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Polysaccharide-Based pH-Responsive Intelligent Halochromic Food Packaging Materials: A Review

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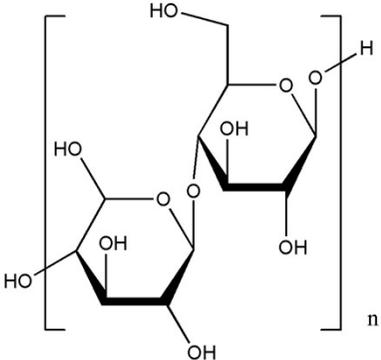
Table S1 Structure, sources, properties, and food applications of major polysaccharide types

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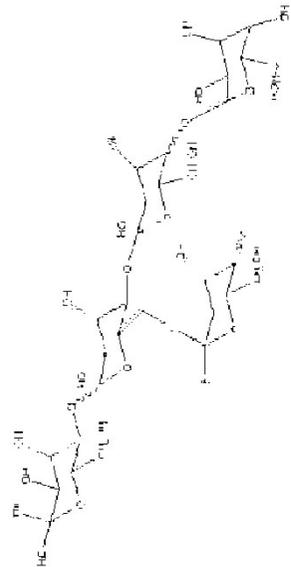
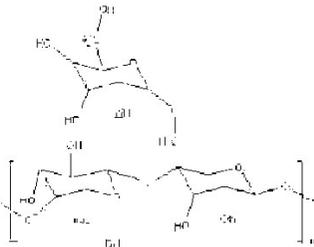
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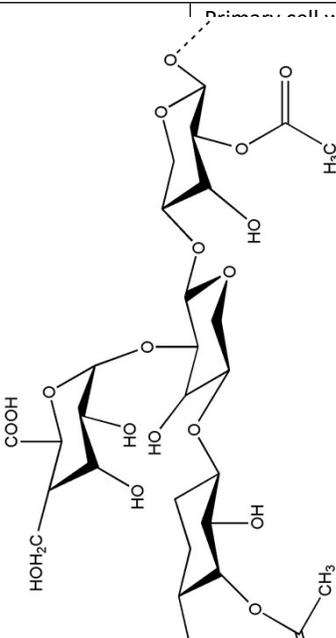
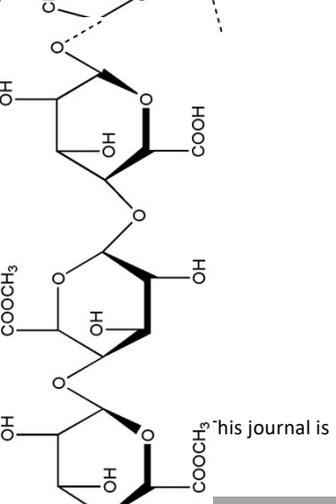
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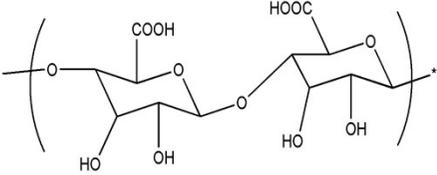
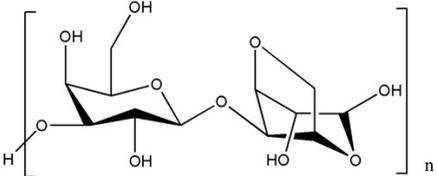
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Example	Polysaccharide type (s)	Molecular structure	Sources	Properties	Food applications	Ref(s)
Cellulose	Plant-based Animal-based Algae-based Microbial-based		Plant cell wall Marine organisms in the tunicate family Algae and fungi Gram-negative bacteria	A network of hydrogen bonds packed together to form high crystalline polymer Insoluble in almost all aqueous solutions Due to poor solubility, the production cost is high for cellulose-based packaging materials Cellulose derivatives, such as cellulose esters and ethers have high solubility, making them ideal for manufacturing PPMs	Packaging of fruits, vegetables, dairy products and flesh and fish	1-4

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Starch	Plant-based		<p>Wheat Corn Cassava sweet potato Rye starch.</p>	<p>Ability to become a thermoplastic material when exposed to heat and shear Nontoxic Odourless Edible Good oxygen barrier Good moisture absorption Poor mechanical strength. Structural tunability, Antibacterial, barrier Antimicrobial properties</p>	<p>Use to manufacture intelligent packaging for fish, meat, fruit juice and cooking oil. Potato starch packaging materials are also available.</p>	2, 5-10
Galactomannans	Plant-based		<p>Endosperm of dicotyledonous seeds of various plants, in the Leguminosae famil</p>	<p>Resistance to the pH and ionic strength changes High thermal stability Gas barrier properties Better blending with plasticizers and other Polym.</p>	<p>Edible films and coatings</p>	11-15

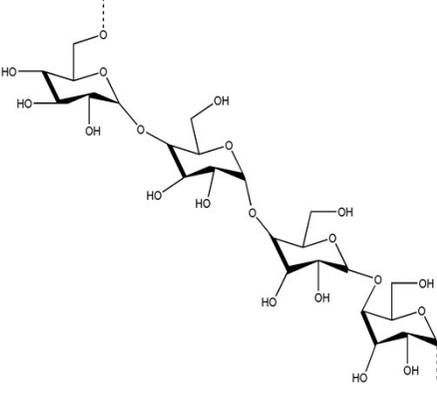
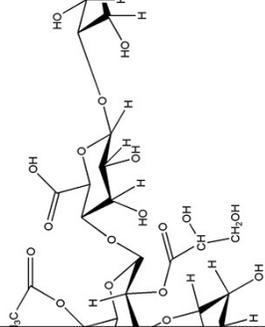
<p>Hemi-cellulose</p>	<p>Plant-based Microbial-based</p>	 <p>Primary cell wall of all of plants, cell wall of algae, and almost all fungi. Structure of plants</p>	<p>Low gas permeability Flexibility Water resistivity Sensitivity to the humidity Low mechanical properties</p>	<p>Biofilms are used for food packaging and coatings</p>	<p>16-18</p>
<p>Pectin</p> <p>4 <i>J. Name.</i>, 2012, 00, 1-3</p>	<p>Plant-based</p>	 <p>found in apples</p>	<p>Film forming, gelling and thickening capabilities. High stability High water resistance Barrier for lipid oxidation Water barrier properties</p>	<p>Edible films (pullulan: pectin – 50:50)</p>	<p>2, 15, 19-22</p>

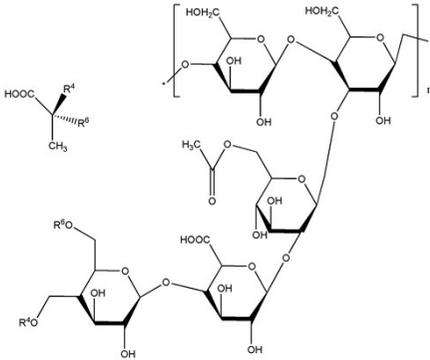
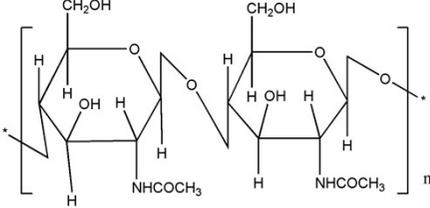
Alginate	Algae-based Microbial-based	 <p>The diagram shows the repeating unit of alginate, a linear polysaccharide. It consists of two pyranose rings linked by a 1,3-glycosidic bond. The left ring is a 4-O-acetylated-D-glucopyranose unit, and the right ring is a 3-O-mannuronate unit. The units are enclosed in large parentheses with a subscript 'n' and a star symbol, indicating a polymer chain.</p>	Marine algae exopolysaccharide of specific bacterial strains like <i>Pseudomonas aeruginosa</i> brown seaweeds (<i>Ascophyllum Nodosum</i> , <i>Laminaria Digitata</i>)	Good film-forming characteristics in the presence of plasticizers Barrier for lipid oxidation. Low cost Nontoxicity Thickening agent	Edible films for vegetables, fruits, seafood, and poultry	12, 15, 23, 24
Agar	Algae-based	 <p>The diagram shows the repeating unit of agar, a linear polysaccharide. It consists of two pyranose rings linked by a 1,3-glycosidic bond. The left ring is a 3-O-mannuronate unit, and the right ring is a 4-O-acetylated-D-glucopyranose unit. The units are enclosed in large square brackets with a subscript 'n' and a hydrogen atom (H) on the left, indicating a polymer chain.</p>	Marine algae	Homogeneous Transparent Flexible Good physical properties	Bioactive plastic, sustainable packaging	2, 25, 26

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Carrageenan	Algae-based		Red sea weeds of the Rhodophyceae family	<p>Good barrier for oxygen Avoid lipid oxidation Antimicrobial and antioxidant properties High stability and moisture resistance</p>	<p>Used to form edible films and coatings for fresh-cut fruits. Camelia oil incorporated carrageenan/glucomannan edible coatings used for avoiding lipid oxidation and increasing the shelf life of chicken. Bioactive films for lamb meat preservation by adding glycerol, water, and olive leaf extract.</p>	8, 12, 27
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Pullulan	Microbial-based		Aureobasidium pullulans	<p>Excellent film-forming ability Able to form flexible thin films Antimicrobial activity High mechanical strength</p>	<p>Edible bio coatings and films for minimally processed <i>Foods</i>, chicken, ready-to-eat meat, and fresh food items. Fruit preservation</p>	15, 28, 29
Gellan gum	Microbial-based		Prepared by <i>Sphingomonas elodea</i> bacteria	<p>Thermal stability and biocompatibility Mechanical strength Gelling temperature can be modified by incorporating other natural Polym.</p>	<p>Form pH-responsive packaging films to monitor the freshness of fish by incorporating anthocyanin and pullulan to improve the properties</p>	30-32

Xanthan gum	Microbial-based		Xanthomonas campestris microorganisms	Barrier properties Antimicrobial and mechanical properties. Reduce the growth of aerobic microbes Reduce oxidation	Excellence for manufacturing active and intelligent packaging Packaging for meat products like pork cuts and fish products like carp.	4, 33-35
Chitin	Animal-based Microbial-based		The cell walls of fungi, yeast, crabs, prawns, lobsters, insects, and fish scales	Insolubility in common solvents Chitin nanoparticles/nanocrystals are excellent for fabricating the packaging materials	Chitin nanoparticles incorporated edible nanocomposite films	36-39

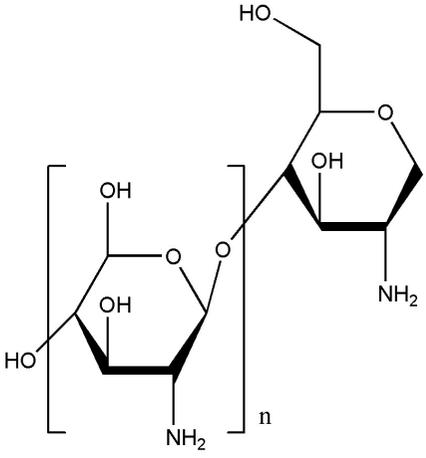
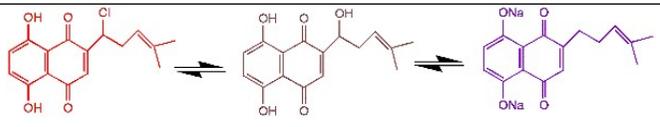
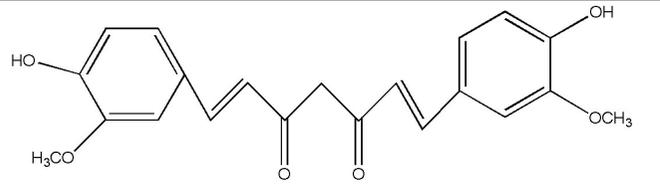
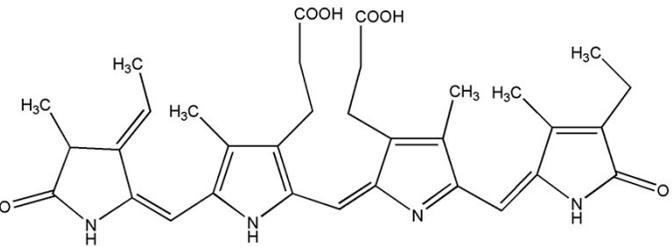
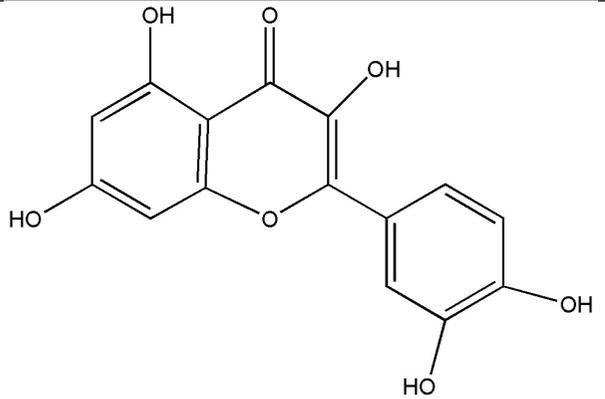
Chitosan	Animal-based Microbial-based	 <p>The diagram shows the chemical structure of a chitosan repeating unit. It consists of two pyranose rings connected by a 1-4 glycosidic bond. The left ring is a 2-amino-2-deoxy-D-glucopyranose unit, shown in its cyclic form with a hydroxyl group at C2 and an amino group at C2. This unit is enclosed in brackets with a subscript 'n'. The right ring is a 2-amino-2-deoxy-D-glucopyranose unit, also in its cyclic form, with a hydroxyl group at C4 and an amino group at C2. The glycosidic bond connects the C1 of the left ring to the C4 of the right ring.</p>	Deacetylation of chitin	Antimicrobial properties Water vapour permeability Good gas barrier properties Excellent mechanical properties	Smooth uniform films, Composite coating for maintaining fruit edibility of dragon fruit. Enhance the storability of Wolfberries, papayas, tomatoes, mangoes, carrots, pomegranates, bananas, and other fruits and vegetables	4, 15, 40-42
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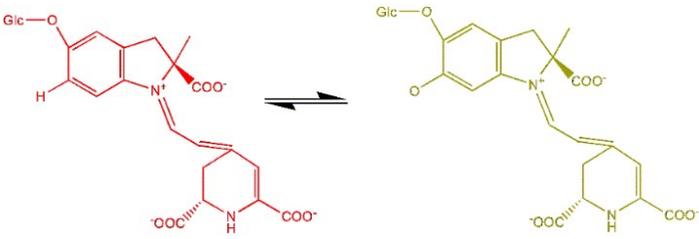
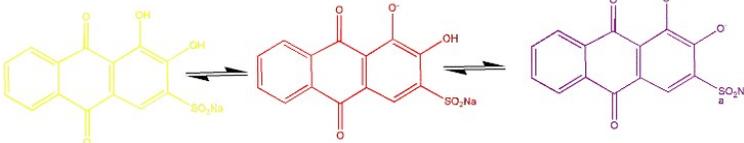
Table S2 The structure, pH responsiveness and properties of the most common natural halochromic dyes used in pH detection

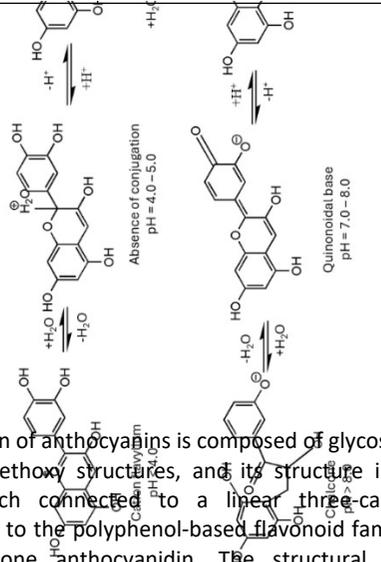
Colorant	Structure	Properties	Source	Ref(s)
Shikonin	 <p>The molecular structure of shikonin consists of two basic parts a chiral 6-carbon side chain and a naphthazarin moiety. The chemically reactive naphthazarin core of shikonin is sensitive to heat, light, acid, and base conditions. The color change occurs because of the instability and breakdown of the chromophore molecular structure.</p>	<p>Antiviral properties. Antimicrobial properties. Antioxidant properties. Anti-inflammatory properties. The color of the dye changes from red to purple to blue under acidic, neutral, and alkaline conditions, respectively.</p>	<p>Extracted from gromwell roots (Lithospermum erythrorhizon)</p>	<p>43-49</p>
Curcumin	 <p>The molecular structure of curcumin consists of a seven-carbon chain with an α-β unsaturated β-diketone moiety bordered by two aromatic rings, which contain ortho-methoxy phenolic hydroxyl ($-\text{OH}$) groups. The molecular configuration of curcumin can be influenced by pH, temperature, and polarity. The β-diketone structure of curcumin causes its reduced solubility in aqueous media, poor light transmission, and precipitation of curcumin crystals. This limitation can be overcome by adjusting the pH of the medium.</p>	<p>pH responsiveness Nontoxic Eco-friendly Bioactive properties Good molecular stability within the pH range of 3 to 7, with a colour range from yellow to orange. At an acidic pH range, the bis-keto form of curcumin gives a yellow color. At a neutral pH, it turns orange. However, at a basic pH range of 8-12, curcumin undergoes a loss of a proton, converting from its keto form to an enol form, which results in an intense red color.</p>	<p>Rhizomes of turmeric (Curcuma longa L.).</p>	<p>48, 50-56</p>

<p>Phycocyanin</p>	 <p>Phycocyanin is a phycobiliprotein composed of two homologous subunits as α and β, which bind to three phycocyanobilin chromophores. Phycocyanobilin is a tetrapyrrole molecule consisting of four pyrrole rings connected to methine bridges, forming an extended conjugated double bond system. This structure provides blue color to the phycocyanin.</p>	<p>Water-soluble Fluorescent Thermolabile Sensitive to pH changes Anti-inflammatory properties. Anticancer properties. Antioxidant properties. Antimicrobial properties. Antidiabetic properties.</p> <p>It appears in dark blue under a neutral or mildly acidic pH range (pH 5-7). At high acidic pH levels, the color changes to a greenish hue, and at highly alkaline pH levels, the color turns purple.</p>	<p>blue-green algae/ spirulina (<i>Arthrospira platensis</i>) biomass</p>	<p>57-62</p>
<p>Quercetin</p>	 <p>Quercetin is a flavonol and its basic skeleton is composed of three ring structure with two benzene rings connected to a heterocyclic ring. Additionally, it contains five hydroxyl groups, which are key to the biological activity and the derivative formation of quercetin.</p>	<ul style="list-style-type: none"> • High antioxidant properties • Anti-inflammatory properties • Antimicrobial properties • Protecting food against UV light and oxidative damage. • The color stability of quercetin is within the pH range of 1-6 and undergoes autooxidation at pH levels of 7 or above. As the pH increases, the light-yellow color of the quercetin halochromic dye changes to a bright yellow color. 	<p>Extracted from herbs, fruits, vegetables, and red wine</p>	<p>51, 61, 63-66</p>

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<p>Betalain</p>	 <p>Betalains are water-soluble chromoalkaloids that contains nitrogen in their structures. Betacyanins (Red-purple pigments) and betaxanthins (yellow-orange pigments) are two main subgroups of Betalains. Betacyanins consist of betalamic acid chromophore and cyclo-3,4-dihydroxyphenylalanine. The second group, betaxanthins composed of betalamic acid with amines or amino acids. This pigment is highly sensitive to environmental conditions like pH, light, temperature, enzymatic actions, water activity, and oxygen exposure. The noticeable color shift of betalains occurs because of the stepwise degradation of betacyanins into colorless cyclo-DOPA 5-O-(malonyl)-β-glycoside and yellow betalamic acid in alkaline solutions.</p>	<ul style="list-style-type: none"> • Water-soluble pigment • Anti-inflammatory properties. • Antioxidant properties. • Antitumor properties. • Anticancer properties. • Antimicrobial effects • Betalain has a three times higher dyeing capacity compared to anthocyanin and is also highly stable within a pH range of 3-7. It undergoes a color change at alkaline conditions from dark red to yellow. 	<p>Red beetroots, various plants belong to Caryophyllales and also from mushrooms such as Hygrophorus, Hygrocybe, and Amanita. Some betalain sources toxic to humans.</p>	<p>67-72</p>
<p>Alizarin</p>	 <p>Alizarin belongs to the anthraquinone dye family, and the basic structure of this halochromic dye is based on anthraquinones. The chemical structure of Alizarin made up with three coplanar, six membered carbocyclic rings joined by two -OH substitutions located at the 1 and 2 positions of the anthraquinone core. Those -OH groups are phenolic and undergo ionization in response to pH changes, which gives the dye its halochromic nature.</p>	<ul style="list-style-type: none"> • Antitumor properties. • Antibacterial properties. • Antioxidant properties. • Non-cytotoxic properties. • At acidic conditions (pH 2-4), the color appeared yellow, and at pH 5-7, the monoanionic molecule resulted in a red color, the purple color intensified at alkaline conditions. 	<p>Roots of madder plants</p>	<p>48, 73-76</p>

<p>Anthocyanin</p>	 <p>The basic composition of anthocyanins is composed of glycosylated polyhydroxy and poly-methoxy structures, and its structure involves two aromatic rings which connected to a linear three carbon chain. Anthocyanin belongs to the polyphenol-based flavonoid family, and it is composed of aglycone anthocyanidin. The structural changes of anthocyanin in response to pH cause its color to change. For instance, at the pH 1-4 range, the predominant form of anthocyanins, flavylium cations, appeared red. Hydration and deprotonation in the pH 4-5 range generate the carbinol pseudo base, which has a colorless appearance. At pH 6-8, anthocyanins display a purple and blue hue because of the quinonoidal base form. A yellowish hue occurs at pH 8, indicating the formation of chalcone. Figure 14 illustrates the structural changes and the corresponding color of anthocyanin as the pH value varies.</p>	<ul style="list-style-type: none"> • Water soluble colorant • Rich in antioxidants • Anti-inflammatory properties. • Anti-obesity properties. • Anti-diabetic properties. • Antimicrobial properties. 	<p>Extracted from various sources, including fruits, flowers, and vegetable, with over 500 anthocyanin varieties identified. It shows color variation according to the extracted sources in response to pH changes (pH 2-12 range). Ex: strawberry red grapes red cabbage purple sweet potatoes blueberry Hibiscus sabdariffa blackberry Clitoria ternatea</p>	<p>48, 77-89</p>
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