

Multifunctional starch-based barrier materials

PRADNYA RAO, CHINMAY SATAM, ASHOK GHOSH, AND PETER W. HART

ABSTRACT: Natural and renewable polymer-based barrier materials play an inevitable role in a sustainable economy. Most commercially available barrier materials are either based on multiple layers of synthetic polymers or petroleum-based chemicals. Tremendous amounts of research are being done in academia and industry to replace these synthetic barrier materials with natural and environmentally friendly materials.

The current work summarizes the application of starch-based materials for various barrier applications, such as water vapor, oxygen, liquid water, oil, and grease. Also, exotic starch-based barrier materials for the application of sound, ultraviolet, and thermal barrier applications are reviewed. The potential of starch-based materials to offer antimicrobial and antiviral properties is discussed. Finally, commercially available starch-based barrier materials have been summarized.

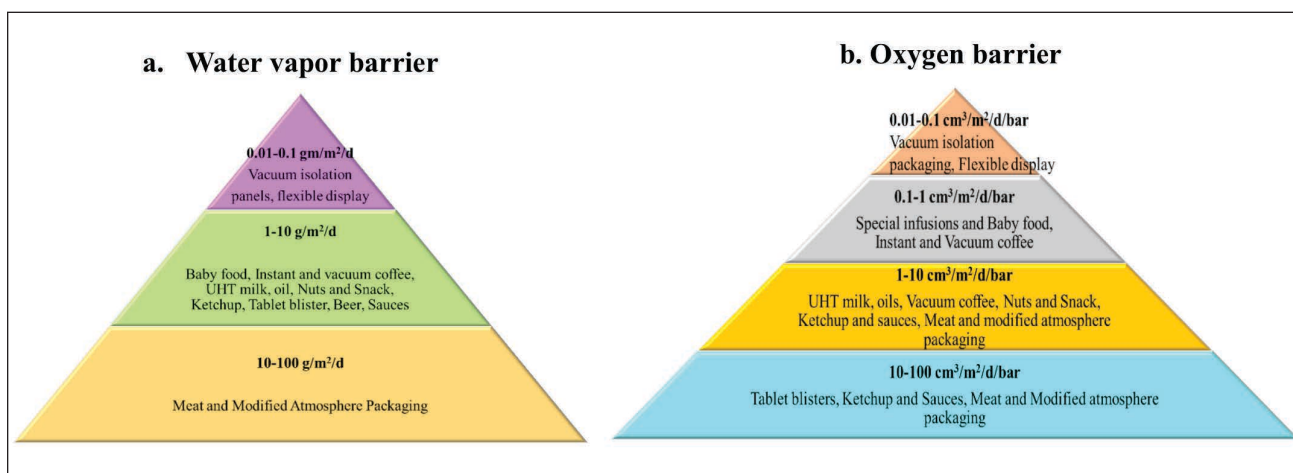
Application: This paper highlights ways for industry to explore new ways of using starches for traditional and non-traditional barrier applications

Synthetic materials, such as polystyrene, polyurethane, high-density polyethylene, and many other petroleum-based polymers, are being used in various barrier applications for packaging, coatings, and food-service. The requirements and performance of barrier materials depend on the end use application. **Figure 1** displays a hierarchical summary of various applications and their barrier requirements for water vapor and oxygen [1].

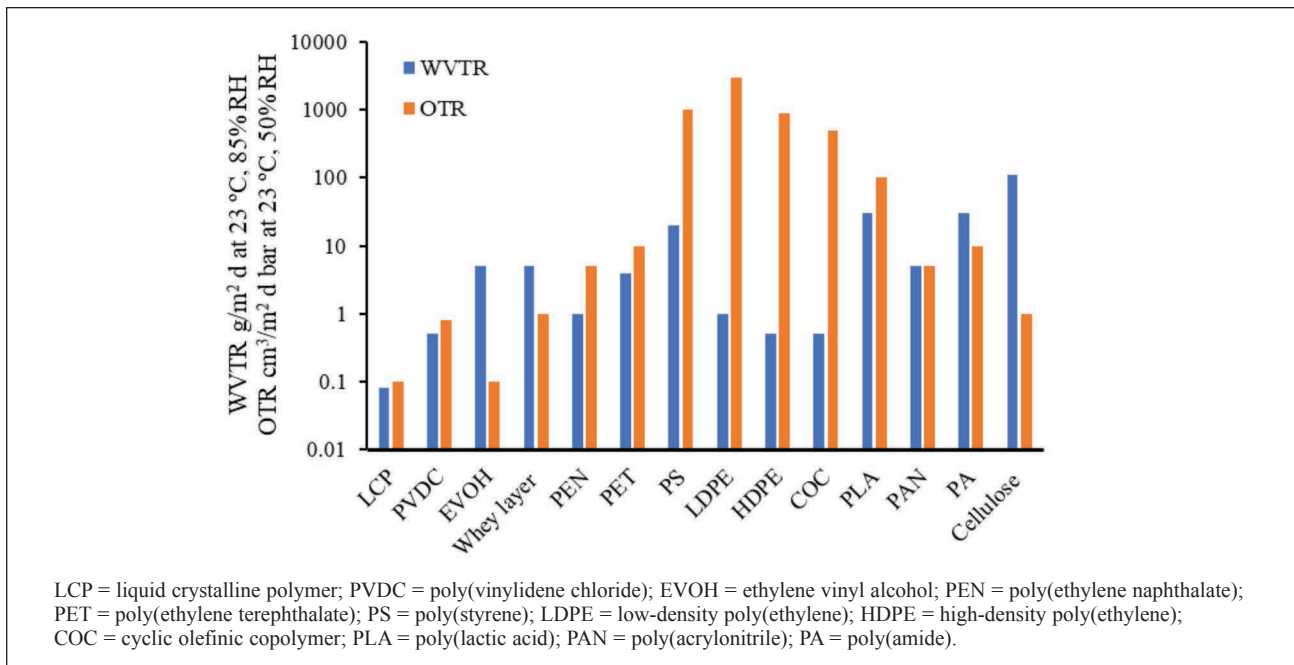
Barrier properties are an important characteristic of any packaging application, along with other properties like good mechanical strength and optical properties. Good barrier properties are required to ensure a long product shelf life. Barrier properties of materials may be defined as a reduction in the transmittance of gases, such as oxygen, carbon dioxide, and water vapor, as well as fluids. Barriers may even imply reduction in light, sound, or reduction in

proliferation of pathogenic microbes. Barrier properties may also be used to protect products from heat, light, microbes, sound, etc. Permeating matter undergoes three physical phenomena, referred to as adsorption, diffusion, and desorption [2-4]. Barriers interfere with some or all these phenomena. **Figure 2** summarizes the water vapor and oxygen barrier potentials of various polymeric materials [1]. It is rare to have all desired barrier properties in one single material while maintaining a cost competitive position. In addition, it is also extremely difficult to achieve these barriers using only bio-sourced polymers. Thus, most commercially available packaging barriers are based on multiple layers of synthetic polymers.

Starches, the main storage carbohydrates for plants, naturally occur in fruits, legumes, tubers, cereals, and seed. Commercially used starches are extracted from maize, po-



1. Water vapor (a) and oxygen (b) barrier requirements for various products. Adapted from Schmid et al. [1]



2. Barrier potential of various polymeric materials (WVTR = water vapor transmission rate; OTR = oxygen transmission rate; RH = relative humidity). Data from Schmid et al. [1].

tato, wheat, rice, and cassava. Chemically, starches are homopolymers of glucose and contain two main components: amylose and amylopectin [5]. Amylose is highly linear and exhibits only slight branching with glucose units joined by α -(1–4) bonds. Amylopectin is a branched starch component consisting of glucose units joined by α -(1–4) bonds in the lineal sections, and by α -(1–6) bonds in the branching points; it is responsible for the starch crystalline lamella, although its branching points are actually part of the amorphous section. The presence of both amorphous and crystalline regions in starch makes it semicrystalline [5]. Depending on the biological origin, starchy materials contain various proportions of amylose and amylopectin, giving them distinct properties that are source dependent, as well as season dependent [5]. Commonly used starches include waxy starches that contain 100% amylopectin and the high-amylose starches with approximately 53% to 70% amylose [6]. These polysaccharides, in their gelatinized form, have been applied in various ways to achieve select barrier properties [5,7,8]. Starches are hydrophilic and the resultant barrier properties tend to degrade at higher moisture levels. Approaches to reduce moisture dependency of starch-based barrier formulations can be broadly divided into three categories: (1) physical blends with other materials like bioplastics, pigments, etc. [9]; (2) chemical modifications [10]; and (3) combinations of physical and chemical modification [11,12]. Overall, the relative proportions of branched and linear polysaccharides from various starch sources harvested at different times of the year impact the final packing of molecules in barrier films, affecting barrier properties. In addition, chemical modification can fur-

ther influence the hydrophilicity of starches, affecting packing of molecules and transport of substances through the film matrix, which leads to a variety of film-forming starches that exhibit barrier properties that may find applications in specialized applications. Plasticizers aid in processing of starches by interrupting the hydrogen bonding of materials. For instance, starches are not inherently thermoplastic but may become so on addition of the appropriate plasticizer and water [13,14].

The focus of this paper is to review starch and starch-based materials, such as films, coatings, foams, nanocrystals, etc., for some commonly encountered barrier applications like resistance to liquids, water, oil, and grease. More exotic applications where starch-based materials have been used as thermal, ultraviolet (UV), sound proofing, and antimicrobial barriers are also discussed. While barrier properties achievable through starch-based materials are discussed, properties of native starches themselves are not covered. Variability of starches according to sources and season is not reviewed. Additionally, where possible, information on barrier testing is provided for each kind of barrier. It is left to the reader to explore the vast breadth of information on barrier testing methodologies in the source references or elsewhere.

TYPES OF BARRIER MATERIALS

Water vapor barrier

Water vapor barrier is an important barrier property for packaging materials to maintain product quality. It is usually measured as a function of the permeability of water vapor through the material. Native starches such as corn,

Starch-based Materials	WVTR, g-mm/m ² /d	RH, %	Temperature, °C	Form	Reference
Hydroxypropylated and oxidized potato starch, citric acid	1.0*	50	23	Coating	[16]
Hydroxypropylated, starch, citric acid	1.1-2.9*	50	23	Coatings	[17]
Starch, chitosan, glutaraldehyde crosslinked	9.3-16.4	50	25	Film	[18]
Oxidized corn starch, bentonite	8.9	65	22	Film	[19]
Potato starch, sodium hypochlorite	10.4	75	25	Film	[18]
Potato starch, nano SiO ₂	70	90	25	Film	[20]
PBS, starch, and Cloisite 30B clay	8.8	90	38	Pellet Composite	[21]
Starch and acrylated epoxidized soybean oil	14.1	90	38	Films	[22]
Crosslinked wheat starch, sodium montmorillonite, TiO ₂	105	97	23	Film	[23]
* Water vapor transmission rate (WVTR) value normalized with respect to coating thickness instead of film thickness. PBS = poly(butylene succinate); SiO ₂ = silicon dioxide; TiO ₂ = titanium dioxide.					

I. Water vapor barrier properties of starch-based materials.

potato, pea, oat, and other types of starches are generally hydrophilic. Development and application of these materials as barriers under high relative humidity (RH) conditions tends to be challenging, especially when compared to plastics whose usage is more widespread and more convenient [14]. Starches tend to exhibit decent water vapor barrier properties at low RH conditions; however, this barrier breaks down at high RH conditions. The reason for high moisture sensitivity (especially at high RH) of these starches [15] is the presence of hydroxyl groups in the starch molecule, which lead to increased water vapor transport through the formation of hydrogen bonds with water molecules. The development of starch-based materials with low water vapor permeability (WVP) is a significant challenge and an area of active research. Hydrophilicity of the final material is key to controlling WVP. Reducing the hydrophilicity of a starch molecule is achieved by a chemical reaction that can block the hydroxyl group or the addition of a hydrophobic polymer to the starch. Chemical modifications [10] and physical blending strategies [9] have thus been successfully employed to improve the water vapor barrier properties of starch-based materials. Selected examples of blended and chemically modified starches, along with their barrier properties, are presented in **Table I**.

Water vapor barrier performance is measured as the transmission rate of water vapor across a film of material. A gradient of water vapor RH is established across the film, and measurements are taken at steady state. Several instruments and standards are available for testing. A commonly used method is the TAPPI “jar method” (TAPPI Standard Test Method T 464 om-2012 “Water vapor transmission rate of paper and paperboard at high temperature and humid-

ity”). The water vapor transmission rate (WVTR) of a starch like amylo maize is on the order of 53 g-mm/m²/d at 33% RH [24] and increases exponentially with increasing RH. As such, two commonly used testing conditions are: (a) 50% RH at 23°C, and (b) 90% RH at 38°C. The high humidity and temperature condition is referred to as “tropical” condition and is utilized for exploring the performance of the materials in the most extreme conditions that are detrimental for food preservation. One of the most common chemical modification approaches utilizes esterification chemistry to attach hydrophobic groups or polymers to the starch backbone. Such esterification modifications have been found to reduce WVTR of starches by up to 70% under certain conditions [25], while other treatments like hydroxypropylation can reduce WVTR by 40%–60% over pure starch [26]. Ultimately, the success of these methods does depend on several factors, including the type of chemical modification used, processing techniques, reaction conditions, and the availability of free hydroxyl groups, as well as their interaction with the surrounding matrix after a chemical reaction. As seen in Table I, blending [20,21] with hydrophobic materials can provide a rather effective means of achieving similar or greater WVTR reductions to chemical modifications. Here, the WVTR improvements and response ultimately depend on the structural arrangement of the starch and the hydrophobic modifier. The compatibility of such additives can be affected by the nature and quantity of the plasticizers. The blends of oxidized starches, plasticizers [24], and various clays, such as bentonite, [19] montmorillonite, [16] and kaolin [27], have been demonstrated to provide significant reductions in the WVTR. The addition of clay creates a tortuous path that makes

Material	Cobb60, g/m ²	Contact Angle, °	Solubility, %	Form	Reference
Starch, chitosan, silver nanoparticles	10	-	-	Coatings	[32]
High molecular weight cationic starch/high molecular weight anionic starch	19	-	-	Coatings	[33]
Hydrophobic non-crystalline starch and silver nanoparticles	20	-	-	Coatings	[34]
PLA, starch, and cellulose foam	-	121	-	Foam	[35]
Corn starch and ZnO nanoparticles	-	121	-	Film	[36]
Thermoplastic starch and 6% CNF with plasma treatment and ultrasonication	-	127	-	Composite Film	[37]
poly(dimethylsiloxane), montmorillonite, starch	-	159	-	Film	[38]
Maize starch, PLA, cardanol	-	-	7.5	Film	[39]
Starch, glycerol and PBAT	-	-	10.5	Film	[40]
Potato starch and sodium hypochlorite	-	-	12.7	Film	[41]
Starch, stearic acid	-	-	13.5	Film	[42]
Wheat starch, TiO ₂	-	-	16.9	Film	[43]
Etherified maize starch, sorbitol	-	-	17.1	Film	[44]
PLA = poly(lactic acid); ZnO = zinc oxide; CNF = cellulose nanofibrils; PBAT = poly(butylene adipate terephthalate); TiO ₂ = titanium dioxide.					

II. Liquid water barriers achieved with starch-based materials.

water vapor difficult to permeate through the film or coating [28-31]. The sheer number of available modifiers, like lignin, cellulose nanofibers (CNF), soybean oil, etc., and the availability of chemically modified starches combined with the simplicity of blending methods provides a convenient route to obtain materials that can match low density poly(ethylene) (LDPE) or high-density poly(ethylene) (HDPE), even under conditions of high humidity (90% RH) and temperature of 25°C–30°C.

Liquid water barrier

Liquid water barrier is important, especially for the packing of liquid products such as juice, milk, and other beverages. Hydrophobicity or super-hydrophobicity is generally required to achieve a barrier to the liquid water. Surface roughness affects the resistance to liquid water of the material as well as the penetration [32] of the coating material used to impart water resistance. Resistance to liquid water is characterized using methods such as contact angle measurements, Cobb water test, water absorption, and water solubility of starch-based films. The Cobb60 test measures contact with water for 60 s and is shorter than the more common industrially used Cobb values for 30 min (Cobb1800). More rigorous methods exist, like hot water Cobb, which test the limits of the barrier properties of the final packaging.

We compared values among similar Cobb test methods,

with the most commonly reported among them being Cobb60. Various starch-based materials and their resistance to liquid water as characterized by these tests are shown in **Table II**. Addition of metal nanoparticles was generally found to increase the contact angle [36] and reduce Cobb60 values by increasing surface roughness and decreasing water solubility [45]. Higher contact angles are a good indicator of surface hydrophobicity, with values above 90° being considered hydrophobic surfaces. The addition of fatty acid and plasticizers have also been shown to increase the liquid water barrier property by a reduction in the solubility of the starch-based films or coatings [42,44]. However, the excess addition of these materials can cause the separation of crystals of fatty acids [46] and the disintegration of the starch matrix [47]. One point to note is that, in general, no single property like contact angle or solubility can truly indicate water resistance, and they must be considered together to enable comparison of water barrier performance of materials under consideration. The true potential of starches is demonstrated by the fact that just pure starches have the potential to replace 70% of polystyrene in packaging applications and still have similar water barrier properties to polystyrene [47,48]. Blending with other natural polymers such as lignin [49], poly(lactic acid) (PLA) [35,50], and essential oils [51] only enhances the liquid water barrier of the composites over pure starch.

Material	OTR, cm ³ -mm/ m ² /d/kPa	RH, %	Temperature, °C	Form	Reference
Starch, chitosan, lauric acid	0.12	0	23	Film	[54]
Starch, PBS	0.01	0	23	Pellet composite	[21]
Hydroxypropylated starch, kaolin	0.1	50	23	Coatings	[55]
Corn starch, sodium caseinate	0.004	53	25	Film	[56]
Amylomaize, sorbitol, sunflower oil	20	64	20	Coating	[8]

OTR = oxygen transmission rate; RH = relative humidity; PBS = polybutylene succinate.

III. Oxygen barrier of starch-based materials.

Oxygen gas barriers

Oxygen barriers are important in packaging applications, especially when the product is susceptible to oxidative degradation. Starches are known to have excellent oxygen barrier properties. However, like the water vapor barrier properties, higher humidity adversely affects the oxygen barrier of the starch-based films. Oxygen barrier properties depend on cohesive energy densities, free volume, and the degree of crystallinity in the matrix. It is known that higher crystallinity of natural materials helps to reduce oxygen penetration [52,53]. Oxygen barrier properties are usually measured as a permeability of oxygen through the film. A gradient of oxygen is established across the film and measurements are taken when the transmission of oxygen across the film reaches steady state. Generally, such values are measured at different relative humidities and temperature conditions. Several instruments and methods (such as ASTM D3985 – 95 “Standard test method for oxygen gas transmission rate through plastic film and sheeting using a coulometric sensor”) are available for measurement of oxygen permeability (OP). Commonly used testing conditions are: (a) 50% RH at 23°C, and (b) 90% RH at 38°C (tropical conditions).

Table III summarizes starch-based materials that can have low OP. As a reference, corn starch films have an OP of 140 cm³-mm/m²/d/kPa at 64% RH [8]. It was shown that the low permeability of these systems was due to the low solubility of gases in the matrix [57]. Linear starches like amylose have significantly better oxygen barrier performance, possibly due to better packing of the linear chains [57]. It was also found that chemical modifications of starches adversely affected the oxygen barrier property of pure starch-based material [58]. Starch blended with various natural materials and biopolymers such as chitosan [58], polybutylene succinate (PBS) [21], or proteins [56] were found to improve the oxygen barrier properties of the film. The type and concentration of plasticizer can also significantly affect the oxygen barrier properties of the starch-based films and coatings. Compiled data from various sources in Dole et al. [57] shows an increase in starch OP with increasing glycerol concentration as well as increased permeabil-

ity of amylose plasticized with glycerol. Garcia et al. [8] reported a decrease in OP for corn and amylo maize films plasticized with glycerol and sorbitol, with sorbitol plasticized films showing lower OP [8]. An 11-fold decrease in permeability was observed when sorbitol was added to high amylose starch [8]. Etherified maize starch films plasticized with sorbitol showed nondetectable oxygen permeation but also higher deformation at break, along with reduced tensile strength at break [44]. While there is discrepancy on the exact influence that plasticizers have on gas permeability of starch films and coatings, it is clear that plasticization effects must be carefully considered for package design.

Oil/grease barrier

Oil and grease resistance (OGR) is an important barrier property used in numerous applications, including takeout containers or packaging boxes for foodservice products. The primary components of grease and oils are triglycerides, which are fatty acids connected to the glycerol backbone. In triglycerides, the glycerol part is polar and hydrophilic, whereas long chains of fatty acids are non-polar and hydrophobic, which makes grease and oils amphiphilic [59]. The amphiphilic nature of OGR influences its penetration through the paper and paperboard. Hence, OGR of packaging materials is a function of the material; its interaction with oil and grease [59]; the length of the fatty acid chain; and the type of fatty acids present, such as saturated or unsaturated [60].

An attempt has been made to summarize the vast literature on starch-based OGR materials and is shown in **Table IV**. OGR is measured in terms of kit value on a scale of 1–12 [67]. The higher the kit number, the better the OGR properties of the material. Kit 12 value is the gold standard for OGR, with chemically modified (anionic and cationic) starches showing kit 12 barrier [33]. Cobb values measured for 60 s contact time (Cobb60) are another way of analyzing OGR. In this case, higher Cobb values indicate higher uptake of oil and are detrimental. Both of these methods are dependent on temperature and properties of the penetrant. In general, it is best to utilize such methods for rapid

Material	Kit	Cobb Oil 60, g/m ²	Form	Reference
Soluble starch, silver nanoparticles	3	-	Coatings	[61]
Cationic starch, chitosan	7	-	Coatings	[62]
Starch, chitosan, silver nanoparticles	7	-	Coatings	[32]
Hydrophobic starch ester	7.5	-	Coatings	[63]
Hydroxypropylated starch, kaolin	8.1	-	Coatings	[64]
Modified starch, sodium alginate	12	-	Coatings	[65]
High molecular anionic starch, high molecular cationic starch	12	1.8	Coatings	[33]
Corn starch, corn zein	12	4.8	Coatings	[66]

IV. Oil and grease barrier of starch-based materials

comparison between candidate OGR materials. The final OGR properties must be rigorously tested. Hydroxypropylated starches have also been used extensively for their OGR properties [55,64,68]. Blending starches with metal nanoparticles [32, 34, 61] and clay also exhibited enhanced OGR properties similar to other barriers. A possible reason for this improvement due to blending could be the creation of a tortuous path [28-31] in the matrix and an increase in surface roughness [60]. It must be noted that some metal nanoparticles like zinc oxide (ZnO) are truly multifunctional in their application, as they not only provide OGR as noted previously but also antimicrobial, UV filtering, and mechanical properties, while also being defined in the Generally Recognized as Safe (GRAS) category by the FDA [69].

UV barrier

Radiation energy from light such as UV or visible light triggers the photochemical reaction in photosensitive products and causes degradation. Light-sensitive materials undergo oxidative degradation when exposed to UV light, which leads to deterioration of the material. Therefore, UV barriers are in demand for various packing and storage grade materials.

Starches are often components of UV barrier coatings employing nanoparticles like titanium dioxide (TiO₂) that are known to be active UV barriers. **Table V** summarizes UV barrier materials with starch as a component. The values indicate transmission of UV light of a particular wavelength or range, with lower transmission indicating better barrier performance. As with previous approaches, blending is actively employed for enhancement of UV barrier properties of starches. Interestingly, some organic natural materials like chicken skin gelatin and potato starch [70] and chemically modified starches like octenyl succinate starch and hazelnut skin extract [51] have been shown to possess excellent UV barrier properties, with transmittance as low as 0.01% in the wavelength range of 200–280 nm,

and are comparable to approaches using inorganic nano-sized metal particles like TiO₂ that are already known to be good UV barrier additives [23,45,36,73].

Thermal barriers

The thermal resistance of a barrier material is a function of resistance to heat flow. It is an important barrier property to preserve the thermal condition of the inside of packaging while limiting the transfer of heat from the outside. This helps to maintain the temperature of a product in a perfect condition for longer duration. The thermal conductivity of the material characterizes its ability to conduct heat. Materials with lower thermal conductivities are better thermal barriers.

Various factors affect the thermal conductivity, such as temperature difference, path length, type of materials, and a cross-section through which heat flows. Also, there are other factors that have an impact on the thermal conductivity of the materials, including random alignment of the polymer chain; the amorphous nature of the material; heavy and increased side chains in the polymer backbone [74]; and small pore size less than the mean free path of air molecules [75].

Starch-based aerogels are a combination of starches and air and have demonstrated effective thermal barrier properties. These starch-based packing foams are great thermal barrier materials, as shown in **Table VI**. They are most often produced using thermoplastic starches. While starches themselves are not thermoplastic in nature, addition of water and/or plasticizers can lead to thermoplasticity in starches [13,14]. It is important to distinguish between thermoplastic starches (which relates to processing methodology) and the final product, which is a starch-air composite (starch aerogel). All thermal conductivities reported in Table VI are at room temperature. Among different types of starches, pea starch aerogels showed the lowest thermal conductivity [76]. High amylose corn starch-based foams

Material	Transmittance, %	At Wavelength, nm	Form	Reference
Crosslinked wheat starch, sodium montmorillonite, TiO ₂	0.45	240	Film	[23]
	1.03	300		
	1.01	360		
	23	600		
Octenyl succinate starch, hazelnut skin extract Chicken skin gelatin, potato starch	0.22	280	Film	[51]
	0.01	200–280	Film	[70]
	245	350		
	40	400		
Potato starch, nano SiO ₂	18–27	300–800	Film	[20]
Starch, cellulose nanofibers	20–30	300–800	Film	[71]
Corn starch and ZnO nanoparticles	37–50	200–400	Film	[36]
Starch, corn hulls	0.4	206	Film	[72]
TiO ₂ = titanium dioxide; SiO ₂ = silicon dioxide; ZnO = zinc oxide.				

V. Ultraviolet (UV) barrier of starch-based materials.

Material	Thermal Conductivity, W/m/K	Form	References
Pea starch	0.021–0.023	Aerogels	[76]
High amylose corn starch	0.024	Foam	[77]
Konjac glucomannan, potato starch, gelatin, wheat starch	0.047	Aerogels	[78]
Starch, geopolymers, gypsum	0.06–0.09	Composite material	[79]

VI. Thermal barrier of starch-based materials.

showed lower thermal conductivity, which can replace commercially available polystyrene foam (polystyrene foam thermal conductivity = 0.035–0.037 W/m/K [77,80,81]). There are numerous materials like plates, foams, and aerogels that are developed based on potato starch and have shown average thermal conductivity as low as 0.046 W/m/K [78,79]. Thermal conductivity may be further lowered to 0.04 W/m/K by using composite materials using gypsum, geopolymer, and starch [79].

Antimicrobial barriers

Several synthetic and bio-based materials are currently being used for antimicrobial applications [82–84]. The general mechanism of antimicrobial materials can be segregated into three categories: barrier by (i) direct disinfection [84]; (ii) indirect disinfection [82]; or (iii) modifying the bacteria, virus, or fungi [85,86]. Preventing microbial colonization, growth, and spread is the key factor in limiting the spread of infections. In general, antimicrobial materials can be divided into three parts such as anti-bacterial, antiviral, and antifungal materials.

The demand for new antimicrobial products is increasing due to the increase in the spread of infections. Pandemics, such as COVID-19, escalate the need for antimicrobial or antiviral materials. It is extremely difficult to develop a universal product that could regulate or stop the entire source of infectious diseases due to the varied nature of infectious agents and mechanisms of action. **Table VII** summarizes some starch-based antimicrobial materials of interest. While pure starch by itself is not an antimicrobial agent and can in fact promote microbial growth (being a convenient food source), it can be blended with other active antimicrobial components to make an effective antimicrobial barrier. For instance, it has been shown that starch enhances the absorption efficiency of fabric when blended with salt for the deactivation of the SARS-CoV-2 virus that causes COVID-19 [86]. A very common strategy is to blend starches with nanoparticles (especially silver) or essential oils to create antibacterial and antifungal films and coatings. Another valid strategy is to use chemically modified starches either by themselves or with other materials. For instance, dialde-

Material	Virus/Bacteria/Fungi	Form	Reference
Antiviral			
Starch and salt treatment	SARS-CoV-2	Dip coating on the fabric	[86]
Dialdehyde starch	Bacteriophage MS2 and PRD1, human polio virus	Aqueous suspensions	[87]
Antibacterial			
Corn starch, <i>Zataria multiflora</i> Boiss essential oil	<i>E. coli</i> , <i>S. aureus</i>	Film	[88]
Corn starch, <i>Mentha pulegium</i> essential oil	<i>E. coli</i> , <i>S. aureus</i>	Film	[88]
Oxidized corn starch, bentonite, chitosan	<i>Pseudomonas syringae</i> pv <i>tomato cell cultures</i>	Film	[19]
Carboxymethyl starch, ZnO nanoparticles	<i>P. aeruginosa</i> , <i>B. cenocepacia</i> , <i>S. maltophilia</i> , <i>A. baumannii</i>	Coatings	[89]
Thermoplastic starch, TiO ₂ , and vancomycin	<i>S. aureus</i>	Film	[90]
Potato starch, nano SiO ₂	<i>E. coli</i>		[20,21]
Hydroxypropylated maize starch, chitosan, ZnO nanoparticles	<i>E. coli</i> , <i>S. aureus</i>	Film	[12]
Native starch, CNF, silver nanoparticles	<i>S. aureus</i> , <i>K. pneumoniae</i>	Coatings	[91]
Starch, chitosan, silver nanoparticles	<i>E. coli</i> , <i>S. aureus</i>	Coatings	[32]
Hydrophobic noncrystalline starch, silver nanoparticles	<i>E. coli</i> , <i>S. aureus</i>	Coatings	[34]
Starch, ZnO nanoparticles	<i>E. coli</i> , <i>S. aureus</i>	Coatings	[73]
Starch, silver nanoparticles	<i>E. coli</i> , <i>Salmonella</i>	Film	[45]
Antifungal			
Cassava starch, bovine gelatin, cinnamon essential oil	<i>F. oxysporum</i>	Film	[92]
Cassava starch, bovine gelatin, clove essential oil	<i>C. gloeosporioides</i>	Film	[92]
Starch, aloe vera gel	<i>F. oxysporum</i>	Film	[93]
Wheat starch, ZnO nanoparticles	<i>T. viride</i>	Coatings	[94]
Starch, Mexican oregano, or cinnamon essential oil	<i>A. niger</i> , <i>P. digitatum</i>	Film	[95]
Starch, chitosan, silver nanoparticles	<i>P. expansum</i>	Coatings	[32]
ZnO = zinc oxide; TiO ₂ = titanium dioxide; SiO ₂ = silicon dioxide; CNF = cellulose nanofibrils.			

VII. Starch-based antimicrobial materials.

hyde starches as aqueous suspensions are effective against certain types of viruses [87], while oxidized starches blended with bentonite and chitosan can form antibacterial films [18].

Sound barriers

Soundproofing, or sound-absorbing, materials are used in various places such as buildings, automobiles, etc. These materials help reduce or block sound propagation to pro-

vide greater comfort and safety. In general, the average sound absorption coefficient is measured on a scale of 0 to 1, with 0 being no sound absorption capacity and 1 being 100% sound absorption.

Wang et al. [96] used a freeze-drying technique to generate an aerogel of potato starch, wheat straw, gelatin, and polysaccharide konjac glucomannan that had a sound absorption coefficient of 0.352, and a low density of 0.047 g/cm³. This represents a 56% improvement over con-

Trademark	Owner	Materials	Application
Biopur	Biotec GmbH	Thermoplastic/disaggregated starch, aliphatic polyester, hydrophobic protein, PVAc	Foam trays
Aqua-Novon	Novon International Inc.	Modified PVA, gelatinized starch	Shaped articles
Bioflex	Biotec GmbH	Starch, epoxide-containing (including cellulose acetate)	Replacement for single-use plastic bags
Biopac	Biopac Ltd.	Starch, fatty acid release agent	Cups, plates, trays
BioPar	Biop AG Biopolymer GmbH	Starch, aliphatic copolyester, hydrolyzed PVAc	Pellets
Bioplast	Biotec GmbH	Starch, PLA, copolyester, PHA	Trays, cups, bags
Bioska	PlastiRoll Oy	Starch, PVA	Packing films, bags
Bio-Solo	Indaco Manufacturing Ltd.	Starch, additives, PE	Commercial bag
Biostarch	Biostarch	Starch, PVAc, polyol elasticity enhancer, >20% amylose	Resin, films, pallets
Clean Green	StarchTech Inc.	Starch	Void fill packing materials, peanuts
Cornpol	Japan Corn Starch	Starch ester	Food containers, films
Eco-Foam	National Starch & Chemical	Starch, hydroxypropylated starch, PVAc	Foamed starch
Eco-Lam	National Starch & Chemical	Starch, PET, PP	Recyclable packing laminates
Envirofil	EnPac/Dupont/ConAgra	Starch, PVA	Loose-fill
Enviromold	STOROPack Inc.	Starch	Loose-fill
BioBag International AS	Polargruppen	Starch, cellulose, vegetable oil	Bioplastics, bags, cups
EverCorn	EverCorn Inc.	Hydrophobic starch, biodegradable plasticizer	Moldable products, films
FLO-PAK BIO 8	FP International	Starch, corn, or wheat	Void fill packaging
GreenFill	Green Light Products Ltd.	Starch, PVA	Void fill
Loose Fill	STOROPack	Expanded PS, starch	Void fill
Solvitose	Avebe	Potato starch	Heat-sealable barrier coatings-oil grease, oxygen
Paragon	Avebe Bioplastics	Starch, PLA, or PHA	Biodegradable plastic mulch
Plantic	Plantic Technologies	Starch, nano-clays	Oil, gas, and aroma barrier film
Vegemat	Vegeplast S.A.S	Starch, corn fiber, proteins, lipids, starch, polyester	Coating, coated film
Sconacell	Buna SOW Leuna	Starch acetate, poly(alkene glycol), carboxylic acid	Packing film for tobacco products, soap, paper tissues, toilet articles

PVAc = poly(vinyl acetate); PVA = poly(vinyl alcohol); PLA = poly(lactic acid); PHA = poly(hydroxyalkanoate); PE = poly(ethylene); PET = poly(ethylene terephthalate); PP = poly(propylene); PS = poly(styrene).

VIII. Starch-based commercially available barrier and packaging materials and end products. Adapted from McKeen [98].

ventional fiber cotton sound-absorbing board commonly used in building applications that has an average sound-absorption coefficient of 0.225. Bumanis et al. [79] made a gypsum, geopolymer, and starch-based composite material that provided effective sound barriers for building applications [79]. Low-density compression bonded loose fill (CBL) starch foams showed an average sound absorption coefficient of 0.5–0.6. These low density CBL starch foams exhibit higher sound absorption coefficients across a wider range of frequencies than their denser counterparts. Low-density regular packing and stacking (RPS) starch foams were found to exhibit a sound barrier with a sound absorption coefficient of 0.7–0.8 [97]. These results from various studies open an avenue for starch-based materials to be used for soundproofing or sound reduction applications.

Commercially available starch-based barriers

There are several starch-based barrier products currently available in the market under various brand names that utilize a blend of starch with some or all environmentally friendly materials. These starch-based commercial materials are being extensively used in the foodservice industry in applications such as coatings, films, and bioplastics. They are versatile and have been successfully used for making trays, cups, spoons, and other moldable components that possess oil, grease, water vapor, or oxygen barrier properties. One of the most ubiquitous applications of starches are as foams and loose fill packaging for making packing peanuts used to prevent damage to items from shock during packaging and transportation. **Table VIII** summarizes the vast array of commercially available starch-based materials used for packaging or barrier applications. The demand for these products is growing. In fact, they have already replaced or have the potential to replace petrochemical and synthetic materials due to their biodegradability, non-toxicity, and recyclability in several commercial applications. There are, however, some limitations [99] to broader commercialization efforts due to the brittleness, sensitivity to humidity, and high viscosity that starches exhibit.

CONCLUSIONS

The global need for bioplastics is expected to increase from around USD 10.5 billion in 2020 to USD 27.9 billion by 2025, at a compound annual growth rate (CAGR) of 21.7% [100]. While this estimate might be overly optimistic by some sources [101], we cannot deny that bioplastics are growing and there is a lot of industry interest and investment in their growth. There is no single material that can provide all barrier properties and there is a huge scope for research and development in the area of bio-based polymers for barrier applications. While starch-based products are inherently sustainable and eco-friendly, achieving desired barrier properties, characteristics, and easy processability can be quite challenging, making them less convenient than petroleum or synthetic materials. Starch-based materials also

have their own unique limitations, such as a tendency to retrograde, low mechanical properties, high viscosity at lower solids, and relatively poor water vapor barrier and an undesirable humidity response. The hydrophilic nature of starches is one of the most significant limitations to potential barrier applications of this promising polymer, especially at high RH conditions. They are highly susceptible to humidity changes and absorb more water with increasing humidity. Plasticizer type has been demonstrated to dramatically alter the water uptake and morphology and thereby glass transition temperature (T_g) and mechanical properties of the material.

Another key issue is the lack of uniformity in properties when dealing with naturally sourced materials that often show large variation between species of plants. The reason for such variations is that starch properties are complex variables governed by numerous parameters that influence the property of starch molecules, which in turn may influence the type of additives (like plasticizers) that are required for workability. These in turn affect the T_g and crystallinity. Crystallinity, in particular, is dependent on the ratio of amylose to amylopectin. The ratio of amylose and amylopectin can equally be a result of processing methods as well as the source of the starch and the season in which the plant was harvested (seasonality). While pure amylose films are known to be stronger than pure amylopectin films [102], processing high amylose starch is challenging, which is specifically due to its tendency to rapidly retrograde.

A major disadvantage of starch-based materials when dealing specifically with aqueous application is the requirements of large amounts of water for processing and high viscosities of the resulting suspensions at relatively low solids, requiring high energy for processing and drying.

To overcome these issues, starches are often blended with biodegradable polymers, such as poly(hydroxybutyrate) (PHB), poly(lactic acid) (PLA), poly(vinyl alcohol) (PVA), and poly(caprolactone) (PCL) [39,103–107], that allow the final material to still maintain an eco-friendly and green tag. Blending with plasticizer, fillers, and composites [21,43,55,108–112] has also been carried out alongside chemical modifications [113–114] to improve the barrier properties of starches. Starch is readily biodegradable and compostable; hence, starch is an attractive additive for blending with various polymers. While the starch may not necessarily help in degradation of the polymers that are not prone to biodegradation like olefins [14], it definitely increases the biodegradable material percentage of the composite. When combined with compostable/biodegradable polymers, it can lead to a purely compostable/biodegradable replacement to plastics, as discussed in Table VIII. These processes can result in new materials with optimal properties, and it is important to consider processability, characterization, and ease of end-use application when assessing the potential of these new materials. Starch-based materials — apart from

their potential as gas, liquid, and oil barriers — have also been evaluated as soundproofing, UV, antiviral, and antimicrobial barriers. Future research on starch-based materials should be directed at overcoming issues related to their low resistance to liquid water, water vapor, poor mechanical properties, difficulties in processability, and high viscosities at lower solids. **TJ**

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ABOUT THE AUTHORS

We chose to study this topic because we are interested in further improving the sustainability of packaging products. It is one of the areas we continue to explore in improving the sustainability of packaging products manufactured by WestRock.

The current work offers a literature review on application of starch as a barrier material. In reviewing work done by others, it is always a challenge to ensure that the procedures and units are taken into consideration when comparing data. This was accomplished by careful data analysis.

Researchers have been working with starch for a very long time, and it is gratifying to see that the research community continues to find new ways to use starch in old and new applications.

Mills and product development can use this infor-



Rao



Satam



Ghosh



Hart

mation to explore new revenue streams. Our next step is to continue developing work streams with biodegradable barrier materials.

At the time of this work, Rao was a co-op intern at WestRock in Furnish Science and a Ph.D. candidate in Chemistry at the University of Maine, Orono, ME, USA. Satam is a scientist; Ghosh is senior R&D engineer; and Hart is director of Fiber Science and Innovation at WestRock, Richmond, VA, USA. Email Ghosh at ashok.ghosh@westrock.com.