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## EUCHEUMA COTTONII SEAWEED-BASED BIOPLASTICS: EFFECTS ON RAW MATERIAL BIOCHEMISTRY, EXTRACTION METHODS, MANUFACTURING PROCESSES, MECHANICAL PROPERTIES, APPLICATIONS, AND CHALLENGES FACED (A REVIEW)

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### ABSTRACT

The global plastic pollution crisis, characterized by the annual production of over 400 million metric tons of petroleum-based polymers - of which less than 10% is recycled-highlights the urgent scientific and policy-driven need for biodegradable, bio-based materials. *Eucheuma cottonii* (synonym, *Kappaphycus alvarezii*), a red macroalga (Rhodophyta) commercially cultivated in Indonesia and the Philippines, represents one of the most strategic feedstocks for bioplastic production in the Asia-Pacific region. This potential is driven by its high kappa-carrageenan content (25-45% dry weight), rapid biomass growth, and suitability for processing in infrastructure-limited environments.

This review provides a critical synthesis of the current literature on *E. cottonii*-based bioplastics, integrating empirical findings from two reference studies, Consebit et al. (2022) and Nurdin et al. (2022), alongside 92 global references published between 2011 and 2025. The scope of analysis includes, (i) biochemical and structural characterization of kappa-carrageenan and its film-forming mechanisms, (ii) alkaline extraction methods using KOH and advanced extraction technologies, such as ultrasonic-assisted extraction (UAE), microwave-assisted extraction (MAE), enzymatic extraction, and deep eutectic solvents (DES), (iii) solvent-casting-based film fabrication techniques and the functional roles of glycerol, sorbitol, chitosan, and sodium benzoate as additives, (iv) mechanical property evaluation, in which the average tensile strength significantly decreased from 17.97 N (F1, 10

mL glycerol) to 6.21 N (F3, 30 mL glycerol; ANOVA:  $F=115.32$ ,  $p=1.63 \times 10^{-5}$ ), corresponding to tensile strengths of approximately 17.97 MPa to 6.21 MPa according to ASTM D882 standards, (v) biodegradation kinetics, with complete soil decomposition occurring within 2–12 days, (vi) modulation of water solubility via polyelectrolyte complexation with chitosan; and (vii) nanocomposite reinforcement strategies through the incorporation of cellulose nanocrystals (CNC), ZnO nanoparticles, nanoclay, and soy protein integration.

Significant methodological limitations were also identified, including variability in tensile strength measurements (spring scale versus ASTM D882), absence of FTIR, SEM, and TGA/DSC characterization, as well as water vapor permeability (WVP) analysis, all contextualized against international benchmarks. Overall, this review emphasizes that *E. cottonii* constitutes a promising bioplastic feedstock with a strong regional foundation and untapped commercial potential. Optimal utilization requires research aligned with international standards and the development of large-scale commercialization pathways.

**KEYWORDS:** *Eucheuma cottonii*, kappa-carrageenan, seaweed-based bioplastics, biodegradable packaging, glycerol as plasticizer, chitosan composites, tensile strength, biodegradability, nanocomposites, Southeast Asia, sustainable packaging.

## 1. INTRODUCTION

### 1.1 Global Plastic Pollution Crisis: Scale, Trends, and Regulatory Context

The proliferation of petroleum-based synthetic plastics represents one of the most extensive and intractable environmental crises in the modern era. Global plastic production surpassed 400 million metric tons per year in 2022 and is projected to double by 2040 in the absence of significant systemic interventions [1]. Of this total, less than 10% is mechanically recycled, while the remainder is directed to incineration (12%), formal landfills (25%), or, most critically, unmanaged disposal into natural environments (63%) [2].

The consequences of this accumulation are severe and multidimensional. Approximately 8 million tons of plastic enter the marine environment annually [2], causing lethal and sublethal effects on over 800 species through ingestion and entanglement. At the molecular level, macroplastics undergo gradual fragmentation into microplastics and nanoplastics, which have been detected in human placental tissue [3], as well as in blood, lung tissue, and marine organisms across multiple trophic levels [4]. These pervasive contaminations have substantial

ecological and toxicological implications, and emerging evidence indicates significant epidemiological concerns.

From a policy perspective, the European Union Single-Use Plastics Directive (EU 2019/904) banned several single-use plastic products starting in July 2021 and established a broader framework for producer responsibility [5]. By 2024, over 127 countries had enacted restrictions on single-use plastics [1]. Nevertheless, regulatory frameworks, while necessary, are insufficient alone to curb the accelerating trend of plastic pollution. Comprehensive support through the development of technically feasible, economically competitive, and ecologically credible bio-based alternative materials is urgently required.

## **1.2 Limitations of First and Second Generation Bioplastics**

First and second generation bioplastics, including starch based polylactic acid (PLA), microbially derived polyhydroxyalkanoates (PHA), and thermoplastic starch (TPS) from cassava and corn have demonstrated commercial feasibility at the proof of concept stage. However, these materials still face fundamental sustainability limitations [6],[7]. A primary concern is competition for arable land. The production of PLA from corn starch requires approximately 2.65 tons of corn per ton of polymer produced. This process is land and freshwater intensive, placing such bioplastics in direct competition with food production systems, which themselves are under increasing pressure from climate change and population growth [6], [8].

Furthermore, PLA undergoes significant biodegradation only under industrial composting conditions-specifically at temperatures  $\geq 58^{\circ}\text{C}$ , controlled humidity, and the presence of active microbial inoculants. When disposed of in open environments, whether terrestrial or marine, PLA can persist for years without substantial degradation [7]. A global analysis conducted by Jin et al. [8] indicates that large-scale transition to bioplastics without diversification of feedstock sources could increase agricultural land demand by up to 35% and freshwater consumption by 22% relative to the 2020 baseline. These impacts underscore a misalignment with sustainable development principles, highlighting the urgent need for the development of alternative bioplastics that are more environmentally friendly and resource efficient.

### 1.3 Macroalgae as Feedstock for Third-Generation Bioplastics

Third-generation bioplastics, particularly those derived from macroalgae (seaweed) and microalgae, offer a sustainable alternative that addresses the land and freshwater constraints typically associated with terrestrial plant based biopolymers, while providing biochemically rich feedstocks that are widely distributed globally [9],[10]. Macroalgae are photosynthetic organisms exhibiting the highest growth rates on Earth. Species of *Eucheuma* and *Kappaphycus* can yield biomass ranging from 15 to 30 tonnes of dry weight per hectare per year under optimized tropical cultivation conditions, significantly surpassing the productivity of conventional terrestrial energy crops [9].

The advantages of seaweed cultivation include the absence of freshwater irrigation, fertile land, or synthetic agricultural inputs. Essential nutrients are entirely sourced from dissolved inorganic compounds present in seawater, including nitrogen, phosphorus, and trace elements [11]. Beyond serving as a feedstock, seaweed aquaculture delivers substantial ecosystem services. These include photosynthetic carbon dioxide sequestration using dissolved inorganic carbon, bioassimilation of nutrient surpluses to mitigate coastal eutrophication, provision of structured habitats for juvenile marine organisms, and enhancement of income and employment opportunities for coastal fishing communities in developing tropical countries [10],[12]. According to Faghali et al. [11], global seaweed forests are estimated to sequester between 61 and 268 megatons of carbon per year, underscoring their role as highly effective blue carbon sinks.

#### 1.4 *Eucheuma cottonii* (Strategic Significance in Southeast Asia)

Among the various macroalgal species investigated for bioplastic applications, *Eucheuma cottonii* (formally reclassified as *Kappaphycus alvarezii* by Doty [13], although the term *E. cottonii* remains widely used in commercial contexts) occupies a strategically significant position in the Asia-Pacific region. *E. cottonii*, one of the most extensively cultivated red seaweeds globally, produces over 10 million tons of fresh seaweed annually. Indonesia and the Philippines collectively account for approximately 75-80% of global production [13]. In Indonesia, the principal cultivation sites are located in Kupang Regency (East Nusa Tenggara), Muna Regency (Southeast Sulawesi), and Pinrang Regency (South Sulawesi). In these regions, roughly 1.2 million smallholder farmers practice *E. cottonii* aquaculture using off-bottom and floating long-line methods [14].

The commercial value of *E. cottonii* is primarily determined by the kappa-carrageenan content in its cell walls, a sulfated galactan biopolymer comprising 25-45% of the dry biomass weight. Kappa carrageenan has long been employed as a high quality food-grade thickening and stabilizing agent in a wide range of food and beverage products, including dairy, meat, and infant formula [15],[16]. Additional advantages of *E. cottonii* as a bioplastic feedstock include the availability of established cultivation infrastructure, a well-developed supply chain, and regulatory approval for food-contact applications. Collectively, these factors position *E. cottonii* as a highly viable bioplastic feedstock, in contrast to other macroalgal species that are less characterized and require both regulatory development and supply chain establishment from the ground up [17],[18].

### **1.5 Objectives and Structure of the Review**

Although the number of experimental studies on *E. cottonii*-based bioplastics is increasing, the field still lacks a comprehensive and critically evaluative synthesis that integrates regional laboratory findings with global knowledge, identifies methodological limitations, and maps research priorities based on empirical evidence. This review is designed to systematically address these identified gaps, drawing on two primary reference studies [17,18] as its empirical foundation, while contextualizing findings within 92 global references published between 2011 and 2025.

#### **The specific objectives of this review include:**

- a. To biochemically characterize the potential of *E. cottonii* as a bioplastic source through structural analysis of kappa-carrageenan and film-forming mechanisms.
- b. To critically evaluate the extraction methodologies and fabrication processes employed.
- c. To synthesize data related to mechanical properties, barrier characteristics, and biodegradation rates, contextualized against international standards, including the conversion of tensile strength to MPa units in accordance with ASTM D882.
- d. To identify current applications and development prospects, spanning food packaging, biomedical, and agricultural sectors.
- e. To present key technical, methodological, and commercial challenges based on global literature.
- f. To formulate evidence-based future research directions, encompassing nanocomposite development, integration of biorefinery concepts, and regulatory pathway mapping.

This review has been systematically prepared to provide a comprehensive and analytical overview of the current state of *E. cottonii* bioplastics, while offering strategic guidance for their sustainable development in line with international standards.

## 2. MATERIALS AND METHODS

### 2.1 Review Methodology and Literature Search

This study employed a narrative review methodology, utilizing a systematic literature identification process based on the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) framework, adapted to suit the context of a narrative review. The literature search was limited to publications from the last ten years, while retaining seminal references considered scientifically relevant.

Inclusion criteria required that publications be peer-reviewed and indexed (SINTA, Scopus, or ScienceDirect), focus on primary or secondary research related to seaweed-derived biopolymers, and provide sufficient quantitative data or methodological detail to enable critical evaluation. Conversely, unverified web content and conference abstracts lacking comprehensive methodological descriptions were excluded from the analysis.

The literature search strategy was conducted systematically using structured Boolean search strings to ensure comprehensive coverage of topics related to *Eucheuma cottonii* based bioplastics. Information sources included both international and national databases, namely Scopus, ScienceDirect, Google Scholar, SINTA, Garuda, and One Search. The search strings were organized into major categories aligned with the research focus, as presented in Table 1.

**Table 1. List of Search Strings.**

Topic / Keywords	Search Strings
Seaweed Species	<i>Eucheuma Cottonii</i> , <i>Kappaphycus Alvarezii</i> .
Bioplastic / Biodegradable Film	Bioplastic, Biodegradable Film, Carrageenan Film, Packaging Film, Seaweed Bioplastic.
Sifat mekanik & permeabilitas	Mechanical Properties, Tensile Strength, Biodegradability, Water Vapour Permeability, Kappa-Carrageenan.
Mechanical Properties & Permeability	Nanocomposite, Chitosan, Cellulose Nanocrystals, Film, Seaweed Polysaccharide.
Kemasan berkelanjutan / pangan	Sustainable Packaging, Food Packaging.
Plasticizer & modifikasi film	<i>Eucheuma cottonii</i> , (Glycerol, Sorbitol, Plasticizer.

## 2.2 Prosedur Eksperimental dalam Penelitian Utama

The reference study by Consebit et al. (2022) [17]. Reported the fabrication of *Eucheuma cottonii*-based bioplastic films at Sultan Kudarat State University using a cook-and-cast method. The film formulation consisted of 50 g of dried *E. cottonii*, 30 g of corn starch, 60 mL of distilled water, 5 mL of acetic acid as a starch dispersant, and glycerol at three different levels: 10 mL (F1), 20 mL (F2), and 30 mL (F3). All components were combined in a pot and heated with continuous stirring until a homogeneous viscous solution was obtained. The mixture was then immediately poured onto a flat surface lined with aluminum foil and air-dried at ambient temperature for 3–4 days. Tensile strength of the films was measured using a spring scale in triplicate for each formulation, while biodegradability was assessed through a 12-day soil burial test, recording gravimetric mass loss every three days. Data analysis was performed using one-way ANOVA at a significance level of  $\alpha = 0.05$ .

Meanwhile, the study by Nurdin et al. (2022) [18] reported the alkaline extraction of carrageenan from *E. cottonii* biomass obtained from Tablolong, Kupang Regency, Nusa Tenggara Timur, at Universitas Muhammadiyah Kupang. A total of 30 g of *E. cottonii* was processed through a two-step protocol. The first step involved alkaline treatment at 60°C for 60 minutes using 0.6% KOH, followed by a hydrothermal lysis step at 90°C for 60 minutes with distilled water. The resulting extract was filtered through cloth and subsequently concentrated by freezing at -18°C for 24 hours to form a carrageenan gel.

### Three film variants were then prepared:

- a. ERL, consisting of carrageenan extract with the addition of 6 drops of glycerol and 6 drops of sorbitol.
- b. ERL + Chitosan, consisting of 50 mL of carrageenan extract combined with 0.5 g of chitosan dissolved in 25 mL of 1% acetic acid and 4 drops of glycerol.
- c. ERL + Sodium Benzoate, consisting of 50 mL of carrageenan extract with 0.5 g of sodium benzoate, 4 drops of glycerol, and 4 drops of sorbitol.

The films were dried in an oven at 50°C for 24 hours using glass molds and subsequently characterized qualitatively based on appearance, texture, and flexibility. Further evaluations included biodegradability assessment via soil burial tests and solubility tests in water at 15°C, 30°C, and 80°C.

**Table 2. Comparative Summary of Materials, Equipment, and Analytical Methods in Primary Studies on *Eucheuma cottonii* Based Bioplastics.**

Parameter	Consebit et al. 2022 [17] - Philippines	Arizal V, et al. 2022 [18] - Indonesia
Biomass	Dried <i>E. cottonii</i> , 50 g	Dried <i>E. cottonii</i> , 30 g; Tablolong, Kupang, NTT
Extraction agent	Direct thermal extraction (pan cooking, ~80–100°C)	KOH 0.6% / 60°C / 60 min → DW / 90°C / 60 min
Formulation additives	Cornstarch 30 g; Acetic acid (vinegar) 5 mL; Glycerine 10/20/30 mL	Glycerol, sorbitol; chitosan 0.5 g; natrium benzoat 0.5 g
Casting / Drying	Flat surface lined with foil; air-dried 3–4 days	Glass molds; oven-dried at 50°C for 24 h / ambient temperature
Tensile test	Spring scale; n = 3; ASTM (not converted to MPa)	Qualitative only, no quantitative measurements
Biodegradation	Soil burial, 12 days; mass loss (%) recorded at 3-day intervals.	Soil burial (pot); evaluated via EM4 inoculum; visual assessment
Solubility	Not performed	Water at 15°C, 30°C, 80°C; dissolution time
FTIR/SEM/TGA	Not performed	Not performed
WVP / O <sub>2</sub> barrier	Not performed	Not performed
Statistics	One- way ANOVA, $\alpha=0.05$	Descriptive qualitative, no inferential statistics

### 3. RESULTS AND DISCUSSION

#### 3.1 *Eucheuma cottonii*: Biochemical Basis and Bioplastic Potential

##### 3.1.1 Classification and Regional Cultivation

*Eucheuma cottonii* is classified within the order Gigartinales, family Solieriaceae, and belongs to the class Florideophyceae (Rhodophyta). The cultivation of this seaweed thrives optimally under tropical conditions, with water temperatures ranging from 26–30°C, salinity of 28–34 ppt, and active photosynthetically available radiation (PAR) of 100–250  $\mu\text{mol photons m}^{-2} \text{s}^{-1}$  [14,15]. Under optimized aquaculture conditions, biomass doubling times are routinely achieved within 15–25 days [9].

Data from the Food and Agriculture Organization (FAO) [13] confirm that Indonesia and the Philippines are the primary global producers, with Indonesia generating approximately 9.8 million tons of fresh biomass in 2020.

Regional cultivation areas in Kupang, Nusa Tenggara Timur, are particularly relevant to this study. The Tablolong coastal zone in Kupang Regency has been identified as a high-productivity area for *E. cottonii* [18],[19],[72]. Furthermore, Zhang et al. [72] reported that global seaweed production experienced rapid growth between 2000 and 2020, with a compound annual growth rate (CAGR) of approximately 5.2%, indicating an increasingly abundant availability of *E. cottonii* biomass as a sustainable industrial feedstock.

### 3.1.2 Kappa-Carrageenan: Structure and Film-Forming Properties

Kappa-carrageenan ( $\kappa$ -CG) is a high-molecular-weight sulfated galactan, with molecular weights ranging from 200 to 800 kDa, composed of alternating residues of  $\beta$ -(1 $\rightarrow$ 4)-D galactose-4-sulfate and  $\alpha$ -(1 $\rightarrow$ 3) 3,6 anhydro L galactose [17],[20]. The presence of sulfate ester groups ( $-\text{OSO}_3^-$ ) at the C4 position of the D-galactose units, with a degree of sulfation of approximately one mole per disaccharide unit, results in a high negative charge density ( $\sim 1.2$  meq/g). These characteristics confer hydrophilicity, polyelectrolyte behavior, and significant reactivity toward cationic species [20],[40],[46].

The gelation mechanism of  $\kappa$ -CG, which underlies its film-forming capability, occurs through a reversible coil to double helix conformational transition that is thermally induced [59],[81]. At temperatures above the helix to coil transition point ( $\sim 65$ - $85^\circ\text{C}$  for dilute  $\kappa$ -CG solutions), the polysaccharide chains adopt a random coil configuration, forming a homogeneous solution with stable viscosity suitable for casting processes. Upon cooling below the gelation temperature, the polysaccharide chains reorganize into double helices, stabilized by hydrogen bonding between 3,6 anhydrogalactose residues and electrostatic interactions mediated by potassium ions ( $\text{K}^+$ ). This sequential process—beginning with dissolution, helix formation, helix aggregation, and culminating in network formation—governs the extraction and development of the  $\kappa$ -CG film structure during evaporative drying [59],[81],[86].

From a materials engineering and mechanical engineering perspective, this gelation mechanism can be modeled using viscoelastic rheological approaches, wherein the Young's modulus ( $E$ ) of the film correlates with the crosslink density of the polymer network and the volume fraction of the crystalline phase formed [21],[59].

### 3.1.3 FTIR Spectroscopic Markers of Kappa Carrageenan

Fourier Transform Infrared (FTIR) spectroscopy is the principal analytical method employed to verify the successful extraction of carrageenan and to characterize interactions between

additives and the polymer matrix in bioplastic films. The characteristic spectral “fingerprint” of  $\kappa$ -carrageenan ( $\kappa$ -CG) includes several diagnostic bands, notably, a broad -OH stretching band at 3200-3600  $\text{cm}^{-1}$ , an asymmetric sulfate ester S=O stretching band around 1250  $\text{cm}^{-1}$  indicating the presence of sulfated polysaccharides, a 3,6-anhydrogalactose band at approximately 928  $\text{cm}^{-1}$  specific to  $\kappa$ -CG and absent in  $\lambda$ -carrageenan, and a galactose 4 sulfate band at around 844  $\text{cm}^{-1}$  [21],[37],[46].

It should be noted that the studies by Consebit et al. [17]. And Arizal et al. [18] did not perform FTIR characterization; thus, verification of carrageenan extraction efficiency and understanding of additive–matrix interaction mechanisms cannot be inferred from these studies. The absence of FTIR data constitutes a significant methodological limitation, restricting the interpretative capacity of their findings within the context of global bioplastic material literature.

Consequently, future research on *E. cottonii* based bioplastics should incorporate FTIR analysis as a fundamental characterization protocol. Successful extraction of  $\kappa$ -CG can be confirmed by identifying characteristic bands at  $\sim 928 \text{ cm}^{-1}$  (3,6-anhydrogalactose) and  $\sim 844 \text{ cm}^{-1}$  (galactose 4 sulfate). Interactions between polymers and plasticizers, such as glycerol, can be analyzed by observing the broadening of the -OH stretching band at 3200-3600  $\text{cm}^{-1}$  due to hydrogen bonding between glycerol hydroxyl groups and the carrageenan backbone. Additionally, the formation of polyelectrolyte complexes (PECs) between chitosan and carrageenan can be confirmed by shifts in the  $\text{NH}_2$  band at  $\sim 1570 \text{ cm}^{-1}$  and the sulfate S=O band at  $\sim 1250 \text{ cm}^{-1}$  [21],[37].

Systematic FTIR data not only provide empirical evidence regarding polymer structure and interactions but also establish an internationally recognized spectroscopic basis, which is a critical criterion for journal reviewers to assess the novelty and mechanistic validity of *E. cottonii*-based bioplastic research.

## **3.2 Extraction and Processing of Kappa-Carrageenan**

### **3.2.1 Alkali (KOH)-Based Extraction Process**

The extraction of carrageenan from *E. cottonii* is governed by alkali based chemical modifications. In the living algal tissue, the predominant biologically synthesized precursor is  $\mu$ -carrageenan ( $\mu$  CG). Treatment with KOH or NaOH facilitates a base-catalyzed elimination reaction, resulting in the formation of the characteristic 3,6-anhydrogalactose

bridge of  $\kappa$ -carrageenan. This chemical transformation simultaneously enhances gel strength, reduces sulfate content, and increases the gel to sol transition temperature [39],[87]. Wibisoni and Hendrawati [39] reported that increasing the KOH concentration from 0.1% to 2% gradually elevates the 3,6-anhydrogalactose content, strengthens the gel (from ~350 to ~800 g/cm<sup>2</sup>), and improves product purity. However, the concomitant rise in pH necessitates careful post-treatment washing to neutralize residual alkali, as unremoved alkali can induce carrageenan chain depolymerization upon heating [39],[87].

In the Kupang study [18], a two-step protocol at 60°C and 90°C was applied as a pragmatic adaptation of industrial-scale alkali treatment to a small laboratory scale. The hydrothermal lysis stage at 90°C is based on a thermodynamic mechanism: as the temperature approaches the carrageenan gel-to-sol transition (~70-85°C), the aggregated helical structure softens, and the polysaccharide cell walls relax, thereby facilitating more efficient biopolymer extraction [86]. Subsequently, a freeze thaw purification step (-18°C for 24 hours), a classical method for semi-refined carrageenan (SRC) production, concentrates the biopolymer by exploiting the freezing of free water, causing the carrageenan-rich phase to precipitate at the bottom of the container. This approach allows separation of the biopolymer without the need for chemical precipitants while preserving the molecular weight integrity of the carrageenan [18],[39].

### 3.2.2 Advanced Extraction

Although conventional alkali-thermal extraction is technically well established, this method is energy-intensive and poses a risk of polymer chain cleavage when conducted at high temperatures for prolonged periods. Several advanced extraction modalities offer higher efficiency with minimal polymer degradation.

Ultrasound-Assisted Extraction (UAE) utilizes high frequency ultrasonic cavitation (20-60 kHz) to disrupt cell walls and accelerate biopolymer release, reducing extraction time by 50-70% while increasing yield by 10-25% without compromising polymer chain integrity [9]. Microwave Assisted Extraction (MAE) operates via rapid volumetric heating due to dielectric relaxation of polar molecules, enabling extraction completion within minutes while preserving gel strength [9],[22]. Enzyme-Assisted Extraction employs enzymes such as cellulase and agarase to selectively degrade non-carrageenan structural cell wall components, facilitating biopolymer accessibility without exposure to thermal stress [19],[23]. Meanwhile, Deep Eutectic Solvents (DES), composed of hydrogen bond donors and acceptors, provide an

environmentally friendly extraction medium that is biodegradable and features tunable polarity to match specific extraction requirements [37].

These advanced methodologies are still relatively underutilized in *E. cottonii* production regions in Indonesia and the Philippines, presenting strategic opportunities for local capacity development [9],[19]. From a process engineering perspective, the application of UAE and MAE also has the potential to significantly reduce specific energy consumption compared to conventional thermal extraction, thereby enhancing operational efficiency and the sustainability of bioplastic production.

### **3.3 Fabrication Techniques and Formulation Design**

#### **3.3.1 Solvent Casting Fundamental Principles and Limitations at Industrial Scale**

Solvent casting is the predominant method for fabricating seaweed-based polysaccharide films at the laboratory scale, due to its operational simplicity, precise control over composition, and low capital requirements [21],[45],[63]. However, this technique presents intrinsic limitations that constrain its applicability for industrial-scale production. The drying process, which typically requires 24 to 96 hours, is incompatible with continuous production lines; achieving uniform film thickness over large surface areas is challenging; and inter-batch reproducibility is highly sensitive to environmental humidity and temperature conditions [45],[65].

In industrial contexts, continuous extrusion is the standard for conventional plastic film production. To adapt carrageenan for extrusion processes, thermoplastic processing at elevated temperatures (160-200°C) is required, which can only be achieved through the addition of sufficient plasticizers or blending with other thermoplastic polymers such as PLA [24],[74].

From a process engineering perspective, the development of pilot scale extrusion methods for carrageenan, based films constitutes a research priority in mechanical engineering. This necessitates a comprehensive understanding of polymer melt rheology, extruder screw geometry design, temperature profile distribution along processing zones, and film cooling kinematics post-extrusion. Characterization of critical process parameters-including melt viscosity ( $\eta$ ), extruder back pressure ( $\Delta P$ ), shear rate ( $\dot{\gamma}$ ), and the Deborah number ( $De$ )-is essential to define safe and optimal processing windows for carrageenan-plasticizer matrices.

### 3.3.2 Glycerol as a Plasticizer: Mechanism, Concentration Effects, and Alternatives

Glycerol ( $C_3H_8O_3$ , molecular weight 92.09 g/mol, boiling point  $290^\circ\text{C}$ , log P-1.76) is a trihydroxy alcohol classified as Generally Recognized as Safe (GRAS) by the U.S. Food and Drug Administration (FDA) and approved as a food additive by EFSA and BPOM [43]. The plasticizing mechanism of glycerol in polysaccharide matrices is well understood: the three hydroxyl groups of glycerol form hydrogen bonds with the -OH,  $-\text{OSO}_3^-$ , and -O- functional groups along carrageenan chains. These interactions competitively disrupt intermolecular hydrogen bonds connecting chain to chain, thereby increasing the free volume fraction of the polymer matrix, lowering the glass transition temperature ( $T_g$ ), and enhancing chain segment mobility under ambient conditions. Collectively, these phenomena manifest as reductions in Young's modulus, decreased tensile strength, and increased elongation at break [25],[26],[59],[84].

The relationship between glycerol concentration and the mechanical properties of *E. cottonii*-based films has been empirically demonstrated by Lyra Consebit et al. [17]. The addition of glycerol significantly reduced the average tensile force: from  $17.97 \pm 0.57$  N in F1 (10 mL glycerol), to  $9.47 \pm 1.50$  N in F2 (20 mL), and  $6.21 \pm 0.57$  N in F3 (30 mL). One-way ANOVA confirmed highly significant differences ( $F = 115.32$ ,  $df = 2,6$ ,  $p = 1.63 \times 10^{-5}$ ,  $p < \alpha = 0.05$ ).

- a. However, tensile force measurements reported in Newtons using a spring scale cannot be directly compared with international literature, which typically reports tensile strength in MPa normalized by the specimen's cross-sectional area. According to ASTM D882 (Standard Test Method for Tensile Properties of Thin Plastic Sheeting), proper testing procedures include
- b. Measurement of specimen dimensions (thickness and width) using a micrometer with a minimum resolution of 0.001 mm.
- c. Testing on a calibrated Universal Testing Machine (UTM) with a constant crosshead speed of 25 mm/min.
- d. A standard gauge length of 50 mm.
- e. Reporting tensile strength ( $\sigma$ , MPa = N/mm<sup>2</sup>), elongation at break ( $\epsilon$ , %), and Young's modulus (E, MPa).

Assuming a typical cross-sectional area of 1 mm<sup>2</sup> (film width 10 mm and thickness 0.1 mm), the estimated tensile strengths are approximately F1  $\approx 17.97$  MPa, F2  $\approx 9.47$  MPa, and F3  $\approx$

6.21 MPa. These values fall within the range reported for  $\kappa$ -carrageenan films by Arizal et al. [27] (18.1–32.4 MPa for glycerol variations of 0–40%). However, definitive confirmation requires actual dimensional measurements. Therefore, revalidation using a UTM in accordance with ASTM D882, with precise recording of specimen thickness ( $t$ ) and width ( $w$ ) at each gauge point, is essential to integrate the data into global material databases and to meet the standards for publication in high-impact international journals [45].

### 3.3.3 Chitosan as a Polyelectrolyte Complexation Agent

Chitosan, derived from the deacetylation of chitin (degree of deacetylation typically >75%; pKa ~6.2-6.5; molecular weight 50-300 kDa), is a cationic polysaccharide obtained through the N-deacetylation of crustacean chitin using alkaline treatment [40],[44],[70]. Under conditions of pH below its pKa, the primary amino groups ( $-\text{NH}_2$ ) of chitosan undergo protonation to form  $-\text{NH}_3^+$ , enabling electrostatic interactions with the sulfate groups ( $-\text{OSO}_3^-$ ) of  $\kappa$ -carrageenan. This interaction serves as the principal driving force in the formation of polyelectrolyte complexes (PECs), characterized by reduced polymer chain mobility, increased matrix density, and enhanced hydrophobicity [40],[70].

In the study conducted in Kupang [18], the addition of 0.5 g of chitosan dissolved in 1% acetic acid to 50 mL of carrageenan extract significantly improved the hydrolytic stability of the films. Pure ERL films dissolved within 4 hours at 30°C, whereas ERL films supplemented with chitosan required 2-3 days to dissolve across all tested temperatures (15°C, 30°C, and 80°C). From the perspective of materials science and mechanical engineering, PEC formation increases the effective cross-linking density within the polymer network, directly contributing to higher elastic modulus and enhanced resistance to plastic deformation.

### 3.3.3 Sodium Benzoate Antimicrobial Chemical Additive

Sodium benzoate ( $\text{C}_6\text{H}_5\text{COONa}$ ; molecular weight 144.1 g/mol; water solubility 628 g/L at 20°C) is a preservative additive with antimicrobial activity, approved under Indonesia BPOM Regulation No. 36/2013 [43]. And by the U.S. Food and Drug Administration (21 CFR §184.1733) for concentrations  $\leq 0.1\%$  in food products. Its antimicrobial mechanism involves the diffusion of undissociated benzoic acid molecules (pKa 4.2) into microbial cells at low pH.

In the study conducted by Arizal et al. [18], the incorporation of 0.5 g sodium benzoate into 50 mL of carrageenan extract produced films that were transparent, smooth, and highly

flexible, with rapid dissolution capability across various testing temperatures. The combination of antimicrobial activity and rapid biodegradability renders the ERL + NaB films particularly suitable for single-dose, water-soluble packaging applications, where fast dissolution upon contact with water is a critical requirement [43],[65].

**Table 3. Tensile Strength Data of *Eucheuma cottonii*-Based Bioplastic Formulations. Data are reported by Consebit et al. (2022) [17].**

including estimated tensile strength values converted to MPa based on an assumed typical cross-sectional area of 1 mm<sup>2</sup>, in accordance with ASTM D882 standards.

Formulation	Glyceron	Trail 1	Trail 2	Trail 3	Mean	Estimated (Mpa)	Anova
F1	10 MI	18,63	17,63	17,65	17,97	~17,97	P<0,05
F2	20 MI	9,80	10,78	7,84	9,47	~9,47	-
F3	30 MI	5,88	5,88	6,86	6,21	~6,21	-

\* The estimated tensile strength ( $\sigma$ , MPa) was calculated assuming a cross-sectional area of 1 mm<sup>2</sup> (width 10 mm  $\times$  thickness 0.1 mm). ANOVA results:  $F = 115.32$ ,  $df = 2,6$ ,  $p = 1.63 \times 10^{-5}$ . Measurements were performed using a spring scale; accurate MPa conversion requires film dimensional data according to ASTM D882.

Analysis of variance (ANOVA) revealed a significant difference among the formulations ( $F = 115.32$ ,  $df = 2,6$ ,  $p = 1.63 \times 10^{-5}$ ), indicating that glycerol content exerts a substantial effect on the tensile strength of the films. Measurements were performed using a spring scale, and film dimension data are required to accurately estimate tensile strength in MPa in accordance with ASTM D882 procedures. These findings empirically confirm that increasing glycerol concentration reduces the tensile strength of *E. cottonii* bioplastic films, which is highly relevant for mechanical design considerations and the optimization of biodegradable film formulations [17,28].

### 3.4 Biodegradation: Kinetics, Mechanisms, and Global Comparison

The biodegradability of *E. cottonii*-based bioplastic films was assessed in the reference studies using the Soil Burial Test, a method that is practical and readily implementable in laboratories with limited facilities. While this approach provides qualitative and semi-quantitative information regarding degradation, it possesses significant limitations, including its inability to distinguish between mass loss due to abiotic leaching of soluble components and true microbial mineralization. Results are also highly influenced by environmental variables such as soil type, pH, moisture content, microbial community composition, burial

depth, and ambient temperature, which are often neither controlled nor systematically reported in the reference studies [64].

Internationally standardized methodologies, such as ASTM D5988 (aerobic biodegradation in soil), ISO 14855 (controlled composting at 58°C), and ASTM D6400 (compostability specifications), provide more reproducible, environmentally relevant, and globally comparable data. These approaches are recommended as the preferred standard over non-standardized soil burial observations for future *E. cottonii* bioplastic research [29],[30],[31].

Despite methodological limitations, biodegradability results from both reference studies demonstrate consistency and reliability. In the study conducted in Kupang [18], the ERL and ERL + NaB films underwent complete degradation in soil within only 2 days, whereas the ERL + Chitosan film exhibited approximately 50% degradation over the same period, reflecting enhanced microbial resistance due to the crystallinity and hydrophobic properties of chitosan [18,40]. The study in the Philippines [17] reported average mass losses after 12 days of 56.25% (F1), 49.50% (F2), and 55.00% (F3). One-way ANOVA analysis revealed no significant differences among formulations ( $F = 0.04$ ,  $df = 2,9$ ,  $p = 0.96 > \alpha = 0.05$ ), indicating that variations in glycerol concentration did not significantly affect soil degradation kinetics. These findings carry practical implications: glycerol levels can be adjusted to optimize mechanical performance without compromising the end-of-life biodegradability of the bioplastic.

Contextualization against global benchmarks confirms the exceptional biodegradability of *E. cottonii* formulations. Krishnan et al. [32]. Reported that marine bacterial consortia could degrade  $\kappa$ -carrageenan-based bioplastics by up to 85% within 21 days in marine environments. In comparison, commercial PLA requires approximately 6 months under industrial composting conditions at temperatures  $\geq 58^\circ\text{C}$  [7]. LDPE exhibits less than 1% degradation over 100 years [48], whereas PET can persist for more than 450 years [33],[36],[57].

**Tabel 4. Comparative Analysis of Mechanical Properties and Biodegradability of *E. cottonii* Bioplastics versus International Benchmark Materials.**

Material	Tensile Strength	Biodegradability	Degradation Time	Reference
<i>E. cottonii</i> + starch (F1, glycerol 10 mL)	17,97 N (~17,97 MPa)	Ya	<12 days (soil)	[17]
<i>E. cottonii</i> ERL (glycerol + sorbitol)	NQ	Ya	2 days (soil)	[18]
<i>E. cottonii</i> ERL + chitosan	NQ	Parsial	~50% / 2 days	[18]
<i>E. cottonii</i> + starch (TS = 1.1 MPa)	1,1 Mpa	Ya	11 days	[35]
Carrageenan + Alginate + Starch (F6)	3,05 N/mm <sup>2</sup>	Ya	90% / 30 days	[37]
$\kappa$ -Carrageenan + CNF 10%	~18–24 MPa	Conditional	Several weeks	[21]
Carrageenan PLA biocomposite	75,37 MPa	Conditional	≥6 months	[24]

\* Estimates based on an assumed cross-sectional area of 1 mm<sup>2</sup>; NQ = Not Quantified in the reference study.

### 3.5 Nanocomposite Strategies: Efforts to Bridge the Performance Gap

#### 3.7.1 Cellulose Nanocrystals and Cellulose Nanofibers

The incorporation of cellulose nanocrystals (CNC, rod-shaped, 3-20 nm width, 100–300 nm length, elastic modulus ~100-150 GPa) and cellulose nanofibers (CNF, highly entangled morphology with a high aspect ratio) has emerged as the most comprehensive and well-documented strategy to enhance both mechanical properties and barrier performance in seaweed-based bioplastic films [21],[38],[46],[55].

In the study by Toledo-Jaldin et al. [21], CNF derived from palm oil waste was evaluated at loadings of 0, 5, 10, and 15% (v/w) in films based on *Kappaphycus alvarezii*,  $\kappa$ -carrageenan ( $\kappa$ -CG), refined carrageenan (RC), and semi-refined carrageenan (SRC), plasticized with 0.9% glycerol. The main results demonstrated that the addition of 10% CNF increased the

tensile strength of the films by 25-45% across all types of seaweed-derived films. Furthermore, water vapor permeability (WVP) decreased by 20-35%, which was attributed to the tortuosity effect, whereby the non-permeable crystalline cellulose phase impedes water vapor diffusion. Thermal stability also improved, as indicated by an increase in degradation temperature (TGA) of 15-25°C, resulting from hydrogen bonding between the hydroxyl groups of cellulose and the sulfate/hydroxyl groups of carrageenan.

Similarly, Kokkuvayil et al. [22] reported that CNCs with particle sizes <100 nm incorporated into a  $\kappa$ -CG matrix at 8% (w/w) loading increased tensile strength from 23.43 MPa (pure  $\kappa$ -CG film) to 39.89 MPa, corresponding to a 70% enhancement. In addition to tensile strength improvement, WVP decreased and elongation at break increased, indicating that CNC functions as the most effective single reinforcing agent for  $\kappa$ -CG-based packaging films.

From a composite mechanics and materials engineering perspective, this tensile strength enhancement can be modeled using the modified Halpin-Tsai equation, wherein the composite modulus ( $E_c$ ) depends on the filler modulus ( $E_f$ ), matrix modulus ( $E_m$ ), and particle aspect ratio ( $l/d$ ). This modeling approach provides a systematic predictive basis for optimizing nanocomposite composition prior to fabrication, thereby supporting the design of materials with superior mechanical performance.

### **3.7.2 Zinc Oxide Nanoparticles and Active Nanofillers**

Zinc oxide nanoparticles (ZnO NPs) with particle sizes ranging from 20 to 100 nm, incorporated into the carrageenan matrix at concentrations of 1-3%, have been demonstrated to confer antimicrobial activity through the generation of reactive oxygen species (ROS) and direct disruption of cell membranes, while simultaneously enhancing the film's ultraviolet (UV) blocking capability and improving its mechanical properties [39,49]. A study by Muncke et al. [39]. Reported that the addition of 3% ZnO NPs in  $\kappa$ -carrageenan films increased tensile strength by 43%, reduced water vapor permeability (WVP) by 9%, and inhibited the growth of *Escherichia coli* (ATCC 25922) and *Staphylococcus aureus* (ATCC 29213), with inhibition zone diameters of 18 mm and 22 mm, respectively [40],[42].

Furthermore, regulatory frameworks governing nanoparticle migration from materials intended for food contact, such as EC Regulation 10/2011 for the European Union and FDA 21 CFR §178.3297 for the United States, impose stringent limits on nanoparticle type, particle size, and specific migration levels (SML). Compliance must be verified through

migration testing prior to the approval of seaweed-based nanocomposite bioplastics for food contact applications [26],[39],[43],[49].

### 3.7.3 Carrageenan-PLA Biocomposites

The strategy of blending  $\kappa$ -carrageenan with polylactic acid (PLA) through melt processing has been investigated as an approach to simultaneously enhance hydrophobicity [44],[45],[46], tensile strength, and processability of seaweed-based bioplastics. Yadav et al. [24] reported that carrageenan-PLA composites processed at 50°C exhibited a tensile strength of 75.37 MPa, significantly higher than that of pure carrageenan films. This enhancement was attributed to the formation of strong hydrogen bonds between oxygen atoms in carrageenan and hydroxyl groups in PLA (bond length 1.88 Å), validated by a <sup>1</sup>H NMR peak shift at 5.3 ppm and density functional theory simulations [41].

This approach provides a promising pathway for producing carrageenan-based films with mechanical properties competitive with conventional synthetic packaging. However, the improved mechanical performance is generally associated with a reduction in ultimate biodegradation rates under open environmental conditions, highlighting the need for a comprehensive environmental impact assessment through Life Cycle Assessment (LCA) [8],[24].

### 3.6 Comparative Analysis: Extraction Methods, Plasticizers, and Nanofillers

To provide a global perspective and emphasize the novelty of this review, Table 4 presents a comprehensive comparison of extraction methods, types of plasticizers, and nanofillers applied in  $\kappa$ -carrageenan-based bioplastic studies at the international scale. This comparative analysis indicates that, although Indonesia and the Philippines account for over 75% of global *E. cottonii* production, both countries lag in the implementation of advanced extraction technologies and material characterization in accordance with international standards [28],[47],[48].

In contrast, publications from Malaysia, India, and Morocco have successfully employed ultrasonic-assisted extraction (UAE), microwave-assisted extraction (MAE), and deep eutectic solvents (DES) techniques [17],[18]. These findings underscore the critical importance of technology transfer and the enhancement of research capacity in major producing countries to improve the competitiveness and material quality of *E. cottonii* based bioplastics.

**Table 5. Comparison of Extraction Methods, Plasticizers, and Nanofillers in Selected International Studies on  $\kappa$ -Carrageenan Based Bioplastics.**

Study / Countr	Extraction Method	Plasticizer	Nanofiller	Maximum Tensile Strength (MPa) $\sigma$	WVP ( $\times 10^{-10}$ $\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}\cdot\text{Pa}^{-1}$ )
Consebit et al. [17] / Filipina	Direct thermal extraction	Glycerol	-	~17,97	TT
Arizal et al. [18] / Indonesia	KOH 0,6% alkali	Glycerol, Sorbitol	-	NQ	TT
Toledo-Jadldin et al. [21] / Malaysia	Refined alkali	Glycerol 0.9%	Palm CNF 10%	~24	~4,2
Kukuvayil et al. [22] / Maroko	Refined alkali	Glycerol	CNC 8%	39,89	~3,8
Tenorio-Garcia et al. [37] / Spanyol	Commercial alkali	Glycerol	ZnO NPs 3%	~28	~5,1

\*Maximum tensile strength estimated by conversion from Newton assuming a cross-sectional area of 1 mm<sup>2</sup>; TT = not available; NQ = not quantified.

### 3.7 Applications of Seaweed-Based Bioplastics

#### 3.7.1 Food Packaging and Edible Coatings

Food packaging represents the most extensive application sector and the nearest commercial prospect for *E. cottonii*-based bioplastics. Carrageenan films function as semi-permeable barriers that regulate gas exchange ( $\text{O}_2$ ,  $\text{CO}_2$ , ethylene) and moisture transfer, while the inherent film-forming capability of this material allows its direct application as an edible coating on food surfaces [15],[25],[59]. Sedayu et al. [59] demonstrated that optimal formulations achieving  $\text{O}_2$  permeability below  $0.5 \text{ cm}^3\cdot\mu\text{m}/\text{m}^2\cdot\text{day}\cdot\text{kPa}$  can extend the shelf life of fresh products by 3-7 days, highlighting the functional potential of these bioplastics to enhance both food safety and quality [49], [50].

#### 3.7.2 Biomedical and Pharmaceutical Applications

The biocompatible, biodegradable properties, along with the intrinsic biological activity of  $\kappa$ -carrageenan, render it a promising material for biomedical applications [15],[43],[66]. Sudhakar and Bargavi [51],[52],[66] successfully fabricated carrageenan-based scaffolds and

bio-membranes that are stimuli-responsive, demonstrating pH sensitive swelling behavior while maintaining cytocompatibility with fibroblast cell lines [53],[54]. Furthermore, Tenório Neto et al. [40]. Reported that chitosan/ $\kappa$ -carrageenan polyelectrolyte complexes can serve as pH responsive drug delivery matrices, where the drug release rate can be modulated between 30% and 90% over a 24hour period by adjusting the mass ratio of  $\kappa$ -carrageenan to chitosan. This highlights the versatility of this biopolymer for designing controlled pharmaceutical systems.

### 3.7.3 Agricultural Applications

Biodegradable seaweed-based biopolymer mulch films present a strategic solution for tropical agriculture. Krishnan et al. [19] reported that the application of carrageenan-based mulch films reduced weed growth by 60-80% compared to open soil, maintained soil moisture levels between 15-30%, and fully degraded within a single cropping season without leaving any residues. Furthermore, Sofianto et al. [35]. demonstrated that *Kappaphycus alvarezii*-based films combined with microbially induced  $\text{CaCO}_3$  exhibited a water vapour permeability (WVP) of 2.05 g·m/m<sup>2</sup>s Pa, which is lower than that of conventional biodegradable mulch films (2.68 g·m/m<sup>2</sup>s Pa), while tensile strength increased by 84.92% relative to traditional mulches.

The environmental advantages are particularly notable, as these biodegradable films eliminate the generation of microplastics after degradation, in contrast to conventional polyethylene mulch, which fragments into microplastics and can persist in agricultural soils for decades [3],[4],[55],[35].

## 3.8 Critical Methodological Assessment

A rigorous methodological evaluation of all reference studies and the broader literature is essential to ensure a valid and responsible synthesis of research findings. In the context of *E. cottonii*-based bioplastics, three principal methodological shortcomings substantially affect scientific validity and the potential for commercial application. Addressing these issues is expected to significantly enhance the impact of future research.

### a. Non-standardized Tensile Strength Measurement

In the study by Consebit et al. [17,56], a spring scale was employed to measure tensile force in Newtons, but without normalization to the specimen's cross-sectional area (width × thickness). International standards for mechanical characterization of bioplastic films,

specifically ASTM D882 (Standard Test Method for Tensile Properties of Thin Plastic Sheeting), prescribe specimen preparation with defined gauge dimensions, a loading rate of 25 mm/min using a Universal Testing Machine (UTM), and reporting of tensile strength (MPa = N/mm<sup>2</sup>), elongation at break (%), and Young's modulus (MPa). Without such normalization, the reported value of 17.97 N for F1 cannot be scientifically compared; for example, a film with a cross-sectional area of 1 mm<sup>2</sup> corresponds to a tensile strength of ~17.97 MPa, whereas a film with a 5 mm<sup>2</sup> cross-section corresponds to only ~3.59 MPa—a fourfold difference attributable solely to geometry rather than material properties [57,58,59]. Budiman et al. [45,56] emphasize that non-standardized mechanical testing represents a systemic issue in the Southeast Asian seaweed bioplastics literature.

### **b. Absence of Spectroscopic and Thermal Characterization**

The reference studies did not include Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), Thermogravimetric Analysis (TGA), or Differential Scanning Calorimetry (DSC). The absence of these analyses represents a significant methodological limitation, given that each technique provides essential evidence for confirming the chemical structure, thermal stability, and microstructural characteristics of seaweed-based bioplastic films. FTIR analysis is required to:

1. confirm the presence of  $\kappa$ -carrageenan through the detection of the characteristic absorption bands of 3,6-anhydrogalactose at approximately 928 cm<sup>-1</sup> and galactose-4-sulfate at around 844 cm<sup>-1</sup>.
2. verify glycerol incorporation through the broadening of the –OH stretching band in the range of 3200–3600 cm<sup>-1</sup>.
3. detect the formation of polyelectrolyte complexes (PECs) between chitosan and carrageenan through shifts in the –NH<sub>2</sub> band (approximately 1570 cm<sup>-1</sup>) and the sulfate S=O band (approximately 1250 cm<sup>-1</sup>) [21],[39].

TGA enables the characterization of the multistage thermal decomposition behavior of biopolymer–plasticizer systems. In glycerol-plasticized carrageenan films, thermal degradation typically proceeds in three stages:

1. 60–200°C, corresponding to the evaporation of physically adsorbed and bound water.
2. 210–290°C, associated with glycerol volatilization and the onset of polysaccharide chain decomposition.
3. above 290°C, corresponding to extensive carbonization [21],[41].

These data are essential for determining the safe processing window, which is critical for the development of pilot-scale melt extrusion processes [21],[41],[51],[74]. DSC complements TGA by measuring the glass transition temperature ( $T_g$ ) and the enthalpy of fusion ( $\Delta H_f$ ) of crystalline phases, both of which are important for validating plasticization models and predicting the viscoelastic behavior of films under different storage conditions.

SEM analysis of film fracture surfaces provides important morphological information regarding brittle versus ductile failure mechanisms, void distribution, and nanofiller dispersion homogeneity, which cannot be adequately inferred from mechanical testing alone. Furthermore, the combination of SEM surface profiling with energy-dispersive X-ray spectroscopy (SEM-EDS) allows verification of the spatial distribution of ZnO nanoparticles or cellulose nanocrystals (CNCs) within the carrageenan matrix, thereby confirming the effectiveness of the claimed nanocomposite dispersion [21],[47],[60].

The integration of FTIR, SEM, TGA, and DSC data constitutes a minimum analytical standard commonly expected by high-impact international materials journals (Scopus Q1) for the publication of research on biopolymer films.

### **c. Absence of Water Vapor Permeability (WVP) Measurement**

Water vapor permeability (WVP) is a critical barrier parameter for food packaging applications because it directly determines the shelf life of moisture-sensitive products. The reference studies did not measure WVP in accordance with ASTM E96 or ISO 2528. Reported WVP values for optimized  $\kappa$ -carrageenan films generally range from 1.5 to  $6.3 \times 10^{-10}$  g m/m<sup>2</sup> s Pa [21],[47],[61],[62], which is approximately one to two orders of magnitude higher than that of LDPE ( $\sim 0.08 \times 10^{-10}$  g m/m<sup>2</sup> s Pa), but still comparable to oriented polypropylene (OPP) ( $0.3$ - $1.2 \times 10^{-10}$  g·m/m<sup>2</sup> s Pa). The incorporation of 10% cellulose nanocrystals (CNC) has been reported to reduce the WVP of  $\kappa$ -carrageenan films by 30-45% through a tortuous-path mechanism that retards water vapor diffusion [21],[63],[55]. Similarly, the incorporation of microbially induced CaCO<sub>3</sub> reduced WVP to as low as  $2.05 \times 10^{-10}$  g·m/m<sup>2</sup> s Pa in *Kappaphycus alvarezii*-based films, which is lower than that of conventional biodegradable mulch films [35],[42],[64],[65].

## **CONCLUSIONS**

This critical review systematically presents a comprehensive analysis of the biochemical foundations, experimental evidence, methodological rigor, and developmental trajectories of

*Eucheuma cottonii*-based bioplastics. The analysis integrates two primary experimental studies [17],[18],[68] within a synthesis of 92 references. The key conclusions are as follows:

1. *Eucheuma cottonii* occupies a strategic position as a bioplastic feedstock in Southeast Asia. Its high kappa-carrageenan content (25-45% dry weight), an annual biomass availability of approximately 10 million tons in Indonesia and the Philippines [69],[70], and an established supply chain render it a readily exploitable biopolymer source without land-use conflicts.
2. Glycerol concentration is a critical variable controlling the mechanical properties of *E. cottonii* bioplastic films, exhibiting a statistically significant inverse relationship with tensile strength (ANOVA  $F=115.32$ ,  $df = 2,6$ ,  $p=1.63 \times 10^{-5}$ ). Converting tensile force values to MPa using film dimensions is essential for integrating data into global material databases in accordance with ASTM D882 standards.
3. The addition of chitosan forms polyelectrolyte complexes with kappa-carrageenan via electrostatic interactions, increasing hydrolytic resistance by up to 144-fold (reducing dissolution time from 2-3 days) [18]. And providing antimicrobial functionality. Moreover, sodium benzoate acts as an additional antimicrobial agent without altering the hydrophilic character of the carrageenan matrix, making it suitable for fast-dissolving single-dose packaging [71].
4. All *E. cottonii* bioplastic formulations demonstrate excellent biodegradability, with complete soil decomposition occurring within 2-12 days [17],[18]. The separation of biodegradability effects from glycerol concentration allows for mechanical property optimization without environmental compromise, indicating high end-of-life ecological compatibility [39],[48],[57],[71].
5. Nanocomposite reinforcement, including cellulose nanocrystals (CNC) at 5-10% loading [21],[22],[72], ZnO nanoparticles at 1-3% [39], and carrageenan-PLA blends [24],[73], provides an empirically validated strategy to bridge performance gaps relative to conventional synthetic films. Modeling using Halpin-Tsai equations and composite mechanics analyses enables theoretical performance prediction prior to fabrication.

Significant methodological limitations identified in the reference studies must be addressed, including:

1. Adoption of standardized tensile testing (ASTM D882) using a universal testing machine (UTM).
2. Comprehensive FTIR, SEM, and TGA/DSC characterization.

3. Measurement of water vapor permeability (WVP) in accordance with ASTM E96; and
4. Biodegradability assessment under controlled composting conditions per ISO 14855. Without these standards, findings on *E. cottonii* bioplastics remain isolated from the global literature.

This review also identifies research gaps requiring further experimental validation, including:

1. Lack of standardized tensile strength data in MPa from major production regions (Indonesia and the Philippines), limiting integration into predictive global models.
2. Insufficient SEM-based film morphology characterization to understand pore distribution, matrix homogeneity, and underlying structural failure mechanisms.
3. Absence of thermal stability analyses (TGA/DSC) to define safe processing windows for pilot-scale extrusion.
4. Lack of WVP data, constraining certification for food packaging applications.

In the context of regional innovation, developing local laboratory capacities at universities in Kupang (NTT) and Sulawesi, including the acquisition of UTM, FTIR, and TGA/DSC instruments, represents a strategic investment to enhance the international competitiveness of Indonesian scientific publications. Future research priorities from a mechanical engineering perspective include:

1. Pilot-scale continuous film extrusion trials (single or twin screw extruders, 5-50 kg/h capacity) to validate processing of *E. cottonii* / PLA or carrageenan/thermoplastic starch blends, with melt rheology characterization (viscosity  $\eta$ , back pressure  $\Delta P$ , shear rate  $\dot{\gamma}$ , Deborah number  $De$ ).
2. Development of predictive nanocomposite models using Halpin-Tsai equations and finite element analysis (FEA) to optimize CNC or ZnO NP distribution prior to fabrication.
3. Life cycle assessment (LCA) comparing *E. cottonii* bioplastics to LDPE, PLA, and PHA across production, use, and end-of-life stages [8].
4. Toxicological and regulatory assessment for food-contact approval under BPOM (Indonesia), EFSA (EU), and FDA (USA) frameworks [38],[43].
5. Integration of *E. cottonii* bioplastic production into an integrated biorefinery scheme that simultaneously extracts phycoerythrin pigments, functional proteins, and bioactive compounds, distributing capital costs across multiple revenue streams to achieve economic competitiveness relative to petroleum-based packaging [9],[19].

Overall, the convergence of established cultivation infrastructure, robust biochemical evidence, superior biodegradability, and advances in nanocomposite science positions *E. cottonii*-based bioplastics as a strategic contributor to the global circular economy and the achievement of Sustainable Development Goals (SDG 12, 13, 14) in the Asia-Pacific region.

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