Review

A Review on the Applications of Coffee Waste Derived from Primary Processing: Strategies for Revalorization

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Abstract: Coffee is an extremely popular beverage worldwide. To obtain it, the berry must be depulped, fermented, washed, dried, and roasted, producing residues: pulp and husk, mucilage, and parchment. Recently there has been an interest in generating high-value products. In this article, advances in the valorization process are critically reviewed, including an overview of the composition of residues derived from primary processing, uses in food, biocomposite, and biofuel production (thermochemical conversion). With an increasing production of coffee projected in the coming years, there is an urgent need to balance it with the appropriate use and industrial application of coffee wastes and by-products, which are renewable resources rich in carbohydrates, proteins, pectin, and bioactive compounds (polyphenols). The applications described above, together with those that will undoubtedly be developed in the future, represent promising opportunities to take advantage of agro-industrial residues derived from primary processing of Coffea spp. and develop more efficient and sustainable systems through biorefinery approaches and the circular economy.

Keywords: coffee waste; biorefinery; biocomposite; biofuel; circular economy

1. Introduction

Coffee is an important food commodity globally due to the infusion prepared from roasted and ground beans, with approximately 60 tropical and subtropical countries producing coffee extensively. However, 90% of such countries are underdeveloped, while consumption usually occurs in industrialized economies [1]. According to the Food and Agriculture Organization of the United Nations (FAO) [2], world coffee production is so important that a cultivated area of about 10 million hectares is estimated, thus supporting more than 25 million families [3]. The world production of green coffee was estimated to be 10,688,153 tons in 2020 [2], which is mainly distributed between Arabic (Coffea arabica) and Robusta (Coffea canephora) varieties. These coffee varieties represent about 60% and 35% of the world production, respectively. The remaining percentage is made up of species of Coffea liberica [4]. The Arabic variety predominates with Colombian soft beans in Colombia, Kenya, Tanzania, Bolivia, Costa Rica, and Zambia, while the Robusta variety (with Brazilian natives) is more frequent in Brazil, Ethiopia, Paraguay, Vietnam, Thailand, and Indonesia. These coffee varieties are traded in the most important stock exchanges worldwide, with Brazil, Vietnam, and Colombia being the most prominent producers (Figure 1). Specifically, they are responsible for more than 50% of the world coffee production, with more than three million hectares planted [1,2].
The coffee fruit, berry, or cherry is a bean of approximately 10 mm [5,6] comprising the exocarp (pulp), mesocarp (mucilage), silver skin, and hulls or parchment, which is a layer that surrounds the two seeds (endosperm) (Figure 2). The primary transformation process of coffee generates substantial quantities of solid and liquid organic wastes that are estimated to amount to ten million tons per year [7].

![Coffee fruit structure](image)

**Figure 2.** Coffee fruit structure.

Specifically, coffee waste mainly derives from the commercial product (bean) separation from the fruit, generating residual materials mainly in the form of pulp and peel, wastewater, and parchment. The transformation process of coffee starts from harvest, and all postharvest operations directly affect the quality of the final product. First, the harvest is conducted by taking the red cherries from the shrubs. Then, the cherries are subjected to a process that can be wet, semi-dry, or dry, depending on the resources available at the production site. The traditional process takes place in wet conditions. In wet processing, after harvesting, cherries are put into a tank where the green cherries are separated from the ripe ones by density. Afterwards, depulping is carried out, leaving the pulp and peel as residual fraction, while the mucilage is removed through a fermentative process (12–18 h). Subsequently, the grains are washed with abundant water and dried afterwards in solar dryers or ovens, to reach a 10–12% final moisture content. However, due to the
remarkable volume of highly polluted wastewater generated, cleaner and more efficient technologies are currently being promoted to reduce the environmental impact of coffee wet processing [8]. In semi-dry processing, grains are subjected directly to drying after pulping, until the required moisture content is reached. Pulp and skin are subsequently separated from the grain. In contrast, the dry process is characterized by cherries being dried directly, and then the husk (skin, pulp, and mucilage) is removed; it is estimated that 1 kg of husk is produced per kg of grain [9,10]. Figure 3 shows a flowchart of the three processing methods, including the primary residues generated in each one [11–14].

In general, processing every 60,000 tons of dried coffee beans produces approximately 218,400 tons of fresh pulp and mucilage or mesocarp [15]. In terms of wastewater from wet processing, 40–45 L/kg of coffee are generated [16]. This wastewater contains a chemical oxygen demand (COD) as high as 3000–7000 mg/L [17–19]. In wet processing, residual pulp and wastewater are usually generated in rural areas where the postharvest process is carried out, leading to proliferation of microorganisms, diptera, and odor emissions that might affect the surrounding populations, water sources, and/or agricultural soils [20]. On the other hand, semi-dry and dry processing also generate polluting wastes, especially in transforming parchment, obtained when coffee is threshed or peeled. It is estimated that 1 kg of parchment is generated per kg of coffee processed [6,14].

These facts have led to the recent concern about the environmental impact of coffee processing, which has promoted a remarkable increase in research studies carried out to recycle and valorize the residual streams generated in such an important agro-industrial process. Figure 4 shows the annual number of publications on Web of Science (https://www.webofscience.com/wos/alldb/basic-search, accessed on 5th of September 2022) containing the terms “waste coffee” and “by-product coffee”. Notably, the numbers of...
such publications quadrupled in the last 10 years, indicating that the use of coffee by-products and residues has acquired marked importance under sustainable development and the bio-economy. In this context, as described before, current production and consumption patterns generate massive quantities of waste that must be adequately managed to minimize their negative environmental impacts and, in turn, contribute to developing more sustainable processes [21].

![Figure 4. Annual number of published papers related to coffee wastes and by-products (source: Web of Science, accessed on 5 September 2022).](image)

In fact, the environmental sustainability of agro-industrial production systems of coffee is being reviewed and evaluated due to the programs that are being developed worldwide. In this sense, FAO suggests avoiding non-recycled biowaste as one of the five pathways to a sustainable, green, and circular bio-economy. In addition to the United Nations’ 2015 Agenda for Sustainable Development, FAO sets out 17 Sustainable Development Goals for 2030, among which the following ones can be highlighted: Good health and well-being; clean water and sanitation; affordable and clean energy; sustainable cities and communities; responsible consumption and production; climate action; life below water; and life on land [22,23].

The implementation of new alternatives to manage wastes and by-products derived from primary processing of coffee might allow the transformation of the linear economy into a bio-economy or circular economy, in such an important agro-industrial sector. In this context, sustainable use or exploitation of biological resources is promoted, recirculating or reusing residual fractions derived from the primary productive process [24]. Therefore, biorefining is important as an industrial system aiming at the sustainable and efficient use of biomass, valuing the potential of varied materials, and delivering multiple bioenergy sources and valuable bioproducts for the cosmetic, pharmaceutical, food, or energy industries, among others [25].

Consequently, with an increasing production of coffee projected in the coming years, there is an urgent need to balance it with the appropriate use and industrial application of coffee wastes and by-products, which are renewable resources rich in carbohydrates, proteins, pectin, and bioactive compounds (polyphenols) [5,6,12,16,19]. Therefore, it is imperative to seek appropriate management alternatives for such residual fractions. This review article presents a novel description of the different uses that might be currently applied to coffee wastes and by-products, to propose systems of use that allow their appropriate management in different parallel production lines of food, cosmetics, pharmaceuticals, en-
ergy, and bioremediation. Specifically, it seeks to explore the possibility of adding value to coffee wastes and by-products through the implementation of combined and concatenated environmentally friendly processes within the frame of the circular economy.

2. Characterization of Wastes and By-Products from Coffee Processing

The exocarp of coffee bean, also known as the pulp, is generally green in unripe fruit but turns violet-red or deep red as it matures due to anthocyanins. Typically, the pulp is removed by mechanical movements during pulping and constitutes about 29–43% (w/w) of the fruit [12,26]. The mesocarp, which is usually removed by using large volumes of water, is a translucent, colorless, thin, viscous, and highly hydrated mucilage (pectin) hydrogel that represents about 11.8% (w/w) of the fruit. In its composition, mesocarp contains 85% moisture, while the dried fraction may contain 4% of reducing sugars, 36% pectin, and 1% ash [26]. Additionally, coffee fruit consists of a thin, yellowish endocarp called parchment that covers the grain and is visible until the moment of drying. Parchment hardens during the fruit ripening and delimits the coffee seed size [27]. During the transformation process of the grain, parchment is removed by mechanical movements called threshing. Parchment represents approximately 6.1% (w/w), while the dried, roasted, ground, and infused seed constitutes up to 55% of the fruit (w/w) [12]. The silver skin is also part of the structure of the coffee fruit; this is removed during roasting and accounts for around 4.2% of the coffee bean [28]. Considering these figures and the production of coffee described previously, the generation of the most representative wastes and by-products derived from its primary processing can be as high as 3000, 816, and 421 thousand tons per year of pulp, mucilage, and parchment, respectively, in Brazil (while Vietnam might generate: 1360, 373, and 192 thousand tons per year; and Colombia: 670, 184, and 95 thousand tons per year, respectively).

Table 1 shows the characterization of the pulp, mucilage, husk, and silver skin as reported in the scientific literature. As can be seen, pulp and husk have a higher percentage of carbohydrates (associated with cellulose, hemicellulose, and lignin), which indicates that they might have a high potential in fermentation processes. However, it might be necessary to carry out chemical, biological, and physical hydrolysis to increase the concentration of reducing sugars available. In the case of caffeine and chlorogenic acid, they have been identified in all the residual fractions derived from primary processing of coffee, which confirms their potential in the pharmaceutical, cosmetic, or food industries. But specific and efficient extraction processes need to be designed. Moreover, the protein content is variable in pulp, husk, and silver skin, while digestibility evaluations or characterization of the specific type of protein available are necessary. In all cases, the presence of a wide variety of nutrients in coffee wastes and by-products is evident, confirming the convenience of evaluating their extraction or recycling.

Table 1. Composition of different fractions of coffee grain (in percentages).

| Variable          | Pulp             | Husk             | Silver Skin |
|-------------------|------------------|------------------|-------------|
| Dry matter        | 23 ± 1           | n.a.             | n.a.        |
| Carbohydrates     | 35–50 ± 2,3      | 57.8 ± 3         | 44 ± 3      |
| Protein           | 5.2–12.0 ± 2,3   | 9.2 ± 3          | 16.2–18.6 ± 3 |
| Fiber             | 18–30 ± 2,3      | 31.9 ± 3         | 60–80 g     |
| Ash               | 1.5–8.9 ± 1,3,4  | 6 ± 3            | 0.5–1.0 ± 1,4 |
| Caffeine          | 0.28–1.20 ± 3    | 1.2 ± 3          | 1.4 ± 3     |
| Tannins           | 1.8–8.6 ± 3      | 4.5–9.0 ± 3      | 0.02 ± 3    |
| Chlorogenic acid  | 10.7 ± 3         | 12.59 ± 3        | 15.82 ± 1,3 |
| Cellulose         | 24.5–63.0 ± 3,4  | 43 ± 3           | 40–49 ± 1,4 |
| Hemicellulose     | 29.7 ± 4         | 7 ± 3            | 25–32 ± 1,4 |
| Lignin            | 23.7 ± 4         | n.a.             | 33–35 ± 1,4 |
| Moisture          | n.a.             | n.a.             | 8.2 ± 3     |

n.a.: Data not available. 1 [26], 2 [6], 3 [29], 4 [5], 5 [30].
3. Use of Coffee Waste in Animal Feeding

Given the nutrient content of the residual streams is derived from primary processing of coffee fruit, their use as complement in animal feeding could be an interesting alternative. However, uses in animal feeding are limited. Pedraza-Beltrán et al. [31] evaluated the effect of including different fractions of coffee pulp in the diet of dairy cows (10%, 15%, and 20%). They monitored the impact on body weight, yield, and milk composition and reported that coffee pulp could replace 20% of conventional food without significant differences in those variables under the study conditions [31]. In addition, Didanna [32] confirmed that it is possible to include coffee pulp in cattle food (10–30% in weight). But the author described how the presence of phenolic compounds led to little weight gain in ruminants, so higher percentages would not be recommended. In birds, the inclusion of more than 10% coffee waste showed toxic symptoms, while the inclusion of 13% did not affect the growth of fish (Tilapia). Specifically, caffeine concentrations between 2.4 and 4.6 g/kg led to low nutrient digestibility mainly due to anti-nutritive factors that can affect the acceptability and palatability of the pulp. The anti-nutritive substances include tannins, other polyphenols, caffeine, and potassium, which might cause mortality in small animal species at high concentrations. They can also generate mortality in ruminants fed only with coffee pulp or mixed at high ratios in their diet. In fact, caffeine has been found to stimulate motor activity and high energy consumption and, therefore, a decrease in weight gain, directly affecting meat production. Nevertheless, Rathinavelu and Graziosi [33] mentioned that silage under appropriate conditions might become an alternative for converting those “anti-nutritive” substances. The final product derived from the fermentation process might be used for animals with different gastric behaviors (monogastric and polygastric animals) or in aquaculture and, in some cases, as a substrate for some plant species growth. In this context, with a favorable cost factor, since the cost of silage is meagre and its benefits to the final product might be attractive, the optimal combination to use coffee wastes and by-products depends on each specific animal species. However, it is still necessary to conduct additional in vitro and in vivo studies to further evaluate their gastrointestinal dynamics and the effect of the controlled intake of some anti-nutrient compounds (caffeine and tannins). Furthermore, it is necessary to generate alternatives in the elaboration of food to elucidate and determine whether hydrolysis or processing operations could be designed to diminish anti-nutrient effects, thus allowing a more comprehensive application of waste and by-products derived from coffee primary processing.

4. Extraction of Biocomponents

Although the physico-chemical characteristics of residual streams derived from primary coffee processing might vary depending on the manufacturing process (wet, semidry, or dry), other uses are currently being promoted instead of landfilling or application as animal feeding (i.e., use of pulp as fertilizer or soil amendment, and the shell as fuel for roasting coffee grain). However, an interesting potential use for the most abundant residual fraction (coffee pulp) focuses on the extraction of anthocyanins, caffeine, and phenolic compounds, which might add significant value to the coffee processing chain [11,13,34].

A wide variety of fruits and vegetables provide a range of nutrients and different bioactive compounds, including phytochemicals (phenolics, flavonoids, and carotenoids), vitamins (vitamin C, folate, and provitamin A), minerals (potassium, calcium, and magnesium), and fiber [35]. Specifically, coffee is considered a source of antioxidants belonging to the family of hydroxycinnamic acids, where caffeic, cumaric, chlorogenic, synaptic, and ferulic acids are included, in addition to other biochemically active compounds with considerable interest as functional ingredients (caffeine, cafestol, kahweol, nicotinic acid, and trigonelline) [34]. The smell derived from roasting is attributed to furans, pyrazines, phenols and ketones, pyrroles, hydrocarbons, carboxylic acids, esters, alcohols, and aldehydes, among others [36]. On the other hand, polyphenols are made up of phenolic compounds and flavonoids, which have recently gained significant interest due to their therapeutic activity as anti-inflammatory, antimicrobial, and antihypertensive agents. Flavan-3-ols,
flavanols, and anthocyanidins can be found within phenolic compounds, as well as epicatechin, catechin, and rutin [37,38]. In this context, caffeine is one of the main active compounds in coffee and, in turn, one of the most ingested substances in the world. Due to its rapid absorption and union with proteins, it easily crosses intracellular barriers and is well distributed in organs and tissues [39]. Hence, its marked importance in different sectors such as pharmaceuticals and food is evidenced. The concentration of caffeine depends on variables such as coffee variety, method of preparation of the infusion, and degree or level of roasting [34], becoming one of the main biotechnological products obtained from the coffee bean. However, its concentration in residues and by-products derived from primary coffee processing might be up to two to ten times lower than in beans [12]. Table 2 shows different extraction methods and biocomponents obtained from residual fractions of coffee fruit, as described in the scientific literature. As can be seen, the considerable potential of coffee pulp to promote the circular economy in coffee processing is demonstrated when this is subjected to different biorefinery processes to extract valuable active components. Specifically, a high potential of pulp, husk, and silver skin products for the extraction of biocomponents such as polyphenols, caffeine, and pectin has been identified. However, most of the reviewed studies focused on the extraction of biocomponents at the laboratory scale, and there is an urgent need to move from the technology readiness level (TRL) 1–4 to more realistic environments (5–9). Consequently, further efforts are required to make the processes viable on an industrial scale, especially in terms of yield and efficiency, without affecting the bioavailability of the compounds and that such “technology packages” can be transferred and implemented by the coffee processing companies. Additionally, such extraction systems might be considered as an essential pre-stage for subsequent biorefinery processes, because biocomponents such as caffeine, polyphenols, and organic acids present in coffee wastes and by-products might have inhibitory effects in biotechnological applications.

| Fraction | Extraction Procedure | Biocomponents | Further Results | Reference |
|----------|----------------------|---------------|-----------------|-----------|
| Pulp-husk | Extraction of caffeine with supercritical CO₂ at different husk humidity, milling, pressure, temperature, time, and flow rate. | Caffeine: Maximum extraction yield: 84% (at 373 K, 300 bar, 197 kg CO₂/kg of waste). | Wetting of coffee husks was required, but not milling. | [40] |
| Evaluation of three aqueous extracts of *Coffea arabica* L. pulp: CPE1: washing, pulp removal, drying, and blending. CPE2: washing, selection, removal, drying, and blending. CPE3: washing, selection, removal, freezing, drying, and blending. Subsequent extraction with water at 92 ± 3 °C, for 2 min. | Total polyphenols and caffeine: CPE1 extract contained the highest concentration of total phenols: 17.4 mg GAE/L (CPE2: 10.47 mg/L, CPE3: 7.61 mg/L). IC50 was 18 µg/mL for ABTS and 82 µg/mL for DPPH. Caffeine in CPE1, CPE2, and CPE3: 0.69%, 0.77%, and 0.68%, respectively. | Antibacterial activity against *S. aureus*, *S. epidermidis*, *P. aeruginosa*, and *E. coli* was demonstrated. | [41] |
| Evaluation of the potential of supermolecular solvents (SUPRAS: decanoic and octanoic acid in ethanol (9.5, 19, 24, 33, 38% v/v), water (85.5, 76, 71, 62, 57% v/v), and 5% of amphiphile) to extract biocompounds from coffee pulp. | Phenolic compounds: Extraction yields ranged from 6.40 ± 1.14% (maceration + 100% ethanol) to 9.61 ± 1.73% (sonication + 30% ethanol). Concentration of phenolic compounds: 32.98–44.49 mg GAE/g, and DPPH: 100–142 µmol TE/g. The extract inhibited IL-8 release by approximately 50%. | Cytotoxicity activity showed prevention of IL-8 liberation for the epithelial gastric cell. In addition, abundant derivates of quinic and procyanidins acids were also found in the extract. | [13] |
| | Caffeine: Extraction of 5.6 mg caffeine/g, 0.9 mg/g protocatechuic acid at a solvents:sample ratio of 4:1 v/w. | Various bioactives were detected, showing high antioxidant capacity (45% for DPPH and 91% for ABTS). | [42] |
### Table 2. Cont.

| Fraction | Extraction Procedure | Biocomponents | Further Results | Reference |
|----------|----------------------|----------------|----------------|-----------|
| Pulp-husk | Extraction of pectin and polyphenols from *Coffea canephora* pulp by simple precipitation. Acid solutions (5N nitric, 5N hydrochloric, and 2N sulfuric) and 2% ammonium oxalate were evaluated; for polyphenol extraction ethanol was used (4 °C for 1 h; centrifugation at 8000 × g rpm for 10 min). | Pectin and total polyphenols: Maximum pectin extraction achieved using metal salts and ethanol at 6.0% and 6.7%, respectively. The equivalent weight of extracted pectin (1180.5 mg/g) was higher than that of commercial pectin (724.8 mg/g). The methoxyl content of commercial pectin and crude pectin was 9.3% and 5.6%, respectively. The main polyphenols were gallic acid, vanillin, catechin, ethylcatechin, coumaric, caffeic, and ferulic acids. | Efficient method for the simultaneous extraction of pectin and polyphenol. The polyphenol fraction showed good antioxidant activity with phosphomolybdate radicals, FRAP, DPPH, and ABTS, respectively. | [43] |
| | Extraction with water (121 °C, 20 min), concentration, and drying. Quantification of biocomponents by HPLC and measurement of inhibitory activity on hyaluronidase. | Uronic acid: High inhibitory effect of the extract against hyaluronidase, possibly due to acid polysaccharides such as uronic acid. | An inhibitory potential of the hyaluronidase enzyme was identified. | [44] |
| | Extraction with choline chloride/glycerol (DES) (biomass/solvent ratio: 1:8, 1:16, 1:32), after pre-treatment at 60 °C, 115 °C, and 150 °C. Monitoring carbohydrates, monomeric sugars, inhibitors HMF, furfural, gallic, and coumarin. | Reduced sugars: The highest yield was 0.24 g glucose/g biomass with pre-treatment at 150 °C and biomass: solvent ratio of 1:16. The hydrolysis yield was found to be 72% at 150 °C. | Low formation of fermentation inhibitors. | [45] |
| | Ethanolic extraction at different liquid/solid ratios (10–50), with variable ethanol purity (50–100%), at three temperatures (45 °C, 60 °C, 75 °C). | Total polyphenols and caffeine: Extraction yield ranged from 0.26 to 2.92 gallic acid/kg (the highest values at a liquid/solid ratio of 30, 100% ethanol, and 60 °C). Caffeine yield ranged between 0.77 and 1.44 g/kg with 41 liquid–solid ratio. | Antioxidant activity varied within the range 2.72–12.83 μmol TE/g (ratio 10, 75% ethanol, 75 °C). | [46] |
| | Intermediate pyrolysis and production of bio-oils and biochars (at 280 °C, 400 °C, and 500 °C, with limiting oxygen for 10 min). | Caffeine and phenols: The oily fraction contained caffeine, acetic acid, pyridine, and phenols. The highest total phenolic content and antioxidant capacity were obtained at 280 °C: 6.09 and 3.02 mg GAE/g bio-oil. The resulting biochar at the same temperature showed high calorific value: 22 MJ/kg. | Overall potential of up to 487 and 242 tons of GAE/year. | [47] |
| Husk-pulp-silver skin | Evaluation of DHP, ACT, and ENZ in dietary fiber and phenols. Suspension of 8.0 g waste in 200 mL water (stirred for 30 min), for DHP at 50–100 MPa; for ACT at 10–30% (v/v) of acetic anhydride (30, 60, 120 min); and ENZ with 5 and 15 U of cellulase (30–120 min). | Dietary fiber and phenols: Effect of DHP at the highest pressure (100 MPa) or chemical effect at a minimal concentration of acetic acid anhydride (10%). DHP decreased TRP by 25%, antioxidant capacity by 8% (ORAC), and 34% FRAP. In addition, chlorogenic acid content was reduced by the DHP process by 60%. | Reduction of phenols-caffeine content and antioxidant capacity were inherent to modifying techniques, while DHP only caused a minor impact. | [48] |
| | Evaluation of the potential use of coffee silver skin as an alternative dietary fiber (DF) food ingredient. | Dietary fiber: DF yield of 60% was reached (14% of DF was soluble). | Batch culture fermentation induced preferential growth of probiotic bifidobacteria. | [49] |
| | Extraction with subcritical water (25–270 °C) and evaluation of antioxidant activity with HORAC and DPPH. | Total polyphenols: The highest yield was 29% (at 210 °C). The antioxidant activity increased with temperature: 2629 μmol TE/g HORAC, 379 ± 36 μmol TE/g silver skin extract (DPPH). | Correlation of antioxidant activity with protein and phenols concentration in the extracts. | [50] |

**ABTS:** 2,2-azinobis(3-ethylbenzothiazoline-6-sulphonic acid) diammonium salt. **ACT:** acetylation. **DES:** Deep eutectic solvent. **DHP:** dynamic high-pressure. **DPPH:** 2,2-diphenyl-1-picrylhydrazyl. **ENZ:** enzymatic hydrolysis with cellulase. **FRAP:** ferric reducing antioxidant power. **GAE:** gallic acid equivalent. **HMF:** hydroxy-methyl-furfural. **HPLC:** high-performance liquid chromatography. **HORAC:** hydrophilic oxygen radical absorbance capacity. **IC50:** inhibitory concentration 50. **ORAC:** oxygen radical absorbance capacity. **TE:** trolox: 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid equivalent. **TRP:** total reducing power.
5. Production of Biofuels

5.1. Bioethanol

Ethanol has different applications in cosmetics, beverages, and perfumery, and as a solvent, antiseptic, and biofuel. In recent years, renewable energy technologies (i.e., wind, solar, hydropower, and biomass) are being promoted to address environmental issues such as climate change, air quality degradation, and energy insecurity. Specifically, the worldwide generation of renewable energy for 2020 rose to around 2799 GW, representing a growth of 0.3% in renewable generation capacity with respect to the previous year; biomass fuels accounted for 10–14% of world energy [51]. It is worth noting that the demand for renewable fuels, particularly bioethanol, is projected to increase 3.4-fold by 2035 worldwide [52]. In this context, coffee pulp biomass can become an alternative for ethanol generation for energy, food, pharmaceutical, or cosmetic purposes, due to its composition of carbohydrates and cellulose [53] (Table 2). In fact, the valorization of coffee pulp through alcoholic fermentation has been studied, with several authors reporting laboratory-scale trials at 30 °C. Bonilla-Hermosa et al. [54] mixed pulp with process wastewater (12% w/v) enriched with yeast extract (1 g/L) and 0.3 g/L of Hanseniaspora uvarum. A final ethanol concentration of 0.55 g/L and conversion efficiency of 94.81% was reported. Gouvea et al. [55] inoculated pulp and S. cerevisiae at 30 °C and 100 rpm, reaching an ethanol production of 8.49 ± 0.29 g/100 g (d.b.) (13.6 ± 0.5 g ethanol/L). On the other hand, Harsono et al. [56] performed fermentations using dehydrated coffee pulp with mucilage and S. cerevisiae as fermentative microorganism. The fermentation medium contained 1.0:0.6 of yeast: NPK 15–15–20. The authors reported a yield of ethanol of 0.474 g/g after 48 h of fermentation. Further, Mussatto et al. [57] evaluated ethanol production using hydrolyzed parchment with sulfuric acid (14 g/g, 170 °C, 45 min). The fermentation was performed with S. cerevisiae as inoculum (10%). The bioreactor was incubated at 30 °C and 200 rpm for 48 h. As a result, the authors reported a yield of ethanol ≤1 g/L due to the initial low concentration of sugars contained in the hydrolysate. However, they identified the fermentative process as an alternative for yeast growth. Furthermore, a research study carried out by Gurram et al. [58] characterized the chemical composition of coffee pulp and compared the results with other bio-ethanol feedstocks. The authors performed the simulation for bioethanol production using AspenPlus simulation software and considering the characterization of coffee pulp (10,000 tons coffee pulp/d). Sugars were extracted with water (Soxhlet) and subsequent acid hydrolysis with (72%) sulfuric acid. The sugar profile showed the presence of arabinose, galactose, and glucose, while the sugar and ethanol yields were found to be 2100 and 1050 tons/day, respectively. However, the yields obtained on a real industrial scale were lower due to the heterogeneity of carbohydrates and their reduced potential in fermentation to obtain ethanol. A more recent study conducted by Da Silveira et al. [59] analyzes the composition of raw and fermented coffee pulps using chromatographic methods (HPLC-DAD-MS/MS) to evaluate the use of alcoholic fermentation for coffee pulp detoxification. Using a commercial strain of S. cerevisiae (10^7 CFU/g), the coffee pulp was inoculated at a ratio of 2.5 g/kg pulp without aeration or agitation at 28°C to stabilize hydroxycinnamic acid. Glacial acetic acid (1.0%) and sodium metabisulfite (0.5%) were also added. The fermentative process allowed detoxifying the pulp from caffeine by 50%, while significantly reducing the amount of residues by 64%.

Therefore, although ethanol production is a promising alternative to valorize organic wastes derived from the primary processing of coffee beans, it is still necessary to improve the process yield. Specifically, the efficiency of the hydrolysis of lignocellulose matter should be increased, with further pre-treatments to increase the extraction of bioavailable sugars to be explored. Moreover, the exploration of new microbial strains with a higher level of efficiency in biotransformation processes might also be carried out (i.e., microorganisms producing cellulase enzymes) [60]. In this context, environmentally friendly and sustainable production methods should be incorporated to avoid the parallel generation of additional negative impacts during the biofuel production process. Such improvements would enable
the production of bioethanol as a complement to fossil fuels, contributing to strengthening the coffee production sector by reducing its carbon footprint.

5.2. Biogas

Anaerobic digestion is one of the most promising biotechnologies for valorizing biodegradable organic substrates. Biomethanization allows the production of renewable and rich methane biogas and a digestate with applicability as an organic amendment in agricultural soils [61–65]. The compositional characteristics of coffee residues, specifically the content of sugars and volatile solids, make them interesting sources for biomethanization and power generation. However, the presence of non-biodegradable and inhibitory substances in residual fractions, such as saponins, lignin, or polyphenols, might require, in some cases, thermal, enzymatic, bacterial, and/or fungal pre-treatments combined with co-digestion to increase the methane yield and the efficiency of the biomethanization process [66–68]. Table 4 summarizes the most relevant research studies on the topic, including single and combined digestion of coffee wastes and the implementation of different pre-treatments. Proposals with inoculums that promote the initial hydrolysis and incorporate technologies to carry out other types of pre-treatment are an alternative to improve methane production. Nevertheless, evaluation of the efficiency of the process at an industrial scale is essential, in addition to carrying out further research that incorporates other types of agro-industrial waste that could increase the global yield, while promoting centralized management systems. These processes could then be incorporated into the productive stages of commercial coffee and other industries by supplying renewable energy and mitigating the environmental impact traditionally caused by the primary manufacturing of coffee.

### Table 3. Biomethanization of wastes and by-products derived from primary coffee processing.

| Type of Digestion          | Main Products/Byproducts                                                                 | Methods                                                                                                               | Reference |
|----------------------------|-----------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------|-----------|
| Single digestion           | Methane yield: 240–280 L<sub>STP</sub> CH<sub>4</sub>/kg VS (76–89% of the theoretical value). Reduction of 30–73% in TS and 75–80% in VS. Inverse linear correlation between methane yield and hydrolysis constant rate was observed. | Biomethanization of wastes from instant coffee: barley, rye, malted barley, and chicory (7 g TS waste/g sludge, pH 7, 57 °C, 150 rpm), at lab-scale. | [69]      |
|                            | Methane yields: 159.4 ± 1.8, 244.7 ± 6.4, 31.1 ± 2.0, 294.5 ± 9.6 L/kg VS, respectively. Parchment presented a low potential in methane production due to its composition. | Hohenheim biogas yield test to evaluate methane potential from husk, pulp, mucilage, and parchment. Inoculum: sludge from a digester with 5% VS and 6.16% TS. | [70]      |
|                            | Higher methane production from pre-treated parchment: 13–164%. Pre-treatment increased the biodegradability of inhibitory biocompounds. | Hydrolysis of parchment with Ca(OH)<sub>2</sub> (55 °C, 30 d) and co-digestion with cow dung (0–100%) at lab scale (55°C, 100 rpm, 168 h). | [71]      |
| Pre-treatments and co-digestion | Lower concentration of polyphenols after pre-treatment (23.9–30.5%). The concentration of protein in <i>Streptomyces</i> sp. increased by 5–20% after adding pulp. | Coffee pulp inoculated with <i>Streptomyces</i> sp. strains as pre-treatment to improve the nutritional value and availability of bio-compounds in subsequent bio-methanization. | [72]      |
|                            | Methane yield: 0.023 L<sub>STP</sub>/kg manure·d; 0.025 L<sub>STP</sub>/kg<sub>treated</sub> coffee pulp·d; 0.019 L<sub>STP</sub>/kg<sub>coffee pulp with cow dung</sub>·(60:40)·d. The digestate obtained improved the water holding capacity of soils and stimulated seed germination. | Fungal pretreatment of coffee pulp with <i>Mycotypha</i> sp. and subsequent co-digestion with bovine manure (60:40). Pre-treated pulp and manure were also digested individually at semi pilot scale (60 d). | [64]      |
|                            | Biogas production: 1.1 L<sub>STP</sub> CH<sub>4</sub>/L·d, under continuous mode. Batch digestion of alkaline pre-treated waste allowed 1.14 L<sub>STP</sub>·L<sub>biogas</sub>·d of biogas, containing 65% methane. Process yield: 83%. | Evaluation of acidic and alkaline pre-hydrolysis on the biomethanization of coffee pulp and rejected grain. Lab and semi pilot scale assays (35 °C and 40 d). Inoculum: mixture of bovine and chicken manure and sludge from an anaerobic digester. | [73]      |
Table 4. Biomethanization of wastes and by-products derived from primary coffee processing.

| Type of Digestion                  | Main Products/Byproducts                                                                 | Methods                                                                 | Reference |
|-----------------------------------|------------------------------------------------------------------------------------------|------------------------------------------------------------------------|-----------|
| Pre-treatments and co-digestion   | High methane content in the biogas (45%), equivalent to 6.44 kJ/L. Marked decrease in the concentration of inhibitors after the pre-treatment: 90% in caffeine, 78% in total phenols, and 66% in tannins. | Pre-treated pulp with ethanol/water (50/50, v/v) at room temperature. Subsequent co-digestion with bovine manure and rumen fluid (30 d, 30–40 °C), at lab scale. | [74]      |
|                                   | Methane production: 36 L<sub>STP</sub>/kg (at LSR: 10 L/kg; pH: 11; SAOL: 18.5 g O<sub>2</sub>/kg). Adding powdered activated carbon enhanced methane yield up to 86 L<sub>STP</sub>/kg, while two-stage digestion produced 49 L<sub>STP</sub> CH<sub>4</sub>/kg and 19 L<sub>STP</sub> H<sub>2</sub>/kg. | Pre-treatment of coffee husk with ozone and evaluation of BMP and the BHP, at lab scale. LSR: 10–20 mL/g; pH 3.7, and 11; SAOL: 6.8–81 g O<sub>2</sub>/kg. Inoculum: bovine manure and anaerobic sludge (1:1, w/w). Loads: 0.7 g COD/g VS inoculum for BMP; 1.8 g COD/g VS inoculum for BHP. | [75]      |
|                                   | Daily production of 23 tons of coffee pulp can generate 1886 m<sup>3</sup> STP CH<sub>4</sub>/d. Efficiency of the methanogenic reactor: 95%, with biogas containing 80% methane. Hydrolysis of pulp in the acidogenic reactor: 23%, at a rate of 0.001 g COD/L·d. | Digestion of fresh coffee pulp derived from wet processing in an acidogenic reactor (35 °C; pH 6); and co-digestion of pulp and mucilage in a methanogenic reactor (35 °C; pH 7). Inoculum derived from coffee fermentation tank. | [76]      |
|                                   | Biogas yields: 330 L/kg<sub>CP</sub>; 670 L/kg<sub>BM</sub>; 1170 L/kg<sub>CP</sub>·CM<sub>W</sub>. But biogas contained isocyanic acid, bromomethane, hydrogen sulfide, and hydrogen iodide. | Batch co-digestion of CP+BM (mixed at 60:40 ratio), and CM+CP+W (80:20 CM+W, 80:20 CP+W, 40:40:20 CP+CM+W), at lab-scale (35–45 °C, 8 months) | [77]      |
|                                   | Methane production: 85.1, 11.3, and 60 L<sub>STP</sub>/g COD from pulp/manure, pulp/rumen fluid, and pulp/manure/rumen fluid, respectively. Caffeine, tannins, and free phenols inhibited the process. | Co-digestion of coffee pulp, bovine manure, and bovine rumen fluid, at lab scale. The digester was filled with coffee pulp and water (1:2 ratio) and the inoculum cow dung, rumen fluid, and cow dung/rumen fluid mixture (1:1 ratio). (pH: 6.8–7.2, 30–40 °C, 40 d). | [78]      |

BMP: biochemical methane potential; BHP: biochemical hydrogen potential; BM: Bovine manure; CP: coffee pulp; CM: cow manure; COD: chemical oxygen demand; LSR: liquid:solid ratio; SAOL: specific applied ozone load; TS: total solids; VS: volatile solids; STP: standard temperature and pressure conditions (0 °C, 1 atm). Water: W.

5.3. Hydrogen

The promotion of hydrogen as a source of energy has become one of the international focal points in recent years, where the world’s major economies have focused their attention [79]. Globally, in addition to the Sustainable Development Goals, different international policies are focusing on production strategies specifically targeting green hydrogen production. In this sense, the EU plans to install 2.4 GW of renewable hydrogen electrolyzers, capable of producing 10 million tons hydrogen annually by 2030 [79,80]. Biological production is an alternative to produce green hydrogen. Dark fermentation with microorganisms belonging to the genus Clostridia is quite promising because it can be carried out under ambient conditions with low energy requirements in which waste biomass containing polysaccharides (hemicellulose and most importantly cellulose) can be used to obtain fermentable sugars for hydrogen production [81,82]. In the process, producing biogas rich in hydrogen or methane from organic waste is widely considered a clean and environmentally friendly source of energy [83]. Authors such as Renaudie et al. [84] have recently evaluated hydrogen production by dark fermentation of coffee silver skin. The trials were conducted in a 1 L reactor with the addition of 31 g COD/L and 62 g COD/L. The pH was initially adjusted to 7.0 ± 0.2 and maintained above 5.7 during fermentation by automatically adding 1 M NaOH. The temperature was maintained at 37 °C. The liquid medium was stirred at 220 rpm and swept with nitrogen at a flow rate of 50 mL/min. Hydrogen yields of up to 24.1 mL H<sub>2</sub>/g COD were achieved, with Enterobacteriaceae sp. (27.5%) and Clostridium sp. (5.1%) as the most relevant hydrogen-producing bacteria. Acetate was the primary metabolite produced in the mixing liquor, while butyrate, ethanol, and propionate were produced at lower amounts. Furthermore, Villa et al. [85] evaluated the fermentation conditions to produce H<sub>2</sub> from coffee wastewater, pulp, and husk. They performed the bioprocesses in batch reactors with bioaugmentation of a microbial consortium (bacteria and fungi) under different conditions, using a rotating central composite design and response surface: pH
(4.82–8.18), pulp and husk concentration (6.95–17.05 g/L), and reactor headspace factor (33.18–66.82%). The highest hydrogen yield (3 L H₂/L·d) was obtained at pH 7.0, 7 g/L pulp-peel, and 33% headspace. The main metabolites contained in the liquid fraction were found to be butyric acid (3838 mg/L), isobutyric acid (506 mg/L), methanol (226 mg/L), and butanol (156 mg/L), while taxonomic identification reported Clostridium sp. (87.9%), Lactobacillus sp. (1.7%), Kazachstania sp. (18.6%), and Saccharomyces sp. (16.3%) as the main microorganisms involved in the process.

5.4. **Biodiesel**

Transesterification is another alternative for energy production, where monoalkyl esters of long chain fatty acids obtained from vegetable oils and animal fats are produced, with or without catalysts [86,87]. Biodiesel production might be promoted if the Kyoto Protocol and clean development frameworks are accepted worldwide. In that context, residual feedstocks could be used for economically viable biodiesel production [86]. Emma, Alangar, and Yadav [86] extracted oil from coffee husk for conversion by transesterification. For the extraction, they used a screw-type expeller press machine and mixed the coffee husk with 10% of defective coffee bean oil. First, esterification was carried out by mixing oil with methanol and sulfuric acid. The mixture was heated to a constant temperature of 62 ± 2 °C at 700 rpm for 2 h. Subsequently, the generated diglyceride was heated to 64 °C and mixed with sodium hydroxide and methanol (700 rpm, 2 h). The biodiesel obtained was measured for physicochemical parameters to be compared to regular diesel (700 mL of biodiesel was generated from 1000 mL of coffee husk oil). The results showed that the fundamental properties of the fuel produced are comparable to those of diesel. Specifically, the brake thermal efficiency showed acceptable results compared to normal diesel, while the exhaust emission characteristics of the tested biodiesel showed better results than normal diesel. Although further studies are still required, the performance, combustion, and emission characteristics of a diesel engine fueled with coffee husk biodiesel are being currently investigated, resulting in a promising alternative fuel that can be used in an internal combustion engine without major modifications.

6. **Pyrolysis and Combustion**

Another alternative to valorize wastes and by-products derived from primary coffee processing is their use in direct thermochemical conversion processes. This is how agro-industrial waste can be used to generate thermal energy, electricity, secondary fuels, or porous materials with adsorptive properties. In this sense, Mendoza et al. [7] evaluated the viability of parchment to produce bio-oil by means of pyrolysis. In the production of charcoal, parchment presented higher thermal degradation, so its direct combustion was recommended due to the high content in cellulose and calorific value (approximately 18.30 MJ/kg, on a dry basis). Nevertheless, the high ash content in parchment generates secondary steam cracking and reduces bio-oil yield. Saenger et al. [88] evaluated the complete combustion characteristics of coffee husks and the emission of polluting gases using a conventional combustion furnace and a fluidized bed combustion chamber at a pilot scale. Parchment was reported to have high volatile matter and low carbon and ash content. In terms of emissions, sulfur dioxide emissions were not detected during combustion. Although a significant concentration of NOx was detected in the flue gases (450–525 mg/Nm³), generating energy through direct combustion of residual biomass derived from primary coffee processing might be a promising alternative. In fact, direct combustion of parchment is a frequent practice. Burning residual biomass does not imply a net increase of carbon dioxide in the atmosphere in the medium term, as such substrate derives from photosynthetic plants. However, not much-added value and energy generation are achieved from parchment due to its rapid combustion and low calorific value. To the best of our knowledge, the direct combustion of the other by-products and wastes derived from the coffee process (i.e., pulp or mucilage) has not been sufficiently explored due to their high moisture content (above 80%), which would require prior drying that might lead
to a negative energy balance. Therefore, it would be interesting to evaluate the suitability of applying different drying methods as pretreatment of residual biomass and, in such a manner, determine the characteristics and potential that it might have for subsequent combustion in energy terms.

Santana et al. [89] have recently used defective coffee grains in high-pressure reactors (at 150, 200, and 250 °C, for 40 min) to produce hydrochar. Methylene blue adsorption assays were performed and analyzed by Langmuir and Freundlich adsorption isotherms. They reported that hydrochar could be applied as a technological resource in the agronomic and environmental fields due to the development of oxygenated functional groups on its surface. Hydrochar also presents high magnesium content and adequate structural characteristics in terms of porosity to control soil biodegradation processes. Moreover, Menéndez et al. [30] evaluated the possibility of valorizing parchment and silver skin through pyrolysis. They compared the effect of microwave and traditional heating in pyrolysis and the composition of the gases produced in the two methods. Molecular nitrogen was used in the pyrolysis at a 60 mL/min flow rate. The tests were performed at laboratory scales at different temperatures and using 15 g of pelletized coffee hulls as substrate. The porous texture of the resulting chars was characterized. Compared with other chars, they had a relatively low real (helium) density (dr), which might denote the presence of closed porosity. Nevertheless, the apparent (Hg) density (da) was also quite low, reflecting the relatively high porosity of the chars (ranging from 42.5% to 55.5%). Significant differences in the BET specific surface area were also reported and attributed to the higher gasification of the char in the case of microwave pyrolysis at 1000 °C (21.8 m²/g against 1.4 m²/g for conventional pyrolysis). The authors also reported significant differences between the methods in hydrogen and carbon monoxide products, which are energy-rich gases, compared to the traditional procedure that only produces carbon dioxide. Further studies carried out by Kumar, Weldon, and Lynam [90] evaluated the effect of hydrothermal carbonization at various temperatures (180, 200, 230, and 260 °C) on the bioproducts of coffee hulls, the low-density residues from coffee bean roasting. They found that the treatment of coffee hulls increased the energy density of the solid product at increasing temperatures.

In parallel, other alternatives such as obtaining charcoal or other systems that could increase the calorific power of coffee pulp and peel have been reported in the scientific literature. Remón et al. [91] evaluated the hydrothermal treatment of coffee pulp to produce biofuels and platform molecules. The hydrothermal processes were developed in a lab-scale reactor at 200–320 °C and 120–180 bar for 20–180 min, whereas the solid/water ratio was fixed at 5–15%. The authors reported the generation of four main products: a gaseous stream, an aqueous fraction, a solid biofuel (hydrocarbon), and a liquid biofuel (biocrude), whose yields depended on the processing conditions and varied by 4–10%, 41–57%, 10–42%, and 10–26%, respectively. Biocrude included a set of alkanes, carboxylic acids, ketones, phenols, and nitrogenous species, with varying amounts of C (54–71 wt.%), H (6–7 wt.%), O (18–34 wt.%), and N (3–5 wt.%), and a calorific value ranging from 23 to 32 MJ/kg. Hydrochar contained different proportions of C (57–72 wt.%), H (4–6 wt.%), O (20–35 wt.%), and N (2–3 wt.%), and had a calorific value between 22 and 29 MJ/kg. Process optimization showed that up to 45% of the coffee pulp could be simultaneously converted into energy-rich liquid (20% biocrude) and solid biofuels (24% hydrochar) during the treatment of a 15% coffee pulp suspension at 320 °C and 162 bar for 1 h. It is worth noting that Setter and Oliveira [91] evaluated the properties of briquettes produced from coffee peels by adding kraft lignin for briquette production. The ratios of kraft lignin to coffee peel mass were 10%, 20%, and 50%. Briquetting was carried out at 120 °C and 15 MPa for 15 min. Physico-mechanical characteristics such as bulk density and compressive strength were determined. Afterwards, slow pyrolysis of briquettes was conducted at 350–450 °C for 1 h. Product yields were calculated at the end of the pyrolysis process. Pyrolysis at 350 °C of briquettes with 50% lignin favored biochar production, while temperature did not affect chemical and energy properties. In the case of bio-oil, 350 °C and 10% lignin produced a
higher yield. In general, the thermochemical process improved the energetic properties of the substrate and FTIR analysis showed that the biochar becomes more aromatic and carbonaceous in relation to the raw agro-industrial waste. Moreover, Torres, Urvina, and Lassa [91] proposed a model for the gasification of coffee pulp. Samples were dried and fed to a 20 kW-moving bed gasification system with four zones: biomass drying, pyrolysis, oxidation, and subsequent reduction of partially converted biomass. Different operational parameters of the gasifier were evaluated, including feeding rate, temperature, moisture content, and equivalence ratio with biomass flow: 17.1 to 27.3 kg/h, airflow range of 12.4–15.9 kg/h; equivalence ratio: 5.3 to 7.7, which led to reduced temperatures varying between 556 °C and 605 °C. The obtained biochar was subjected to elemental analysis and the results were then modelled in Aspen HYSIS. The authors developed a model for obtaining biochar with thermodynamic properties, predicting the mole fractions of various syngas components (CH$_4$ and H$_2$ through the CO$_2$ hydrogenation reaction), and the converted carbon.

7. Other Uses

Other uses of wastes and by-products derived from the primary processing of coffee have been recently reported. It is worth noting that such alternatives might be of significant interest in different sectors, especially in productive systems that tend to be sustainable.

7.1. Adsorptive Properties of Biochar–Bioremediation

Konnhe et al. [92] evaluated the use of biochar derived from rice husk, coconut peel, and coffee husk in the adsorption of nitrates and nitrites from slaughterhouse wastewater. The desorption efficiencies of the adsorbed nutrients were also explored to determine the applicability of the enriched biochar as a slow-release fertilizer. The biochar was characterized by elemental analysis and physical–chemical tests, scanning electron microscopy for morphological characterization, and functional groups by Fourier transform infrared spectroscopy. Adsorption evaluation was performed in slaughterhouse wastewater at 26 °C, and the initial pH of the sorption solution was 7.35 ± 0.15; 1.5 g of biochar was added to 50 mL of water (stirred at 120 rpm). Samples were taken at 30, 60, 90, and 120 min and nitrates and nitrites were analyzed in the waste solution by spectrophotometry. Coconut peel biochar was found to be the most porous, having the highest adsorption capacity for N-NO$_3^-$ (12.97 mg/g) and N-NO$_2^-$ (0.24 mg/g). The adsorption capacity of rice peel charcoal was 12.315 mg N-NO$_3^-$ /g and 0.233 mg N-NO$_2^-$ /g, while coffee husk charcoal allowed the removal of 12.08 mg N-NO$_3^-$ /g and 0.218 mg N-NO$_2^-$ /g. In terms of nitrite desorption, efficiencies as high as 22.4%, 24.4%, and 16.8% were found for rice husk, coconut husk, and coffee husk charcoals, respectively, while the figures increased up to 80.73%, 91.39%, and 83.62% for nitrate. Moreover, Quyen et al. [93] evaluated the use of coffee husks as bioadsorbent for heavy metals such as Pb$^{2+}$ and Cd$^{2+}$ from wastewater. The biochar was obtained from 100 g of pyrolyzed biomass at 500 °C, 150 mL N$_2$/min. Before pyrolysis, sodium hydroxide was added for chemical modification of the biochar. The bioadsorbent removed 89.6% of Pb$^{2+}$ and 81.5% of Cd$^{2+}$ ions from the wastewater, which confirms its suitability to remove micropollutants from aqueous streams.

7.2. Compost

The use of residual organic matter for its incorporation in agricultural soils after composting has provided favorable results. Composting allows a considerable decrease in the need for inorganic fertilizers and the reduction of water for irrigation due to the higher water retention capacity achieved by those agricultural soils supplemented with compost. In fact, composting is a cost-effective technology that can be used at the industrial level to recycle industrial organic waste and obtain an amendment with fertilizing properties [94]. Sathianarayanan and Khan [95] evaluated the quality of vermicompost and inhibition of *Rhizoctonia solani* (through radial growth at 12, 24, 48, and 72 h) using coffee husk as compostable substrate. Vermicomposting was performed at laboratory scale by inoculating
Eudrilus eugeniae and Trichoderma viride. They reported high content of nitrogen (248 mg/kg) and phosphate (276 mg/kg) in the vermicompost was obtained, and marked inhibition capacity was seen against R. solani (61%) [95]. On the other hand, Kassa and Workayehu [96] evaluated the physico-chemical properties of compost and the influence of composting time using different mixtures of parchment, pulp, bovine manure, and the legume Millettia ferruginea (MF): (1) coffee husk + pulp (control); (2) coffee husk + pulp + cow dung; (3) coffee husk + pulp + MF; (4) coffee husk + pulp + cow dung + MF; (5) coffee husk + pulp + effective microorganism (EM) obtained from an Agricultural Research Organization. Coffee residue amended with M. ferruginea and/or cow dung showed the best results under the study conditions. Specifically, a final product with marked cation exchange capacity, high pH, and total nitrogen concentration, C/N ratio of 13, and available phosphorus was obtained after 70 d of composting time [96]. Furthermore, Dzung et al. [97] studied effects of coffee pulp compost on soil fertility and uptake of nutrients in leaves, growth, and production yield of coffee plants. Co-composting of coffee pulp (875 kg) with 10% (w/w) cow manure, 2% lime, 0.5% urea, and water (60%), was carried out in 1.2 m piles. The effect of the initial inoculation of Trichoderma sp. and Streptomyces sp., and the addition of Azotobacter sp. and Bacillus sp. after 90 d was also evaluated. Specifically, composting process reduced the C/N ratio of coffee pulp from 40.0 to 13.6, while the application in land cultivated with coffee allowed an increase in the growth rate of the branches and the production yield of up to 14%, compared to the control [97].

Nevertheless, further composting production processes should be evaluated to increase the efficiency of this type of valorization process applied to coffee wastes and, in turn, increase the added value of the final product obtained, thus promoting its real implementation in medium and large cultivated areas.

7.3. Solid State Fermentation and Enzyme Production

The use of enzymes in the industry has wide use due to their capacity to function as catalysts with high specificity and environmental efficiency that can increase the profitability of the production processes. In fact, enzymes have multiple applications in industrial sectors such as those devoted to renewable energy production and bioremediation technology [98]. Specifically, the enzyme market is on the rise, with an annual growth of 6.8% between 2020 and 2027 [99]. Consequently, the use of coffee waste and by-products as substrates for fermentation processes conducted by mushrooms, fungi, yeasts, or bacteria for the production of enzymes is a promising alternative, with solid state fermentation being the most frequent procedure [98].

Shankar et al. [100] performed saccharification of lignocellulosic agricultural residues: corn husk (CHu), peanut husk (PH), and coffee cherry husk (CCH), using the crude lignocellulolytic enzyme consortium produced by Sphingobacterium sp. ksn. in mineral salt medium. A consortium of lignocellulolytic enzymes, i.e., cellulase, xylanase, pectinase, mannanase, and laccase was reported in the study. Different protein concentrations (10–60 mg protein), substrate concentration (2.5%, 5.0%, 10.0%, 12.5%, 15.0% w/v of the crude enzyme), temperature (30, 40, 50, and 60 °C), pH (4–9), and time (0–30 h) were evaluated. Once the treatments were completed, reducing sugars were quantified by HPLC. Maximum saccharification was found at 50 mg protein in the enzyme cocktail with 10% (w/w) substrate concentration after 24 h. The amount of total sugars produced was 63, 49, and 31 g/L for CH, CCH, and PNH respectively [100].

Further solid-state fermentation processes of coffee waste have been reported, using fungi such as Rhizopus spp., Aspergillus spp. [19], Streptomyces spp. [72], Lentinula edodes, and Pleurotus spp. [101–103]. Murthy and Manonmani [102] evaluated the bioconversion efficiency, period of mycelium colonization, first fructification, fruit weight, and variation in caffeine and tannins derived from the production of Pleurotus florida at laboratory scale using dried coffee leaves, husk, parchment, silver skin, and coffee spent ground, with or without wheat bran, at four combinations: 10%, 20%, 25%, 50% (at 25–27 °C). The maximal substrate yield required a period of mycelium colonization, while the first fructification
took 16 d. The total weight of the fungus was found to be 114 g, which means a biological efficiency of 220%, while the concentration of caffeine and tannins decreased by 2.6% and 1.3%, respectively [102]. In the same line, Sabogal-Ótálora et al. [104] evaluated biological pre-treatment of coffee husk (Castilla variety) with the white rot fungus *Pleurotus ostreatus* CECT 20.311 (PL), combined with steam explosion. Polyethylene bags were filled with 150 g of dried coffee husk. Moisture was adjusted to 67% (w/w) with water. The units were sterilized (20 min, 121 °C), and the inoculum was added and incubated at 30 °C. Then, steam explosion was performed at 160, 180, and 200 °C for 10 min, with materials treated and untreated with white-rot fungi. The time required by the biological pre-treatment was found to be 3 weeks, with biodegradation percentages as high as 10.94% of cellulose, 10.73% hemicellulose, and 7.15% lignin. The overall yield of reducing sugar production with the biological pretreatment was 4.19 g/100 g coffee husk (22% more than the original material). Specifically, the biological treatment resulted in 2.1 times higher yields of reducing sugars than those obtained after the same steam explosion treatment conditions without fungal pre-treatment (13 g/100 g coffee husk) [104]. Furthermore, Brand et al. [105] evaluated the detoxification in coffee husk (reduction in caffeine and tannins) by inoculating *Rhizopus* sp. (Rh), *Phanerochaete* sp. (Ph), and *Aspergillus* sp. (As). A reduction in the concentration of caffeine (87%, 70.8%, 92.0%), and tannins (65%, 45%, 65%) was observed for Rh, Ph, and As, respectively. Consequently, such solid-state fermentation might be a promising pre-treatment for animal feeding or subsequent anaerobic digestion [105].

Santos et al. [106] extracted and stabilized chlorogenic acids from coffee pulp during solid-state fermentation using commercial yeast strains. They fermented coffee pulp with *S. cerevisiae* to produce an extract rich in chlorogenic acid with potential in pharmaceutical and functional ingredient markets. The evaluation was carried out at laboratory (0.4 kg), semi-pilot (12 kg), and pilot-scales (90 kg), with 2.5 g yeast/kg, at 28 °C in the dark, with and without prior extraction using ultrasound: 42 kHz, 10 min, 250 W/kg. In all the scales, an extract rich in chlorogenic acid was obtained (300–400% more than the initial concentration) in less than 24 h of fermentation. However, the addition of sodium metabisulphite (at a concentration of 0.5%) was required as the stabilizing agent. In parallel, Moreira et al. [107] fermented coffee husk and pulp with *Rhodotorula mucilaginosa* to extract carotenoids. Pre-treatment with KOH (0.06% w/v) and subsequent sterilization were carried out and two growth media were evaluated: (i) pulp extract (PE) (6.68% w/v) + peptone (10.04 g/L) + yeast extract (3.00 g/L) + glucose (2.00 g/L) + Tween 80 (0.5 w/v); (ii) coffee husk extract (HE) (8.36% w/v) + glucose (6.36 g/L) + peptone (3.68 g/L) + Tween 80. Inoculum: 10^7 cell/mL; 28 °C; 160 rpm; 5 d in the dark. The biomass yields (in g/L) were found to be 7.92 from PE, 8.69 from HE, 8.60 from media control. The results regarding carotenoids (in µg/L) were 16.362 for PE, 21.358 HE, and 591 from control. It is worth noting that the pigments obtained might eventually replace the artificial pigments commonly used in the food and pharmaceutical industries [107].

Among others, the fermentative processes described allow reducing the concentration of some anti-nutrients contained in coffee waste, such as tannins. In addition, the substrates derived from coffee by-products have been reported to be suitable for the cultivation of medicinal mushrooms, obtaining a biological efficiency comparable to the figure achieved using standard substrates [108]. Consequently, such processes can be used as a pre-treatment for subsequent biotechnological alternatives and, in turn, might be a method for obtaining metabolites of marked interest and value in different food and non-food industries [64,105].

### 7.4. Colorants

As mentioned in the section on biocomponents, coffee waste and by-products are a source of pigments such as anthocyanins [11,13,34]. Current demands for colorants are focused on natural or naturally derived products that do not have a potential hazard associated with their consumption [109]. In that context, Prata et al. [110] investigated the potential of coffee peel as a source of anthocyanins. The extraction was evaluated
with 1.5 mol/L of HCl in methanol for 18 h, and the sample was concentrated in a rotary evaporator at 35 °C. The extract was then subjected to sonication and vacuum. A total of 19.2 mg of pigment per 100 g of fresh peel was obtained, with the main anthocyanin being cyanidin 3-rutinoside. Furthermore, Parra-Campos and Ordóñez-Santos [111] optimized and evaluated the pigment extraction process for coffee exocarp in French meringue. The color was measured on fresh samples using a colorimeter with CIEL*a*b coordinates. Coffee exocarp (15–85% w/v) was mixed with acidified ethanol (36–65% v/v) for 18 h; after which the samples were filtered and quantified by spectrophotometry. Meringues were prepared with inclusion of 0, 3, 6, and 9 g/100 g of fresh dough compared to a control pigment. The 60% solvent extraction conditions resulted in the highest concentration of anthocyanins (0.145 mg cyanidin 3-glucoside/g of coffee fresh exocarp). The 3% extract in meringues had the smallest total color difference (ΔE), as compared to the control [111].

In general, due to the characteristics of the pigments, extraction techniques by means of solvents such as water and ethanol can be viable and allow obtaining pigments from coffee wastes and by-products. Consequently, we should continue exploring the use of “green techniques” and later concentration and stabilization of pigments that in addition to their color characteristics are associated with medicinal and functional properties of interest in the food, cosmetic, and pharmaceutical industries [34,112,113].

7.5. Agglomerates and Building Materials

Coffee husk and parchment can also be used to manufacture agglomerates for insulation in construction as sustainable materials that can be incorporated in a sector that presents a remarkable demand. Bekalo and Reinhardt [114] evaluated the mechanical and physical performance of particle boards made of coffee husk and parchment mixed with polymers. Three types of substrate were studied: coffee husk, coffee hulls, and wood for a single-layer mat construction, and three different resin types: PMDI, Kauramin 534, and Kaurit 390. The process consisted of sieving, glue spreading, layup and forming, hot pressing, specimens cutting and conditioning, and testing flexural strength and stiffness, internal joint strength, and hygroscopic properties. The modulus of rupture and flexural properties showed values of 10–17 N/mm² and 1886–2747 N/mm², respectively. The results show that a partial replacement of wood up to 50% could be replaced with the biomass substrate. Consequently, using coffee husks and parchment to produce building boards or coatings is a promising alternative valorization method [114]. In addition, Farias et al. [115] evaluated the production of glass-ceramic materials using coffee husk ash as a K₂O precursor. The formulation contained (wt.%) SiO₂ (54.39), Na₂CO₃ (13.78), Al₂O₃ (11.26), CaCO₃ (9.02), MgCO₃ (8.37), coffee husk ash (1.67), H₃BO₃ (0.90), and Y₂O₃ (0.61). It was mixed in a ball mill for 4 h followed by melting (1500 °C, 1 h), grinding, and pressing (20 MPa for 40s; 850–900 °C for 3 h). Glass-ceramics with coefficients of thermal expansion ranging between 9 and 10·10⁻⁶ °C⁻¹ were obtained. The results indicate the feasibility of obtaining glass-ceramics through a more environmentally friendly method using coffee peel ash instead of commercial potassium carbonate [115]. The use of coffee wastes for such applications might even be projected to generate social impacts and improve the housing conditions of vulnerable sectors, especially coffee producers, and thus improve their living conditions and facilities to carry out better production processes that, in turn, would lead to a higher added value of the final product. However, most of the results available in the literature derive from preliminary assays with scarce real implementation at pilot or full scale.

8. Discussion and Future Perspectives in Primary Coffee Waste Management

According to the scientific literature, multiple sources of biomass have been identified in the transformation of coffee (cherry to parchment coffee) with wide possibilities of valorization. Among them, the generation of energy takes importance due to the current needs for alternative sources of energy in the framework of climate change and the fulfillment of the Sustainable Development Goals [25,116,117]. Furthermore, the largest coffee producers are located in developing countries with high infrastructure and technology
needs. In this regard, Zabaniotou and Kamaterou [118] mention the need to increase their TRL, as this is related to the risk in the implementation of the proposals for exploitation of wastes and by-products derived from coffee manufacturing. TRLs are classified from 1–9, with 1 being basic research and 9 corresponding to a proven system in a commercial environment [118,119].

Within the proposals put forward in this review, several alternatives allow the transition from waste to by-products, improving the economic income of the coffee producers, as well as the social, environmental, and economic conditions of such an important agronomic chain. Figure 5 shows different routes that might be taken for the transformation and valorization of residual biomass derived from primary coffee processing. First, by-products such as husk and pulp might be subjected to the extraction of biocompounds due to their polyphenol and caffeine content; the need in this aspect focuses on optimizing and scaling up extraction and stabilization processes of those biocomponents to allow their use in industry, i.e., to advance in TRL. Second, the exhausted pulp remaining from the extractive process might be used in subsequent biotechnological processes to obtain renewable energy in the form of ethanol or methane. Several authors indicate that the presence of polyphenols and caffeine generates an inhibitory effect in such bioprocesses, which can become attractive alternatives after previous extraction [76,78,120]. Further fermentative processes might be carried out to generate compost or valuable metabolites, and even diverse types of crops could be cultivated. Third, the original biomass and even the exhausted biomass derived from previous stages might be used to produce agglomerates, biochar, or heat through combustion (after drying), or for bioremediation processes in which several authors report remarkable adsorption capacity for different water-polluting metals and compounds that favor eutrophication processes [90,91]. In this context, Shah et al. [117] mention how different challenges must be considered when planning biorefinery approaches:

(i) Financial issues: the transition from waste to by-product or raw material must be identified in combination with the required technologies or logistical systems necessary for supply during the transformation process.

(ii) Technical aspects: to achieve an efficient production system, i.e., according to the nature of the waste, the transformation route should aim at the generation of added value as well as the generation of energy.

(iii) Governmental policies: subsidies and tax incentives favor investment and installation of technologies and production systems. Additionally, they mention the need for training and consolidation of databases that favor access to information by the different parties.

(iv) Social aspects: increasing the knowledge of consumers promotes market acceptance and establishes a pattern of demand and the need for biorefineries.

Policies regarding the different alternatives for the use or implementation of biorefinery systems are framed within several aspects that each country or geographical area must adopt. They should generate policies or regulations that allow actions to favor the generation of initiatives that add value to the waste recovery process: mitigation of climate change and the policies or recommendations that are deployed through the summits, pacts, or international meetings of countries. As a complement, the Sustainable Development Goals identify and postulate the problems as well as a possible roadmap that allows to break the issues down for their fulfillment. However, the lack of a universal definition of food waste impacts their efficient reuse for technological and commercial exploitation. Specifically, Zabaniotou and Kamaterou [118] affirm that the circularity of a microeconomy within the context of coffee production naturally raises issues related to scalability. The collection of coffee waste requires storage space, proximity between participating agents, proximity to additional production facilities where the biomass is to be used, and other logistical issues. Consequently, the inclusion of biorefinery systems within a circular economy might require the effort of different sectors involved in the whole chain: federations, producer associations, government, public companies, non-governmental organizations, and academia. Their integration and active participation are essential in the
consolidation of a participatory mapping in which the environmental and social-economic benefits compared to traditional production are clearly identified.

Figure 5. Possible biorefinery routes for valorizing wastes and by-products derived from primary coffee processing.

9. Conclusions

As a conclusion, the high consumption of coffee in the world drives the production to meet its current demand. Thus, within the tendencies toward clean and sustainable production to minimize climate change and migrate to renewable energies and products, the exploitation of what is currently considered waste becomes a by-product in the context of biorefinery. Such approach might allow the generation of other products with an added value even more important than the products derived from the original production process. In this context, research has shown that the integrated use of coffee waste and by-products derived from primary processing might increase the feasibility of the full process. Large-scale applications are likely to be forthcoming. However, it is still necessary to:

1. Improve the processes of extraction and stabilization of some biocomponents (i.e., polyphenols) that may have a high potential in the pharmaceutical, cosmetics, and food industries;
2. explore the commercial caffeine extraction and purification, as there is a considerable presence of this compound in coffee wastes;
3. perform further pre-treatments with more efficient microorganisms (using metabolic engineering) that allow the reduction of non-extracted compounds that are considered as anti-nutrients in animal or human food (i.e., tannins);
4. improve aerobic fermentation and biomethanization processes, either by pre-treatment, co-digesting, or by obtaining or isolating more efficient microorganisms; and
5. promote an integrated implementation of the steps in the treatment of coffee residues, in which the waste obtained in one step can be used as raw material in the next, according to the definition of a biorefinery approach.

Author Contributions: Conceptualization, J.A.S. and A.F.C.; writing—original draft preparation, J.A.S.-J.; writing—review and editing, J.A.S.-J., J.A.S., M.d.l.Á.M. and A.F.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Asociación Universitaria Iberoamericana de Postgrado, for funding the interchange research stay of Johanna Andrea Serna-Jiménez between Andalusian and Iberoamerican Universities. We also wish to express our gratitude to the funding received through Projects PID2020-117438RB-I00 (MICINN and AEI), UCO-FEDER-1262384-R (AT21_00189), and PYC20_RE-048 (Regional Government of Andalusia and FEDER).

Data Availability Statement: Not applicable.

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

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