

# Sustainable Food Technology

Accepted Manuscript

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## Sustainability Spotlight Statement

View Article Online  
DOI: 10.1039/D5FB00637F

**Chlorophylls** are the most abundant natural pigment which can be extracted and utilized in making functional packaging. This review investigated the recent advances in **chlorophylls** included packaging in improving and monitoring the shelf life of packed food. The area of this study is directly aligned with sustainability development goal 3, good health and well-being. The presence of **chlorophylls** in packaging could be beneficial in developing smart packaging materials.



1 **A review on chlorophyll-based active and intelligent packaging: chemistry, stability and**  
2 **applications in food freshness monitoring**

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11  
12 **Abstract**

13 Chlorophylls, a natural bioactive pigment that has recently gained significant attention as  
14 multifunctional component in sustainable food packaging due to its antioxidant, antimicrobial, and  
15 pH-sensitive properties. The incorporation of chlorophylls into biodegradable films, coating and  
16 encapsulated systems provides dual functionality acting as an active agent that extends shelf life  
17 by reducing oxidative damage and microbial spoilage and serving as an intelligent indicator that  
18 signals freshness through colorimetric or fluorescence changes. Despite these advantages,  
19 chlorophylls are inherently unstable and degrades rapidly under light, heat, oxygen. Additional  
20 challenges including large scale processing limitations and the high cost of advanced extraction  
21 and stabilization techniques restricts its practical implementation. This review highlights the  
22 chemical properties of chlorophylls, conventional and non-conventional extraction methods,  
23 stabilization strategies, and encapsulation approaches along with their integration into  
24 biopolymeric matrices within scalable and regulatory frameworks. Therefore, chlorophylls-based  
25 packaging represents a sustainable approach with strong potential to reduce microbial spoilage,  
26 enable real-time quality monitoring and contribute to food waste reduction.

27 **Keywords** Chlorophylls, sustainable, active & intelligent packaging, microencapsulation,  
28 bioactive compound, antioxidant

29



## 30 1. Introduction

31 Packaging is a crucial aspect in the food industry for the protection of food from external  
32 conditions<sup>1</sup>, such as temperature, humidity, and light, and maintaining quality, integrity, freshness,  
33 and safety during its shelf life by controlling gas and vapor exchange with the atmosphere  
34 preventing degradation while preserving sensory and nutritional qualities. Beyond protection,  
35 packaging significantly influences consumer perception as the appearance of the product are often  
36 associated with its freshness and overall quality ultimately affecting consumption patterns.  
37 Traditionally, food packaging has used materials such as paper, glass. Paper is lightweight,  
38 recyclable, and cost-effective while glass is inert, and provides excellent barriers to oxygen and  
39 moisture, suitable for acidic or fatty products. Further, plastic being versatile with excellent barrier  
40 properties ideal for wide range of foods<sup>2</sup>. In recent years there has been a growing emphasis on  
41 environmentally friendly and sustainable packaging alternatives. This shift has driven research and  
42 innovations in biomaterials for food packaging, leading to the rise in the application of advanced  
43 techniques such as coatings, antimicrobial, antioxidant packaging and modified atmosphere  
44 packaging<sup>3,4</sup>.

45 Active and intelligent packaging has garnered attention for its ability to extend functionality  
46 beyond passive containment. Active packaging interacts with the food component to extend the  
47 shelf life mitigating the microbial spoilage<sup>5</sup> while intelligent packaging monitors the  
48 environmental factors<sup>6</sup> through visible color changes or variations in fluorescence provide  
49 non-invasive information on food freshness, spoilage<sup>7</sup>, or changes in the package  
50 microenvironment<sup>8</sup>. Addressing issues such as temperature fluctuations, microbial contamination,  
51 and package integrity these packaging systems contribute to reduced food waste, improved  
52 traceability, and decreased risk of food borne illness<sup>9,10,11</sup>.

53 Chlorophylls are naturally derived pigment with antioxidant properties and nutritional value. It  
54 eliminates free radicals and protecting the cells from oxidative damage. Several studies reported  
55 the promising antioxidant, anti-inflammatory and anti-cancer properties of chlorophylls and its  
56 derivatives but oxidation of chlorophylls phenolic compounds can contribute to pigment loss is  
57 one of the challenges. Despite these properties, chlorophylls are sensitive to light and oxidation,  
58 but advanced techniques such as encapsulation with different carriers, such as  
59 carboxymethylcellulose, can increase its stability and performance<sup>17,18,19</sup>.



60 There have been recent advancements in biodegradable films and smart packaging showing the  
61 potential of natural extracts and nanoparticles as functional additives. Numerous studies concluded  
62 the efficiency of chlorophylls -based packaging, highlighting the pigment's amphiphilic structure,  
63 pH sensitivity and antioxidant activity which has the potential advantages in both active and  
64 intelligent applications. For example, sodium iron chlorophyllin incorporated into chitosan/ gelatin  
65 films enhanced antimicrobial and antioxidant activity, improved UV barrier properties, and  
66 effectively delayed spoilage in fresh cut chilli peppers, showed its potential as an active packaging  
67 agent<sup>12</sup>. Similarly, chlorophyllin incorporated into photoactive coatings generated reactive oxygen  
68 species (ROS) enabling effective inactivation of *L. monocytogenes* demonstrating both  
69 antimicrobial activity and light responsive packaging behavior<sup>13</sup>. Further, another study reported  
70 that cornstarch chlorophyllin composite films under light exposure generated ROS resulting in  
71 lower microbial and nitrogen levels compared to control samples and maintaining acceptable  
72 quality of shrimp up to 4 days<sup>14</sup>. Moreover, chitosan based films incorporating chlorophylls with  
73 curcumin exhibited enhanced barrier and mechanical properties, exhibited significant  
74 antimicrobial activity and extended the shelf life of the cherries and pork<sup>15</sup>. The integration of  
75 chlorophylls pigment has significantly expanded the functionality of food packaging<sup>16</sup>. These  
76 natural components are especially found in plant extracts such as green tea and basil and are  
77 explored for smart packaging, due to its unique chemical and functional properties.

78 The aim of this review is to comprehensively analyze recent advancements in chlorophylls-based  
79 smart packaging for maintaining and monitoring food quality and freshness, focusing on key  
80 properties of chlorophylls and further its relevance in the packaging, active and intelligent  
81 packaging. It also discusses the development and characterization of chlorophylls -incorporated  
82 films and coatings, while analyzing their applications in both active and intelligent packaging.  
83 Furthermore, the review also highlights the existing knowledge gaps and areas require further  
84 development for commercial optimization and broader adoption.

## 85 **2. Chlorophylls chemistry and properties**

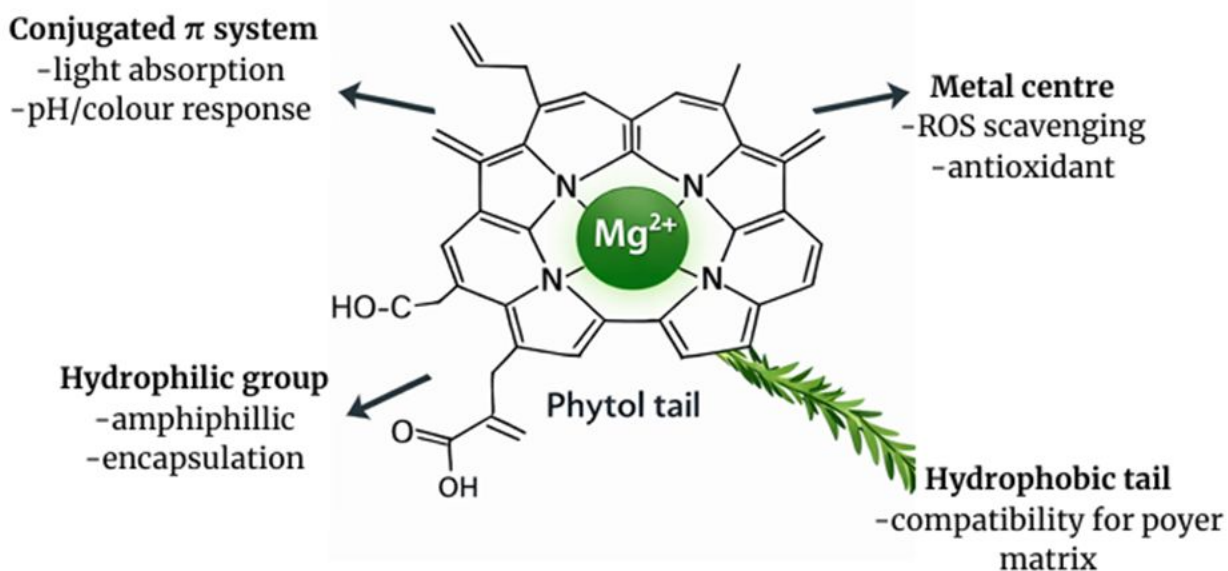
86 Chlorophylls has a unique chemical structure and properties which are fundamentally responsible  
87 for its characteristic color, potent antioxidant activity, and suitability for packaging technologies



88 aimed at maintaining and monitoring food freshness. Figure 1 shows the molecular structure of  
89 chlorophylls, highlighting the porphyrin ring with the central magnesium ion and the phytol tail.

## 90 2.1 Molecular structure of chlorophylls

91 Chlorophylls are a complex organic molecule made of a porphyrin ring which is hydrophilic, a  
92 central magnesium ion, and side chain containing the phytol chain, which is strongly hydrophobic.  
93 The porphyrin ring, a macrocyclic conjugated tetrapyrrole structure which contains four nitrogen  
94 atoms in pyrrole groups; responsible for absorbing light energy, while the magnesium ion acts as  
95 an electron acceptor. Various conjugated double bonds impart an advantage to the ability of  
96 chlorophylls to absorb visible light <sup>19,21,22</sup>.



97

98 **Figure 1** Molecular structure of chlorophyll<sup>23,24</sup>

99 There are several forms of chlorophylls, including chlorophyll a, b, c, d, and e, with chlorophyll a  
100 being the most common in plants. Chlorophylls efficiently absorb light in the red and blue regions  
101 of the spectrum, exhibiting peak absorption around 430 and 662 nanometers, respectively.  
102 chlorophyll b, which mainly functions to protect chlorophyll a from excessive light, absorbs light  
103 optimally near 453 nanometers. Other chlorophylls forms such as chlorophyll c, d, and e are found  
104 in diverse organisms like algae and differ in their absorption spectra and biological roles. For



105 instance, chlorophyll c absorbs primarily in the blue-green region, chlorophyll d in the red region,  
106 and chlorophyll e in the far-red region of the spectrum<sup>19</sup>. Structurally, chlorophylls molecules vary  
107 chiefly in the degree of saturation in their pyrrolic rings and in functional groups attached to the  
108 macrocycle. For example, chlorophyll b differs from chlorophyll a by having an aldehyde group  
109 instead of a methyl group at the C7 position, which results from enzymatic oxidation of the methyl  
110 group catalyzed by oxygenase. chlorophyll c contains a fully unsaturated phytyl system  
111 (with a double bond between C17–C18), whereas chlorophylls a, d, and f contain partially  
112 saturated phytyls. These structural differences significantly affect the absorption properties:  
113 chlorophyll a, d, and f show approximately balanced absorption intensities in blue, red, and green  
114 regions, while chlorophyll c absorbs weakly in the red and more strongly around 450 nm. This  
115 variety in the absorption spectra are mirrored in their distinctive green hues, chlorophyll a appears  
116 bluish-green, chlorophyll b bright green, chlorophyll c yellowish-green, chlorophyll d bright forest  
117 green and chlorophyll f emerald green. This diversity makes these pigments promising candidates  
118 as natural colorants<sup>19</sup>. Chlorophyll molecules possess two distinct regions: a hydrophilic  
119 macrocycle and a hydrophobic phytyl tail. The most hydrophilic segments of the macrocycle  
120 include the cyclopentanone ring and a propionic acid ester group at the C17 position. This  
121 amphiphilic nature influences solvent selection for chlorophyll extraction, impacting yield and  
122 purity important for technological applications<sup>22</sup>.

123 Chlorophyll's distinctive structure positively influences its function in the packaging. For example,  
124 the hydrophilic porphyrin macrocycle promotes the antioxidant activity allowing chlorophylls and  
125 its derivatives act as singlet oxygen quenchers and free radical scavengers. This protective effect  
126 against lipid oxidation attributes not only to their free radical scavenging capacity but also to their  
127 ability to chelate pro-oxidant metal ions such as Fe<sup>2+</sup> and Cu<sup>2+</sup> which retards lipid peroxidation in  
128 food <sup>5</sup>. On the other hand, chlorophylls acts as a photosensitizer, absorbing visible light and  
129 transferring the energy to molecular oxygen to produce singlet oxygen and other reactive oxygen  
130 species<sup>25</sup>.

## 131 2.2 Extraction methods

132 Chlorophylls are widely extracted for industrial applications from stinging nettle, spinach, algae,  
133 silkworm excreta, alfalfa, pine needles, other pasture grasses and plant harvest by-products<sup>26</sup>.



134 Several methods were used to extract chlorophylls of natural ingredients from the plants, such as  
 135 aqueous extraction, enzyme-assisted extraction, alcoholic/organic solvent extraction, and  
 136 supercritical/subcritical extraction<sup>27</sup>. Several methods were used to extract chlorophylls from the  
 137 plants, such as aqueous extraction, enzyme-assisted extraction, alcoholic/organic solvent  
 138 extraction, and supercritical/subcritical extraction<sup>26</sup> summarized in table 1. Extraction can be  
 139 carried out in various ways, involving different solvents and preparation methods. Variables such  
 140 as pressure, temperature, contact efficiency, and time are critical. Since chlorophylls are sensitive  
 141 to extreme light exposure, pH, and temperature, solvent selection must consider factors like  
 142 density, viscosity, heat of evaporation, cost, environmental and health effects. The solvent should  
 143 not react with or damage the extracted compound and must not be corrosive. Different methods  
 144 are tailored for different microalgae species<sup>28</sup>.

145 **Table 1** Extraction of chlorophylls from different source and extraction method

Extraction method	Raw material/food	Yield	Purity	Conditions	References
Solid- liquid extraction	Spinach (solid phase)	NA	Chlorophyll a/b ratio 4.8 in ethyl acetate phase	25 °C, 30 min, 600 rpm agitation; mild energy (centrifugation + vacuum drying); solvent recovery-condenser	29
Extraction by methanol (1:50) atmospheric	Cucumber	Chlorophyll a: 480.14 mg/100 g Chlorophyll b: 342 mg/100 g	Chlorophyll a/b ratio $\approx$ 1.4 (480/342); CV: 52% chlorophyll a, 48% chlorophyll b	1:5 ratio (1 g:50 mL methanol); centrifugation 3000 rpm, 10 min; room temp.	30
Enzyme-assisted extraction (Pectinex Ultra SP-L)	Spinach pulp	50.747 mg TCC/100 g spinach pulp (optimized); 39% higher Zn-chlorophylls derivative yield vs non-enzymatic	NA	8% enzyme (Pectinex Ultra SP-L), 45 °C, 30 min; followed by ethanol extraction (2.5:1 ratio, 60 °C, 45 min)	5
Subcritical fluid extraction (R134a +	<i>Laminaria japonica</i> Aresch (seaweed)	Chlorophyll a: 2.326 g/kg (optimized)	NA	Optimized: 324.13 K (51.13 °C), 17 MPa, 4.73% ethanol cosolvent; RSM	31



ethanol cosolvent)				Box–Behnken design	
Subcritical CO <sub>2</sub> extraction	<i>Nigella sativa</i> seeds	Volatile oil: 66.6 wt.% of oleoresin	NA	70 bar, 30 °C, 2 hours	32
Supercritical CO <sub>2</sub> extraction with a static modifier	<i>Scenedesmus obliquus</i> (microalga)	Chlorophyll a: 0.848 mg/g biomass; chlorophyll b: 0.356 mg/g biomass; chlorophyll c: 0.018 mg/g biomass	NA	200–250 bar; 40–60 °C; CO <sub>2</sub> flow rate varied; 7.7% v/v ethanol cosolvent	33
Ultrasound-assisted green solvent extraction	<i>Scenedesmus obliquus</i> , <i>Arthrospira platensis</i> (microalgae)	Optimized ultrasound + ethanol/ionic liquid solutions; carotenoid/chlorophyll extracts potent peroxyl scavengers (5.94–26.08× α-tocopherol)	NA	Ultrasound extraction; ethanol + ethanolic ionic liquid solutions; optimized extraction time, repetitions	34

146

147 Over the years, several approaches have been reported to remove chlorophylls from botanical  
 148 crude extracts (CEs). Most rely on solid-phase extraction (SPE) with stationary phases such as  
 149 Diaion HP-20 for initial fractionation. Charcoal has also been used to remove chlorophylls or other  
 150 pigments entirely from CEs without fractionation of other constituents. However, these methods  
 151 often eliminate other unspecified, potentially bioactive compounds along with chlorophylls.  
 152 Detailed protocols vary by author, and preservation of the original phytochemical profiles is  
 153 generally not assessed<sup>35</sup>.

154 Liquid-liquid or countercurrent separation (CCS) methods are effective for isolating large  
 155 quantities of target compounds from complex matrices. CCS offers ease of use, affordable cost,  
 156 reduced solvent consumption, and efficient equipment performance, making it valuable in natural  
 157 product research. CCS has been applied to isolate plant pigments like anthocyanins and  
 158 carotenoids from chlorophylls -enriched extracts obtained from grass, spinach, and other plant  
 159 materials. However, CCS suitability for chlorophylls removal remains under evaluation. In CCS,  
 160 both the stationary and mobile phases are liquids forming a biphasic solvent mixture. This mixture  
 161 solubilizes extracts entirely while balancing partitioning of target compounds. Phytochemicals in



162 both phases can be recovered with solvent evaporation, making CCS effectively loss-free when  
163 analytes are non-volatile. A recent study implemented CCS to selectively remove chlorophylls  
164 from botanical CEs to produce chlorophylls -free "degreened" Knock-Out Extracts (chlorophylls  
165 -KOE). This method enables subtraction of assay interference compounds while optimally  
166 recovering other phytochemicals. The study produced chlorophylls -KOE from three  
167 botanicals: *Epimedium sagittatum*, *Senna alexandrina L.*, and *Trifolium pratense L.*, evaluating  
168 reproducibility and selectivity with HPLC, UHPLC-UV/MS, LC-MS, and <sup>1</sup>H NMR  
169 spectroscopy<sup>35</sup>.

170 Supercritical/subcritical CO<sub>2</sub> extraction selectively isolates components suitable for heat-sensitive  
171 products. Traditionally, chlorophylls extraction uses liquid solvents followed by evaporation and  
172 drying at high temperatures, risking degradation. Supercritical/subcritical CO<sub>2</sub> extraction operates  
173 at low temperatures, preventing thermal degradation. CO<sub>2</sub> is inert, non-toxic, non-flammable,  
174 inexpensive, and easily separated from extracts without heating, producing solvent-free extracts.  
175 Stability tests on chlorophylls from *katuk* leaves showed decreased content at high temperatures,  
176 indicating supercritical/subcritical CO<sub>2</sub> as the preferred extraction method to preserve  
177 chlorophylls<sup>27</sup>. Enzyme-assisted extraction is gaining attention for eco-friendly processing.  
178 Targeted enzymes like pectinases, cellulases, and hemicellulases degrade cell walls, increasing  
179 solvent pre-treatment efficiency, reducing solvent use, or increasing bioactive compound yield.  
180 Used widely in juice processing and beer clarification, enzymatic pretreatment enhances  
181 extractability. Enzyme-assisted extraction of Zn-chlorophylls derivatives from spinach pulp under  
182 optimized conditions (8% enzyme concentration, 45°C, 30 min) increased yield by 39%<sup>5</sup>.

## 183 **2.3 Key properties of chlorophylls relevant to packaging**

184 The chemical structure of chlorophylls determines its bioactivity, influences potential health  
185 benefits<sup>19</sup>. As mentioned earlier, chlorophylls are among the most prominent bioactive  
186 compounds, having positive health impacts and high antioxidant activity. They are used as natural  
187 food coloring agents and have wound healing and anti-mutagenic properties<sup>20</sup>.

### 188 **2.3.1 Antioxidant activity**

189 Chlorophylls is a strong antioxidant that has the capability to scavenge free radicals and chelate  
190 metal ions (e.g., Fe<sup>2+</sup>), hence they prevent the oxidative damage to lipids and cells. This activity  
191 inhibits a major cause of food spoilage which is lipid peroxidation. It also protects against oxidative



192 DNA damage and reduces reactive oxygen species (ROS) formation, helping in preserving food  
193 freshness by delaying spoilage processes. Natural chlorophylls possess antioxidant properties  
194 making them promising candidates for preventing or mitigating the formation of reactive species.  
195 Lanfer-Marquez et al., (2005)<sup>36</sup> demonstrated that Cu-chlorophylls in displayed higher antioxidant  
196 activity compared to natural chlorophylls, highlighting the influence of the chelated metal in the  
197 porphyrin ring on the strength of the antioxidant capacity<sup>19</sup>. Additionally, a study showed that the  
198 kale chlorophylls which were microencapsulated in isolated whey protein, resulted in increased  
199 antioxidant activity by 20% as assessed through the DPPH method<sup>37</sup>.

### 200 **2.3.2 Photoactivity and reactive oxygen species generation**

201 Chlorophylls are very unstable compound and the stability is highly affected by pH, temperature,  
202 heat, and light<sup>21</sup>. When exposed to irradiation, it can generate singlet oxygen ( $^1O_2$ ) through  
203 photosensitive reactions causing discoloration<sup>20</sup>. The chlorophylls content decreased faster under  
204 light than in the dark because of singlet oxygen. Samples with added lipids showed lower and  
205 slower degradation of chlorophylls than in samples without lipids. High pressure high temperature  
206 processing results in the degradation of chlorophyll *a* and chlorophyll *b*. Both chlorophylls were  
207 highly degraded at 117°C<sup>21</sup>. This singlet oxygen generation thereby can prolong the food shelf life  
208 causing microbial inactivation; a property that can be exploited in active packaging to extend shelf  
209 life.

### 210 **2.3.3 Color properties and sensitivity to pH, light, and oxygen**

211 As mentioned in the earlier section, chlorophylls is highly unstable compound, sensitive to light;  
212 therefore, it becomes difficult to keep its molecules intact with a green color; a photosensitive light  
213 harvesting pigment with special electronic properties<sup>38</sup>. Furthermore, it is susceptible to heat,  
214 oxygen, and chemical degradation. The pH has an effect on chlorophylls degradation as well<sup>39</sup>.  
215 Koca et al., (2006)<sup>40</sup> studied the effect of pH on the chlorophylls degradation and visual green  
216 color loss in blanched green peas were studied at 70, 80, 90 and 100°C in buffered solutions of pH  
217 5.5, 6.5 and 7.5. The rate constants of green color loss and chlorophylls degradation decreased  
218 with increasing pH, indicating that the green color was retained at higher pH conditions. It was  
219 found that chlorophyll *a* degraded faster than chlorophyll *b* at all pH values for each temperature  
220 applied. The results revealed that chlorophyll *a* was more susceptible to thermal degradation than  
221 chlorophyll *b* in acidic conditions<sup>40</sup>.

### 222 **2.3.4 Stability challenges and encapsulation**



223 The chlorophylls incorporation into the food matrix is challenging because of its instability  
224 towards light, oxygen and pH and poor bioavailability. Encapsulation is an excellent process to  
225 enhance its bioaccessibility, digestibility, and controlled release<sup>21</sup>. For example, the encapsulation  
226 carriers like maltodextrin (MD) and whey protein isolate (WPI) can provide multi-layered  
227 protection. The MD/WPI walls acts as light barriers through scattering and UV absorption, oxygen  
228 diffusion barriers limiting the access of reactive oxygen species, and pH buffers via hydrogen  
229 bonding between WPI amine groups and MD hydroxyls with chlorophylls's porphyrin nitrogen.  
230 Freeze-dried microcapsules of chlorophylls (MD/WPI from *Ulva intestinalis*) retained 38.12 %  
231 green color after light exposure versus 1.84 % for unencapsulated chlorophylls, whereas the  
232 chemical stability and retention rates increased significantly ( $p < 0.05$ ). Spray-dried microcapsules  
233 showed DPPH scavenging of 67.5% and freeze-dried 79.1%. Thus, chlorophylls can be protected  
234 from bad light conditions and have a longer shelf-life during storage via microencapsulation and  
235 possible hydrogen bonding with MD and WPI complexes<sup>41</sup>.

236 Emerging stabilization technologies such as core-shell nanoparticle encapsulation using zein,  
237 casein, or whey protein isolate as wall materials offers enhanced protection for chlorophylls in  
238 food packaging applications. Encapsulation efficiencies of chlorophylls retention after 10 days  
239 were 83.6–96.3% and 39–97.8% at pH 3.0 under light/acidic stress; compared to 40% for free  
240 chlorophylls. These technologies therefore offer superior photostability and controlled delivery for  
241 active packaging applications<sup>42</sup>.

### 242 **3. Chlorophylls based active & intelligent packaging development and key properties**

243 The active and intelligent smart packaging based on chlorophylls is a promising method which  
244 leverages the natural bioactivity and colorimetric responsiveness of chlorophylls extracts from  
245 various sources including plants or algae. Active compounds as intelligent indicators enhance the  
246 stability and function of packaging or coating matrices. These pigments can change color because  
247 of environmental stimuli, such as temperature, pH, oxygen, light or microbial activity, providing  
248 visual cues to consumers about the freshness, spoilage, ripeness, or contamination of food<sup>43</sup>. The  
249 packaging is typically developed by incorporating chlorophylls or microencapsulated chlorophylls  
250 into biodegradable polymer matrices like alginate, chitosan, gelatin, or pectin. Common  
251 fabrication methods include solution casting, extrusion<sup>44</sup> designed to preserve the functional and  
252 colorimetric properties of chlorophylls during processing.

#### 253 **3.1 Film/coating fabrication using chlorophylls**



254 As mentioned earlier in the introduction part, chlorophylls have currently emerged to be a  
 255 promising bioactive compound for the development of active and intelligent packaging films and  
 256 coatings because of its unique chemical, antioxidant, and colorimetric properties. For example,  
 257 edible coatings are revolutionizing food preservation by offering a sustainable and effective  
 258 solution to key industry challenges. They are produced from natural biopolymers such as proteins  
 259 (e.g., gelatin, zein), polysaccharides (e.g., starch, chitosan, alginate), and lipids, these coatings  
 260 form a thin, edible layer on food surfaces. These biodegradable and edible matrices reduce  
 261 moisture loss, protects against oxidative damage, and limits microbial growth, thereby extending  
 262 shelf life while preserving food quality. Enhanced with natural additives like essential oils and  
 263 antioxidants, these coatings offer antimicrobial benefits and contribute to health<sup>45</sup>. Figure 2 shows  
 264 the overall process for chlorophylls extraction, incorporation into the matrix, and subsequent  
 265 analysis leading to the development of chlorophylls-integrated film and food packaging  
 266 application. Lv et al. (2023)<sup>16</sup> mentioned a study film containing chlorophylls and chitosan having  
 267 the potential for generating color within the colorimetric temperature range of 50–75 °C. The  
 268 system will undergo an irreversibly color change from green to yellow when exposed to this  
 269 temperature range.



270  
 271 *Figure 2 Schematic representation of chlorophylls-based active film formation*<sup>46 47</sup>

### 272 3.1.1 Casting method



273 A lab or pilot scale method, also known as solvent casting, is a rather simple and one of the most  
274 common techniques of edible film formation<sup>48</sup>. It involves manufacturing films from biopolymers,  
275 and includes the following steps: (i) solubilizing the biopolymer in a suitable edible and non-toxic  
276 solvent, such as ethyl alcohol or water. The solubilization step ensures an even dispersion of the  
277 biopolymer in the solvent. This solubilization step is crucial as the film formation depends on the  
278 polymer's solubility rather than melting; (ii) casting of the solution in a predefined mould, where  
279 it forms a gel structure (cohesive film adhering to the mould) as the solvent evaporates with time;  
280 (iii) drying the cast solution layer, which is necessary to form a cohesive film, but with the moisture  
281 content maintained at 5% to 8% to prevent wrinkling and tearing of the film during peeling.  
282 Moreover, optimization of the drying temperatures and methods for a particular film is done to  
283 produce high-quality edible films<sup>49</sup>. A study investigated the formation of fractal structures in  
284 chlorophylls films prepared by casting technique using two different chlorophylls concentrations  
285 solution. Results were based on DLA model analyzed on optical microscopy showed that  
286 chlorophylls cast films formed distinct fractal aggregates at both 0.50 g/L and 0.12 g/L  
287 concentrations. Further image analysis showed a consistent fractal dimension for about 1.55 for  
288 all samples which suggests fast aggregation. There was no effect of change in concentration on the  
289 fractal dimension, suggesting that aggregation dynamics were stable across the tested range<sup>50</sup>.

### 290 3.1.2 Extrusion

291 The extrusion method is one of the underexplored techniques for manufacturing edible films, but  
292 is now attracting more attention, particularly for starch/protein combinations. Extrusion forms  
293 films through a thermomechanical process<sup>49</sup>. There are various steps involved in extrusion process;  
294 preparation of formulations using different composition of raw materials and their mixing,  
295 blending the mixture in an extruder in order to pelletize all film-forming ingredients, cutting  
296 extrudates into pellets through the pelletizer, drying pellets in a hot-air oven, followed by extruding  
297 the pellets into sheets through the second extruder, and finally blowing the mixed resins into a film  
298 by a blown film extruder. Extrusion process often provides the films with acceptable mechanical  
299 properties and good thermal stability<sup>51</sup>.

300 In comparison with widely used solvent casting method, extrusion of corn starch and poly  
301 (butylene adipate-co-terephthalate) (PBAT) blends with intact *Chlorella pyrenoidosa* biomass  
302 utilizes the amphiphilic nature of chlorophylls and algal lipids to improve compatibility for  
303 hydrophilic starch and hydrophobic PBAT phases. The results indicated that films containing a



304 higher content (5.0%) of intact *Chlorella pyrenoidosa* biomass exhibited superior tensile strength  
305 ( $4.37 \pm 0.24$  MPa) and elongation ( $88.43 \pm 6.8\%$ ) compared to films with disrupted biomass  
306 suggesting better dispersion and interactions between the phases which enhances homogeneity  
307 because of amphiphilic compatibilization further confirmed by SEM. Furthermore, these films  
308 displayed lower water vapor permeability ( $5.19 \times 10^{-11}$  g m<sup>-1</sup> s<sup>-1</sup> Pa<sup>-1</sup>), enhancing their barrier  
309 properties. Films produced by blown extrusion from starch, PBAT, and *Chlorella pyrenoidosa*  
310 microalgae biomass have the technological potential to be used as packaging for food products.  
311 Starch and PBAT blends are widely studied<sup>52</sup>. Overall, the films produced by blown extrusion  
312 blending starch or biodegradable polymers incorporating microalgae biomass containing  
313 chlorophylls have shown good mechanical and antioxidant properties.

### 314 3.1.3 Encapsulation

315 Encapsulation technology has been extensively used to enhance the stability, specificity, and  
316 bioavailability of essential food ingredients<sup>53</sup>. Microencapsulation is a nanotechnology method  
317 which can be used to prevent damage to bioactive compounds by protecting the encapsulated  
318 compounds during edible film processing. The enrichment of alginate-based edible film with  
319 chlorophylls microcapsules enabled to blend into the alginate-based edible film where they  
320 migrated and slowly released compounds which were able to prevent or retard some microbial  
321 growth on the fish bubble snack product tested. The enrichment increased the film thickness,  
322 improved the surface texture of the film, increased the resistance of the coated food (fish bubble  
323 snacks) to the growth of the mold *Rhizopus sp.*, and increased resistance to the proliferation of *E.*  
324 *coli* but not of *S. aureus*. At room temperature the antifungal effect was twice as strong as for the  
325 same product without enrichment of the edible film coating. The antifungal properties of the  
326 enriched edible film extended the shelf life of the product tested<sup>54</sup>.

327 Another study showed the encapsulation of chlorophylls with MD and WPI as carriers applying  
328 both spray drying and freeze-drying methods was done in order to increase the stability of  
329 chlorophylls extracted from *Ulva intestinalis* algae. The optimum combination of wall and core  
330 materials to achieve the highest response including encapsulation efficiency (EE) and chlorophylls  
331 content (CC) was obtained by response surface methodology and central composite design. The  
332 optimal chlorophylls microcapsule obtained was chosen for subsequent tests containing solubility,  
333 moisture content, and antioxidant properties. The results showed that the highest EE and CC were  
334  $90.27 \pm 0.21\%$ ,  $55.36 \pm 0.36$  µg/mL, and  $90.46 \pm 0.62\%$ ,  $85.85 \pm 0.43$  µg/mL, respectively in SD



335 and FD. The microcapsules produced by the freeze dryer (FD) had higher antioxidant activity ( $79.1$   
336  $\pm 0.24$ ) than the microcapsules produced by the spray dryer (SD) ( $67.5 \pm 0.16$ ). The highest  
337 solubility ( $95.32\%$ ) and the lowest moisture content ( $3.7 \pm 0.05$ ) were related to the SD. Freeze  
338 drying method (FDM) had the highest EE ( $91.2\%$ ), CC ( $89.67 \mu\text{g/mL}$ ), and antioxidant properties  
339 ( $79.1\%$ )<sup>55</sup>.

#### 340 **3.1.4 Compression molding**

341 Either thermo-compression or ultrasonic compression binds the film-forming materials into a  
342 desirable shape and thickness. An ultrasonic welder is used for welding the film materials; post  
343 compression, the welded materials are cut and processed to elaborate sustainable edible packaging  
344 systems. This technique has not yet gained popularity for manufacturing edible films but is a fast  
345 and economical method and needs to be adapted to suit the edible film packaging industry<sup>48</sup>.

346 The modern packaging, embedded with sensors, not only interacts with the food product but also  
347 assesses its surrounding conditions, providing stakeholders with real-time information and  
348 insights, detecting freshness, pathogens, pH levels and other environmental changes, offering more  
349 insight than traditional measures such as weight or appearance. Compared to other natural food  
350 colorants, the color of chlorophylls is relatively stable, making it a less sensitive and observable  
351 colorimetric indicator in intelligent biodegradable packaging. This suggests that it may be less  
352 applicable for providing real-time information<sup>56</sup>. The study by Chavoshizadeh et al.,(2020)<sup>17</sup>  
353 introduces a wheat gluten-based biodegradable film incorporating chlorophylls, highlighting its  
354 role in enhancing the shelf life of sesame oil and indicating expiration dates. The film reduces oil  
355 oxidation, evident from the halved peroxide value and changes color from green to yellow in  
356 response to oil quality after a prolonged storage period. Integrating chlorophylls into edible films  
357 or coatings has a significant effect enhancing their physical and functional properties. For  
358 example, incorporated chlorophylls microcapsules into an alginate-based edible film showed  
359 enhanced film thickness and improved surface texture which ultimately resulted in a smoother,  
360 more homogeneous film without agglomeration<sup>54</sup> (discussed in table 2). This resulted in enhanced  
361 morphology of the surface, improving the barrier properties of the film while protecting the food  
362 product. Moreover, scanning electron microscopy (SEM) for microstructure showed that  
363 chlorophylls -enriched films exhibited a clearly different and smoother surface compared to  
364 control films without chlorophylls. Also, Fourier transform infrared spectroscopy (FTIR) analyses  
365 showed interactions between chlorophylls microcapsules and the film matrix. The films based on



366 their function demonstrated the enhanced antimicrobial activity by slowly releasing chlorophylls  
367 compounds that inhibited the growth of molds such as *Rhizopus sp.*; and bacteria such as  
368 *Escherichia coli* on food products, thereby extending shelf life<sup>54</sup>.

### 369 370 **3.2 Key properties of chlorophylls based smart packaging**

#### 371 **3.2.1 Color**

372 Films enriched with chlorophylls microcapsules exhibits pronounced greenish intensity, and the  
373 value increases with higher microcapsule content. A study by Dewi et al., (2022) showed that a\*  
374 color values shift to more negative numbers (e.g., from  $-0.41 \pm 0.09$  to  $-11.25 \pm 0.24$ ), reflecting  
375 the deepening greenness imparted by chlorophylls encapsulation<sup>54</sup>. Moreover, microencapsulation  
376 or film matrix stabilization significantly enhance the green color stability under environmental  
377 stress (heat, light, pH change) shown by the study in which freeze-dried microcapsules retain up  
378 to 38% of chlorophylls color after light exposure, while unencapsulated chlorophylls may retain  
379 as little as 1.84%<sup>41</sup>. The intensity and stability of green color can also act as a freshness or shelf-  
380 life indicator for intelligent packaging due to visual changes upon degradation. Also review papers  
381 focused on chlorophylls integration into smart packaging systems with color changes serving as  
382 effective freshness and shelf-life indicators, emphasizing their application potential<sup>25</sup>.

383 Chlorophylls pH-driven color change occurs because of the loss of  $Mg^{2+}$  from the porphyrin ring,  
384 the bright green color of the chlorophylls turns to olive brown <sup>57</sup>. Resulting in absorption bands  
385 shift from  $\sim 430/662$  nm (green) to  $\sim 410/650$  nm (yellow/brown) species <sup>58</sup>. Koca et al. (2006)<sup>40</sup>  
386 observed that chlorophyll a degraded faster than chlorophyll b across pH 5.5–7.5 at higher  
387 temperatures, and due to reduced pheophytinization, the green color loss rate constants decreased  
388 at higher pH.

#### 389 **3.2.2 Mechanical properties**

390 Chlorophylls often act as a natural filler or plasticizer modifying the tensile strength, elongation,  
391 modulus, and impact resistance. For example, in polypropylene blends, low chlorophylls content  
392 (0.1–0.25 wt%) reduce rigidity and ductility slightly, but when there is an optimal concentration  
393 (0.5 wt%), it acts as a plasticizer increasing elongation and impact resistance while maintaining  
394 tensile strength comparable to pure polypropylene. Higher chlorophylls concentrations (>0.5  
395 wt%) typically reduce mechanical strength due to filler agglomeration disrupting polymer chain  
396 cohesion<sup>43</sup>. Also, Dewi et al., (2022)<sup>54</sup> showed in their study the chlorophylls microencapsulation



397 improved tensile strength by interacting with film matrices and enhancing polymer network  
398 bonding while increasing film thickness.

### 399 **3.2.3 Barrier properties**

400 Chlorophylls incorporation leads to thicker films with improved moisture and solubility barriers,  
401 promoting longer shelf life and quality retention in packaged foods. Study discussed in table 2 in  
402 which cornstarch-Chlorophylls in composite films added with coconut oil, oregano essential oil,  
403 and beeswax showed significant reduction in moisture content (~12.58%), water solubility  
404 (~15.41%), swelling ability (~29.30%), and water vapor permeability ( $\sim 1.78 \times 10^{-10} \text{ gm}^{-1}\text{s}^{-1}\text{Pa}^{-1}$ )  
405 were observed. These changes enhance the ability of packaging films to maintain food quality by  
406 limiting moisture-induced spoilage and degradation<sup>59</sup>.

### 407 **3.2.4 Antioxidant and antimicrobial properties**

408 The antioxidant and antimicrobial performance of packaging films depends on many factors, such  
409 as the molecular interaction between natural pigments and polymer substrates, the quantity of  
410 added natural pigments, structural modifications, and environmental conditions<sup>16</sup>. Moreover, the  
411 encapsulation of chlorophylls has shown to preserve the antioxidant/antibacterial properties of the  
412 film during a long time. It is noteworthy that chlorophylls changes color in nitrate media, which  
413 gives the film the ability to be used in the future as a smart film in identifying nitrate compounds  
414 used in food<sup>18</sup>. For example, biodegradable films based on wheat gluten modified with  
415 chlorophylls /polypyrrole showed that the addition of both increased the antioxidant activity of the  
416 films as mentioned in the table 2<sup>17</sup>. Chlorophylls have a notable antibacterial property when it is  
417 incorporated into various food packaging films. For example, López-Carballo et al. (2008)<sup>60</sup>  
418 developed gelatin films incorporating water-soluble chlorophylls in salts which showed substantial  
419 reduction of *Staphylococcus aureus* and *Listeria monocytogenes* growth, showing that s  
420 derivatives act as effective photosensitizers with antimicrobial effects under light exposure.

421

## 422 **4. Application of chlorophylls -based packaging film/coating**

423 Chlorophylls, a naturally occurring green pigment essential for the photosynthetic process in  
424 plants, has garnered attention in sustainable packaging research due to its biocompatibility, eco-  
425 friendly characteristics, and functional versatility<sup>20</sup>. It exists in several forms, primarily classified  
426 into six types i.e., chlorophyll a, b, c, d, e, and f. Among these, chlorophyll a and b are predominant  
427 in higher terrestrial plants, with chlorophyll a serving as the principal photosynthetic pigment

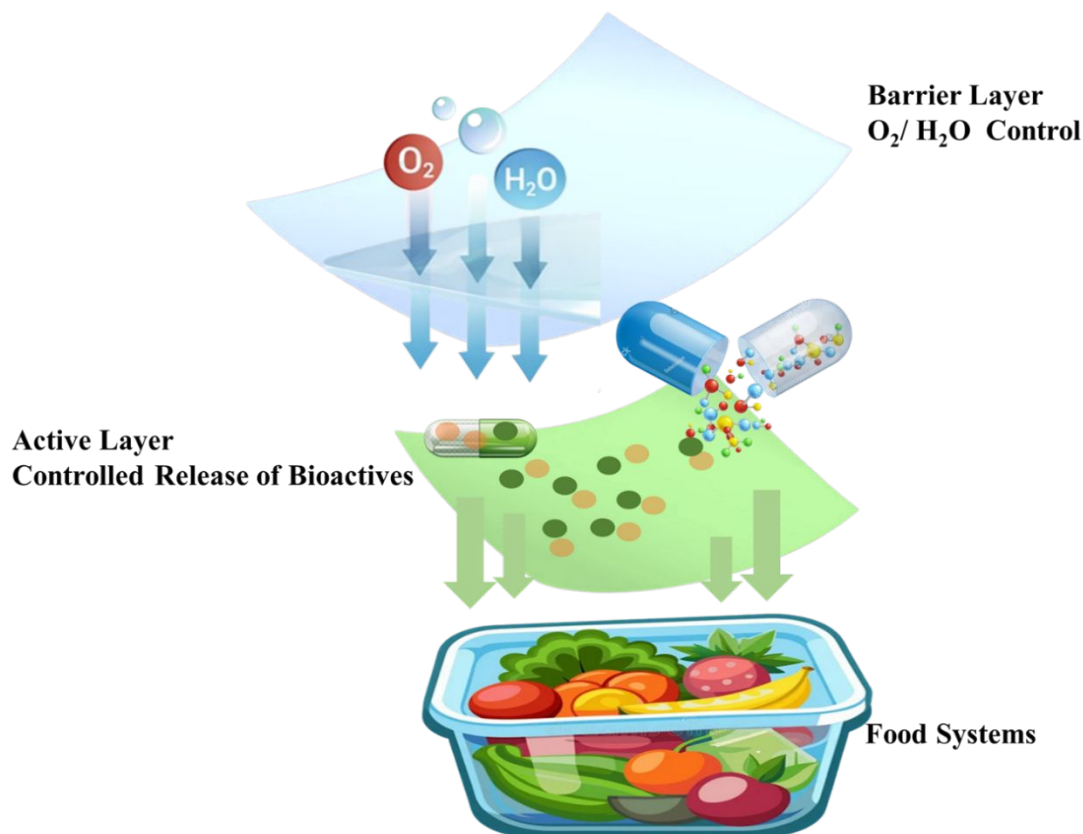


428 found across plants, algae, cyanobacteria, and other phototrophic organisms, while chlorophyll b  
429 functions as an accessory pigment predominantly located in green algae and higher plants<sup>61</sup>. The  
430 presence of extensive conjugated double bonds in these molecules contributes to their significant  
431 antioxidant capacity<sup>37</sup>.

#### 432 **4.1 Active packaging**

433 Chlorophylls are widely distributed across various plant-based sources, particularly in green  
434 vegetables and fruits. Its distinctive physicochemical and biological properties including strong  
435 light absorption, pH sensitivity, antioxidant activity, and chromatic responsiveness render it a  
436 valuable component in the formulation of both active and intelligent biodegradable packaging  
437 systems<sup>62</sup>. Furthermore, its molecular structure, characterized by a porphyrin ring coordinated with  
438 a central magnesium ion (notably in chlorophyll a and b), is inherently organic and  
439 biodegradable<sup>63</sup>. Natural pigments or dyes for sustainable food packaging application. This  
440 structural composition allows for environmental degradation through natural enzymatic and  
441 microbial pathways, enhancing its appeal for environmentally sustainable applications. Figure 3  
442 illustrates a multilayer smart packaging system which consist of a barrier and an active layer. The  
443 active layer has dual functions: as an active releasing system that delivers antimicrobial agents and  
444 antioxidants to the food, and as an active scavenging system regulating and responding to key  
445 environmental factors such as oxygen, carbon dioxide, moisture, ethylene, and odor. This  
446 combination can help to maintain food quality while extending shelf life by actively interacting  
447 with the packaged food environment.





**Figure 3** Multilayer smart packaging system consisting a barrier and an active layer

448

449

450

451 The incorporation of chlorophylls into biopolymer-based packaging films has been shown to  
 452 markedly influence their physical (e.g., thickness, opacity, and ultraviolet light-blocking  
 453 capability), mechanical (such as tensile strength and elongation at break), and barrier properties  
 454 (including resistance to water vapor and gas transmission). Insights into the edible and  
 455 biodegradable ulvan-based films and coatings for food packaging<sup>64</sup>. These alterations are strongly  
 456 dependent on the nature of the polymer matrix as well as the method of film fabrication. Numerous  
 457 studies have explored such formulations to enhance the functional attributes of environmentally  
 458 sustainable packaging materials. For instance, Dewi et al., (2022)<sup>54</sup> developed an alginate-based  
 459 film embedded with chlorophylls microcapsules derived from *Caulerpa racemosa*, reporting  
 460 improved barrier performance, particularly in reducing moisture and gas permeability, which  
 461 consequently extended the shelf life of packaged fish products. Similarly, Ukwatta et al. (2025)<sup>59</sup>  
 462 investigated chlorophyllin-doped corn-starch films combined with different lipid additives and  
 463 observed that the addition of chlorophylls in enhanced the water vapor barrier properties. This



464 enhancement was attributed to increased matrix density, making the films more effective for  
465 packaging applications involving light- and moisture-sensitive food items.

466 Chlorophylls -based packaging materials exhibit high environmental degradability, as their  
467 constituent compounds including chlorophylls pigments and biopolymeric matrices like starch and  
468 chitosan are susceptible to enzymatic breakdown by naturally occurring microbial populations,  
469 particularly bacteria and fungi<sup>65</sup>. The degradation processes typically involve the breakdown of  
470 the porphyrin ring structure in chlorophylls, resulting in simpler intermediates like pheophytin and  
471 chlorins, which are eventually mineralized into carbon dioxide, water, and biomass<sup>66</sup>. These  
472 degradation products are non-toxic to ecosystems and may even serve as beneficial nutrients within  
473 the soil. Hence, the unique physicochemical and biological attributes of chlorophylls can be  
474 effectively leveraged in the development of active and intelligent packaging systems. Such  
475 packaging systems offer a sustainable and innovative approach to enhancing food preservation,  
476 monitoring product quality, and reducing environmental impact within the packaging sector<sup>67</sup>.

477 Recent developments have underscored the effectiveness of chlorophylls-derived compounds as  
478 antimicrobial agents within active packaging systems, emphasizing their potential to prolong shelf  
479 life and improve food safety. These packaging films are specifically designed to suppress or delay  
480 the growth of spoilage and pathogenic microorganisms that compromise food quality and safety<sup>68</sup>.

481 In a study conducted by Dewi et al., (2022)<sup>54</sup>, an alginate-based edible film was formulated using  
482 chlorophylls extracted from *Caulerpa racemosa*, a species of green seaweed. The antimicrobial  
483 efficacy of the film was evaluated by wrapping fish snacks, and the results indicated a notable  
484 reduction in microbial proliferation, particularly targeting spoilage organisms during storage. The  
485 observed antimicrobial effect was attributed to the chlorophylls component, which exhibited both  
486 antioxidant and antimicrobial functionalities, thereby positioning it as a promising natural  
487 preservative for use in biodegradable packaging systems. The inclusion of antioxidants such as  
488 chlorophylls in packaging materials plays a critical role in inhibiting oxidative degradation in food  
489 products, especially those rich in lipids and proteins<sup>25</sup>. These compounds function through various  
490 mechanisms, including neutralization of singlet oxygen, scavenging of free radicals, reduction of  
491 hydrogen peroxide, and chelation of pro-oxidant metal ions<sup>37</sup>. Such multifunctional activities  
492 contribute to the stabilization of food quality during storage and further enhance the utility of  
493 chlorophylls-based systems in active packaging applications. Plant-derived botanical extracts have  
494 been widely utilized as natural antioxidants in the development of active packaging materials<sup>69</sup>. In



495 a study conducted by Micó-Vicent et al., (2020)<sup>70</sup>, chlorophylls-containing hybrid nano pigments  
496 sourced from broccoli processing residues were integrated into polyester-based bio nanocomposite  
497 films. This incorporation led to a marked improvement in the antioxidant functionality of the  
498 resulting packaging films. Additionally, the films exhibited improved thermal stability, color  
499 uniformity, and mechanical performance, thereby indicating their suitability for active food  
500 packaging applications. This study underscores the dual advantage of valorizing agricultural waste  
501 while integrating naturally occurring antioxidant compounds into packaging matrices to prolong  
502 food shelf life and mitigate environmental burden. Chlorophylls-based materials thus offer a  
503 promising and sustainable alternative for active packaging technologies. Beyond packaging,  
504 chlorophylls pigments are currently utilized across multiple sectors including cosmetics,  
505 pharmaceuticals, and the food industry<sup>71</sup>. Within the food sector, chlorophylls (designated as E-  
506 140) is primarily employed as natural colorants<sup>56</sup>. However, their functionality may be further  
507 extended to serve as natural pH-responsive indicators for monitoring food freshness, presenting  
508 additional value in the development of intelligent packaging systems.

#### 509 **4.2 Intelligent packaging**

510 Intelligent packaging in the food sector integrates inherent bio functional properties with real-time  
511 monitoring capabilities to enhance food quality and safety management<sup>72</sup>. Chlorophylls, owing to  
512 its chemical and optical sensitivity, serves a dual role in such systems i.e., functioning not only as  
513 a packaging component but also as a responsive indicator capable of providing visual or  
514 quantifiable feedback on product freshness, quality, and safety<sup>73</sup>. One of the most widely employed  
515 mechanisms in this context is colorimetric sensing, where chlorophylls-based compounds exhibit  
516 perceptible color changes in response to environmental factors such as pH fluctuations,  
517 temperature shifts, or the presence of spoilage-associated gases like ammonia or oxygen<sup>74</sup>.

518 The pH-induced chemical transformation in the chlorophyll's macrocycle determines the  
519 colorimetric properties of the chlorophylls -based intelligent indicators. Chlorophylls consists of a  
520 porphyrin (tetrapyrrole) ring complexed with a central  $Mg^{2+}$  ion, giving this pigment its green  
521 coloration and visible region absorption <sup>75</sup>. Moreover, under acidic environment, the protonation  
522 of the porphyrin nitrogen atoms results in the removal of  $Mg^{2+}$  and formation into pheophytin  
523 resulted in the visual color change from green to olive or yellow-brown <sup>76</sup>. While, in alkaline  
524 conditions, deprotonation occurs and changes in the porphyrin structure result in the color shift  
525 and the electron delocalization, further modifies the optical behavior <sup>77</sup>. These structural

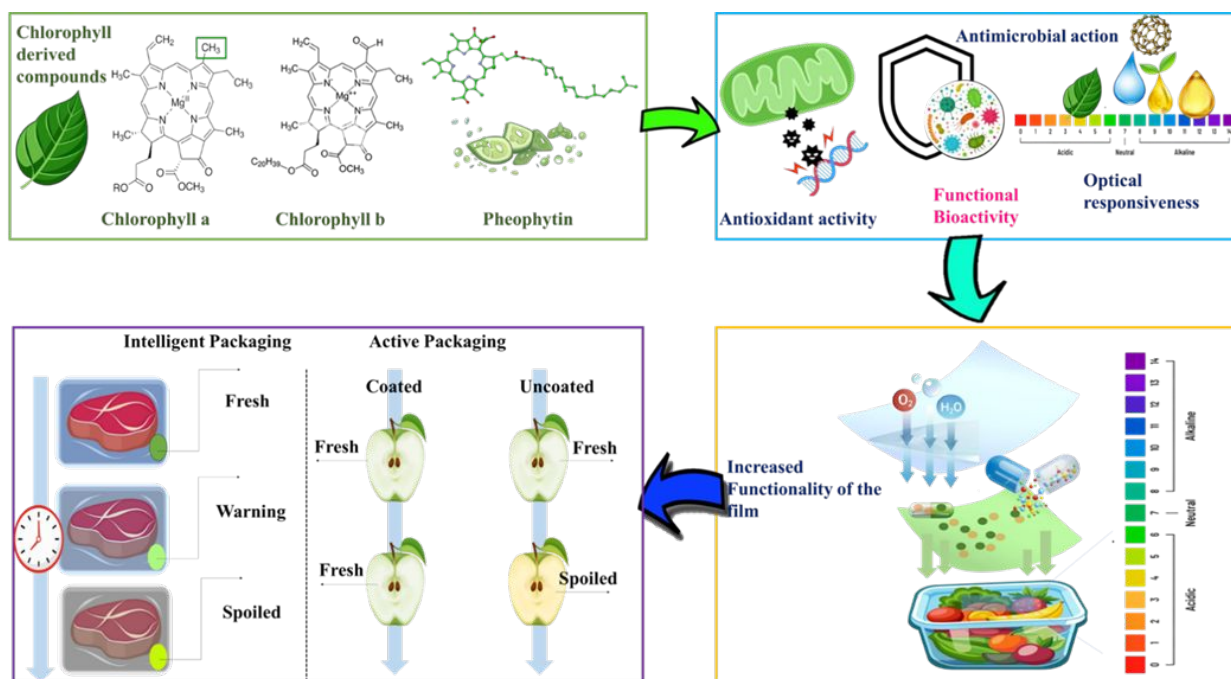


526 modifications impact absorption directly, i.e., native chlorophylls exhibits strong UV-Vis  
527 absorptions near 430 nm (Soret band) and 660-665 nm (Q band)<sup>78</sup>. Moreover, the peak broadening,  
528 lower intensity, and shift in wavelengths caused by the pheophytin produced through acidic  
529 environments strengthen the physicochemical basis concerning the use of chlorophylls as a  
530 halochromic indicator in smart packaging devices.

531 These alterations enable visual detection of food spoilage without the need to open the package,  
532 offering a user-friendly freshness assessment tool for both manufacturers and consumers. For  
533 example, Yu et al. (2024)<sup>79</sup> developed a chlorophylls-infused colorimetric film that demonstrated  
534 distinct color transitions under varying pH conditions. These pH-responsive films were effectively  
535 applied to monitor spoilage in actual food systems. Chlorophylls functioned as the primary  
536 halochromic pigment, undergoing progressive discoloration in response to acidic or alkaline  
537 environments typically resulting from microbial activity, thereby serving as a reliable indicator of  
538 food degradation<sup>80</sup>. Similarly, in a study by Mohammadian et al. (2020)<sup>81</sup>, chlorophylls was  
539 incorporated as a natural pigment in packaging films designed to function as temperature-sensitive  
540 indicators. While the primary focus was pH and gas sensitivity, the study observed visible color  
541 variations at elevated temperatures, correlating with the microbial spoilage activity. In another  
542 study, Kılıç (2024)<sup>82</sup> fabricated a thermoplastic starch-based film enriched with chlorophylls -rich  
543 *Aronia* extract, which demonstrated a gas-responsive colorimetric behavior. Exposure to  
544 ammonia, a volatile compound typically generated during the initial stages of spoilage in protein-  
545 rich food products such as fish, induces a noticeable color change in the film, shifting its hue from  
546 green to brown<sup>83</sup>. Fig. 4 illustrates the utilization process of extracted chlorophylls from plants to  
547 develop bioactive compounds which are further incorporated into intelligent and active packaging  
548 systems. In intelligent packaging these chlorophylls-based indicators visually signal food  
549 freshness by changing color, whereas in active packaging, chlorophylls coatings help maintain  
550 food freshness by slowing spoilage compared to uncoated packaging, thereby extending shelf life  
551 through bioactive protection. Collectively, these studies confirm the efficacy of chlorophylls as  
552 functional indicator in intelligent packaging systems aimed at real time spoilage detection<sup>84</sup>.



553



554

555

556 **Figure 4** Schematic illustration of chlorophylls extraction from plants and its incorporation into  
 557 intelligent and active food packaging systems.

558 Chlorophylls also possess intrinsic fluorescence properties that can be exploited in the design of  
 559 fluorescent sensing systems for intelligent packaging. Its fluorescence is highly responsive to  
 560 environmental variations, making it a viable noninvasive indicator for detecting food spoilage and  
 561 microbial contamination<sup>85</sup>. Upon exposure to light, chlorophylls emits red fluorescence in the  
 562 spectral range of approximately 680 to 740 nm<sup>86</sup>. During food degradation, metabolic by products  
 563 such as organic acids, ammonia, However, during food degradation, metabolic by-products such  
 564 as organic acids, ammonia, and reactive oxygen species alter local pH and oxidative conditions,  
 565 leading to structural modifications in chlorophylls that reduce fluorescence intensity or shift  
 566 emission wavelength. Such fluorescence variations provide measurable signals corresponding to  
 567 the degree of food spoilage<sup>87</sup>. For instance, Xue et al. (2025)<sup>88</sup> designed a dual channel pH  
 568 responsiveness fluorescent bio ink system that integrated chlorophylls natural fluorescence to  
 569 generate visual cues corresponding to pH fluctuations during spoilage. Similarly, Herppich  
 570 (2021)<sup>89</sup> evaluated chlorophylls fluorescence imaging (CFI) as a non-destructive tool for assessing  
 571 the physiological status and quality of fresh produce demonstrating its potential for real time  
 572 contactless evaluation in intelligent packaging.



573 When incorporated into biopolymer-based films and coatings, chlorophylls intrinsic properties  
574 including pH sensitivity, antioxidant capacity, and color responsiveness can modulate physical  
575 parameters such as tensile strength, flexibility, and barrier performance, all critical for packaging  
576 functionality and product protection. Table 2 summarizes the diverse chlorophylls-incorporated  
577 biomaterials used in active and intelligent food packaging systems, highlighting applications  
578 across various food products, functional properties such as antioxidant, antimicrobial,  
579 photoactivity, and freshness indicator alongside their physical attributes (e.g., barrier performance,  
580 mechanical strength, biodegradability). Empirical studies reported outcomes, including prolonged  
581 shelf life, minimized food spoilage, and visual cues for product freshness. Nevertheless, broader  
582 commercial application requires addressing challenges related to physicochemical stability and  
583 cost-effective extraction methods. Future advances in microencapsulation techniques, polymer  
584 matrix optimization, and integration with sensing technologies are expected to enhance the  
585 functional performance facilitating sustainable, intelligent packaging solutions.

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589



590 **Table 2** Summary of chlorophylls-based biomaterials in food packaging applications: functional properties and physical characteristics

Biomaterial	Food product	Functional Properties	Packaging	Physical properties	Limitations	References
Cornstarch + chlorophylls in + lipids (coconut oil, oregano essential oil, beeswax)	Shrimp	Photoactive antibacterial; ROS generation; shelf-life extension	Active	Increased Tensile strength; Improved water vapour permeability; rough surface	Starch's susceptibility to moisture; lipid composition affects performance	59
Pectin + chlorophylls (leaf extract) + CMC/silica nanoparticles	-	Antioxidant; Antimicrobial (against <i>E. coli</i> and <i>S. aureus</i> ); Thermally stable	Active	Increased tensile strength, increased flexibility, thickness, improved	Decreased film flexibility; enhanced opacity by the addition of nanoparticles	18
Chlorophylls microcapsules (gum arabic/maltodextrin)	-	Antioxidant activity; UV protection	Active	Improved thermal stability, high encapsulation efficiency (77.19%)	spray-drying-induced chlorophylls degradation; high surface oil with gum arabic blends	90
Wheat gluten + chlorophylls extract-based film	Sesame oil	UV blocking; Color change (indicator)	Active/ Intelligent	Increased thickness; increased water vapor permeability (WVP)	weak mechanical properties; uneven nanocomposite dispersion affecting functional uniformity; higher water vapor permeability and moisture sensitivity	17
Polyester-based film (INZEA) + chlorophylls hybrid nanopigments	-	Strong antioxidant potential; pigment stabilization; UV resistance	Active	Improved Young's modulus, and increased thermal stability	Limited biodegradability; reduced transparency	70



Biomaterial	Food product	Functional Properties	Packaging	Physical properties	Limitations	Reference
Chlorophylls orella-k-carrageenan composite films	-	Rapid biodegradation; UV barrier	Active	High opacity	High opacity affects in consumer visibility	83
WPI + chlorophylls microcapsules	-	High antioxidant potential; pH- responsive indicator	Active	Low moisture content, enhanced water solubility	-	91
Chitosan + chlorophylls a + 2-HP- $\beta$ - Cyclodextrin	-	High antioxidant potential; Generates reactive oxygen species (ROS) under light; antimicrobial activity	Active	Amorphous structure, modified surface roughness, good light absorbance	Absence of controlled release of bioactive compounds analysis; light dependent ROS	92
PVA + ZnO + AgI + Chlorophylls (from spinach)	-	Photocatalytic pollutants degradation; strong activity	Active	-	-	93



## 592 **5. Conclusion, challenges, and future perspective**

593 Chlorophylls based active and intelligent packaging has emerged as a sustainable, multi-functional  
594 food packaging technology. The chlorophylls molecular structure imparts high antioxidant  
595 activity, photo-responsiveness, pH sensitivity, and spontaneous biodegradability. These naturally  
596 occurring properties enables chlorophylls to perform the dual function in packaging which is an  
597 active ingredient which slows down food spoilage through antioxidant and antimicrobial activities;  
598 and secondly an active indicator that provides immediate visual cues about food freshness through  
599 colorimetric and fluorescent signals. Chlorophylls or microencapsulated chlorophylls addition in  
600 biodegradable polymer matrices enhances physical and functional characteristics of films. Some  
601 of the examples include improved thickness, improved barrier properties towards moisture and  
602 gases, and changed mechanical behavior, typically resulting in smoother and more homogeneous  
603 film microstructures. Active packaging films incorporated with chlorophylls have been shown to  
604 be effective scavengers of free radicals and inhibitors of spoilage bacteria which results in extended  
605 shelf life for various foods including seafood and meat. Concurrently, chlorophylls pH,  
606 temperature, and gas sensitivity (such as ammonia) provide a measurable and often observable  
607 color transition from green to yellow, brown, or other colors characteristic of spoilage, thus arming  
608 consumers, and supply chains with non-invasive freshness sensors.

609 Chlorophylls is inherently unstable during conventional processing and storage conditions as it is  
610 sensitive and degrades rapidly when exposed to light, heat, oxygen, and fluctuation in pH, which  
611 damages its functional shelf life and color stability. Enhancing its stability through application of  
612 advanced encapsulation methods such as micro- and nano-encapsulation by biopolymer carriers is  
613 critical but currently making manufacturing more complex and costly. Moreover, cost-effective  
614 large-scale extraction and purification procedures that preserve functionality of chlorophylls must  
615 be optimized; and compatibility with various polymer matrices can affect film homogeneity,  
616 mechanical resistance, and barrier properties, which requires proper formulation and control of  
617 processing.

618 Future advancements should focus more on encapsulation to preserve chlorophylls bioactivity and  
619 enable controlled release of antimicrobial and antioxidant molecules, thereby optimizing shelf-life  
620 extension potential. Refining fabrication processes (extrusion, solution casting) may increase film  
621 robustness and scalability without compromising functionality. Combinations with digital sensing  
622 technology like fluorescence-based biosensors and IoT-based smart labels can potentially advance



623 real-time monitoring of food quality. As such, future research efforts should focus on developing  
624 active, and intelligent packaging from chlorophylls to offer sustainable alternative that is driving  
625 food preservation and transparency of freshness together. This will ultimately result in safer,  
626 fresher, and more sustainable global food supply chains with reduced food waste and  
627 environmental impact.

628

### 629 **Conflict of interest**

630 Authors declare there is no conflict of interest in this work.

### 631 **Data availability statement**

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

### 634 **Funding**

635 No funding received.

### 636 **Acknowledgment**

637 None

### 638 **Authors contribution**

639 Harshita Ranjan: Writing – Original draft, Software, Methodology, Investigation, Data curation,  
640 formal analysis. Manisha Joshi: Writing – Original draft, Investigation, Software. Swarup Roy:  
641 Conceptualization, Writing – review & editing, Visualization, Validation, Supervision.

642

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**Data availability statement**

View Article Online  
DOI: 10.1039/D5FB00637F

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

