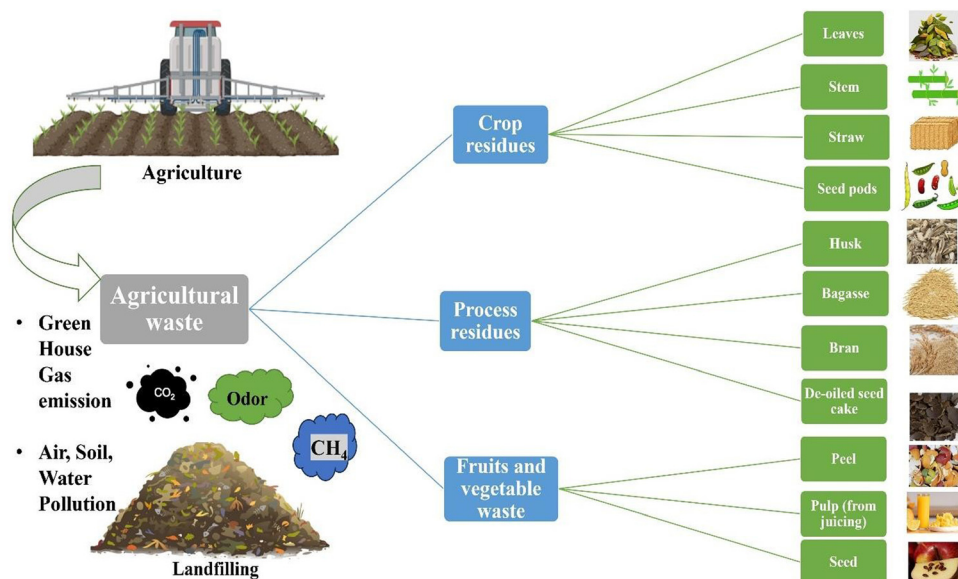


Fig. 5 Agricultural waste types and their negative impacts [image created by the first author, Dr Zeba Tabassum, based on information from ref. 19].

other half is discarded. Additionally, the practice of burning rice residue (parali) in the northwestern region of India leads to significant air pollution and poses serious public health risks.<sup>23</sup> In recent years, there has been a notable rise in the adoption of natural fibres largely due to escalating environmental concerns. These fibres offer a blend of cost-effectiveness and eco-friendliness. Rice husk, a byproduct of agriculture, is now being incorporated as a natural fibre in the manufacturing of biocomposites.<sup>34</sup> A biodegradable, cost-effective, and durable recycled high-density polyethylene (HDPE) polymer was reinforced with rice husk fibre and processed into a foam structure using extrusion, internal mixing, and hot-pressing techniques. Analysis of the foam's structure revealed that a 50 wt% filler

content led to well-dispersed cells with the smallest size (58.3  $\mu\text{m}$ ) and highest density ( $7.62 \times 10^{11} \text{ sel cm}^{-3}$ ), resulting in enhanced mechanical properties. The study showed that the alkali-treated composite foam exhibited the highest tensile strength and Young's modulus (10.83 MPa and 858 MPa, respectively) compared to the other treatments, with increased strength noted in flexural and impact tests following chemical treatment.<sup>25</sup> According to a recent publication, the research team aimed to enhance the ductility and gas barrier properties of poly(lactic acid) (PLA), and developed composites using functionalized mesoporous silica using the solution casting method. The mesoporous silica, derived from rice husk through sol-gel synthesis, was modified with biosurfactant sucrose





Fig. 6 Lignocellulosic fibres from agricultural residues [the image is created by the 1st author, Zeba Tabassum, and the data presented are based on information from ref. 24].

Table 1 Agricultural waste-based fillers for enhancing the performance of bioplastics

S. no.	Filler	Matrix	Mechanical attributes	Other properties	Ref.
1	Rice husk fibre	High-density polyethylene (HDPE)	Enhanced mechanical properties, highest tensile strength and Young's modulus were 10.83 MPa and 858 MPa, respectively	—	25
2	Rice husk fibre	Corn starch	20% rice husk fibre content (and 0.05% benzalkonium chloride) lead to a 61% increase in tensile strength	Optimal moisture levels, transparency, and effective antimicrobial properties against food-spoiling bacteria were found, extending the shelf life of packaged strawberries for over 11 days	26
3	Rice husk	Poly(lactic acid)	Significant enhancements in tensile strength, yield stress, and tensile modulus (by approximately 55%, 88%, and 89%, respectively)	Oxygen transmission rate data indicated a reduction of approximately 52%	27
4	Rice husk ash	Cassava starch	The addition of 20–40% ash increased flexural tensile strength	20–50% ash content led to improvements in thermal stability, density, and biodegradability while reducing their water absorption capacity	28
5	Oat hull fibres	Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate)	At an 8 v/v% concentration, the inert fillers were able to improve the mechanical attributes, increased the Young's modulus by nearly 12%	Cost reduction	29
6	Corn husk fibre	Corn starch	Better tensile strength, and Young's modulus	Exhibited increased thermal stability, improved degradability and crystallinity index	30
7	Corn stover	Poly(butylene adipate-co-terephthalate) (PBAT)	Enhanced the modulus	Accelerated the crystallization. However, it also led to decreased thermal stability and increased moisture absorption	31
8	Corn stover	Poly(lactic acid and poly(butylene adipate-co-terephthalate) blend	Good tensile strength (9.7 MPa), flexural strength (18.1 MPa), elongation at break (61.8%)	Exhibited surface hydrophobicity	32
9	Corn stalk fibre	Poly(lactic acid (PLA)	A decrease in tensile strength (ranging from 22.9% to 51.1%) and bending strength (ranging from 18.9% to 36.6%) was observed. Furthermore, decreasing filler fibre diameter of 1 mm exhibited the best mechanical and thermomechanical properties	—	33

palmitate and anthocyanin. The investigation assessed how varying nanofiller weight percentages impacted the structural, morphological, thermomechanical, and barrier properties of PLA. Morphological examination confirmed the uniform dispersion of the filler within the PLA matrix up to 5 wt%. These composites also exhibited reduced UV-visible light transmission

compared with that of pure PLA. Mechanical testing revealed significant enhancements in tensile strength, yield stress, and tensile modulus (approximately 55%, 88%, and 89%, respectively). A reduction of approximately 52% in the oxygen transmission rate (OTR) indicates a significant improvement in the barrier properties of the packaging material. This reduction



means that the packaging material is less permeable to oxygen, which is crucial for preserving the freshness and shelf life of oxygen-sensitive products, such as food and pharmaceuticals.<sup>27</sup>

Corn starch is a promising alternative to traditional packaging materials; however, its high hydrophilicity and low mechanical strength limit its applications. To address these issues, rice husk fibre, an agricultural byproduct, was utilized as reinforcement, and benzalkonium chloride was incorporated as an antimicrobial agent to create a sustainable and biodegradable packaging film with antimicrobial capabilities. The study investigated how the variations in rice husk fibre amount (ranging from 10% to 50%) affected the mechanical, thermal, moisture, morphological, biodegradability, and optical properties of the bio-composite films. Benzalkonium chloride was utilized as an antimicrobial agent (concentrations varied from 0.05% to 0.2%). Optimal results were observed when incorporating 20% rice husk fibre content and 0.05% benzalkonium chloride into the polymer matrix, resulting in a 61% increase in tensile strength. This blend also maintains optimal moisture levels, transparency, and water solubility, which are essential for practical and aesthetic packaging applications. Additionally, the optimized film demonstrated effective antimicrobial properties against food-spoiling bacteria, thereby significantly enhancing their preservation capabilities. When tested, the film extended the shelf life of packaged strawberries by more than 11 days, highlighting its potential use in food packaging to reduce spoilage and waste. This combination of strength, antimicrobial activity, and preservation effectiveness makes it a promising solution for sustainable food packaging.<sup>26</sup>

Srivastava and team studied how varying amounts of rice husk ash affected the physical, morphological, and mechanical properties of cassava starch-based foams produced *via* thermal expansion. The process involved blending cassava starch with different concentrations of rice husk ash (ranging from 0% to 60%), water, and glycerol. This mixture was then expanded using a thermohydraulic press machine. Incorporating 20–50% ash content led to improvements in the thermal stability, density, and biodegradability of the starch-based foams while reducing their water absorption capacity. These filled foams exhibited finer pores in their internal structures. The addition of 20–40% ash increased the flexural tensile strength, but beyond 50%, the mechanical resistance decreased. The researchers concluded that rice husk ash serves as an effective filler for enhancing the properties of biodegradable starch-based foams.<sup>28</sup>

Wheat straw, an abundant agricultural byproduct, is frequently disposed of or burnt after harvest, contributing to significant environmental issues, such as air pollution and greenhouse gas emissions. Despite its availability, it is often used in low-value applications, such as animal feed or mulching. However, a large portion of wheat straw remains underutilized or wasted, which represents a missed opportunity for more sustainable and valuable applications. Innovative approaches to repurpose this biomass could help address environmental concerns while adding economic value. Potential uses include bioenergy production, bioplastics,

paper manufacturing, and biodegradable packaging.<sup>10</sup> However, there is growing interest in using wheat straw as a reinforcing fibre in bio-composite production. Extracting cellulose from wheat straw can yield customized biopolymers that are ideal for packaging, leveraging non-food resources economically. Recent studies have explored wheat straw-derived cellulose and its nanostructured forms for potential applications in food packaging. Various extraction methods, such as microwave-assisted extraction, fractionation, mechanical fibrillation, steam explosion, microfluidization, and enzymatic hydrolysis, have been mentioned by scholars.<sup>35</sup>

Oat hull fibres, an agricultural waste product, were used as fillers in a poly(3-hydroxybutyrate-*co*-3-hydroxyhexanoate) matrix to create a fully bio-based polymer–matrix composite. The compounding process involved a twin-screw extruder, avoiding the use of chemical solvents and employing a common industrial technology.<sup>29</sup> Salinization was utilized to enhance the bond between the matrix and filler. The compounds were subjected to structural, microstructural, mechanical, and thermal analyses. The results indicated that even at an 8 v/v% concentration, oat hull fibres served as inert fillers and slightly enhanced the mechanical properties of the biopolymer. Specifically, they increased the Young's modulus by nearly 12% without compromising the tensile elongation at break. In this way, Giubilini and coworkers presented an innovative approach that reduces material costs and the volume of poly(3-hydroxybutyrate-*co*-3-hydroxyhexanoate) while valorising agricultural wastes.<sup>29</sup>

Maize is the second most produced crop globally, with a production of 1.13 billion tonnes, after sugarcane. Approximately 40% of the grain produced from corn consists of by-products such as corn cobs, corn husks, and corn stover (as depicted in Fig. 7). Recent research in this field suggests that these lignocellulosic fibre by-products can be effectively utilized as reinforcing materials, rather than synthetic ones, such as carbon and glass fibres, because the former are more affordable, more environmentally friendly, and have better mechanical qualities. The bioplastic composite reinforced with these waste fillers as a supplement for bioplastics exhibit enhanced mechanical, thermal, and barrier characteristics.<sup>36</sup>

Ibrahim and colleagues investigated corn waste-generated filler's impact on bio-based packaging films. The composite films were created using a casting method, incorporating different levels of corn husk fibre (ranging from 0% to 8%) and fructose as a plasticizer in corn-starch-based films. The results showed that as the fibre concentration increased (from 2% to 8%), the tensile properties of the films improved by 88.88%. Overall, adding husk fibre enhanced the film performance. Analysis of the film structure revealed a consistent and compatible matrix-reinforcement arrangement, leading to better tensile strength, Young's modulus, and crystallinity index. The films also exhibited increased thermal stability, with higher onset decomposition temperatures than those of the pure film. Fourier transform infrared analysis indicated stronger intermolecular hydrogen bonding with higher fibre content. Furthermore, the films had reduced density and moisture content and showed improved biodegradation in the soil burial tests. Using corn husk residues in these composites addresses waste disposal concerns and adds value to waste management practices.<sup>30</sup>



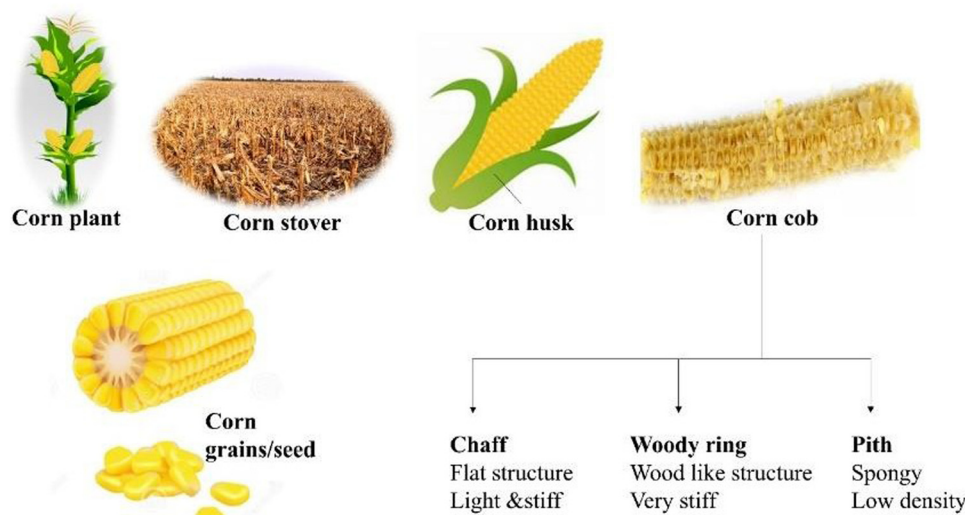


Fig. 7 Different corn processing residues as potential fillers [the image is created by the 1st author, Zeba Tabassum, using data from ref. 36].

To create environmentally friendly and fully biodegradable composites, poly(butylene adipate-co-terephthalate) (PBAT) was blended with corn stover without any chemical treatment. This blending process enhanced the modulus and accelerated the crystallization of PBAT. However, it also led to decreased thermal stability and increased moisture absorption due to the presence of corn stover. As the amount of corn stover increased, there was a noticeable decrease in the morphology and properties, including poor particle dispersion, reduced crystallinity, lower thermal stability, and decreased strength and toughness. Interestingly, reducing the size of corn stover particles at a constant content level resulted in more uniform dispersion, increased PBAT crystallinity, and significant improvements in thermal stability and tensile properties. This adjustment also improved the compatibility between the materials. The addition of corn stover facilitated PBAT crystallization through heterogeneous nucleation.<sup>31</sup>

In another research effort, scientists incorporated corn stover into a PLA/PBAT matrix using the melt blending technique. They discovered that the color and odor of the resulting bioplastics were significantly influenced by the lignin and hemicellulose components. To address this issue, they implemented an optimized alkaline hydrogen peroxide oxidation process on a large scale commonly used in the pulp and paper industry. This modification notably enhanced the appearance, color, and odor of the bioplastic compared to previous methods. The modified bioplastic demonstrated impressive mechanical properties, including good tensile strength (9.7 MPa), flexural strength (18.1 MPa), elongation at break (61.8%), and surface hydrophobicity with a contact angle of 91.6°, meeting the Chinese National Packaging Standards. These improvements were attributed to the rigid structure of corn stover, its robust fibre network, high cellulose crystallinity, and the hydrophobic nature of lignin after treatment, as confirmed by FTIR, XRD, and SEM analyses. The researchers concluded that these bioplastics, filled with treated agricultural waste, offer an attractive appearance, competitive economics, and biodegradability, positioning them as a sustainable alternative to conventional plastics.<sup>32</sup>

Scholars have examined the impact of filler size, specifically 15 wt% corn stalk fibres, on the mechanical and thermomechanical properties of polylactide matrix composites. Four different lengths of corn stalk fibres were used with diameters of 1 mm, 1.6 mm, 2 mm, and 4 mm. The composites were produced using single-screw extrusion, and the samples were prepared through injection molding. The results revealed that as the diameter of the filler fibres increased in the PLA/corn stalk composites, there was a degradation in mechanical properties compared to the matrix, including a decrease in the tensile strength (ranging from 22.9% to 51.1%), bending strength (ranging from 18.9% to 36.6%), and impact energy absorption (ranging from 58.8% to 69.8%). The 3D images of the composite structures showed weak dispersion of filler particles larger than 2 mm, leading to a significant decline in the mechanical and thermomechanical properties of the composite. The composite with a filler fibre diameter of 1 mm exhibited the best mechanical and thermomechanical properties. Overall, from these studies, scholars have suggested that green composites fabricated with 1 to 2 mm corn stalk fibre fillers offer an alternative to traditional plastic-based materials in certain applications.<sup>33</sup>

### Fruit waste

Fruit wastes mainly fall under the category of agricultural and agro-industrial wastes. Fruit waste encompasses the parts of fruits that are often discarded or left unused, such as peels, seeds, cores, and other remnants, that are typically removed during food processing or preparation. This waste presents a notable challenge in the food industry, as it contributes to environmental pollution and resource depletion. Only a small fraction, approximately 0.5%, of the waste produced by the fruit industry is repurposed into usable products. The majority of this waste is disposed through methods such as open burning, landfilling, and composting.<sup>37</sup> Nevertheless, fruit waste can be repurposed creatively, offering opportunities to mitigate its negative effects and generate value. Different fruit wastes, such as jackfruit skin,



orange peel, apple waste peels and pulp (from juice making), mangosteen and durian peel, pineapple crown leaf, avocado seed, banana peel and inflorescence, are discussed in this article. Table 2 summarizes the fruit waste-based fillers incorporated into bioplastics and their packaging property enhancement.

Scientists have proposed an investigation into creating a cost-effective natural fibre-reinforced polymer for food packaging using tropical fruit wastes, such as jackfruit skin as a filler for poly(lactic acid) (PLA). To enhance its properties, bleaching treatment was employed on the fibres. This treatment resulted in rougher fibre surfaces, as observed in scanning electron microscopy (SEM) images, which are expected to improve mechanical bonding with the matrix.<sup>38</sup> This improvement was also evident in the Fourier-transform infrared spectroscopy (FTIR) analysis, which showed better compatibility between the fibres and the PLA matrix. Although the initial fibre insertion led to lower tensile strength, the use of bleached fibres significantly enhanced the composite's mechanical performance. Thermal characterization revealed that the initially low-thermal stability natural fibre composite exhibited improved thermal behaviour after the bleaching treatment, indicating a longer service life for the packaging materials. Additionally, incorporating 15 wt% thymol into the composites showed antibacterial properties against Gram-positive bacteria. Interestingly, the non-treated fibre composite demonstrated better thymol effects than the treated counterpart. The composite with 30 wt% bleached fibre insertion stands out as a promising option for reducing the production costs of bioplastic products without compromising the overall performance.<sup>38</sup>

Apple waste, consisting of peels and pulps remaining after juice extraction, was utilized as both the foundational material and a strengthening component in the development of novel bioplastics. To attain the optimal stiffness, a higher percentage (40 wt%) of dried apple pulp paired with a minimal quantity (2 mL) of glycerol was employed. Conversely, the highest hardness levels were achieved at the lower concentrations (20 wt%)

of rehydrated apple pulp along with the same minimal glycerol additions. High degradability was observed with 58% weight loss after 1 month.<sup>46</sup>

This study aimed to develop environmentally friendly polymer composites with circular economic goals. The research involved melt mixing poly(butylene succinate-co-adipate), a biopolyester, with orange peels of up to 20% by weight to create active polymer composites.

Experimental findings revealed that orange peels, acting as natural fillers, contain abundant phenolic compounds with antioxidant properties that are readily available from food waste sources. Despite a slight decrease in crystallization temperatures and percentage, the filler had a minimal impact on the thermal stability of these composites. Mechanical testing showed an increase in elastic modulus but a decrease in tensile strength, while elongation at break remained stable. The addition of natural filler significantly enhanced the antibacterial and antioxidant properties, thereby improving the functionality of the base polymer.<sup>20</sup>

A bioplastic incorporating cellulose nanocrystals derived from mangosteen peel as a reinforcing filler within a cassava starch matrix was developed. The process involved several steps, such as delignification, bleaching, hydrolysis, and sonication, to isolate the nanocrystals. The impact of these filler additions on the mechanical properties was evaluated using Fourier transform infrared spectroscopy (FTIR) and a universal testing machine. The results showed that compared to the pure bioplastic without fillers, the addition of nanocrystals had varying effects on the tensile strength, tensile modulus, and elongation at break. FTIR analysis confirmed the absorption pattern of cellulose within the composite bioplastic matrix. With the inclusion of cellulose nanocrystal fillers, the elongation at break and density of the bioplastics increased, with the lowest values of elongation at a break recorded at 13.93% and a density of 952.5 kg m<sup>-3</sup>. This study illustrated that the

Table 2 Fruit waste as filler for bioplastic reinforcement

S. no.	Filler	Matrix	Characteristics	Ref.
1	Jackfruit skin	Poly(lactic acid)	Improved thermal behaviour, antimicrobial property, and compatibility	38
2	Orange peel	Poly(butylene succinate-co-adipate)	Despite a slight decrease in crystallization temperatures and percentages, the filler had a minimal impact on the thermal stability of these composites. Mechanical testing showed an increase in the elastic modulus but a decrease in the tensile strength, while the elongation at break remained stable. The addition of the natural filler significantly enhanced antibacterial and antioxidant properties	20
3	Mangosteen peel-derived cellulose nanocrystal	Cassava starch	Mechanical parameters were influenced, and the elongation at break and density of the bioplastics increased	39
4	Durian peel fibre	Thermoplastic cassava starch	Enhanced the thermal and mechanical properties, accelerated biodegradation	40
5	Nanocrystalline cellulose extracted from pineapple crown leaf	Whey protein isolate	Water solubility decreased. Tensile strength peaked, while elongation at break showed a decreasing trend. An increased glass transition temperature was observed	41
6	Banana inflorescence	Starch	Enhanced water resistance, mechanical strength, and thermal properties	42
7	Banana peel	Thermoplastic starch	10% filler by weight demonstrated the highest tensile and tear strengths, reaching values of 39.303 MPa and 66.388 N mm <sup>-1</sup> , respectively	43
8	Cellulose nanofibres extracted from unripe banana peel	Starch	Biocompatible, biodegradable, and non-toxic	44
9	Avocado seed	Polyethylene/ethylene-vinyl-acetate	Achieved a UL-94 V-1 classification in fire safety tests, indicating improved flame retardancy	45



mechanical properties were distinctly influenced by the presence of nanocrystals in the bioplastic composite.<sup>39</sup>

Durian peel fibres, sourced from agricultural waste, were extracted and blended into thermoplastic cassava starch at varying concentrations (10, 20, 30, 40, and 50 wt%) using compression molding. The addition of these fibres notably enhanced the thermal properties of the bio-composite, leading to increased thermal stability, as indicated by the higher onset decomposition temperature. Furthermore, the incorporation of durian peel fibres accelerated the biodegradation process of the composites. Moreover, the study revealed that introducing durian peel fibres into thermoplastic cassava starch composites improved their tensile and flexural properties, with the 40 wt% fibre content yielding the highest modulus and strength. The tensile and flexural strengths showed substantial improvements, increasing from 2.96 to 21.89 MPa and 2.5 to 35.0 MPa, respectively, compared to the control samples. This enhancement was corroborated by Fourier-transform infrared spectroscopy and scanning electron microscopy analysis, which indicated a strong interaction between the fibres and matrix. Overall, the analysis suggested that a 40 wt% ratio of durian peel fibres offered the most favourable composition among the tested ratios. These results highlight durian peel fibres as a promising green reinforcement resource for biodegradable thermoplastic cassava starch matrices.<sup>40</sup>

A study investigated the impact of bio-based nanocrystalline cellulose (NCC) extracted from pineapple crown leaf (PCL) on whey protein isolate (WPI) films using a solution casting technique. Six WPI films were created with varying NCC loadings from 0 to 10% w/v, and their mechanical, physical, chemical, and thermal properties were analysed. The results revealed that higher NCC loadings increased the thickness of the films but reduced their transparency. The presence of NCC decreased the moisture content and absorption, especially at higher loadings. Water solubility decreased from 92.2% in pure WPI to 65.5% in 10% w/v NCC. The tensile strength peaked at 7% NCC loading, reaching 5.1 MPa, while the elongation at break showed an inverse trend. FTIR analysis showed a slight impact of NCC on the WPI film spectra. NCC also improved the thermal stability of the films, evident in the increased glass transition temperature at higher NCC loadings. Morphologically, films became rougher and more heterogeneous with small particle aggregates due to NCC addition. Overall, incorporating PCL-based NCC as a filler enhanced the water barrier and mechanical properties of WPI films.<sup>41</sup>

Banana inflorescence, also known as banana heart or banana blossom, is not typically considered waste, and in many cultures, especially in Southeast Asia, it is used as a culinary ingredient in various dishes. However, in some cases in which the banana plant is cultivated primarily for its fruit and not for its inflorescence, the banana blossom may be discarded as waste. Nonetheless, efforts are being made to utilize it more efficiently, thereby reducing waste and maximizing the use of this plant in part.<sup>12</sup> Banana inflorescence contains medically active compounds, such as polyphenols, sterols, and fatty acids. The extraction processes generate waste rich in insoluble fibres, such as lignocellulose, typically considered without value outside of

medical applications. However, researchers, such as Pongsuwan and colleagues, explored repurposing this waste using banana inflorescence fibre as a filler in starch-based bioplastic production. Their findings showed good adhesion between the filler and starch matrix, with hydrogen bonding confirmed through Fourier transform infrared spectroscopy, indicating phase compatibility. This approach enhanced water resistance, mechanical strength, and thermal properties of the starch-based bioplastic, showcasing the potential of banana inflorescence waste as an effective filler.<sup>42</sup>

Izzah and team extracted banana peel (BP) through a maceration process and integrated it into the thermoplastic starch (TPS) matrix to create the TPS/BP polymer complex. The TPS/BP composites containing 10% BP by weight demonstrate the highest tensile and tear strengths, reaching values of 39.303 MPa and 66.388 N mm<sup>-1</sup>, respectively. Regarding biodegradability, the composite with 40% BP shows a faster degradation rate compared to those with 5% BP, experiencing an average weight loss of 65.1% over 8 weeks, while the latter shows an average weight loss of 45.2%. These findings collectively highlighted the significant improvement in the physical and mechanical properties of TPS/BP polymer composites, positioning them as a promising option among existing biodegradable polymers.<sup>43</sup>

Ahmed and groups fabricated nano-composite packaging films using starch molecules reinforced with cellulose nanofibres extracted from unripe banana peel. Incorporating 10 g of cellulose nanofibres per gram of dry starch significantly enhances the mechanical and thermal stability of these films. The addition of cellulose nanofibres also leads to a notable reduction in the water solubility and water vapor permeability of the packaging films. The mechanical properties, specifically elongation at a break and tensile strength, show significant improvements with the inclusion of these nanoparticles. Analysis using Fourier transform infrared spectroscopy revealed similar spectra for all tested samples, indicating the similar chemical nature of starch and cellulose. Additionally, differential scanning calorimetry analysis demonstrated improved thermal stability in the packaging films when nanofibres from banana peel cellulose were added. Scanning electron microscopy results further confirmed the uniform distribution of particles within the matrix films, leading to smooth surfaces and well-spaced particles throughout the material, unlike the control samples. These findings suggest that these nano-composite packaging films hold promise for applications in the food packaging industry owing to their biocompatibility, biodegradability, and non-toxic characteristics.<sup>44</sup>

A study performed by Sharma and team aimed to extract starch from avocado seeds (ASS) and produce high-quality starch-cellulose bio-composites. The extraction process yielded 31.24% granules from the ASS, with thermal analysis showing a paste beginning temperature of 75.3 °C and a maximum viscosity of 706 BU at 88.1 °C. ASS demonstrated excellent stability at lower temperatures, with a low syneresis rate of 1.51%. To enhance its properties, cellulose fillers from *Ensete ventricosum* biomass waste were incorporated into the isolated avocado seed starch, resulting in a high-performing bio-



composite film (BCF). Crystallinity percentages were measured at 29.8% for raw ASS, 5.6% for the starch-only biofilm, and 12.3% for the bio-composite with 15% cellulose. The bio-composite with 15% cellulose showed significant improvements in tensile strength (18.0 MPa), Young's modulus (1782.4 MPa), and elongation at break (1.5%). The addition of cellulose greatly enhanced the mechanical properties by ensuring optimal filler dispersion.<sup>47</sup>

In another study, avocado seeds were processed to create a sustainable fire-retarding filler by washing, dehydrating, pulverizing, and then chemically modifying them with phosphoric acid and urea. Various analytical techniques were used to study the characteristics of this modified avocado seed, revealing significantly improved thermal stability. Cone calorimeter tests on polyethylene/ethylene-vinyl-acetate composites containing the modified avocado seed demonstrated a 54% reduction in the peak heat release rate and a 15% decrease in the total heat released, indicating enhanced flame-retardant effects. Importantly, the incorporation of avocado seed did not alter the melting temperature of the polymer blend as observed through differential scanning calorimetry (DSC). The modified avocado bio-composites also achieved a UL-94 V-1 classification in fire safety tests, indicating improved flame retardancy. Tensile tests showed similar tensile strength and modulus between composites with unmodified and modified avocado seeds but with slightly reduced elongation in the latter. Zuluaga-Parra *et al.* suggested that phosphorylated avocado seed could serve as a promising renewable biofiller for polymer composites with enhanced flame-retardant properties.<sup>45</sup>

## Shells and hulls

### Nutshell Waste

Biodegradable polymer composites reinforced with agri-food lignocellulosic biowaste have emerged as cost-effective and sustainable alternatives to conventional composites. In a study by Sarai and associates, the preparation of polylactic acid (PLA) biocomposites with high filler-loading (50 wt%) pecan (*Carya illinoensis*) nutshell (PNS) was investigated, along with the effects of two environmentally friendly physical treatments; ball-milling of the filler and thermal annealing. The incorporation of PNS improved the thermal stability, viscoelastic properties, and crystallinity of the PLA matrix. Moreover, ball-milling of the filler enhanced the melt fluidity of the bio-composites, potentially facilitating their melt processing. The presence of PNS significantly amplified the impact of thermal annealing, with the heat deflection temperature of the bio-composites increasing by up to 60 °C compared to non-annealed samples. These findings highlight the potential of integrating natural fillers with eco-friendly physical treatments to fine-tune the properties of PLA biocomposites, particularly for applications requiring materials that are stiff and lightweight, and exhibit low deformability.<sup>48</sup>

### Walnut shells

Harini and team focused on extracting and characterizing starch from cashew nut shells (CNSs) and cellulose from walnut

shells (WNS) to develop cellulose-reinforced starch films. Additionally, the extraction and characterization of pomegranate peel extract for incorporation with CNS-WNS films were investigated. The CNS starch was identified as moderate amylose starch with an amylose content of  $26.32 \pm 0.43\%$  and a thermal degradation temperature of 310 °C. Walnut shell cellulose exhibited a high crystallinity index of 72% and had thermal degradation temperatures of 319 °C and 461 °C. Films reinforced with 2% WN cellulose showed good oxygen transfer rates and mechanical and physical properties. The thermal degradation temperature of the CNS-WNS starch films ranged from 298 to 302 °C. The surface roughness of these films increased with higher cellulose concentrations. The major active compounds in the hydrophilic extracts of pomegranate peels were identified as hydroxymethylfurfural, benzene, 2-methoxy-1,3,4-trimethyl, and 1,2,3-propanetriol, 1-acetate. Starch films reinforced with 2% WN cellulose and infused with these hydrophilic active compounds from pomegranate peel demonstrated good active packaging properties.<sup>49</sup>

### Peanut hull

Oulidi and associates synthesized new biocomposites by incorporating peanut shell (PSP) from agricultural waste into polyamide (PA6) through *in situ* polymerization at varying concentrations (from 5% to 20%). Fourier-transform infrared spectroscopy (FTIR) analysis indicated strong reinforcement/matrix interactions in the resulting biocomposites. X-ray diffraction (XRD) studies of PA6, PSP, and the biocomposites revealed a uniform distribution of PSP within the PA6 matrix without significant changes in crystallinity. Differential scanning calorimetry (DSC) analysis confirmed the formation of biocomposites with crystals solely in the  $\alpha$  phase, indicating improved thermal and structural properties without the presence of the  $\gamma$  phase. The addition of 20% PSP notably impacted the thermal properties of PA6, as observed in the thermogravimetric analysis (TGA) measurements, showing a significant shift in the decomposition temperature of biocomposites with higher PSP concentrations. Lower concentrations (2% and 10% PSP) had less pronounced effects. The study underscores the potential of natural fibres as sustainable alternatives to inorganic fillers, offering opportunities to develop eco-friendly composites. This statement suggests that scholars are advocating for more research into using agricultural waste in the development of biocomposites. Their work yields promising initial results, indicating the potential of agricultural waste as a viable material in biocomposite applications. The recommendation implies that further investigation could lead to practical and sustainable uses, possibly advancing fields such as construction, packaging, and other industrial sectors where eco-friendly materials are in demand.<sup>50</sup>

### Coconut shells

The impact of filler content and chemical treatment on the biodegradation of poly(lactic acid) (PLA)/coconut shell (CS) biocomposites in a diastase enzyme-containing buffer medium was investigated by Tanjung and coworkers. The coconut shell (CS) underwent two chemical treatments: maleic acid treatment



and silanation using 3-aminopropyltriethoxysilane (3-APE). These treated CS materials were then incorporated into the PLA composites, and their biodegradation behavior was studied. Both types of chemically treated PLA/CS biocomposites exhibited slower biodegradation rates than the untreated biocomposites. This was attributed to the enhanced interfacial adhesion between the filler and the PLA matrix, leading to reduced exposure of the material to enzyme hydrolysis. Scanning electron microscopy images taken after 30 days of biodegradation showed surface roughening in the treated biocomposites, with fewer voids compared to the untreated ones. Differential scanning calorimetry results indicated an increase in glass transition temperature and melting temperature values for the treated biocomposites although crystallinity decreased. The peak corresponding to crystallization temperature disappeared likely due to polymer chain alignment and rearrangement caused by degradation, especially in shorter PLA chains. Fourier transform infrared analysis revealed structural changes in the bio-composites after biodegradation, suggesting the presence of soluble lactic acid, which was confirmed by ultraviolet-visible spectroscopy analysis.<sup>51</sup>

*Polyalthia longifolia* seeds are typically considered a byproduct or waste in some contexts, especially when they fall off the trees, as they are not widely utilized in mainstream industries. A recent study aimed to develop novel polymer composites by incorporating *Polyalthia longifolia* seed (PLS) particulates into epoxy matrices using the open layup molding technique, with varying weight percentages of PLS fillers (10, 20, 30, 40, and 50 wt%). Among the different composites, the 30 wt% PLS-filled epoxy composites showed the highest tensile strength of  $18.5 \pm 0.5$  MPa, compressive strength of 22.5 MPa, flexural strength of 22 MPa, and Shore D hardness of  $92 \pm 0.5$  SHN. The strength of this composite was further improved by adding a single layer of 400 GSM E-glass fiber on both sides, resulting in a maximum strength of  $26.5 \pm 0.5$  MPa. Fourier transform infrared spectroscopy (FTIR) analysis revealed the presence of various functional groups, while the surface morphology and elemental composition of the 30 wt% PLS-filled epoxy composites were examined using field emission scanning electron microscopy (FESEM) and energy dispersive X-ray spectroscopy (EDX). The average length of the fillers was determined to be  $2.25 \pm \mu\text{m}$  via FE-SEM.<sup>52</sup>

Jute-epoxy composites were fabricated with and without varying amounts of rosewood and padauk wood dust fillers using the conventional hand lay-up method. These composites were evaluated for mechanical, thermal, water absorption, and biodegradation properties. The results revealed that padauk wood dust-filled jute epoxy composites exhibited superior ultimate tensile strength (43 MPa) compared to those with rosewood dust (23.6 MPa). Additionally, padauk wood dust composites showed better performance in terms of flexural strength, compressive strength, hardness, and impact energy absorption, which was attributed to stronger adhesion with the matrix. Although the rosewood dust composites displayed higher thermal stability, the biodegradation rates were higher for unfilled jute epoxy composites than for those with wood dust fillers. However, the wood dust filler composites demonstrated improved water absorption resistance.

Overall, wood dust filler-based jute epoxy composites are suitable for lightweight and medium-load applications.<sup>53</sup>

## Agro-industrial waste

### coffee processing waste

Coffee, the second most consumed beverage globally and considered a commodity, generates spent coffee grounds as waste after brewing, accounting for around 90% of the original coffee beans. The environmental concern related to the disposal of waste from coffee fermentation was particularly due to the emission of compounds, like caffeine, tannins, and polyphenols. If these compounds are not properly managed, they can potentially harm ecosystems due to their toxicity. However, spent coffee grounds are rich in carbohydrates, lipids, proteins, and minerals.<sup>54</sup> Carbohydrates, specifically polysaccharides, can be extracted and fermented to produce lactic acid, succinic acid, or polyhydroxyalkanoate (PHA). Alternatively, coffee oil can be extracted from lipids and used to synthesize PHA. Additionally, spent coffee grounds have been successfully utilized as fillers in composite production with various polymer matrices. These composites exhibit reasonable mechanical, thermal, and rheological properties, making them suitable for applications in food packaging.<sup>55</sup> Incorporating coffee waste into polymers presents an opportunity to add value to a waste product while producing more environmentally friendly packaging materials compared to petroleum-based plastics, which are neither renewable-based nor biodegradable (Fig. 8). Researchers have noted the potential of both spent coffee grounds and coffee silver skin as fillers in composites and their potential use as nanomaterials for nanocomposite production.<sup>56</sup>

A study explored the potential of using chemically treated spent coffee grounds as micro biofillers to reinforce Poly-3-hydroxybutyrate-co-3-hydroxyvalerate (PHBV) biopolymer composites. The results revealed successful dispersion and even distribution of the microfiller within the composite.<sup>59</sup> Chemical treatment improved the bonding between the microfiller and the biopolymer, as indicated by the higher water contact angles observed in the composites. Field emission scanning electron microscopy confirmed the effective interaction of the treated spent coffee grounds microfiller, leading to enhanced mechanical properties of the composites. When buried in natural soil, the composites degraded faster compared to pure PHBV polymers, especially at higher levels of spent coffee grounds, indicating their biodegradability. Despite the potential for agglomeration at higher concentrations, the incorporation of these microfillers resulted in improved functional properties, making the green biopolymer composite a promising material for sustainable packaging and various applications.<sup>59</sup> For edible food packaging, researchers utilized natural red seaweed (*Kappaphycus alvarezii*) and coffee waste using the solvent casting technique. The coffee waste had an average particle size ranging from 1.106 to 1.281  $\mu\text{m}$ , with a zeta potential value of  $-27.0$  mV, indicating a strong negative charge. Analysis using scanning electron microscopy (SEM) showed that the coffee filler was evenly distributed in the polymer matrix, enhancing the film's structural properties.





Fig. 8 Various coffee processing byproducts in the production of fillers [image created by the first author, Zeba Tabassum, utilizing data from ref. 55–58].

Fourier-transform infrared spectroscopy results confirmed the successful integration of coffee waste into the film matrix, as evidenced by the presence of an N–H bond. Incorporating coffee filler also improved the film's hydrophobicity, as indicated by an increased water contact angle compared to the neat film. The tensile properties of the biopolymer film were notably enhanced with the addition of 4 wt% coffee powder, achieving an optimum tensile strength of 35.47 MPa. The inclusion of coffee waste in the seaweed matrix resulted in improved functional properties of the fabricated biopolymer film. Therefore, the researchers concluded that the seaweed/coffee biopolymer film has the potential for use in food packaging and various other applications.<sup>60</sup>

Spent coffee grounds were incorporated at different concentrations (1%, 3%, 5%, and 7%) into the biocomposites containing poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and chemically modified kenaf bast fibers with the help of sodium formate as a catalyst. Three variations of PHBV-reinforced bast fiber mats incorporated SCG were prepared: unmodified mats, mats chemically modified with propionic anhydride, and mats modified with propionic anhydride in the presence of a catalyst. These biocomposites were evaluated for physical, thermal, functional, water absorption, thickness swelling, water contact angle, and mechanical properties. Catalyzed propionylated bast fibre-based biocomposites showed improved functional properties, enhanced thermal stability, and superior mechanical performance than propionylated and unmodified biocomposites. Lower concentrations (1% and 3%) of spent coffee ground filler resulted in better properties, making them suitable for sustainable packaging applications.<sup>61</sup>

Spent coffee grounds were also utilized as a reinforcing biowaste filler in thermoplastic starch/alginate films, with wet ball milling employed to reduce the filler particle size. The composite films, containing dark brown spent coffee grounds particles, exhibited a UV transmission barrier 1.5–3.4 times higher than that of the binary matrices. Incorporating fillers created voids within the polymeric chains, resulting in a 2.10% reduction in moisture content and a decrease in water vapor

permeability from  $(10.50 \pm 1.07) \times 10^{-13}$  to  $(9.03 \pm 1.69) \times 10^{-13} \text{ ng s}^{-1} \text{ m}^{-2} \text{ Pa}^{-1}$ . A slight enhancement in thermal stability was noted due to the partial substitution of thermoplastic starch with spent coffee grounds. Despite the stress concentration and plasticizing effects of the fillers, the TPS-Alg/SCG-10 composite exhibited notable tensile strength ( $67.6 \pm 6.1 \text{ MPa}$ ) and tensile modulus ( $4.7 \pm 0.3 \text{ GPa}$ ). Overall, these findings highlighted the significant potential of the developed material for use in food packaging applications.<sup>62</sup>

Zdanowicz and team plasticized corn starch using two selected urea-rich plasticizers, choline chloride or betaine eutectic mixtures, at a molar ratio of 1:5, with the inclusion of spent coffee grounds as fillers. These biomaterials were prepared using a solventless one-step extrusion method and then thermoformed into sheets using compression molding. For the first time, the impact of these materials on the growth of plants in soil was investigated. The addition of coffee filler slightly improved the mechanical properties and reduced the swelling degree of the materials. Dynamic mechanical thermal analysis results showed that the biocomposites were easily thermoformable, and even with a high filler addition, the processability was not affected. Biodegradation tests revealed that the materials completely degraded in soil within 70 days.<sup>63</sup>

As mentioned earlier, polylactic acid (PLA) is a promising biodegradable polymer, albeit with cost challenges. To address this issue, ongoing research focuses on developing bioplastic composites by blending PLA with spent coffee grounds and thermoplastic starch, aiming to reduce costs. Different percentages of spent coffee ground dust (5%, 10%, and 15% by weight) were studied in these bioplastics. The physical and mechanical properties of the bioplastic composite were found to decrease due to the increased aggregation caused by the presence of spent coffee dust. However, the bioplastics showed improved crystallinity, leading to enhanced thermal properties. The most favorable characteristics were observed with a 5% spent coffee ground addition, a water vapor transmission rate of  $1276 \text{ g d m}^{-2}$ , water



vapor permeability of  $1.86256 \times 10^{-7} \text{ g ms}^{-1} \text{ Pa}^{-1}$ , Young's modulus of 420 MPa, elongation of 2.59%, and tensile strength of 5 MPa. Based on these findings, it was concluded by the researchers that the addition of spent coffee grounds is not recommended for enhancing the physical and mechanical properties of bioplastics. However, due to its high content of organic compounds, it remains a promising and cost-effective material that can be utilized in creating various value-added products.<sup>64</sup>

Coffee pulp is obtained as a major byproduct during coffee bean production and contains more than 30% dry weight among all the coffee waste. In an investigation by Malarat and groups, the coffee pulp (CP) was subjected to alkali and bleaching treatments before nanocellulose coffee pulp (NCP) production *via* acid hydrolysis. Transmission electron microscopy (TEM) revealed that the NCP had an average diameter of  $16.03 \pm 4.70 \text{ nm}$ , with an increased crystallinity size and crystallinity index. Glycerol (G) was incorporated into the PVA matrix to act as a plasticizer, resulting in increased flexibility and reduced hardness and brittleness of the PVA nanocomposite films. The PVA/G/NCP nanocomposite films were fabricated by applying the casting method with varying NCP ratios. The inclusion of NCP significantly improved the physical properties of the PVA matrix compared to neat PVA films.<sup>65</sup>

### Industrial byproducts

Researchers have focused on alternative bio-based polymers to replace petroleum-based packaging due to environmental issues, such as non-biodegradable waste and fossil fuel depletion. Sugarcane bagasse, a by-product of the sugarcane industry, is being considered for food packaging to promote industrial symbiosis. Lignocellulosic biomass, with cellulose, hemicellulose, and lignin, is valued owing to its ecological and biodegradable qualities.<sup>12</sup>

Natural frankincense is made from the *Boswellia serrata* tree and has many advantageous qualities that make it useful for use in packaging. Nanocellulose generated from sugarcane bagasse was reinforced in it. FTIR, XRD, and FESEM were used to characterize the biocomposite thin films. Reshmi and colleagues concluded from their study that the antibacterial and antifungal qualities of suggested novel biodegradable plastics with long shelf lives might be created for use in food packaging.<sup>66</sup>

Another study focused on using biochar derived from the oxygen-deficient thermochemical processing of organic wastes as a reinforcing agent in biocomposite films. These films were made by combining sugarcane bagasse pyrolyzed biochar with polyvinyl alcohol (PVA), and their electrical and mechanical properties were evaluated. Different amounts (5 wt%, 8 wt%, and 12 wt%) of biochar produced at pyrolyzing temperatures of 400 °C, 600 °C, 800 °C, and 1000 °C were used to create the bio-composite films. Characterization techniques such as X-ray diffraction, scanning electron microscopy, and Fourier transform infrared spectroscopy were employed. The results showed that higher pyrolyzing temperatures led to a significant enhancement in the electrical conductance of the bio-composite films. The highest electrical conductance of  $7.67 \times 10^{-2} \text{ S}$  was observed with a 12 wt% addition of biochar

produced at 1000 °C. The study indicated a trend where increasing the biochar content improved the electrical properties until a certain threshold, creating a continuous conductive network across the films. However, a notable reduction in tensile strength was observed with higher biochar dosages. The film with 12 wt% biochar produced at 800 °C exhibited the lowest tensile strength of 3.12 MPa. The pyrolyzing temperature had a minor effect on the mechanical strength of the bio-composite films.<sup>67</sup>

Sugarcane bagasse fibre cellulose nanocrystals (SBFCNCs) and microcrystalline cellulose-derived-cellulose nanocrystals (MCC-CNCs) were extracted from sugarcane bagasse fibre and microcrystalline cellulose (MCC), respectively. Both SBFCNC and MCC-CNC were produced using sulfuric acid hydrolysis, followed by freeze-drying, resulting in stable water suspensions with zeta potential values of approximately  $-40.5 \text{ mV}$  for MCC-CNC and  $-42.2 \text{ mV}$  for SBFCNC. Transmission electron microscopy analysis revealed that SBFCNC has a higher aspect ratio (length-to-diameter ratio) of 65 compared to MCC-CNC's aspect ratio of 25. Poly(lactic acid) (PLA) nanocomposites containing MCC-CNC and SBFCNC were prepared using the solvent casting method, producing highly amorphous films as confirmed by differential scanning calorimetry (DSC). The tensile strength of PLA/SBFCNC-10 films exceeded that of PLA/MCC-CNC-10 films. Thermogravimetric analysis (TGA) indicated significantly improved thermal stability of PLA when incorporating MCC-CNC and SBFCNC. In particular, poly(lactic acid)/SBFCNC-15 nanocomposites exhibited enhanced UV shielding properties with a UV blocking ratio ranging from 0.63 to 0.66 across the UVA, UVB, and UVC regions, compared to PLA/MCC-CNC-15 nanocomposites with a UV blocking ratio of 0.38–0.54. Sugarcane bagasse fibre cellulose nanocrystals demonstrate potential as a biofiller, offering improved thermal stability and UV shielding capabilities for environmentally friendly bionanocomposites.<sup>68</sup>

PVA/starch nanocomposite film reinforced with cellulose nanofibre (CNF) derived from sugarcane bagasse was also developed as an alternative to existing biodegradable plastic packaging materials. Polyvinyl alcohol (PVA) was chosen due to its ability to degrade and its excellent film-forming properties among synthetic polymers. The nanocomposite film was prepared using the solution casting method. CNF was extracted from sugarcane bagasse (SCB) through alkaline and mild acid treatment with ultrasonication assistance (reference). Different loadings of CNF (ranging from 1 to 6 wt%) were incorporated into the PVA/starch film to determine the optimal loading for mechanical, thermal, and antibacterial properties. The results showed that the thermal properties, tensile strength, and elongation at the break of the PVA/starch/cellulose nanofibre film improved significantly at a CNF loading of 4 wt%. The reinforcement of CNF led to a substantial enhancement in tensile strength, with a value of 85 MPa representing a 254% improvement. Furthermore, the composite film PVA/starch/CNF, when incorporated with lemon-grass essential oil, exhibited antibacterial properties, inhibiting the growth of Gram-positive bacteria, such as *S. aureus*. This study marked an initial step towards the development of active food packaging with improved biodegradable film properties.<sup>69</sup>



## Fishery waste

### Chitosan (from shrimp shells)

Value-added bionanocomposite films were developed by Salaberria and fellow researchers using oceanic biomass, specifically chitin nanofillers (nanocrystals-CHNC and nanofibres-CHNF) along with chitosan (CS) derived from *Cervimunida johni* (yellow lobster) waste. Two sets of bionanocomposite films (CS/CHNC and CS/CHNF) were produced by incorporating CHNC or CHNF into a CS matrix using the solvent evaporation-casting method.<sup>70</sup> The films were analysed for their chemical composition, structure, mechanical and thermal properties, as well as their ability to inhibit fungal growth (specifically against *A. niger*). This study evaluated and compared how CHNC and CHNF influenced the final properties of CS-based bionanocomposite films. The results indicated that the properties of the bionanocomposite films were influenced by the content and nano-characteristics (size and shape) of the chitin nanofillers embedded in the CS matrix. CS-based films reinforced with CHNF exhibited superior mechanical properties compared to those reinforced with CHNC. Additionally, antifungal testing revealed that CS/CHNF bionanocomposite films displayed a significant inhibitory effect (FGI > 80%) against *A. niger*. These findings emphasize the potential of PHBV-reinforced bast fiber mats incorporating spent coffee grounds (SCG) as promising bionanocomposite films for packaging applications.<sup>70</sup>

## Poultry waste

### Egg shell

In recent years, rising incomes and increased dietary acceptance have led to higher egg consumption, particularly in developing countries, where eggs are recognized as a high-quality protein source, influencing consumer purchasing decisions. Over the past 30 years, egg production has surged by more than 150%, with Asia seeing the most significant growth, as production has increased fourfold. However, the Environmental Protection Agency has ranked eggshell waste as the 15th most significant pollution problem in the food industry. When not properly disposed of in designated locations, eggshell waste becomes a major environmental pollutant and poses health risks.<sup>71</sup>

Nowadays, recovering eggshell powder from food waste and using it as a bio-filler for a biopolymer matrix to create sustainable composites is becoming an appealing recycling technique.<sup>72</sup> Li *et al.* explored a sustainable recycling technique using recovered eggshell powder from food waste as a bio-filler in creating biodegradable composite films. Thermal extrusion and compression molding were used to produce films based on potato starch/gelatin, incorporating varying percentages (0–50%, w/w) of eggshell powder. FTIR analysis confirmed hydrogen bonding interactions between starch/gelatin and the organic compounds on the eggshell filler surface, while SEM imaging revealed a uniform distribution and strong adhesion of eggshell particles within the film matrix. The optimal filler content was determined to be around 30% w/w, significantly enhancing tensile strength, oxygen barrier, water vapor resistance, and water resistance compared to the control films. The composite films exhibited

a tensile strength of 4.54 MPa, elongation at break of 27.23%, water vapor permeability of  $4.16 \times 10^{-10} \text{ g m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ , and oxygen permeability of  $8.15 \times 10^{-15} \text{ kg m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ . Remarkably, these films completely degraded in less than a month, showcasing their strong potential for biodegradable packaging solutions due to their favorable biodegradability and excellent physicochemical properties.<sup>72</sup>

Sharif *et al.* fabricated bioplastic sheets using the solvent casting method, which were made of yam starch (Dorceasea) and reinforced with different amounts of eggshell. Static contact angle analysis and Fourier transform infrared spectroscopy confirmed that the intensity and hydrophilicity of films drastically decreased as eggshell concentrations increased due to the covalent interaction of carbonate with the glycoside group.<sup>73</sup> Furthermore, the film tensile strength increased with the addition of additional filler, or eggshell. It was discovered that the filler-reinforced films were progressively disintegrating in the soil, with 3% eggshell reaching approximately 56% after 21 days. Additionally, they demonstrated a lower rate of transmission of water vapour, which is required for bioplastics to have a long shelf life.<sup>73</sup>

Biodegradable and recyclable thermoplastics, such as poly(lactic acid) (PLA), are derived from sustainable sources such as sugarcane, potatoes, and corn flour. However, PLA tends to have low impact strength and brittleness. To improve its mechanical properties, calcium carbonate (CaCO<sub>3</sub>) fillers were added to PLA.<sup>74</sup> Injection molding was used to incorporate white chicken eggshell and typical limestone powders with particle sizes of 63 μm and 32 μm at 5%, 10%, and 20% concentrations into PLA composites. Tensile strength initially decreased with higher filler content but peaked at 5% loading, with 32 μm powder fillers showing higher strengths than 63 μm fillers. The tensile modulus increased with filler content, reaching a peak at 20% for both particle sizes. Limestone powder-based composites exhibited superior toughness compared to eggshell-based ones. However, the water absorption of PLA/CaCO<sub>3</sub> composites was higher than that of pure PLA.<sup>74</sup>

### Chicken feather

Dutta and his team conducted a review that focused on the progress and efficiency of various biopolymer composites reinforced with fillers derived from chicken feathers. The significant annual production of waste chicken feathers globally presents environmental challenges, with landfilling and incineration being the predominant disposal methods, both of which entail environmental risks (Dutta *et al.*, 2024). New biodegradable polymeric materials with high loadings of chicken feathers (CFs) were developed by a team of scientists. The effects of CF concentration and the type of biodegradable matrix on the physical, mechanical, and thermal properties of the biocomposites were evaluated. The selected biopolymers included polylactic acid (PLA), polybutyrate adipate terephthalate (PBAT), and a PLA/thermoplastic copolyester blend. The biocomposites were produced using a torque rheometer, incorporating CF at concentrations of 50% and 60% by weight. Due to the low tensile strength of CF, the mechanical properties of the resulting materials were compromised. However, the high-loading CF







Fig. 9 Poultry and sea food industry generated fillers [image created by the first author, Zeba Tabassum, using data from ref. 1].

compatibility within the composite. Cell tests indicated non-cytotoxicity with varying OSP/PLF content, and the presence of OSP notably enhanced the composite's antimicrobial properties. Comparing two formulations, one with polyhydroxyalkanoate (PHA), pineapple leaf fibre, and waste oyster shell powder, and the other with MPHA, OSP, and PLF, the latter exhibited higher water absorption capacity. However, both composites demonstrated biodegradability, particularly with increased levels of oyster shell powder and pineapple leaf fibre.<sup>81</sup>

A different research team explored the potential of utilizing waste from mangosteen and durian peels as natural pigments and bio-fillers in polybutyrate adipate terephthalate (PBAT) biopolymer. They varied the weight percentages (ranging from 0 to 30 wt%) of mangosteen powder and durian powder and both cellulose-based nanofillers were added to PBAT to study their effects on the colour, structure, chemical composition, strength, heat resistance, and flow properties. The inclusion of mangosteen and durian peels naturally darkened the green composites without the need for prior burning, offering a sustainable method to repurpose agricultural by-products while adding value to them.<sup>82</sup> The resulting black biopolymer composites were found to be suitable for environmentally friendly food packaging and medical zipper applications. Comparing the two, mangosteen/PBAT composites exhibited better mechanical strength, thermal stability, and coloration, while durian/PBAT composites showed superior thermal and flow properties.<sup>82</sup>

Shaik and colleagues synthesized bioplastics comprising powdered walnut shells and eggshells in different weight proportions and polylactic acid (PLA). To obtain plasticization, five weight percent of epoxidized soybean oil is employed. Following a thorough analysis, the researchers concluded that the properties of plasticized PLA–egg shell composites outperformed those of plasticized PLA–walnut shell composite.<sup>83</sup>

A research project was conducted to explore the feasibility of using rice and corn starch to produce bioplastics, along with studying the impacts of incorporating natural agricultural

wastes, such as rice hulls and eggshells as fillers. Bioplastic samples were developed with varying ratios of starches, plasticizers, and fillers to assess their viability. The results indicated that the physicochemical properties of the bioplastics could be significantly influenced by the choice of plasticizers, types of starches, and the types and amounts of fillers used.<sup>2</sup>

In a different study, the effects of brown eggshells vs ordinary limestone powder on the mechanical properties of PLA composites were examined by a group of scientists. The maximal ultimate tensile and ultimate flexural strengths for eggshell composites with 32  $\mu\text{m}$  fillers were 48 MPa (5–10 wt%) and 67 MPa (10 wt%), respectively. The tensile and flexural moduli increased with the addition of filler and reached their maximums of 3.4 GPa and 4.5 GPa at 20 weight percent, respectively. The study demonstrated that powdered brown eggshell waste can be effectively used as a filler in polylactic acid (PLA) composites.<sup>85</sup>

Sivakumar and his co-workers demonstrated by mixing different proportions of eggshell powder and walnut shell powder with plasticized polylactic acid (PLA), where plasticization is achieved using 5 wt% of epoxidized soyabean oil. The resulting bioplastic granules are then processed *via* injection molding to produce dog bone-shaped samples that undergo mechanical, thermal, and optical microscopy tests. Mechanical assessments, including tensile, charpy impact, and flexural tests, indicate reduced properties compared to pure PLA when agricultural waste fillers are added. However, it is noteworthy that the plasticized PLA–epoxidized soybean oil composite performs better than the plasticized PLA–walnut shell composite in terms of mechanical properties.<sup>86</sup>

This study aimed to explore the use of corn starch and ground walnut shells as biofillers in natural rubber (NR) biocomposites, focusing on enhancing their performance through chemical modifications. This ionic liquid was used to treat the biofillers, possibly improving their dispersion within the NR matrix and enhancing their interaction with the rubber.<sup>87</sup> BmiCl can modify



Table 3 Influence of multiple fillers on the biopolymer matrix

S. no.	Filler 1	Filler 2	Matrix	Characteristics	Ref.
1	Pineapple leaf fibre	Waste oyster shell powder	Polyhydroxyalkanoate	Enhanced antimicrobial properties, non-cytotoxicity, and biodegradability	81
2	Mangosteen peel	Durian peels	Polybutyrate adipate terephthalate	Mangosteen-based composites exhibited better mechanical strength, thermal stability, and coloration, while durian/PBAT composites showed superior thermal and flow properties	82
3	Walnut shell powder	Eggshell powder	Polylactic acid	Plasticized PLA-egg shell composite resulted better than plasticized PLA-walnut shell composite	83
4	Rice hulls	Eggshells	Starch from corn and rice	Fillers influenced the physiochemical properties of the biodegradable plastic	84
5	Limestone powder	Brown egg-shell powder	Polylactic acid	The tensile and flexural moduli rose with the addition of eggshell filler and reached their maximums of 3.4 GPa and 4.5 GPa at 20 weight percent, respectively	85
6	Walnut shell powder	Eggshell powder	Polylactic acid	Mechanical assessments, including tensile, Charpy impact, and flexural tests, indicated reduced properties compared to pure biopolymer	86
7	Walnut shells	Corn starch	Natural rubber	Bio-composites containing starch demonstrated enhanced tensile properties. Ground walnut shells exhibited improved resistance to thermo-oxidative aging, attributed to the presence of lignin in walnut shells, which contain polyphenols with antioxidant properties	87
8	Potato peel powder	Wood dust powder	Banana peel starch and a composite of banana peel starch, cornstarch, and rice starch	The addition of powdered potato peels and wood dust as fillers resulted in increased water absorption capacity. Moreover, improved tensile strength and Young's modulus, with a proportional enhancement observed with increasing filler content	2
9	Sewage sludge	Biochar from wood	Polylactic acid	The biochar-enhanced bio-composites showed enhanced rigidity and water absorption capacity. Particularly, sewage sludge-derived biochar significantly improved the mechanical and thermal properties of the biodegradable polymer-based composites	88

the surface characteristics of the fillers, increasing their compatibility with NR and leading to improved mechanical properties and curing efficiency. A 3-aminopropyl-triethoxysilane (APTES) coupling agent was applied to further enhance the adhesion between the biofillers and the NR matrix. The incorporation of these biofillers, along with BmiCl and APTES, is expected to improve the curing characteristics, such as vulcanization rates, and optimize the mechanical properties, crosslink density, mechanical performance under static and dynamic conditions, hardness, thermal stability, and resistance to thermo-oxidative aging of the resulting NR biocomposites, making them suitable for various applications while maintaining sustainability.<sup>87</sup>

The research delved into the impact of these biofillers, both in their original form and when modified with aminosilane or ionic liquid, on the curing characteristics and functional properties of NR composites. The study identified starch and ground walnut shells as inert fillers that could serve as alternatives to commercial inert fillers, such as chalk. BmiCl and APTES proved effective in aiding vulcanization and enhancing the dispersion of biofillers within the NR elastomer matrix. Bio-composites containing starch, particularly those with APTES and BmiCl, demonstrated enhanced tensile properties due to increased crosslink density and uniform dispersion of starch facilitated by BmiCl. NR filled with ground walnut shells exhibited improved resistance to thermo-oxidative aging, attributed to the presence of lignin in walnut shells, which contain polyphenols with antioxidant properties.<sup>87</sup>

Bioplastic samples were produced using banana peel starch (BPP) and a composite of banana peel starch, cornstarch, and rice starch (COM), incorporating varying amounts of potato peel powder and wood dust powder as fillers. Two different plasticizers, glycerol and sorbitol, were used individually and in a 1 : 1 combination. A total of 12 samples of each bioplastic type

were created with different filler and plasticizer combinations to assess their physical and chemical characteristics, including moisture content, water absorption, and solubility in water and alcohol, biodegradation in soil, tensile strength, Young's modulus, and FT-IR analysis. The addition of powdered potato peels and wood dust as fillers resulted in an increased water absorption capacity in the bioplastic samples compared to those without fillers (except for the control). Moreover, the inclusion of fillers led to improved tensile strength and Young's modulus in both types of bioplastics, with a proportional enhancement observed with increasing filler content. The distinct properties observed in these bioplastic samples make them suitable for various applications. Notably, Shafqat and coworkers claimed that all 24 samples were developed using natural and environmentally safe materials, and they exhibited biodegradability, highlighting their potential as a sustainable alternative to conventional plastics.<sup>2</sup>

Biochar from wood and sewage sludge was added as fillers in poly(lactic acid) and BIOPLAST GS2189 bio-composites, with concentrations of up to 20% by weight. The study revealed that the inclusion of biochar affected various properties, such as mechanical strength, thermal behaviour, and optical attributes. The biochar-enhanced bio-composites showed enhanced rigidity and water absorption capacity. Particularly, sewage sludge-derived biochar significantly improved the mechanical and thermal properties of the biodegradable polymer-based composites.<sup>88</sup>

## Conclusion

Agricultural and industrial waste generation causes significant environmental issues, including pollution, greenhouse gas emissions, and landfill overflow, leading to ecosystem



degradation and health risks. Waste valorization addresses these challenges by transforming waste into valuable resources, reducing pollution, and conserving natural resources. Utilizing waste-derived fillers in bioplastics promotes sustainability and offers eco-friendly and cost-effective solutions aligned with the principles of circular economy. The demand for renewable bioplastics is driven by the environmental harm caused by conventional plastics, which contribute to non-degradable waste and deplete fossil fuels. Waste-based fillers enhance bioplastic mechanical, thermal, and barrier properties, supporting their functional and sustainable use in packaging. Although commercially available nanofillers, such as metal oxides, enhance bioplastics, their toxicity concerns have driven research into safer, waste-derived alternatives. These fillers demonstrate comparable or superior efficacy, reduce reliance on harmful materials, and advance sustainable packaging innovations. Continued research is vital for optimizing this emerging field.

From an extensive literature review, several key parameters affecting the properties and performance of bioplastic composites incorporating waste-derived fillers were noted. The color and odor of the resulting bioplastics were notably influenced by the presence of lignin and hemicellulose components. In many cases, modification or pretreatment of these fillers is necessary to achieve optimal results. For instance, although the initial addition of fibres tended to reduce the tensile strength, the use of bleached fibres significantly enhanced the composite's mechanical performance. Chemical treatments were also found to improve the bonding between the microfillers and the biopolymer matrix, leading to better overall material properties. Biodegradability was another important aspect, with composites degrading faster than pure polymers, especially at higher levels of spent coffee grounds, indicating enhanced biodegradability. However, lower concentrations of fillers, such as spent coffee grounds, resulted in better biocomposite properties, as higher filler concentrations often led to increased aggregation, negatively impacting the physical and mechanical properties of the bioplastic composite. This highlights the trade-off between enhanced biodegradability and optimal physical properties based on the filler concentration. In summary, the incorporation of bio-based and waste-derived fillers into bioplastics offers significant potential for improving the material performance while enhancing biodegradability. However, careful consideration of filler types, concentrations, and pretreatment methods is essential for optimizing the properties and functionality of the resulting bioplastic composites. Furthermore, effective waste management, storage, preservation, and treatment before processing are essential hurdles that must be overcome to advance both research and industrial applications. In addition to optimizing filler parameters, addressing the challenges related to waste handling and preparation is equally critical. Legislative support is urgently required to facilitate the widespread utilization of waste and to promote sustainability and environmental benefits. Governmental bodies and relevant authorities must advocate innovative packaging strategies and develop cost-effective composite materials with enhanced properties for large-scale economically viable

production. Additionally, optimizing production processes and addressing safety concerns related to substance migration are paramount to ensure the success and acceptance of bioplastic packaging. In conclusion, although the potential for waste valorization in bioplastic production is immense, achieving this potential necessitates concerted efforts in research, legislative action, and industrial innovation. By overcoming existing challenges, we can pave the way for a sustainable future using eco-friendly packaging solutions.

## Author contributions

Zeba Tabassum – conceptualization, investigation, writing – original draft, review and editing; Madhuri Girdhar – supervision, review and editing; Abhinav Anand – review; Bhawana Sood – review; Tabarak Malik – resource and review; Anil Kumar – resource and review; Anand Mohan – supervision, review and editing.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Conflicts of interest

The authors declare no competing financial interest.

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