

Review

Valorization of Coconut By-Products Using Eutectic Solvents: A Comprehensive Review on Green Extraction

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Abstract

Coconut (*Cocos nucifera* L.) is one of the most widely cultivated tropical crops, generating substantial by-products during industrial processing. Although rich in lignocellulosic components, phenolic compounds, lipids, and other bioactive molecules, they are often discarded or burned as low-grade fuel, causing environmental pollution and the proliferation of disease-carrying vectors. The sustainable valorization of these by-products is therefore essential to reduce environmental impact and enhance the economic value of the coconut chain. In this context, eutectic solvents (ES) have emerged as powerful potential green alternatives to conventional organic solvents for the extraction and fractionation of biomolecules. ES are formed through hydrogen-bonding interactions between hydrogen-bond donors and acceptors, yielding tunable solvents with low volatility, high selectivity, and biocompatibility. Their application to coconut biomass can enable the recovery of high-value compounds such as lignin, tannins, phenolic compounds, and fatty acids. This review provides a comprehensive overview of ES-based strategies for coconut by-product valorization, highlighting solvent compositions, main by-products explored, and target compounds extracted. Furthermore, it compares ES efficiency with traditional techniques and identifies current research gaps. Finally, the review highlights challenges and future directions for expanding ES application toward full coconut utilization, emphasizing their critical role in advancing sustainable, green chemistry.

Keywords: eutectic solvents; coconut by-products; extraction methods; green chemistry



Academic Editor: Agnieszka
Zgoła-Grześkowiak, Magdalena
Ligor and Tomasz Grześkowiak

Received: 4 May 2026

Revised: 13 June 2026

Accepted: 24 June 2026

Published: 27 June 2026

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1. Introduction

The coconut palm tree (*Cocos nucifera* L.) is considered one of the most important tropical species domesticated by humans. Known as the “Tree of Life,” coconut palms hold significant socio-cultural and economic value in many regions worldwide [1]. From the processing of the coconut fruit, it is possible to generate more than 100 products, by-products, and services with applications across various sectors—including food, cosmetics, textiles, and others—whether derived from the mature or immature fruit, processed or in its natural form [2,3].

In terms of production, according to the Food and Agriculture Organization of the United Nations Statistics Division (FAOSTAT), approximately 65.5 million tons of coconuts were produced worldwide in 2024, with the Philippines, Indonesia, India, and Brazil being

the four largest producers [4]. It is estimated that the international coconut products market generates around \$11.5 billion per year [5], with coconut water, milk, oil, and copra among the most commercially valuable products [6].

As an economically significant and growing market, the production and consumption of coconuts and their derivatives generate substantial by-products, including husks, shells, testa, residual water, and press cake (coconut meal). It is estimated that coconut processing can generate over 3 million tons of by-products annually. However, quantifying coconut waste generation is particularly challenging, as a significant fraction is improperly discarded after consumption as food [7,8]. This improper disposal poses an alarming environmental risk by promoting the proliferation of disease-carrying vectors and microorganisms and contaminating soil and water sources [9].

From a chemical perspective, coconut by-products are valuable sources of biomolecules, including lignin, cellulose, hemicellulose, tannins, phenolic acids, flavonoids, and fatty acids [3,10,11]. These bioactive compounds have high potential for application in pharmaceuticals, cosmetics, and nutraceuticals due to their antioxidant, antimicrobial, and anti-inflammatory properties [12]. Furthermore, the lignocellulosic fraction in these by-products is of great biotechnological interest, as it can be converted into various bioproducts, such as biofuels, biopolymers, and high-value chemicals [3]. Therefore, the valorization of coconut by-products represents a promising strategy for producing high-value materials, thereby minimizing waste generation.

Due to the complexity and variety of compounds present in the different coconut by-products, various fractionation and extraction strategies can be employed to obtain the desired molecules. Bioactive compounds such as phenolic acids, tannins, flavonoids, and others are extracted from a wide range of plant matrices (including coconut by-products) using methods such as maceration, Soxhlet extraction, solvent extraction, and percolation, and more recently through eco-friendlier techniques such as ultrasound-assisted extraction (UAE) and microwave-assisted extraction (MAE) [13]. On the other hand, the fractionation and extraction of the different lignocellulosic fractions—lignin, cellulose, and hemicellulose—can be achieved through various approaches, with methods such as acid or alkali treatment, hydrothermal processing, steam explosion, and organosolv extraction being the most commonly used [14]. Most of these methods have drawbacks, such as the use of larger quantities of solvents, long extraction times, high energy consumption due to temperature and pressure requirements, degradation of compounds during extraction, and the use of hazardous solvents [15–17]. Such drawbacks underscore the need for greener and more efficient extraction strategies that align with sustainable processing principles.

In this context, eutectic solvents (ES) have emerged as a sustainable alternative to conventional solvents in extraction processes. Although the term ‘deep eutectic solvent’ is more commonly found in the literature, the more general term ‘eutectic solvent’ is intentionally used throughout this text. According to Martin et al. [18], ‘deep eutectic solvent’ should be strictly defined as a mixture of pure compounds for which the eutectic point temperature is below that of an ideal liquid mixture. However, most studies cited in this review lack a thorough characterization of the prepared solvents, so they cannot guarantee that the specific eutectic temperature condition is met to classify them as ‘deep’. ES consists of a mixture of two or more components, with at least one acting as a hydrogen bond donor (HBD) and another as a hydrogen bond acceptor (HBA). This type of solvent is characterized by a decrease in the melting point of the mixture compared to its individual components. This condition arises from the interactions between them, especially hydrogen bonding interactions [19]. The growing interest in ES has arisen mainly from their advantages, such as ease of preparation, tunable properties, recyclability, relatively low toxicity compared to other organic solvents, potential biodegradability, low vapor

pressure, and low flammability, among others [20,21]. The nature and properties—and, consequently, their efficiency in different applications—of the ES are determined by the components used in preparation. A wide range of chemicals can be used in ES formulation, including quaternary ammonium salts like choline chloride, organic acids, carbohydrates, poly-alcohols, amides, and short-chain fatty acids [22].

Recently, ES have been successfully applied to extract a wide range of biomolecules from agricultural and food by-products, such as fruit peels [23,24], seeds [25,26], coffee by-products [27], rice straw [28,29], corn cob [30], wheat straw [31,32], and many others. Regarding coconut by-products, most literature studies focus on the extraction of the lignocellulosic fraction. Nevertheless, when compared to other biomass, the number of studies remains limited, both for lignocellulose and other bioactive compounds. The promising results observed in other biomass types suggest that eutectic solvents could be highly effective for the valorization of coconut by-products, enabling the recovery of high-value compounds while reducing environmental impact. This research gap highlights the opportunity for further exploration of eutectic-based extraction strategies in the coconut industry.

Therefore, this review aims to provide a comprehensive overview of recent advances in the use of eutectic solvents for the valorization of coconut by-products. The article presents the main by-products of coconut processing, the molecules that can be extracted from them, and a comparison of the performance of eutectic solvents with conventional solvents. Furthermore, it highlights current challenges, research gaps, and future directions for the sustainable use of eutectic solvents to valorize the coconut production chain fully.

To ensure a transparent and comprehensive overview, a literature search was conducted across the Scopus and Web of Science databases. The search strategy involved combinations of keywords related to the solvents and the specific biomass, including: (“eutectic solvent” OR “deep eutectic solvent”) AND (“coconut” OR “coconut by-product” OR “coconut husk” OR “coconut shell” OR “coconut testa” OR “coconut residue” OR “copra” OR “coconut oil” OR “coconut coir”). No specific start date restriction was applied, and the search included all available literature up to April 2026. The inclusion criteria encompassed peer-reviewed original research and review articles published that directly addressed the application of eutectic solvents to coconut by-products. Document types such as conference abstracts, patents, and studies focused on unrelated biomass sources or those that did not apply eutectic solvents for extraction on coconut by-products were excluded during the screening process.

2. Coconut Fruit: Structure, Processing, and By-Product Generation

The coconut palm (*Cocos nucifera* L.) is a tree, member of the *Arecaceae* family, known for providing food, medicine, fiber, fuel, and shelter for at least 500,000 years for numerous humans [33]. Due to this versatility of applications, the coconut palm has become one of the most important tropical crops in the world, earning the epithets “tree of life,” “tree of paradise,” and “tree of abundance” [34]. Gunn et al. [35] suggest two possible independent geographical origins for coconut cultivation: Southeast Asia and the southern margin of the Indian subcontinent. Initially, the species was spread to regions of Eastern Polynesia, the Indian Ocean, and the Pacific coast of Latin America through the seafaring activities of pre-Columbian Asian populations. From the 15th century onward, Europeans intensified the dissemination of coconut palms along the Atlantic coasts of Africa and South and Central America (especially the Caribbean).

Currently, coconut palms are distributed across nearly all continents, with a cultivated area of approximately 11.3 million hectares in 85 countries [4], thriving particularly well between latitudes 23° N and 23° S [33]. The species’ varieties can be classified into three

main groups: tall, dwarf, and hybrid. Tall varieties can reach up to 30 m in height, have a lifespan of approximately 90 years, bear their first fruits after about 7 years, and are primarily intended for the production of copra, oil, and fiber. In contrast, dwarf varieties reach around 15 m in height and are mainly used for the production of coconut water and sugar [2,36]. Hybrid varieties result from crosses between tall and dwarf types and generally combine traits that make them suitable for both fresh consumption and industrial processing [2].

The coconut, the primary product of the coconut palm, is a fibrous fruit composed internally of five main parts (Figure 1). A central cavity containing the solid endosperm (also called coconut ‘meat’ or kernel) and the liquid endosperm (coconut water) comprises the edible portions of the fruit. These parts are enclosed by three layers: the epicarp (outermost, also known as skin), mesocarp (husk), and endocarp (innermost, known as shell) [3]. Mature and young (green) coconut fruits exhibit some differences in their physical, chemical, and functional properties. In green fruits (6–8 months after pollination), the epicarp has a greener appearance. At the same time, the solid endosperm (meat) is thinner, softer, and more aromatic, with a higher sweetness potential, much like the liquid endosperm (water) [37,38]. In contrast, mature fruits exhibit a more yellowish or brown epicarp, with firmer, thicker meat rich in lipids [39,40]. The liquid endosperm has a lower carbohydrate content, resulting in a less-sweet flavor than in younger fruits [41].

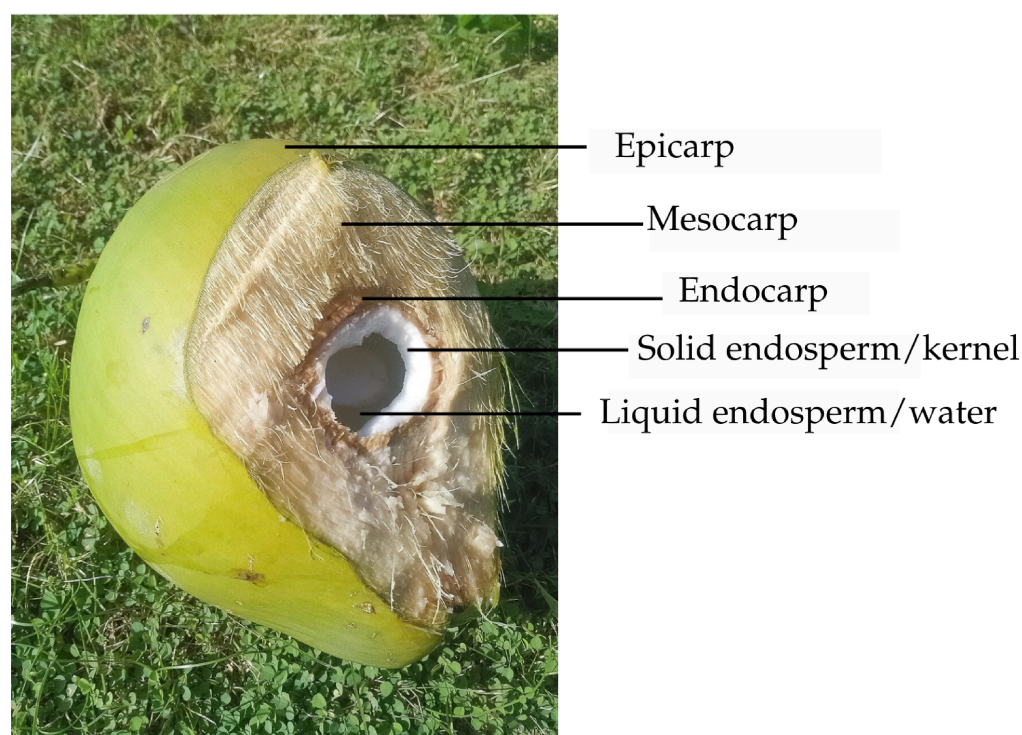


Figure 1. Coconut fruit parts. (Source: Own authorship).

Each part of the coconut fruit can be used to prepare various products. For instance, the immature (green) coconut is highly valued for food and beverage purposes, primarily due to the sweet flavor of its water and its isotonic properties [42]. The shells may be used as raw material for the production of activated carbon [12]. Furthermore, the fibers are frequently employed in the manufacture of bags, clothing, and handicrafts [42], as well as applied as inert substrates for agriculture and as additives in cement within the civil construction industry [5].

From a categorization perspective, coconut products can be divided into four groups corresponding to their respective value chains: food products, oil products, water products,

and by-products. Food products constitute the most diverse class, encompassing all products intended for human consumption—excluding coconut water—such as copra, coconut milk, coconut cream, desiccated coconut, and coconut chips [43]. Oil products include those based on coconut oil (crude or virgin) and its derivatives, such as soaps and cosmetics. Water products represent the commercialization of coconut water itself and certain derivatives, such as nata-de-coco. Finally, by-products involve all materials derived from the non-edible parts of the fruit, specifically the exocarp, mesocarp, and endocarp.

Depending on the fruit's maturation stage, processing aims to yield different products. Generally, green coconuts are primarily utilized for water extraction and the formulation of food products derived from the solid endosperm; they are also extensively marketed as whole fruits, particularly in tropical regions [5] or as diamond-shaped (after trimming) and snowball coconuts (after removing husks, shell, and testa) [44]. In contrast, the mature fruit offers a wider range of industrial applications, where its various components are utilized in both traditional sectors—such as oil extraction, copra production, and charcoal manufacturing—and non-traditional fields, including biofuel and coconut coir pith (dust) production, for example. Figures 2 and 3 present general flowcharts for the processing of green and mature coconuts, respectively, highlighting some of the primary products and by-products generated.

MATURE COCONUT PROCESSING

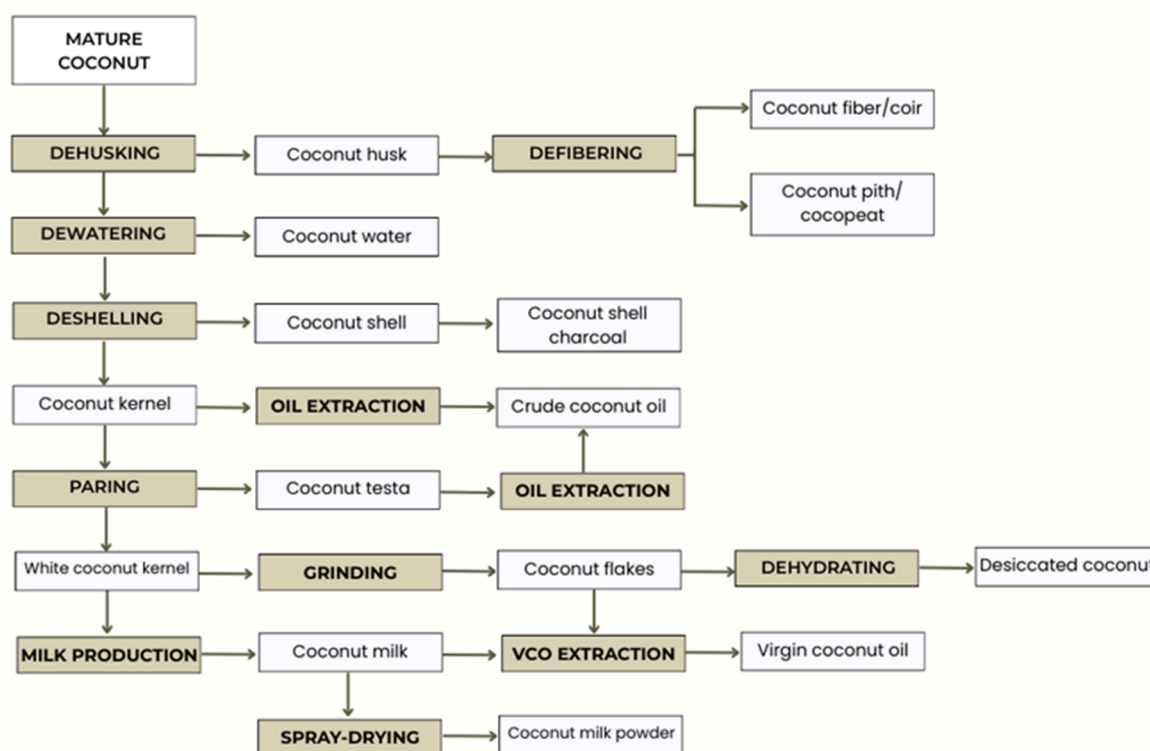


Figure 2. Mature coconut processing (Source: Own authorship).

As an economically relevant market with growing demand, the high volume of production and consumption of coconut and its derivatives generates a large amount of by-products. The type, quantity, and destination of these by-products depend primarily on the coconut's maturity level and the products of interest during processing and consumption. For instance, tender coconuts are generally used for their water and meat. Once the edible parts are consumed, the husks and shells are typically discarded or burned [42].

On the other hand, in mature coconuts, the pulp (kernel) is used in processing, while the water is usually discarded as waste. This occurs because the water from mature coconuts has a considerably less pleasant taste than that of green coconuts, resulting in much lower commercial appeal [45].

TENDER COCONUT PROCESSING

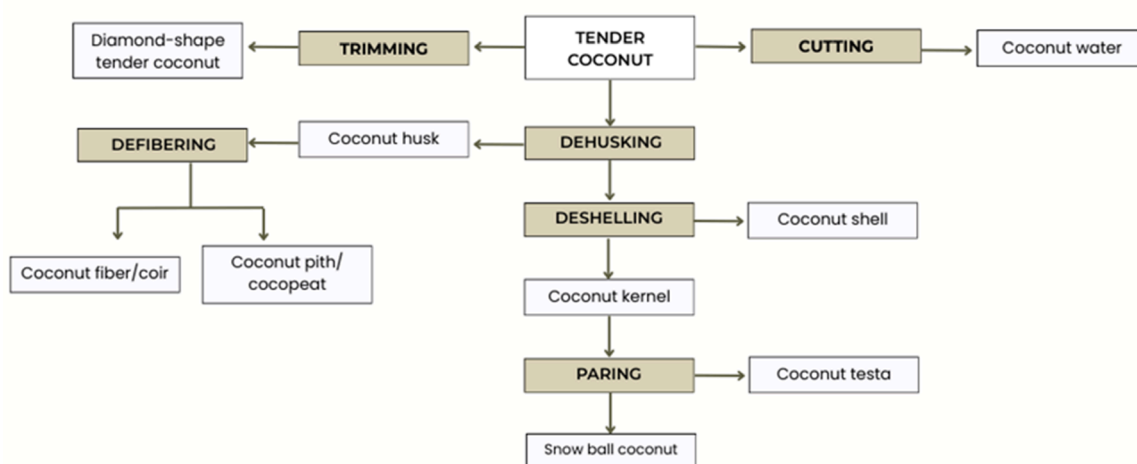


Figure 3. Young/green coconut processing (Source: Own authorship).

The main by-products of coconut processing and consumption include the husk, shell, testa, copra cake, and copra meal. Additionally, other types of by-products may be generated, such as mature coconut water, the epicarp, and coconut flour.

Coconut husk (mesocarp) and shell (endocarp) represent the most abundant by-products from coconut processing, accounting for about 35% and 15% of the fruit's total weight, respectively. It is estimated that industrial processing generates approximately 1.4 million tons of these fractions [46]. Both fractions are primarily composed of lignocellulosic compounds—lignin, hemicellulose, and cellulose—although they also contain other components such as phenolic compounds and tannins [47,48]. Table 1 presents a comparison between shells and husks in terms of lignocellulose composition.

Table 1. Lignocellulosic composition of husk and shell coconut by-products.

Material	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Reference
Husk (young coconut)	30.33–36.95	19.85–25.93	19.45–31.42	[49]
Husk (mature coconut)	37.36–41.30	16.31–25.12	36.16–48.85	
Husk	23.2	15.0	38.8	[50]
Husk	37.6	15.2	41.3	[51]
Shell	25.2	27.7	46.0	
Shell	32.1	39.6	28.3	[52]
Shell	10.4	15.2	33.7	[53]

The coconut husk is the mesocarp of the coconut fruit. It is a fibrous material that is normally processed for fiber extraction, yielding a major fraction of coir fiber (around 75%) and a minor fraction of fine material called coconut powder or cocopeat/coir pith [54]. Traditionally, the coir fibers are used as an agricultural substrate, as a concrete additive,

and in the manufacturing of clothing, bags, and accessories [54,55], while a small amount of coir pith is conventionally used in landfills and for manuring purposes [56,57].

Coconut shell is the hard, woody endocarp of the coconut fruit, lying between the husk and the edible kernel. This coconut by-product is primarily composed of lignocellulosic material—especially rich in lignin—and is recognized for its low ash content, high carbon content, and high calorific value [58]. Due to these characteristics, the shell is frequently used in the production of activated carbon and charcoal [59].

The coconut testa is another common by-product generated from coconut processing, primarily during the production of desiccated coconut, coconut milk, and virgin coconut oil. This fraction consists of a brown skin covering the external surface of the solid endosperm and is typically discarded or utilized in animal feed formulations [60]. The testa constitutes approximately 18% (*w/w*) of the total solid endosperm, resulting in a substantial volume of this by-product annually [61]. For instance, in India alone, the generation potential of this by-product is estimated at approximately 90,000 tonnes per year [62]. Regarding its composition, coconut testa is a lipid-rich by-product, with lipids accounting for approximately 60% of its content, predominantly composed of saturated fatty acids (88.75–91.23%) [63]. Furthermore, coconut testa is rich in phenolic compounds, particularly phenolic acids and flavonoids [10,61].

Other highly significant by-products of the coconut industry are the cakes derived from oil extraction. Both crude coconut oil and virgin coconut oil (VCO) are generally extracted from copra (the dried coconut meat). Oil extraction can be performed through mechanical means, using an expeller or hydraulic press, or via solvent extraction (typically utilizing *n*-hexane) [64]. The resulting by-product is termed copra cake when mechanically extracted and copra meal when obtained by solvent extraction [65,66]. The generation of these by-products is estimated at approximately 2 million tonnes per year [67]. Although these are similar carbohydrate-rich by-products, they exhibit distinct characteristics resulting from the extraction process, particularly regarding the percentage of residual oil [68].

In copra cake, the majority of the content is carbohydrates (32–45% *w/w*), primarily hexoses, including mannose (~80% *w/w*), glucose (~13% *w/w*), and galactose (~6% *w/w*). Additionally, copra cake has a protein content ranging from 18 to 27% *w/w* and residual lipid levels between 10 and 15% *w/w* [69,70]. Conversely, copra meal exhibits a lower residual oil content (~7%) [64], a high protein content (~17%), and a high carbohydrate fraction, primarily in the form of mannan or galactomannan [67,71].

It is estimated that coconut by-product generation is around 3 million tons per year. However, a more reliable quantification is particularly difficult, as a large fraction is burned or discarded in vacant lots and on beaches after consumption—especially in the case of tender coconuts. This improper disposal poses an alarming environmental risk, especially given the high resistance of coconut by-products to decomposition, which can take up to 10 years [7]. The accumulation of these by-products promotes the proliferation of various disease vectors such as mosquitoes, flies, rats, and cockroaches, potentially leading to the spread of diseases like dengue and cholera. Furthermore, other environmental and sanitary impacts, such as the clogging of public drainage systems, air pollution, and water contamination, can result from the inadequate disposal of coconut waste [12].

3. High-Value-Added Compounds from Coconut By-Products

3.1. Lignocellulose

In recent decades, lignocellulosic biomass has emerged as one of the primary natural and renewable sources of organic compounds, serving as a sustainable and promising alternative for the production of biofuels and high-value-added chemical compounds [72]. Their high availability, low cost, and chemical composition have driven a significant

increase in both academic and economic interest regarding the utilization of lignocellulosic biomass [73].

Lignocellulosic biomasses are basically composed of three main fractions—cellulose, hemicellulose, and lignin—which form a complex three-dimensional arrangement through their interactions and bonds, known as the lignin-carbohydrate complex (LCC) or lignocellulose [74]. This complex, which is part of the plant cell wall, is highly compact, stable, and resistant [75]. It is present in various plant materials, including forestry materials, plantation crops, agricultural residues, and agro-industrial waste [76]. In addition to the three main fractions, lignocellulosic biomasses also contain a smaller fraction of ash and extractives, which may include lipids, resins, tannins, terpenes, steroids, and phenolics [77].

Cellulose is a polysaccharide composed of glucose molecules linked by β -1,4-glycosidic bonds. It forms a linear, ordered polymer in which cellobiose (formed by the linkage of two glucose molecules) is the repeating subunit, creating long chains of more than 10,000 units. Structurally, cellulose forms fibrils held together by hydrogen bonds and van der Waals forces between glucose molecules [78]. Within the cell wall, elementary fibrils can associate to form more tightly packed, more resistant microfibrils, thereby increasing fiber recalcitrance. Based on these characteristics, cellulose polymers contain crystalline regions, which are more ordered and rigid, and amorphous regions, which are more disordered and more susceptible to chemical or enzymatic attack [79].

Hemicellulose is the designation given to a group of heterogeneous polymeric polysaccharides formed by ether bonds between pentoses (xylose, arabinose), hexoses (glucose, mannose, galactose, fucose, rhamnose), and gluconic acid [78]. The diversity of monomers, combined with the multiple bonding forms between them, provides hemicellulose with a variety of polysaccharides featuring distinct compositions and structures. These are generally classified as xyloglucans, xylans, mannans, galactans, etc., according to the predominant monosaccharide in their composition [80]. Unlike cellulose, hemicellulose possesses various branches in its structure, leading to the formation of an amorphous polymer with a lower degree of polymerization—approximately 200 polymeric units, making hemicellulose more prone to being broken down into monomeric units [78].

Lignin is a complex, three-dimensional amorphous biopolymer formed by three *p*-hydroxycinnamic alcohol monomers, known as monolignols: *p*-coumaryl, coniferyl, and sinapyl alcohols. These monolignols differ in the degree of methoxylation of the aromatic ring; during lignin synthesis, they are polymerized through coupling reactions to form the polymer subunits known as *p*-hydroxyphenyl (H), guaiacyl (G), and syringyl (S), corresponding respectively to *p*-coumaryl, coniferyl, and sinapyl alcohols [81]. These lignin subunits are linked by various types of ether and carbon-carbon bonds, which include β -O-4 (aryl ether), β - β (resinol), β -5 (phenylcoumaran), 5-5' (biphenyl), β -1' (1,2-diaryl propane), 4-O-5' diphenyl, and α -O-4. Among these linkages, the first four are the most frequent in lignin polymers—especially β -O-4, which can account for 45–85% of the structure of native lignins (protolignins) [82]. Other types of linkages are less common. The composition of lignins, regarding both the subunits (H, G, and S) and the types of bonds present, varies according to the biomass species, the lignin type (native or isolated), and the isolation method [83]. The relative composition of each component within lignocellulosic biomass varies depending on the type of biomass and factors such as climatic conditions and the plant's developmental stage.

Among coconut by-products, coconut shell and husk represent a very rich source of lignocellulosic compounds, as presented in Table 1. Vieira et al. [84] analyzed the characteristics and sustainable applications of coconut waste, especially lignocellulosic by-products from husk and shell. In their work, they not only identify the fundamental role of characterizing lignin, hemicellulose, and cellulose content to determine pretreatment strategies

that ensure the isolation of these fractions but also present several applications for these by-products. These include their use as civil construction materials, biofuel production, application as adsorbent materials, and feedstock for pyrolysis, among others, demonstrating the broad spectrum of potential value recovery for these wastes.

3.2. Phenolic Compounds

Phenolic compounds are molecules defined by the presence of one or more hydroxyl groups attached to an aromatic ring. These compounds are widely distributed throughout plant species, where they act primarily as secondary metabolites with fundamental roles in plant protection [85]. Phenolic compounds can be classified based on various criteria, one of the main ones being the structure of the carbon skeleton, in which phenolic compounds are divided into two major classes: flavonoids (e.g., anthocyanins, flavanols, flavanones, flavonols, flavones, and isoflavones) and non-flavonoids (e.g., phenolic acids, stilbenes, and tannins) [86].

Flavonoids are structurally characterized by a C6-C3-C6 carbon skeleton, consisting of two aromatic rings (rings A and B) connected to a three-carbon heterocyclic ring containing one oxygen atom (ring C) [87]. Flavonoids represent a highly diverse class of phenolic compounds, with various structural modifications to the basic carbon skeleton—such as acylations, hydroxylations, methylations, and glycosylations—resulting in a further division into six main subclasses: flavonols, flavones, flavan-3-ols, anthocyanins, flavanones, and isoflavones [88].

Phenolic acids constitute a prominent class of organic acids derived from either hydroxybenzoic or hydroxycinnamic precursors, distinguished by the presence of at least one carboxylic functional group (-COOH) [89]. Hydroxybenzoic acid derivatives are characterized by a C6-C1 carbon framework, featuring a carboxyl group linked to a benzene ring; this group encompasses essential compounds such as gallic, protocatechuic, and vanillic acids [90]. In contrast, hydroxycinnamic acid derivatives possess a C6-C3 structure, where the cinnamic acid backbone typically undergoes hydroxylation at various positions on the aromatic ring. Key representatives of this subclass include p-coumaric, caffeic, and ferulic acids [91].

Phenolic compounds have garnered substantial attention due to their multifaceted health-promoting effects and wide-ranging industrial applications, particularly within the food, cosmetic, and pharmaceutical sectors [92,93]. These molecules are prized for their potent antioxidant, anti-inflammatory, and antimicrobial activities, which underpin their role in managing chronic diseases through dietary intake [94,95]. In the food industry, phenolics serve as versatile additives, acting as natural antioxidants [96], colorants [97], and preservatives that enhance both the nutritional profile and the shelf-life of products [98]. Beyond traditional uses, their integration into advanced technological platforms—such as hydrogels [99], nanoparticles, and specialized delivery systems [100]—highlights their strategic importance in modern biotechnology.

Numerous studies demonstrate the presence of a wide variety of phenolic compounds within coconut by-products, specifically phenolic acids and flavonoids. Research conducted by Leliana et al. [47,101], Li et al. [102], and Valadez-Carmona et al. [103], among others, has identified a broad spectrum of bioactive molecules across different coconut fractions. These findings highlight the occurrence of key phenolic acids, such as gallic, protocatechuic, chlorogenic, and vanillic acids, as well as flavonoids like catechin, epicatechin, and kaempferol. Furthermore, the identification of high-value compounds such as vanillin underscores the industrial potential of these by-products.

3.3. Tannins

Unlike other phenolic compounds, tannins are polyphenolic polymers with high molecular weight, typically ranging from 500 to 30,000 Da [104]. These compounds can be categorized into two types: hydrolyzable and condensed tannins (also known as proanthocyanidins). Hydrolyzable tannins are derived from gallic acid (gallotannins) or ellagic acid (ellagitannins) linked to carbohydrates—usually glucose—by esterification; these can be broken down into their monomers through hydrolysis reactions. In contrast, condensed tannins are molecules formed through covalent bonds between flavan-3-ols (catechin) and/or flavan-3,4-diols (leucoanthocyanidins). This type of tannin typically exhibits greater resistance to degradation [105,106].

Coconut by-products are rich in tannins, particularly within the mesocarp and endocarp fractions, and predominantly condensed tannins [107]. These tannins perform various biological functions in the fruit, playing critical structural and defensive roles in plant species [108] due to their potent antioxidant and antimicrobial properties [109,110]. The presence of these polyphenols is not only biologically significant but also industrially relevant. Specifically, their antioxidant and antimicrobial activities make them valuable for applications in food preservation and pharmaceuticals. Furthermore, other intrinsic characteristics of these polymers allow them to be utilized as natural precursors for adhesives [111], eco-friendly corrosion inhibitors [112], and natural dyes for textiles—offering sustainable alternatives to synthetic dyes [113], among other diverse applications.

3.4. Lipid/Fatty Acids

Coconut processing generates a significant volume of lipid-rich by-products, primarily from oil extraction processes, including coconut testa, copra cake, and copra meal. Some studies demonstrate that these by-products contain substantial amounts of lipids, which are particularly characterized by a high concentration of medium-chain fatty acids (MCFAs), such as caprylic, capric, and lauric acids [61,70,114]. Beyond their overall lipid content, research has revealed distinct chemical profiles for oils recovered from these by-products compared to conventional kernel oil. For instance, testa oil exhibits a higher proportion of mono- and polyunsaturated lipids than standard kernel oil [115].

Another high-lipid by-product is the residue derived from coconut milk extraction. Coconut milk is typically obtained through the disintegration and cold pressing of the solid endosperm [116], a process that generates a significant secondary material known as coconut milk residue. This by-product retains a substantial oil content, ranging from 24 to 38 wt%, which is primarily composed of lauric (46%) and myristic (19%) acids [117].

These medium-chain fatty acids are of significant industrial relevance, particularly when formulated as medium-chain glycerides (MCGs). MCGs have a broad spectrum of applications across the cosmetic and food industries [118,119]. Specifically, medium-chain triglycerides (MCTs) have gained prominence due to their therapeutic and functional benefits, including applications in treating metabolic and gastrointestinal disorders, parenteral nutrition, and atherosclerosis [119,120]. Beyond clinical use, MCTs serve as effective green solvents for extracting lipophilic bioactive compounds [121], as well as excipients and drug delivery systems in pharmaceutical formulations [122], and even as antimicrobial agents [123]. Such a vast range of applications underscores the importance of coconut by-products as a versatile raw material for high-value industrial production.

4. Extraction with Eutectic Solvents

4.1. General Concepts of Eutectic Solvents and Principles of Green Extraction

Eutectic solvents are solvents composed of two or more components, where at least one is a hydrogen bond donor species, and another is a hydrogen bond acceptor species.

This type of solvent is characterized by a decrease in the melting point of the mixture relative to its individual components—a condition arising from the interactions between them, primarily hydrogen bonding [19].

Since the term was first introduced by Abbott et al. [124], who observed the melting point depression of mixtures of quaternary ammonium salts with urea compared to the pure components, eutectic solvents have emerged as a new class of green solvents suitable for a wide range of applications. The growing interest in eutectic solvents is largely due to their beneficial characteristics, such as ease of preparation, tunable properties, recyclability, relatively low toxicity compared to other organic solvents, potential biodegradability, low vapor pressures, and low flammability, among others [20]. All these features make this type of solvent an excellent substitute for traditional solvents in chemical processes, particularly from environmental and safety perspectives, as eutectic solvents—depending on their composition—frequently follow many of the principles of green chemistry [125].

Eutectic solvents can be classified under different categorizations, but one of the most widely adopted is based on the chemical nature of the components. According to this, they can be categorized into five types: types I, II, and III are formed by quaternary ammonium salts—primarily choline chloride (ChCl)—and metal halides (e.g., AlCl_3), hydrated metal halides (e.g., $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$), and different HBDs (e.g., carboxylic acids, polyols), respectively. Type IV consists of inorganic transition metal salts (e.g., ZnCl_2) with different HBDs [20,126], while type V comprises eutectic solvents composed of non-ionic species (e.g., thymol, menthol) [127].

Beyond the chemical nature of their components, eutectic solvents can also be categorized based on their physicochemical properties, which directly influence their affinity for specific target molecules. Regarding their affinity for water, they are divided into hydrophilic and hydrophobic solvents. Hydrophilic eutectic solvents, often based on components like choline chloride and polar molecules such as sugars, organic acids, etc., are characterized by miscibility in water and polar solvents [128]. Most of the eutectic solvents traditionally used for extraction—especially those based on choline chloride are hydrophilic solvents, as they are highly effective for extracting polar compounds such as certain phenolic compounds [13,129]. On the other hand, as the name suggests, hydrophobic eutectic solvents—typically composed of fatty acids, terpenes (e.g., menthol, thymol), or quaternary ammonium salts with long alkyl chains—are those with water immiscibility and hydrophobic nature [126,130]. Hydrophobic eutectic solvents have emerged as superior green alternatives for recovering non-polar substances, including volatile fatty acids [131], bioactive compounds such as lycopene [132] and specific terpenes [133], which exhibit a higher affinity for non-polar solvents [134].

The characteristics of eutectic solvents enable their use in a variety of processes, especially in green technology. Much of their popularity and the interest they generate stems from their application in the treatment of lignocellulosic biomass, mainly due to their ability to fractionate lignin, hemicellulose, and cellulose [135]. However, several other fields have successfully applied eutectic solvents in processes such as reaction media for inorganic material synthesis [136], extraction of phenolic compounds [129], separation media for azeotropic mixtures [137], pesticide extraction [138], metal removal from marine fish [139], CO_2 capture [140], lithium battery applications [141], and the activation and stabilization of enzymes [142,143], among countless other uses.

4.2. Applications of Eutectic Solvents in Extraction

The application of eutectic solvents for extracting compounds from plant biomass is widely established. Countless studies published over the last two decades have demonstrated the efficiency of these solvents in recovering a diverse array of molecules from

a wide range of biomasses. Among the most common types of target molecules are lignin [144,145], phenolic compounds [146–148], tannins [149,150], polysaccharides such as pectin [151,152] and xylan [153,154], and saponins [155,156], among many others.

Despite the widespread adoption of eutectic solvents for the extraction of plant biomasses, it is striking that so few studies have addressed the extraction of coconut residual biomass. In this review, only 14 studies involving the direct use of eutectic solvents on coconut by-products were identified (Table 2). Most of these efforts have focused primarily on deconstructing the lignocellulosic matrix of the mesocarp (husk/coir) and endocarp (shell) to isolate lignin. Only four of these studies targeted the extraction of other molecules [114,157–159]. Furthermore, only one study utilized a by-product distinct from the mesocarp or endocarp, specifically coconut milk powder [114], which, although often considered a product, serves here as a proof-of-concept for lipid extraction in coconut biomass.

Regarding lignin extraction, certain trends are evident, such as a clear predominance of eutectic solvents based on choline chloride and carboxylic acids, particularly lactic and oxalic acids. This trend is supported by the fact that acidic eutectic solvents generally exhibit higher lignin removal efficiency than neutral or alkaline systems. This occurs due to the ability of carboxylic acids to supply more acidic protons, which effectively promotes the cleavage of linkages within the lignocellulose matrix—specifically the ether and ester linkages of the lignin-carbohydrate complex [160]. Mankar et al. [161], for instance, compared the lignin extraction potential of acidic eutectic solvents against a system containing ChCl and urea during their initial screening. They observed a recovery yield of less than 1% with the urea-based solvent, whereas the acidic solvents yielded near 30%. Upon optimization with a ChCl:lactic acid (1:4) system, an 82% (*w/w*) recovery was achieved, further highlighting the role of acidity in lignin extraction. Zulkiflee et al. [159] also demonstrated this pH influence, comparing different eutectic solvents using glycerol as the HBD. Among the tested systems—ZnCl₂:glycerol (1:7), AlCl₃:glycerol (1:7), ChCl:glycerol (1:2), and Betaine:glycerol (1:2)—the highest cellulose recovery (and, by direct consequence, the highest lignin removal) occurred with AlCl₃:glycerol (1:7), which exhibited a significantly more acidic pH than the other candidates.

Table 2. Extraction of compounds from coconut by-products with eutectic solvents.

Material	Compounds Extracted	Method	Solvents	Optimal Extraction Conditions	Extraction Yield	Reference
Coconut coir	Lignin	MAE + ES	ChCl:lactic acid (1:4)	SLR = 1:30, 20 min, 150 °C	82% lignin recovery	[161]
Coconut coir	Lignin	ES	ChCl:lactic acid (1:10)	SLR = 1:20, 160 °C, 3 h	68.51% lignin recovery	[162]
Coconut coir	Lignin	ES	ChCl:oxalic acid (1:1) ChCl:ZnCl ₂ (1:2)	SLR = 1:20, 100 °C, 24 h	-	[163]
Coconut coir	Lignin	ES	ChCl:urea (1:2)	SLR = 1:20, 120 °C, 1 h	60.93% delignification	[164]
Coconut husk	Lignin	UAE + ES	ChCl-based ES ChCl:glycerol:urea (1:1:1)	Bath: SLR = 1:250, 90 min, 65 °C, 95 W, 50–60 Hz Probe+bath: SLR = 1:250, 35 min, 80% amplitude	Bath: 100 ± 10% lignin recovery Probe+bath: 90 ± 10% lignin recovery	[165]
Coconut husk	Lignin	ES	ChCl:lactic acid (1:2)	SLR = 1:10, 120 °C, 12 h	17% lignin recovery	[166]
Coconut husk	Lignin	ES	ChCl:monoethanolamine (1:4) Betaine:lactic acid (1:4)	SLR = 1:10, 121 °C, 6 h	60.53% lignin recovery 65.81% lignin recovery	[167]

Table 2. Cont.

Material	Compounds Extracted	Method	Solvents	Optimal Extraction Conditions	Extraction Yield	Reference
Coconut husk	Lignin and sugars	ES	ChCl:Lactic acid (1:10)	HT: SLR = 1:20, 210 °C, 78 min ES: SLR = 1:30, 120 °C, 6 h	10.1% TRS 50.2% lignin recovery	[157]
		Sequential HT + ES	Water and ChCl:Lactic acid (1:10)		31.3% TRS 48.8% lignin recovery	
Coconut shell	Lignin	ES	K ₂ CO ₃ :ethylene glycol (1:7)	SLR = 1:15, 130 °C, 1 h	70.7% delignification	[168]
Coconut shell	Lignin	ES	Tetramethylammonium hydroxide-based ES	SLR = 1:15, 50 °C, 3 h	63.33–67.37% delignification	[169]
Coconut husk	Cellulose	ES	ZnCl ₂ :glycerol (1:7) AlCl ₃ :glycerol (1:7) ChCl:glycerol (1:2) Betaine:glycerol (1:2)	SLR = 1:12, 100 °C, 100 min	39.39% (ZnCl ₂ :glycerol) 48.72% (AlCl ₃ :glycerol) 38.04% (ChCl:glycerol) 38.37% (Betaine:glycerol)	[159]
Coconut shell	Phenolics	ES	ChCl:ascorbic acid (1:2)	SLR = 1:10; 20 min, 25 °C	TPC = 3715.67 mg GAE/L	[158]
Coconut milk powder	Lipids and phytosterol	UAE + ES	ChCl:ethylene glycol (1:2):n-hexane (30:70)	SLR = 1:10, 60 °C, 1 h	Lipid content 56.35 g/100 g Phytosterol content 604.11 mg/kg	[114]

SLR: solid–liquid ratio (*w/v*); UAE: ultrasound-assisted extraction; MAE: microwave-assisted extraction; ES: eutectic solvent; HT: hydrothermal treatment; ChCl: choline chloride; GAE: gallic acid equivalent; TPC: total phenolic content; TRS: total reducing sugar.

Despite the effectiveness of acidic systems in achieving high yields, a significant drawback is the extensive condensation of the extracted lignin and the degradation of hemicelluloses. To address this, recent studies have shifted toward alkaline-based eutectic solvents to prioritize structural preservation. He et al. [168,169] demonstrated that using K₂CO₃:ethylene glycol and tetramethylammonium hydroxide-based solvents on coconut shell could achieve delignification rates of approximately 70% under remarkably mild conditions (50–130 °C). Most notably, these alkaline systems preserved up to 88% of the native β -O-4 linkages, as confirmed by Heteronuclear Single Quantum Coherence Nuclear Magnetic Resonance (HSQC-NMR) analysis. This structural integrity is a critical advantage over the acidic treatments discussed previously, as it facilitates subsequent valorization of lignin into high-value aromatic molecules, such as vanillin and monomers, while enabling high recovery of intact hemicelluloses (over 70%). This trend toward the extraction of lignin with a preserved structure has been widely investigated in recent years in several studies [170–172].

The physicochemical properties of the eutectic solvent intricately govern the extraction efficiency of target compounds from coconut by-products. In addition to the influence of solvent pH on extraction yields, another fundamental property is the HBA:HBD molar ratio. An increase in the HBA:HBD ratio typically alters the solvent's supramolecular structure, decreasing its viscosity and enhancing mass transfer across the dense lignocellulosic matrix, thereby improving yields. Zheng et al. [162] observed a gradual increase in lignin extraction from coconut coir by increasing the ChCl:lactic acid molar ratio from 1:2 to 1:5 and finally to 1:10, raising the lignin recovery from approximately 57% to 66%. Mankar et al. [161] also observed a similar effect, where increasing the molar ratio of the same solvent from 1:2 to 1:4 improved the lignin extraction efficiency from 73.9% to 82%. However, an increase beyond this threshold (1:6, 1:8) led to a decrease in extraction efficiency, demonstrating that

the relationship between efficiency and molar ratio is not strictly linear, and an optimum molar ratio is required to achieve the maximum extraction efficiency.

Viscosity modulation is a decisive factor in solvent selection. Although Zulkiflee et al. [159] did not find a direct correlation between cellulose recovery and solvent viscosity, they evaluated several molar ratios of ZnCl_2 :glycerol and AlCl_3 :glycerol solvents (ranging from 1:1 to 1:7) and selected those with the lowest viscosity for application. Another frequently used strategy to modulate the viscosity of eutectic solvents is the controlled addition of water (typically 5–20% *v/v*). This addition modifies the internal hydrogen-bond network of the solvent, impacting diffusion and solute mobility without severely compromising the solvent's affinity for target molecules [173]. Nevertheless, none of the studies covered in this review investigated the effect of water content on eutectic solvents on extraction efficiency.

Beyond the intrinsic properties of the ES, operational parameters, such as extraction temperature and time, also exert a synergistic effect on extraction efficiency. Elevated temperatures are widely recognized to lower solvent viscosity and enhance molecular diffusion, thereby accelerating mass transfer rates [13]. Consequently, several studies compiled in this review consistently report a positive trend, where lignin extraction efficiency increases alongside rising temperatures [161,162,168,169]. However, this behavior is non-linear and typically exhibits an upper threshold. For instance, Mankar et al. [161] observed a distinct decline in extraction efficiency when the temperature was raised from 150 °C to 170 °C. This underscores a commonly observed parabolic trend, in which yields improve up to an optimal thermal point, beyond which higher temperatures lead to a loss in efficiency—often attributed to thermal degradation of target compounds or to undesirable lignin condensation. This specific non-linear optimization trend is frequently associated with extraction time, as also evidenced in the work of Chakane et al. [166].

Regarding the observed yields, a comparative critical analysis of the reported efficiencies becomes a complex task. Studies frequently oscillate between two distinct metrics: 'delignification' (the percentage of lignin removed from the solid matrix) and 'lignin recovery' or 'lignin yield' (the mass of lignin successfully precipitated and isolated from the liquid fraction). Authors such as Zheng et al. [162], Chakane et al. [166], and Wijaya et al. [157] evaluated the treatment efficiency of their coconut by-products based on lignin recovery values—68.51%, 17%, and 50.2%, respectively. In contrast, other authors report only the delignification values obtained, as seen in the works of He et al. [168,169]. Notably, only Agrawal et al. [164] reported results using both metrics, allowing for a more comprehensive assessment. This dichotomy regarding efficiency metrics requires further caution: while high delignification rates indicate the solvent's potency in deconstructing biomass, they do not necessarily translate into high recovery yields, as small phenolic fragments can remain solubilized in the liquid fraction, for instance.

Furthermore, the evaluation of extraction efficiency using the 'lignin recovery' metric has the potential for overestimation. For example, Carbonell et al. (2026) [165] reported a remarkable lignin yield of $100 \pm 10\%$ using a system composed of ChCl :glycerol:urea (1:1:1) combined with UAE in an ultrasound bath. While this result indicates a vastly superior efficiency compared to conventional NaOH extraction (~59%), it should be interpreted with caution. Since the yield is calculated relative to a baseline lignin content, the inherent experimental scatter and the potential co-precipitation of non-lignin components can yield values that theoretically exceed the total measured lignin. In this case, it is essential to evaluate the purity of the lignin recovered in the process—a step that few studies have actually performed.

A better way to evaluate the efficacy of eutectic solvents is to compare them against conventional methods, such as alkaline or organosolv extractions. Agrawal et al. [164] com-

pared the efficiency of eutectic solvents and organosolv treatment in lignin extraction of coconut coir. They tested four organosolv systems (ethanol + FeCl₃, ethanol + FeCl₃ with reflux, diethylene glycol + NaOH, and diethylene glycol + FeCl₃) and three eutectic solvents ChCl:oxalic acid (1:1), ChCl:urea (1:2), and ChCl:lactic acid (1:2). The organosolv treatments could achieve higher delignification results (39–71%) compared to eutectic solvents (39–60%). However, organosolv lignin recoveries were significantly lower (16–42%) than the results observed with eutectic solvents (70–94%), indicating that eutectic solvents not only can promote the removal of lignin from lignocellulosic matrix but also facilitate its recovery after the treatment.

With respect to phenolic compounds, only the study by Aziz et al. [158] presents an extraction approach for this class of compounds from coconut by-products. The authors compared the extraction of phenolics using various eutectic solvents containing ascorbic acid, glucose, sucrose, and lactic acid as HBDs against a conventional aqueous extraction. They achieved a maximum total phenolic content of 3715.67 mg GAE/L at room temperature using a eutectic solvent composed of choline chloride and ascorbic acid (1:2). This system—along with seven other solvents containing ascorbic acid—proved significantly more efficient than conventional aqueous extraction, which yielded only 2427.50 mg GAE/L under similar conditions. Furthermore, GC-MS analysis revealed that the extracts derived from the eutectic solvents contained approximately 26 different compounds, including molecules such as lycopene, astaxanthin, lycopene, and α -carotene. Notably, four specific phenolic compounds were identified—1,2,4-benzenetricarboxylic acid, 1,2-dimethyl ester; 2-nonaprenyl-6-methoxyphenol; trimethyl[4-(1,1,3,3-tetramethylbutyl)phenoxy]silane; and 2',6'-Bis(trimethylsiloxy)acetophenone—which were absent in the aqueous extract.

While the majority of studies focus on the valorization of lignocellulosic fibers, Cui et al. [114] expanded the application of eutectic solvents to the recovery of lipids and phytosterols from coconut milk powder. Unlike the direct extraction methods used for lignin or phenolics, the authors employed a hybrid system composed of choline chloride and lactic acid (1:2) and n-hexane (in a 30:70 ratio). This integrated approach achieved a lipid yield of 56.35 g/100 g, which was lower than that of the other solvents tested (n-hexane, absolute ethanol, dimethyl carbonate, and cyclopentyl methyl ether). However, this hybrid system could achieve a good phytosterols extraction of 604.11 mg/kg, with the best result for squalene, cholesterol, and campesterol extraction in the study. These results demonstrate the capacity of eutectic systems to extract hydrophobic molecules, thereby creating opportunities for the application of hydrophobic eutectic solvents.

Another notable aspect is the integration of ES with non-conventional emerging technologies, such as ultrasound-assisted extraction (UAE) and microwave-assisted extraction (MAE), which have gained significant attention as a strategy in extraction processes, offering advantages such as shorter process times, lower solvent consumption, and potentially greater extraction yields [128,129]. While the combination of ES and these enabling technologies is already a well-established trend for the extraction of phenolic compounds and bioactive molecules across various biomass sources, its application for lignin isolation remains comparatively scarce in the literature. However, recent studies demonstrate the immense potential of this synergistic approach. Specifically in the case of coconut by-products, Mankar et al. [161] observed that combining MAE with ES significantly enhanced lignin extraction efficiency, yielding a six-fold increase compared to the identical process conducted under conventional heating for the same duration. Furthermore, compared with a fully optimized 8 h conventional heating process, the MAE-assisted method achieved nearly twice the efficiency, demonstrating that this association can drastically reduce extraction times. This highlights that while integrated approaches for biopolymer isolation

are not yet a dominant trend, they represent a highly promising frontier for optimizing biomass fractionation.

Despite the relatively limited number of reports focused on the extraction of compounds from coconut by-products using eutectic solvents, the comparison with extraction using traditional methods can be very valuable for deeper analysis. Table 3 summarizes a series of studies involving the use of these methodologies for the extraction of compounds from coconut by-products. Traditionally, the extraction of compounds from coconut by-products has been dominated by conventional methods, such as maceration, Soxhlet extraction, and UAE, using organic solvents such as methanol, ethanol, acetone, and hexane. In the specific case of lignin, other methodologies such as organosolv treatments, acid, and alkaline hydrolysis are considered more traditional methods for the fractionation of lignocellulosic biomass [174].

Table 3. Extraction of compounds from coconut by-products with conventional methods.

Material	Compounds Extracted	Method	Solvents	Optimal Extraction Conditions	Extraction Yield	Reference
Coconut coir	Lignin	Organosolv	Acetic acid 93%: HCl 0.3%	Lignin: SLR = 1:20, 110 °C, 3 h	30.1% (lignin)	[175]
	Nanocellulose	Acid hydrolysis	H ₂ SO ₄ (60% <i>w/w</i>)	Nanocellulose: SLR = 1:20, 60 °C, 6 h	59.8% (nanocellulose)	
Coconut husk and shell	Lignin	Ionic liquid extraction	<i>N,N,N</i> -dimethylbutylammonium hydrogen sulfate ([DMBA][HSO ₄])	SLR = 1:10, 170 °C, 45 min	77% delignification (husk) 82 delignification (shell)	[51]
Coconut coir	Lignin	Organosolv	Ethanol 50% (<i>v/v</i>) + FeCl ₃ 3% (<i>w/v</i>)	SLR = 1:20, 90 °C, 3 h	71.03% delignification	[164]
Coconut husk	Lignin	Organosolv	Ethanol:water (65:35 <i>v/v</i>) + H ₂ SO ₄ 0.5% (<i>w/w</i>)	SLR = 1:10, 190 °C, 1 h	2.79% lignin recovery	[176]
Coconut husk	Lignin	Organosolv	Ethanol 95% (<i>v/v</i>) + NaOH	SLR = 1:20, 80 °C, 6 h	34% lignin recovery	[177]
		UAE	1,4-dioxane 95% (<i>v/v</i>)	SLR = 1:15, 30 °C, 7 h	19% lignin recovery	
Coconut husk	Polysaccharides	Alkaline treatment	5% NaOH	SLR = 1:20, 121 °C, 40 min	TRS = 45 g/100 g DW	[178]
Coconut husk	Oligosaccharides	UAE	Aqueous NaOH solution 1.05% (<i>w/v</i>)	SLR = 1:127, 248 W, 5 min, 30 °C	TC = 14.29 mg/mL TRS = 3.84 mg/mL	[179]
Coconut husk	Xylan	Alkali and steam treatment	20% NaOH (<i>w/v</i>)	SLR = 1:10, 25 °C (alkali) 121 °C, 15 psi (pressure), 60 min (steam)	82%	[180]
Coconut husk	Cellulose	Acid digestion	10% <i>v/v</i> NaOCl + 2 N HCl	1 h, 25 °C	48.10%	[181]
Coconut husk	Cellulose	Steam explosion	Water	121 °C, 20 lbs (pressure), 1 h	45.52%	[182]
		Alkali-acid hydrolysis	0.05 N Nitric acid 0.1 M NaOH	70 °C, 1 h (acid treatment) 45 °C, 3 h (alkali treatment)	80.32%	
Coconut coir	Tannins	Solvent extraction	Water Ethanol	SLR = 1:30, 70 °C, 2 h	8.117 mg/g (water) 4.517 mg/g (ethanol)	[107]
Coconut husk	Tannins	Maceration	Water	SLR = 1:10, 80 °C, 3 h	16.34%	[110]
Coconut husk	Tannins	Solvent extraction	Water+ 5% Na ₂ SO ₃	SLR = 1:15, 80 °C, 2 h	3.37%	[111]
Coconut husk	Phenolics	UAE	50% Ethanol	SLR = 1:20, 70 °C, 5 min, 200 W, 26 kHz	48.05 mg GAE/g	[101]
Coconut shell and husk	Phenolics	UAE	Ethanol	SLR = 1:20, 45 °C, 1 h, 320 W, 37 kHz	39.41% (husk) 39.21% (shell)	[47]
Coconut shell	Phenolics	UAE	Methanol	SLR = 1:24, 33 °C, 15 min,	TPC = 40.99 mg GAE/g TFC = 36.13 mg QE/g TTC = 176.73 mg TAE/g 38% total yield	[48]

Table 3. Cont.

Material	Compounds Extracted	Method	Solvents	Optimal Extraction Conditions	Extraction Yield	Reference
Coconut testa	Phenolic acids and flavonoids	UAE	80% Acetone (phenolics) 80% Methanol (flavonoids)	SLR = 1:10, 60 °C, 1 h	TPC = 167 mg GAE/g TFC = 115 mg GAE/g	[10]
Coconut shell	Phenolics	UAE	50% Ethanol	SLR = 1:50, 30 °C, 15 min, 150 W, 25 kHz	22.44 mg TAE/g	[183]
Coconut shell and coir	Phenolics	Solvent extraction	Methanol, ethanol, and acetone	SLR = 1:20, 20 °C, 8 h	47.89 mg GAE/g (shell) 1035.61 mg GAE/g (coir)	[184]
Coconut testa	Phenolics	Solvent extraction	70% Ethanol	SLR = 1:10, 25 °C, 48 h	TPC = 44.61 mg GAE/g TFC = 67.60 mg QE/g	[185]
Coconut testa	Phenolics	Solvent extraction	0.1 M HCl acidified Ethanol	SLR = 1:10, 75 °C, 90 min	TPC = 154.39 mg GAE/g TFC = 53.65 mg QE/g	[62]
Coconut coir	Phenolics	Solution extraction	Water	SLR = 1:10, 28 °C, 30 min	TPC = 76.04 mg GAE/g TFC = 1.57 mg QE/g TTC = 522.95 mg TAE/g	[186]
Coconut skin	Flavonoids	UAE	60% Ethanol	SLR = 1:40; 50 °C, 90 min, 150 W	366.03–596.38 mg RE/g	[11]
Coconut testa	Phenolic acids	Solvent extraction	Water	SLR = 1:10, 100 °C, 15 min	0.78 mg GAE/g	[187]
Coconut milk powder	Lipids and phytosterol	UAE	n-hexane Dimethyl carbonate Cyclopentyl methyl ether Ethanol	SLR = 1:10, 60 °C, 1 h	Lipid content: 65.66 g/100 g (best result ethanol) Phytosterol content: 644.26 mg/kg (best result cyclopentyl methyl ether)	[114]
Coconut copra cake	Lipids	UAE MAE Sohxlet extraction	Hexane	Sohxlet: SLR = 1:40, 60 °C, 48 h UAE: SLR = 1:30, 60 °C, 30 min, 120 W, 40 kHz MAE: SLR = 1:10, 60 °C, 15 min, 800 W	81.39% (Sohxlet) 75.80% (UAE) 62.97% (MAE)	[188]
Coconut copra cake hydrolysate	Lipids	Solvent extraction	Hexane	SLR = 1:8, 50 °C, 10 min	190 mg/g (80% recovery)	[70]
Coconut testa	Lipids	UAE	80% Methanol	SLR = 1:4, 60 °C, 3 h	76.83% (total oil content)	[189]

SLR: solid–liquid ratio (*w/v*); UAE: ultrasound-assisted extraction; MAE: microwave-assisted extraction; DW: dry weight; GAE: gallic acid equivalent; QE: quercetin equivalent; TAE: tannic acid equivalent; RE: rutin equivalent; TPC: total phenolic content; TFC: total flavonoid content; TTC: total tannin content; TC: total carbohydrates; TRS: total reducing sugar.

Analyzing not only Table 3 but also the wide range of studies available in the literature, a significant quantitative gap is notable between these established protocols and the emerging use of eutectic solvents. While the conventional literature provides an extensive mapping of diverse coconut biomasses, such as husk [180–182], shell [48,51], copra cake and meal [69,188], and testa [61,185], ES research remains in its nascent stages, with most reports focusing on husk and shell. Furthermore, the diversity of target compounds is also a very discrepant factor between the different approaches. While in methodologies using eutectic solvents most studies seek lignin extraction/delignification, with few cases of different target molecules, conventional methodologies have been used to extract various types of target molecules, such as phenolic compounds in general [101,184,186], flavonoids [10,11], phenolic acids [187], tannins [107,110,111], and lipids [70,188,189].

Several factors may account for the limited number of studies and the clear preference for investigating lignin extraction. Initially, eutectic solvents gained significant traction as a pretreatment method for lignocellulosic biomass due to their ability to solubilize

the lignin fraction [20]. This opened a new perspective on pretreatment, where efficient delignification served as a prerequisite for subsequent valorization—primarily for the conversion of polysaccharide fractions into biofuels, such as bioethanol. This trend has become evident in various studies over recent years [102,190,191], including the work of Yerizam et al. [167], in which the authors utilized eutectic solvents to promote the delignification of coconut husk and enhance bioethanol yields. Furthermore, the global shift toward “lignin-first” biorefinery concepts has repositioned this biopolymer from a low-value by-product to a strategic platform for aromatic compounds, such as vanillin and phenols [192].

Within this context, the higher prevalence of studies involving husk and shell is understandable, considering the high lignin content in these by-products. Furthermore, the fact that these two by-products represent the primary waste streams generated during coconut consumption and processing in terms of volume also helps explain the research focus on these specific biomasses [193,194].

The comparison between traditional extraction methods and eutectic solvents (ES) reveals strategic research gaps, particularly concerning underutilized biomasses and specific target molecules within the coconut production chain. Although residues such as the mesocarp (husk) and endocarp (shell) already have a considerable mapping of phenolic compounds through traditional solvents (Table 3), the coconut testa emerges as a substrate with high potential for the application of green technologies. The transition to ES-based extraction is supported by significant efficiency gains; for instance, Aziz et al. [158] demonstrated that the application of these solvents can increase phenolic extraction yields by approximately 1.5 times compared to aqueous methods. This scenario suggests that the data consolidated by conventional methodologies serve as a roadmap to validate the technical superiority of ES in residual fractions that remain largely unexplored.

Furthermore, the scarcity of reports on the use of ES for tannin recovery in coconut by-products represents a great opportunity for scientific innovation. The viability of this approach is corroborated by successes achieved in other lignocellulosic biomasses, such as the work of Xue et al. [195], who achieved a tannin yield 2.1 times higher (104.40 ± 4.3 mg TAE/g DW) in chestnut burr waste using a betaine:urea (1:2) solvent. In parallel, the application of hydrophobic eutectic solvents for the extraction of lipids from copra cake, meal, or testa offers a promising pathway to enhance process selectivity and sustainability. Given the lipophilic nature of these matrices, hydrophobic eutectic solvents can overcome the limitations of conventional volatile organic solvents—such as hexane—allowing for a more efficient recovery of lipids/fatty acids [196,197].

Despite being potential alternatives to conventional solvent processes and presenting clear advantages over them, the application of methods using eutectic solvents still faces some barriers due to certain drawbacks, especially related to the scalability of extraction processes.

The high viscosity presented by eutectic solvents compared to conventional solvents is one of the most commonly noted disadvantages, which can hinder mass transfer, leading to longer extraction times and potentially altering the solubility of the target compounds—a factor that could be detrimental in continuous and industrial-scale processes. In general, this drawback can be overcome by adding water or increasing the temperature [15,198]. The optimization of temperature and water content in ES extraction processes can even lead to an increase in extraction efficiency. For instance, Dai et al. [199] observed higher extraction efficiency of carthamin by adding 25% of water to a solvent composed of ChCl:sucrose.

Another significant disadvantage is associated with the recovery of the extracted compounds and the solvent itself. Due to the low vapor pressure exhibited by ES, recovering the extracted products by simple evaporation of the solvent is not possible, unlike

with conventional solvents, which adds purification efforts when considering industrial applications [200,201]. Among the main strategies for compound recovery are liquid–liquid extraction (LLE), solid–liquid extraction (SLE), chromatographic purification, and precipitation by anti-solvent addition [200]. Among these, SLE is the most common for phenolic compounds, utilizing resins and molecular sieves [129], while the addition of anti-solvents (typically water or ethanol) is especially effective for lignin precipitation [202]. This latter approach can be even more valuable since it does not require specific equipment or additional fees or investments, allowing the development of sustainable processing [129]. Regarding the recycling and reuse of ES, although some studies point to potential positive results [202–204], showing that ES can be reused for several extraction cycles with the loss of extraction efficiency minimally altered—leading to a lower cost per extraction and reducing solvent waste—the large-scale adoption of these strategies still lacks a thorough assessment of economic viability and sustainability [205].

5. Conclusions

Coconut by-products, primarily comprising the husk, shell, and testa, represent a significant portion of the fruit’s total mass and are characterized by a complex lignocellulosic matrix. If mismanaged, these residues pose substantial environmental risks; however, they also constitute a sustainable and low-cost source of high-value compounds, including lignin, tannins, phenolic acids, flavonoids, and lipids. As demonstrated throughout this review, eutectic solvents have emerged as notably efficient and versatile tools for recovering these components, offering a superior alternative to conventional organic solvents. The analyzed studies indicate that ES not only enhances extraction yields but also preserves the chemical integrity and biological activities of the extracts. Furthermore, their tunable properties allow for tailored strategies to recover both hydrophilic and hydrophobic compounds, reinforcing their role within a sustainable biorefinery framework.

Despite this promising potential, the transition from laboratory research to industrial application requires addressing several critical hurdles. Future research should prioritize expanding the scope of ES application beyond the predominant husk and shell to include underexplored biomasses such as coconut testa, copra cake, and meal. Moreover, broadening the target molecules to include specific tannins and lipids through the development of hydrophobic eutectic systems represents a strategic frontier for innovation. Technologically, overcoming the inherent high viscosity of certain formulations and establishing efficient solvent recyclability protocols remains essential for mass transfer optimization. Finally, to consolidate ES as a viable technology, it is imperative to conduct techno-economic and life cycle assessments, alongside more rigorous evaluations of toxicity and biodegradability. Addressing these gaps will be fundamental to transforming coconut waste into high-added-value inputs for the pharmaceutical, food, and materials industries.

Author Contributions: Conceptualization, L.d.S.S., R.P., M.L.L.P., I.I.J. and B.D.R.; formal analysis, L.d.S.S.; investigation, L.d.S.S., I.I.J. and B.D.R.; resources, I.I.J. and B.D.R.; writing—original draft preparation, L.d.S.S. and R.P.; writing—review and editing, L.d.S.S. and B.D.R.; supervision, B.D.R., I.I.J. and M.L.L.P.; project administration, I.I.J. and B.D.R.; funding acquisition, I.I.J. and B.D.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by FAPERJ, grant number E-26/210.780/2025 and E-26/200.492/2026, and by CNPQ, grant number 308830/2022-9.

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Acknowledgments: The authors thank the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES), Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), and Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro (FAPERJ) for the support of this work.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

ChCl	Choline chloride
DW	Dry weight
ES	Eutectic solvents
FAOSTAT	Food and Agriculture Organization of the United Nations Statistics Division
GAE	Gallic acid equivalent
GC-MS	Gas chromatography-mass spectrometry
HSQC-NMR	Heteronuclear Single Quantum Coherence Nuclear Magnetic Resonance
HBA	Hydrogen bond acceptor
HBD	Hydrogen bond donor
HT	Hydrothermal treatment
LCC	Lignin-carbohydrate complex
MCFA	Medium-chain fatty acids
MCG	Medium-chain glycerides
MCT	Medium-chain triglycerides
MAE	Microwave-assisted extraction
QE	Quercetin equivalent
RE	Rutin equivalent
SLR	Solid–liquid ratio
TAE	Tannic acid equivalent
TC	Total carbohydrates
TFC	Total flavonoid content
TPC	Total phenolic content
TRS	Total reducing sugar
TTC	Total tannin content
UAE	Ultrasound-assisted extraction
VCO	Virgin coconut oil

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