

Review

Toward Circular Economy: Potentials of Spent Coffee Grounds in Bioproducts and Chemical Production

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Abstract: With growing concern over environmental sustainability and dwindling fossil resources, it is crucial to prioritise the development of alternative feedstocks to replace fossil resources. Spent coffee grounds (SCGs) are an environmental burden with an estimated six million tons being generated on a wet basis annually, globally. SCGs are rich in cellulose, lignin, protein, lipids, polyphenols and other bioactive compounds which are important raw materials for use in industries including pharmaceuticals and cosmetics. Furthermore, the energy sector has the potential to capitalize on the high calorific value of SCGs for biofuel and biogas production, offering a sustainable alternative to fossil fuels. SCGs are readily available, abundant, and cheap, however, SCGs are currently underutilized, and a significant amount are dumped into landfills. This review explores the potential of SCGs as a source of a value-added compound through various conversion technologies employed in the valorisation of SCGs into biochar, biofuel, and important chemical building blocks. The state-of-the-art, current knowledge, future research to stimulate the creation of sustainable products, and the challenges and economic feasibility of exploring SCGs in a biorefinery context are presented.

Keywords: spent coffee grounds; biorefinery; biochar; cellulose; lignin; polyphenols; biodiesel; bioethanol; circular economy



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1. Introduction

Spent coffee grounds (SCGs) are the residue obtained after brewing coffee using various coffee-brewing methods such as hot water extraction (drip brewing, a French press) or steam extraction (espresso). A considerable volume of SCGs is generated globally each year. Although data on the exact volume of SCGs generated is limited due to the ways the waste is generated and inadequate inventory studies [1]. However, it is estimated that about six million tons of SCGs are generated each year worldwide on a wet basis [2]. For example, in the United States, Starbucks alone generates about 91,000 tonnes of coffee grounds annually [3]. About 50% of the SCGs that are produced globally are generated from small-scale coffee shops, restaurants, cafeterias, or individuals [4]. They are usually discarded and end up in landfills, which poses significant environmental challenges. However, recent developments have seen a rise in technologies and policies aimed at changing this practice and developing SCGs as a viable feedstock for the synthesis of bioproducts, platform chemicals, and value-added energy materials [5]. SCGs are a rich source of lignin, cellulose, hemicellulose, proteins, lipids, and other bioactive substances [6] making them a valuable and cheap feedstock using various technologies for the extraction and refining of these valuable components of SCGs.

The cellulose and hemicellulose fractions, for example, can be hydrolysed to produce sugars, which can be fermented to produce second generation bioethanol. Furthermore, the pharmaceutical, nutraceutical, and cosmetics sectors can also explore the extraction

of bioactive components, such as antioxidants and polyphenols from SCGs for potential commercial application [7]. Some of the other ways to reuse SCGs include as biochar for pollutant removal, as fuel products, and fractionating SCGs into their components in a biorefinery process operation. SCGs can also be repurposed as a natural fertilizer for plants [8], a cleaning agent [9], and their oil can be transformed into biodiesel [10]. By diverting coffee grounds from landfills or incineration, there will be a reduction in the pressure on landfill space and mitigate the associated environmental issues. Therefore, the valorisation of SCGs represents a sustainable and economically viable strategy for reducing waste and harnessing their latent value. SCG upcycle efforts also align with the UN Sustainable Development Goals #6, #7, and #12.

SCG valorisation is also consistent with the concept of a biorefinery, which minimizes waste and reduces the environmental impact by utilizing integrated processes to produce a broad range of goods and energy from a single biomass source [11]. Transforming a cheap waste feedstock such as SCGs into biobased products adds value to this waste and reduces the production costs for biobased chemicals and materials [12]. This review aims to assess the potential of SCGs in biobased material and chemical production with a focus on the lipids, cellulose, lignin, and polyphenol components of SCGs as well as the transformation of these components into value-added products such as bioethanol and biodiesel. Further, the application of SCGs as a feedstock in the production of biopolymer, biochar, and activated carbon is also presented.

Composition and Characteristics of SCG

SCGs typically contain cellulose, hemicellulose, lignin, ash, minerals, fat, phenolic compounds, and protein [13]. Extensive analyses of the proximate composition of SCGs are abundant in the literature. Studies reveal that SCGs are rich in organic matter, with a typical composition comprising approximately 40–50% carbohydrates, 20–25% lipids, 15–20% proteins, and 5–10% moisture content [14]. The chemical composition of SCGs can vary slightly depending on factors such as the type of coffee beans used, the brewing method, and the degree of the roast [15–17]. Research is continuously active in utilizing SCG components as essential building blocks for a variety of value-added products.

Cellulose in SCGs can be found in varying quantities, typically between 10–20%, with Lina et al. reporting a 12.4% cellulose content in SCGs [18]. Other SCG types can be dominant in cellulose over hemicellulose as Marina et al. reported an approximately 23% cellulose and 24% hemicellulose content in their study [19]. Lignin is another important component found in abundance in SCGs. Lignin is a heterogeneous, amorphous polymer made up of phenolic compounds, primarily coniferyl, sinapyl, and p-coumaryl alcohol units, connected by various types of chemical linkages [20]. Lignin content in SCGs [21] is slightly lower when compared to other biomass forms such as wood, which is known for its high lignin content [22]. Nevertheless, SCGs can be a promising feedstock for lignin extraction and transformation into high-valued products and fuels

Hemicellulose is a prominent and diverse polysaccharide component found in large quantities within SCGs, playing a crucial role in their overall chemical composition and physical properties. Hemicellulose monomers in SCGs consist primarily of mannose, galactose, and arabinose [23,24]. SCGs typically contains approximately 37% mannose, 32% galactose and 7% arabinose [18]. When compared to other biomasses such as spent brewer grains that also contain hemicelluloses, SCGs possess a higher content of mannose and no xylose content, while BSG levels contain xylose and arabinose, but no mannose [25]. The presence of arabinoxylans in SCGs adds complexity to the matrix, influencing their texture and overall properties [26].

SCGs have a high lipid content, which can be used as a feedstock for biodiesel production [27]. The diversity of lipids in SCGs makes them a potential resource for various applications, including that of biofuel production and the extraction of valuable bioactive materials. These lipids contain tocopherols that can improve the oxidation stability of the biodiesel. Triglycerides are the predominant lipids and consist of three fatty acid chains

esterified to a glycerol backbone [28]. The nature of the fatty acids, including their length and degree of saturation, contributes to the diversity of triglyceride species present in SCGs. Phospholipids, a crucial component of biological membranes, are also found in SCGs. These molecules contain a phosphate group, which imparts amphiphilic properties, allowing them to interact with both hydrophilic and hydrophobic substances [28]. The phospholipids in SCGs may play a role in encapsulating lipophilic compounds, influencing the aroma and flavour of coffee. Additionally, free fatty acids resulting from triglyceride hydrolysis are present, contributing to the overall lipid content [29]. The specific chemical composition of these lipids can vary depending on the coffee bean type, roasting process, and extraction method.

Coffee beans contain storage proteins like globulins and albumins, which serve as a source of amino acids for the developing coffee plant [30]. These proteins may be present in SCGs to some extent. SCGs may contain insoluble proteins that were not extracted during the brewing process [31]. These unextracted proteins can contribute to the overall protein content of spent grounds. SCGs typically contain between a 13 and 17% protein content and this plays a vital role in their structural composition [32]. Various techniques of extracting this protein from SCGs have been reported, such as alkaline-mediated treatment and isoelectric precipitation, where high levels of polyphenols and antioxidant activity were found in the protein obtained [33]. During the roasting process, the enzymes in the coffee beans play a role in the development of flavour and aroma compounds [34]. Some of these enzymes may remain in the SCGs, although they are likely to be denatured by heat. The Maillard reaction is a chemical reaction that occurs during the roasting of coffee beans, leading to the formation of various compounds, including proteins and amino acids [35]. Some of these products may remain in the spent grounds.

SCGs also contain various phenolic compounds such as chlorogenic acid and caffeine which are also abundant in the coffee grounds. The composition of phenolic compounds in SCGs may vary due to the methods of brewing coffee which influence the overall SCGs' composition. Caffeine and chlorogenic acid are the most extensively studied phenolic components in SCGs due to their antioxidant properties [36]. These phenolic compounds have antioxidant and metal-chelating properties, which can protect against free radical damage and reduce the risk of degenerative diseases [37]. For example, chlorogenic acids (CGAs) are the main phenolic components in green coffee seeds and are associated with the reduced incidence of atherosclerosis, diabetes, and cancer. The CGA in SCGs is highly bioavailable and can be easily absorbed throughout the gastrointestinal tract. Table 1 demonstrates the typical chemical composition of SCGs based on peer-reviewed studies while Table 2 demonstrates their typical chemical composition when compared to other waste biomass types.

Table 1. Typical Composition of some SCGs studies (*w/w%* dry basis).

Components	Cellulose	Hemicellulose	Lignin	Total Extractives	Ash	Protein	Lipids	Total Phenolics	Caffeine	Chlorogenic Acid	Dietary Fibre	References
SCGs	-	-	-	-	6.2	11.5–16.5	15.3–15.9	-	-	-	[38]	
SCGs	-	37.06–40.80	19.84–26.51	51.43–55.78	-	-	-	0.17–4.54	-	-	-	[39]
SCGs	-	-	-	-	-	-	-	-	1.41	1.50	-	[40]
SCGs	12.40 ± 0.79	39.10 ± 1.94	23.90 ± 1.70	-	1–21	17.44 ± 0.10	2.29 ± 0.30	-	-	-	60.46 ± 2.19	[18]

Table 2. SCGs' typical chemical composition range in comparison to the chemical composition range of other agricultural waste biomass types from peer-reviewed literature studies (*w/w%* dry basis, unless otherwise stated) (N/D = Not Determined, mg GAE/g = milligram of gallic acid equivalent per gram of sample).

Biomass Type	Cellulose %	Hemicellulose %	Lignin %	Protein %	Lipids %	Ash %	Total Phenolic Compounds (mg GAE/g)	References
<i>Spent Coffee Grounds (SCGs)</i>	12.40 ± 0.79	37.06–40.80	19.84–26.51	11.5–16.5	15.3–15.9	1.5	0.17–4.54	[18,38–40]
Coffee Husk	39.2	12.6	26.2	8.77	1.06	7.86–9.5	2.12	[41,42]
Coffee Silver skin	N/D	N/D	N/D	16.31–18.9	2.91–3.0	9.47	1.28	[42,43]
Coffee Parchment	N/D	N/D	N/D	1.66	0.18	0.65	0.18	[42]
Brewers Spent Grains (BSGs)	25.4	21.8	11.9	24.0–31.4	10.3–10.6	2.4–3.7	N/D	[44,45]
Whole Corn Stover (Combined Stalks, Leaves, Cobs, and Flower)	37.72	20.62	34.25	N/D	N/D	5.03	N/D	[46]
Sugar Cane Bagasse (SCB)	38.4–47.0	23.2–27.0	19.1–32.4	N/D	N/D	1.0–2.8	N/D	[47–52]
Rice Husk	35.0–35.23	24.39–25.0	12.92–20.0	3–3.75	N/D	17	14.90 ± 0.70	[53,54]
Rice Straw	34	36.06	14.5	N/D	N/D	19.5	N/D	[55]
Peanut Shells	44.8	5.6	36.1	5.4	0.1	N/D	N/D	[56]
Wheat Straw	34.9 ± 1.52	25.17	18.5	N/D	N/D	7.56 ± 0.03	N/D	[57]

2. Creation of Value-Added Products from Spent Coffee Grounds

Spent coffee grounds contain a complex blend of organic compounds, including cellulose, hemicellulose, lignin, lipids, phenolic compounds and proteins. These compounds can be extracted and converted into a wide array of biobased products, including bioethanol, biodiesel, biochar and other bioactive compounds. [58] Table 2 shows the various methods of obtaining some value-added products from spent coffee grounds. The exploration of coffee grounds as a resource for biobased chemicals and materials encourages research and innovation in sustainable materials technology, fostering the development of novel processes and products [59]. The carbohydrate fraction, primarily consisting of cellulose and hemicellulose, offers potential for bioethanol production [60]. The lipid content makes SCGs a source of valuable coffee oil which can be used for biodiesel production while the protein fraction presents opportunities for animal feed protein supplementation [61]. Additionally, SCGs contain varying amounts of minerals, antioxidants, and bioactive compounds, suggesting their potential for diverse applications beyond waste disposal, including as a resource for sustainable and value-added products [62].

2.1. Biodiesel Production from Spent Coffee Grounds

Utilizing SCGs as a feedstock to produce biodiesel has garnered significant attention in recent years, with growing research exploring this innovative approach. SCGs are a valuable source of lipids that can be converted into biodiesel through various modification processes. Several studies have investigated the extraction of lipids from SCGs for biodiesel production. A schematic representation of biodiesel production from SCG oil is presented in Figure 1. The lipids in SCGs can also be extracted using the Soxhlet apparatus with n-hexane as the most efficient solvent reported for fat extraction from SCGs [63]. Other types of solvents that may be used include ethyl acetate, n-heptane and n-Octane. The lipid content in SCGs can vary depending on the bean type, roasting type, and extraction method, with a fat content ranging from 13.4% to 14% being reported [63]. This compares similarly to the lipid content of other oleaginous plants, such as soybeans, which range from 10 to 22% [64]. Studies have also demonstrated that SCGs can yield a substantial amount of oil, making them a promising feedstock for biodiesel production; for example, Tuntiwattapanun and Tongcumpou [65] reported a yield of about 102 mg of biodiesel per 1 g of SCGs, which is promising [65]. However, challenges related to the efficient recovery of solvents and the need for additional purification steps have been highlighted in the extraction and transformation of SCG oil into biodiesel which may increase production costs [63]. Alternative extraction techniques, such as supercritical fluid extraction and the application of enzymes to facilitate oil release from SCGs have also been explored as viable alternative. Supercritical carbon dioxide (SC-CO₂)-assisted extraction has been employed as an environmentally friendly and efficient method for the extraction of lipids from SCGs [66]. For example, a study by Coelho et al. demonstrated that the extraction of oil from SCGs using a supercritical fluid and a co-solvent decreases the time required to obtain the maximum oil yield by half [67]. Enzymatic hydrolysis, on the other hand, utilizes enzymes to break down the complex lipids in SCGs into simpler components, facilitating the subsequent transesterification process [68]. These methods offer potential advantages in terms of reducing solvent use and improving the overall sustainability of biodiesel production from SCGs. After the extraction of oil from SCGs, the oil is converted into biodiesel through a transesterification process. Most studies utilize a two-step transesterification process involving acid esterification followed by alkaline transesterification [69]. Acid esterification helps convert the free fatty acids present in the coffee oil into esters, making it suitable for subsequent alkaline transesterification [70]. These processes typically involve the use of methanol and a catalyst, such as sodium hydroxide (NaOH) or potassium hydroxide (KOH), to facilitate the chemical reactions [71]. Also, biodiesel production from SCGs via transesterification using immobilized lipase resulting in high yields and purity of biodiesel has been reported [72].

Furthermore, researchers are exploring strategies to optimize the biodiesel production process from SCGs. Parameters like the reaction time, temperature, catalyst concentration, and the molar ratio of methanol-to-oil have been investigated to enhance the biodiesel yield and quality, which can hold valuable insights in comparison to parameters used to enhance the biodiesel yield and quality from SCGs [73]. Additionally, efforts have been made to develop cost-effective and sustainable approaches, such as using heterogeneous catalysts and microwave-assisted transesterification [74]. In addition to biodiesel production, some studies have also considered the utilisation of byproducts generated during the biodiesel production process. Yang et al. [75] explored the valorisation of crude glycerol, which is a co-product of transesterification, for the improvement of the overall viability of SCG-based biodiesel production. The crude glycerol can be refined for various applications, contributing to the overall sustainability of the SCG-based biodiesel production process by converting glycerol at rates necessary to prevent a large accumulation of glycerol from biodiesel production [75].

Various challenges associated with the utilisation of SCGs for biodiesel production have been reported. This includes the variation in the oil content within the SCGs which poses a huge obstacle, making consistent and high yields difficult to achieve [76]. Also, the extraction process, whether it is solvent-based, mechanical, or using supercritical fluid, struggles with efficiency and standardization due to this variability challenge [77]. Pretreatment has been explored to investigate its impact on improving oil extraction yield, energy requirement, and processing cost [78]. For example, Akula et al. investigated the effect of pretreatment methods of SCGs on coffee oil yield and they reported a higher yield of 17% coffee oil in pretreated SCGs and a 13% coffee oil yield in untreated SCGs [79].

In addition to the variability in oil yield, the quality and purity of extracted oil are also of concern as they can impact the resulting biodiesel properties. A poor quality and poor purity of oil may necessitate further refining steps to meet stringent biodiesel standards, adding to production costs [80]. Economically, the scalability and cost-effectiveness of large-scale production remain uncertain, given the expense associated with the collection of SCGs, the extraction of oil, and the processing into biodiesel. Environmental implications, energy consumption during extraction and waste management are additional challenges. Some promising methods for improving the overall energy consumption and reducing the associated environmental impact of producing biodiesel from SCGs have been reported [81]. Figure 1 demonstrates the production of biodiesel from transforming spent coffee grounds [82].

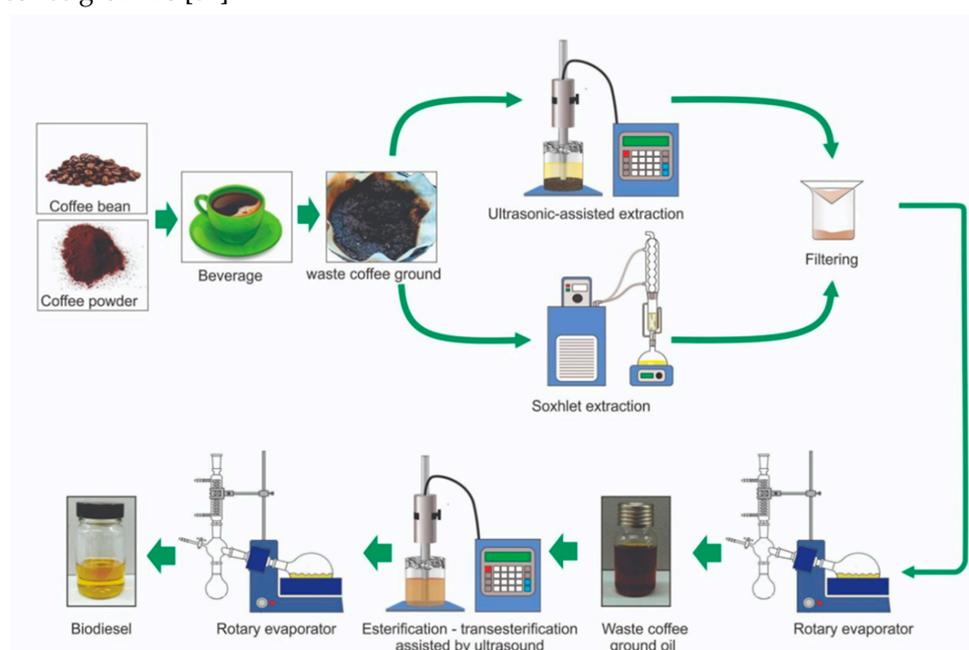


Figure 1. Production of Biodiesel from SCG [82].

2.2. Bioethanol Production from Spent Coffee Grounds

SCGs represent a promising feedstock for the synthesis of bioethanol due to their high carbohydrate content and widespread availability. Several studies have examined the production of bioethanol from SCGs [83]. The conversion of SCGs into bioethanol involves several key steps. Firstly, a pretreatment step is required to remove the lignin, break down the complex nature of the SCGs' biomass, and make the polysaccharides present in the SCGs available for enzymatic action [84]. Various pretreatment methods, including acid [85], alkaline [86], deep eutectic solvents [87], and steam explosion treatments [88], have been explored to improve the accessibility of carbohydrates to enzymes [89]. After pretreatment, the carbohydrates present in SCGs, primarily cellulose and hemicellulose, can be released, and fermented to produce bioethanol [90]. Hydrolysis is an essential process, and various researchers have focused on optimising enzyme cocktails and reaction conditions to enhance sugar yield [91]. Enzyme conversion, microbial fermentation, and physio-chemical conversion techniques are employed to ferment or convert the sugars before distillation. This fermentation product is then distilled or separated to reach the purity levels that are acceptable for product formulation [92]. The schematic diagram for the production of bioethanol from SCGs is shown in Figure 2.

The main sugars from SCG hydrolysis are the hexoses such as glucose, mannose, and galactose, with mannose being reported as the most abundant sugars [93]. Pentose such as arabinose have also been reported to be present in SCGs. The overall economic viability of a lignocellulose-based bioethanol can be improved through an efficient sugar conversion process. This includes the development of robust yeast strains that can ferment these sugars and the optimization of fermentation parameters such as pH, temperature, and sugar concentration [94]. Studies to improve bioethanol's economic potential using pentose-fermenting have been reported [95]. For example, Moremi et al. developed yeasts fermenting both D-xylose and L-arabinose in a medium containing acetic acid. The developed yeast strain produced up to 5.7 g/L of ethanol, with variance between yeast strains, demonstrating the possibility of improving the ethanol yield using improved yeast strains [95]. In addition to the development of various yeast strains, some studies have explored the simultaneous saccharification and fermentation (SSF) processes, where enzymatic hydrolysis and fermentation occur concurrently [96]. This integrated approach can reduce the production time and increase the overall bioethanol yields. The potential of using co-cultures of microorganisms to improve bioethanol production from SCGs has also been investigated [97]. Co-culturing different microorganisms can enhance the fermentation performance and tolerance to inhibitors that may be present in SCGs, such as phenolic compounds. Co-culturing microorganisms has been shown to positively impact bioethanol yields and the efficiency of producing bioethanol in one single system from agricultural wastes [98]. This approach has shown promise in increasing the overall bioethanol yield and reducing the need for detoxification steps, thereby reducing the operational cost [97]. The sequential and co-production of biodiesel and bioethanol with spent coffee grounds were also explored with promising results showing yields of 0.46 g/g for bioethanol and $97.5 \pm 0.5\%$ for biodiesel [99].

Challenges in SCG-based bioethanol production remain. Variability in SCGs' composition due to factors like the coffee bean type and brewing methods can impact the efficiency of the conversion process. Moreover, the presence of inhibitory compounds in SCGs, such as caffeine and tannins, can affect the fermentation performance and require detoxification strategies [100]. Figure 2 showcases the production of bioethanol from spent coffee grounds [101].

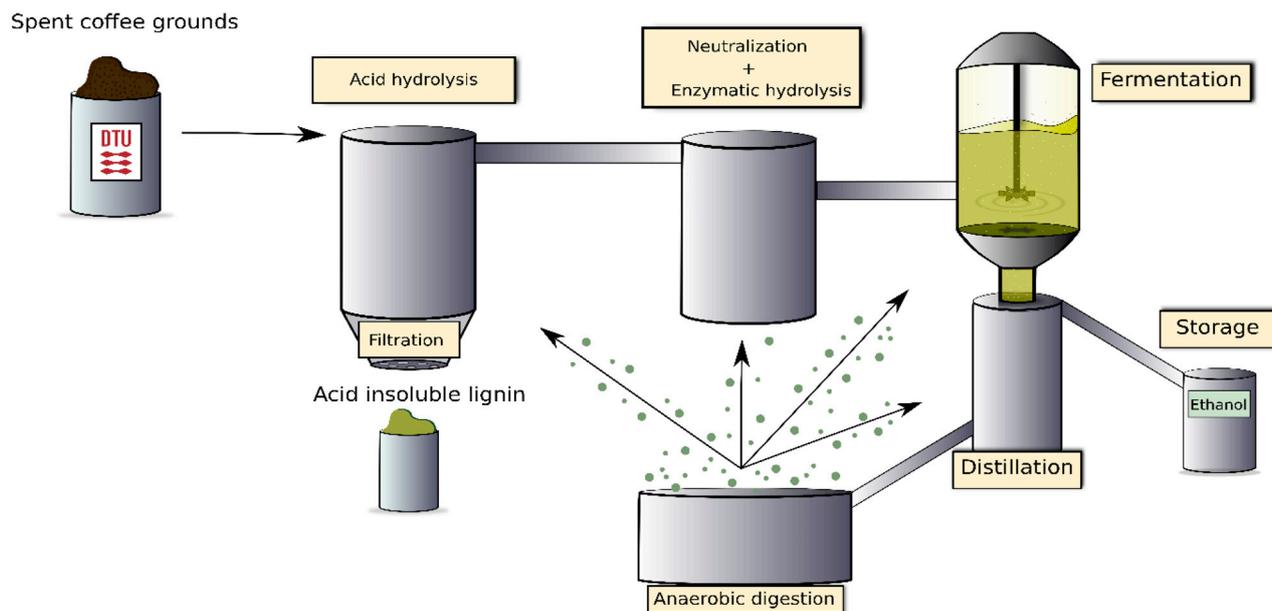


Figure 2. Production of Bioethanol from SCGs [101].

2.3. Production of Biopolymers and Biocomposites from Spent Coffee Grounds

To reduce the dependence on fossil resources and their associated environmental impact, significant research has been carried out on replacing synthetic polymers with biodegradable materials, especially those derived from natural resources. SCGs are rich in cellulose, hemicellulose, lignin, and other bioactive compounds. These components can serve as the building blocks for the development of sustainable materials addressing both the need for waste reduction and the demand for green alternatives [61].

The extraction of biopolymers such as cellulose fibre from SCGs is a critical step in their utilization. Various extraction methods have been explored, each with its advantages and drawbacks. Alkaline treatments, such as sodium hydroxide or potassium hydroxide, and deep eutectic solvents, are commonly employed to remove lignin and hemicellulose, leaving behind purified cellulose. A bleaching process is then used to enhance the purity of the cellulose with chemicals such as sodium chlorite used as a bleaching agent [102]. On the other hand, enzymatic treatments, using cellulase or xylanase enzymes, offer an eco-friendly alternative but may be slower and require further optimization for large-scale applications [103]. Once the cellulose is extracted from SCGs, it can potentially serve as a versatile precursor to produce various biopolymers such as cellulose acetate [104], cellulose nanocrystal and carboxymethyl cellulose (CMC). Each of these biopolymers exhibits unique properties and potential applications.

Cellulose acetate is a thermoplastic polymer derived from cellulose and has garnered attention in applications such as films, membranes, and textiles [105]. Its biodegradability and ability to form flexible films make it a promising alternative to petroleum-based plastics in packaging and other industries [106]. Cellulose nanocrystals (CNCs) are nanoscale particles derived from cellulose and exhibit exceptional mechanical properties [107]. They are known for their high strength, stiffness, and low density, making them ideal candidates for reinforcement in biocomposites. CNC-reinforced materials have been explored in various applications, including automotive parts, construction materials, and biomedical devices [107]. Carboxymethyl cellulose (CMC) is a polymer and a modified cellulose derivative. CMC is a water-soluble cellulose derivative with numerous applications in the food, pharmaceutical, and cosmetic industries [108]. Its water-absorbing properties and biocompatibility make it a valuable ingredient in products like pharmaceutical tablets, food additives, and wound dressings. Biocomposites are materials composed of a polymer matrix reinforced with natural fibres or particles. SCGs have been investigated as a

potential reinforcing agent in biocomposites production. Fang et al. [109] developed Poly (butylene succinate) (PBS)-based biocomposites comprising varying amounts of spent coffee grounds (SCGs). PBS-based biocomposites comprising varying amounts of spent coffee grounds (SCGs) were developed via a reactive extrusion with the polymethyl methacrylate-polyglycidyl methacrylate random (PMMA-r-PGMA) copolymer as a compatibilizer. The PMMA-r-PGMA copolymer was effectively produced through free radical polymerization techniques to change the surface of coffee oil-extracted SCGs (mSCGs). It was reported that mSCGs might operate as a nucleating agent, increasing the degree of crystallinity and the crystallization temperature of composites. The mechanical characteristics of mSCG/PBS composites were superior to those of raw PBS and eSCG/PBS composites. Furthermore, the coefficient of thermal expansion of mSCG/PBS biocomposites decreased, while the heat deflection temperature of the composite with the addition of 20 wt% mSCGs increased by 11 °C [109]. Similarly, spent coffee waste was used to produce sustainable poly (butylene Succinate) biocomposites by Gaidukova et al. [110]. The addition of SCGs was found to significantly increase the elasticity, tensile strength, and storage modulus of biocomposites [111]. This suggests that SCGs can serve as valuable fillers in biocomposites to improve their elasticity.

While the utilisation of SCGs for biopolymers and biocomposites shows great promise, several challenges and limitations must be addressed. Also, achieving a high extraction efficiency of biopolymers from SCGs is essential for cost-effective material production [112]. The further optimization of extraction methods is required to maximise yield while minimising energy consumption. Ensuring compatibility between SCG-derived biopolymers and other polymers used in biocomposites is critical for achieving desirable material properties [113]. Strong interfacial bonding is essential for the success of these materials. Scaling up the production of biopolymers and biocomposites from SCGs to an industrial level presents logistical and economic challenges that must be overcome for widespread adoption [114]. Comprehensive life cycle assessments are necessary to evaluate the environmental impacts and sustainability of SCG-based materials compared to their conventional counterparts.

To address these challenges and further explore the potential of SCGs for biopolymer and biocomposite production, several avenues of research and development should be pursued. Developing extraction methods tailored to specific SCG compositions and intended applications can improve efficiency and material properties [115]. The surface modification of SCG-derived materials can enhance their compatibility with polymers and improve the interfacial adhesion in biocomposites [116]. Exploring the incorporation of SCGs' natural antioxidant and antimicrobial properties into biopolymers and biocomposites can open new applications in packaging and healthcare [117]. Collaborations with coffee producers and retailers can help establish a consistent supply of SCGs and promote the circular economy. Conventional extraction methods, such as solvent extraction, alkaline treatments, and acid hydrolysis, have been widely studied. These methods effectively extract various components of SCGs for application, including the lipids, antioxidants, and cellulose from SCGs that could potentially be utilized in biocomposites. However, they often involve the use of hazardous chemicals and may have limited sustainability, hence the need to use novel methods and processes that are continuously being explored, such as the potential of greener solvents, enzymatic extraction, microwave-assisted extraction, and supercritical fluid extraction.

2.4. Extraction of Phenolic Compounds from Spent Coffee Grounds

Phenolic compounds are a diverse group of secondary metabolites found abundantly in SCGs. The compounds themselves reside in the coffee bean and are left behind after the beans have been exhausted into coffee grounds through extraction processes (hot water and steam). The roasting of coffee beans can affect the levels of polyphenolic compounds found in the original coffee bean, and results have shown both increases and decreases in the polyphenolic content in coffee beans, with decreases primarily seen as the roasting time steadily increased. Antioxidant capacity assays such as FRAP and DPPH were among

the tests performed to conclude this [118]. These compounds, which include phenolic acids, flavonoids, and tannins, are known for their antioxidant properties and have been associated with various health-promoting effects, such as anti-inflammatory, antimicrobial, and anticancer activities [119]. As such, their extraction from coffee waste has gained significant attention. Chlorogenic acids, a subgroup of phenolic compounds, are particularly prevalent in coffee waste and are known for their antioxidant and anti-inflammatory properties [120]. Studies have demonstrated that SCGs contain a substantial amount of chlorogenic acids, making them a valuable source for its extraction. SCGs also contain other bioactive compounds, such as melanoidin (in a favourable concentration range, reporting up to 8.8 mg/L when using SCGs at 8.3 g/L) [121] and caffeine (1.41%) [40,122]. These compounds have potential applications in nutraceuticals, functional foods, and dietary supplements due to their health benefits. Beyond their potential health-related applications, phenolic extracts from SCGs could also be potentially employed in the food and beverage industry as natural preservatives, flavour enhancers, and colourants [123]. Additionally, they find use in cosmetics and skincare products for their skin-protective and anti-ageing properties [124].

The extraction of phenolic compounds from SCGs has been a subject of significant interest. Solvent-based extraction methods have been widely employed, with ethanol and methanol being among the most common solvents [125]. The optimization of extraction parameters, such as the temperature, solvent type, extraction time, and extraction method, has been crucial in maximising the yield of phenolic compounds from SCGs [126].

2.5. SCG Is a Source of Biochar and Activated Carbon

Biochar and activated carbon are two types of carbon-based materials with unique properties. Biochar is a carbon-rich material produced through the pyrolysis of organic materials such as SCGs and has potential benefits in soil improvement, carbon sequestration, and as a sustainable energy source [127]. Biochar has gained attention for its potential as a soil conditioner and carbon sequestration agent. Several studies investigated the effects of SCG-derived biochar on soil properties and plant growth. SCG biochar was reported to show a high removal efficiency and stabilization of trace elements in soil, with further studies being necessary on its biological toxicity [128]. SCG-derived biochar has been found to improve soil quality (soil amendment) by enhancing the soil's water retention, nutrient retention, and cation exchange capacity when compared to peat-growing media [129]. Its porous structure creates a habitat for beneficial microorganisms and promotes soil aeration. These improvements can lead to increased crop yields and decreased fertiliser requirements [130]. One of the potential benefits of using SCG-derived biochar is its ability to sequester carbon in the soil. Biochar's recalcitrant nature allows it to persist in the soil for an extended period, reducing carbon dioxide emissions and mitigating climate change [131]. A study by Islam et al. evaluated the potentials of SCG-derived biochar and its adsorption capacity of silver. The authors reported that the biochar produced at 500 °C offered a maximum surface area of 40.1 m²/g with a yield of 23.48% biochar and the highest silver adsorption capacity of 49.0 mg/g with a 99.9% silver removal efficiency [132]. This removal efficiency demonstrates the great potential of SCG-derived biochar. The chemical composition of SCG-derived biochar may compare differently to other biochar from other biomasses, due to the chemical nature of SCGs. SCGs, in general, typically contain higher levels of nitrogen and potassium when compared to most other biomass types, which may result in differences in the resulting biochar.

Activated carbon is a highly porous form of carbon produced through physical or chemical processes. It is widely used for the adsorption of various substances and purification processes. Chemical activation involves the impregnation of SCGs with chemical agents, typically potassium hydroxide (KOH) or phosphoric acid (H₃PO₄), followed by high-temperature carbonisation [133]. This method has been shown to enhance the porosity and adsorption capacity of the activated carbon produced from SCGs. Physical activation methods, such as steam activation, involve exposing SCGs to high-temperature steam in

the absence of oxygen [134]. This process creates pores within the material, increasing its surface area and adsorption capacity [135]. Researchers have explored different activation temperatures and times to optimize the properties of activated carbon derived from SCGs [136].

Activated carbon is widely used for its high adsorption capacity, making it effective in various applications, including water treatment, air purification, and wastewater remediation [137]. The production of activated carbon from SCGs has been investigated for its potential as a low-cost and sustainable alternative to traditional sources. Studies have shown that SCG-derived activated carbon exhibits a good adsorption capacity for a range of contaminants, including heavy metals, organic pollutants, and dyes [138]. Its performance is influenced by factors such as the activation method, activation agent, and precursor material [139]. The biochar and activated carbon derived from SCGs exhibit a high porosity and surface area, making them suitable for adsorption applications. The specific surface area is a crucial factor in determining their adsorption capacity, and the optimization of production methods can influence these properties [140]. Its adsorption properties can be tailored to target specific contaminants, offering a sustainable solution for treating various types of wastewater.

Researchers have extensively characterized SCG-derived biochar and activated carbon to understand their physical and chemical properties [141]. Key characterization techniques include scanning electron microscopy (SEM), Brunauer–Emmett–Teller (BET) surface area analysis, Fourier-transform infrared spectroscopy (FTIR), and X-ray diffraction (XRD) [142]. An FTIR analysis is employed to reveal the presence of functional groups on the surface of SCG-derived materials, such as hydroxyl, carboxyl, and phenolic groups [143]. These functional groups play a role in adsorption mechanisms and can be modified through activation methods. An XRD analysis is used to investigate the presence of crystalline phases, such as graphite-like structures in SCG-derived biochar and activated carbon [144]. Understanding the crystalline structure is essential for tailoring the materials to specific applications. The biochar and activated carbon produced from SCGs offer numerous applications beyond soil improvement and water treatment. Some potential future directions include SCG-derived carbon materials that could be explored for energy storage applications, such as supercapacitors and batteries, due to their high surface area and conductivity [145]. Research into the catalytic properties of SCG-derived materials for various chemical reactions, including the conversion of biomass into valuable chemicals, is an emerging area of interest [146]. Incorporating SCG-derived carbon materials into composite materials could lead to enhanced properties, such as mechanical strength, thermal stability, and electrical conductivity [147]. Table 3 showcases the extraction of value-added products from SCGs. Figure 3 below shows a refining scheme of the chemical activation of SCGs to activated carbon and bio oil.

Table 3. Extraction and application of value-added products from SCGs.

Coffee Waste Type	Methods/Parameters	Research Findings	Product/Potential Applications	Reference
SCGs	<ul style="list-style-type: none"> Pyrolysis of SCGs at different temperatures (350 and 500 °C) for 2 h each at the heating rate of 10 °C/min 	<ul style="list-style-type: none"> SCGs significantly hamper the compressive strength of SCG-blended concrete. Pyrolyzing the SCGs at 350 °C led to a significant improvement in their material properties. A 29.3% enhancement in the compressive strength of the composite concrete blended with coffee biochar was reported 	Biochar for concrete strength enhancement	[148]
SCGs	<ul style="list-style-type: none"> Alkaline extraction of fibre from SCGs using 50% NaOH at a ratio of 1:10 (<i>w/v</i>) at 60 °C for 8 h with constant stirring (100 rpm). Extracted cellulose fibre was solubilized using 68% ZnCl₂ and crosslinked with salt (CaCl₂) amounts of 0.1, 0.2, 0.3, and 0.4 g for the preparation of biodegradable films. 	<ul style="list-style-type: none"> Films showed significant antioxidant properties and blocked UV IR radiation. The highest tensile strength of 6.8 MPa. The tensile strengths are positively correlated to salt and SCG extract amounts. 	Cellulose fibre is used to produce biodegradable, UV-blocking, and antioxidant films	[149]
SCGs	<ul style="list-style-type: none"> The ultrasound pretreatment was performed with a frequency, nominal power, and amplitude of 35 kHz, 160 W, and 100%, respectively. The ultrasound pretreatment was performed for 1 h at T_{amb} and 80 °C. 	<ul style="list-style-type: none"> A reduction in the lignin content and the increased contact surface due to the powdery nature of the SCGs resulted in the highest methane potential among the LMs investigated. This difference in methane potential can be attributed to the diversity in the coffee species, as well as to the torrefaction and coffee-brewing procedures. 	Methane Production for Organic Chemicals	[150]

Table 3. Cont.

Coffee Waste Type	Methods/Parameters	Research Findings	Product/Potential Applications	Reference
SCGs	<ul style="list-style-type: none"> Plaster, Water, and SCGs were placed in a mould together. 	<ul style="list-style-type: none"> The study found that coffee grounds and gypsum have a good homogeneity and adhesion when mixed. The addition of coffee grounds reduced the density of plaster and improved its thermal qualities, A reduction in flexural strength of the plaster was observed. 	Plastering Composite	[151,152]
Spent Coffee Ground Wastes	<ul style="list-style-type: none"> Pyrolysis of SCGs at different temperatures (30 and 900 °C) at a rate of 10 °C/min with a continuous flow of nitrogen at 50 mL/min 	<ul style="list-style-type: none"> The SCG-biochar showed a high fixed carbon content (82.83%), low volatile matter (12.28%), and low ash content (2.22%). The combustion characteristics of the SCG-biochar were in a form of steady combustion behaviour suitable for energy generation. The cost of SCG-biochar was also found to be competitive at the price of \$7.22 per kg. 	Solid Biofuel	[153]
SCGs	<ul style="list-style-type: none"> Extractions using a powder-to-liquor ratio of 1:50. Temperature was raised from 25 °C to 80 °C at 2 °C/min, and maintained at 80 °C for 60 min. After cooling, extracted solutions were centrifuged at 10,000 rpm for 10 min and were then filtered to separate the solution from the leftover grounds 	<ul style="list-style-type: none"> An ethanol/alkali solvent is effective for extracting natural colorants and bioactive chemicals from SCGs. Silk and wool fabrics dyed with the extracts exhibited an improved antioxidant capacity as well as a natural brownish colour with satisfactory colour fastness, An extraordinarily high level (>90%) of antibacterial activities (against <i>E. coli</i>, <i>S. Aureus</i>, and <i>C. albicans</i>) was achieved by all fabric samples investigated. 	Textiles Dyes—Natural bio-colorant, improved UV protection.	[154]

Table 3. Cont.

Coffee Waste Type	Methods/Parameters	Research Findings	Product/Potential Applications	Reference
Spent Coffee Grounds	<ul style="list-style-type: none"> SCGs mixed with a 5% NaOH solution (1:10 <i>w/v</i>) at 70 °C for 2 h, oven drying at 80 °C for 24 h. Followed by bleaching using a mixture of an acetate buffer and a sodium chlorite solution (1:1 <i>v/v</i>), at 80 °C for 2 h. Treatment was repeated three times to ensure the complete removal of lignin. 	<ul style="list-style-type: none"> The percent yield of the purification was 29.5%. The purified SCGs (PSCGs) have a greater surface roughness leading to a higher specific surface area. PSCGs contain approximately 30% cellulose and, 70% hemicellulose with a trace of nitrogen-containing compounds. At any given filler loading, TESPT-treated PSCGs exhibits greater reinforcement than PSCGs as evidenced by the improved modulus, hardness, and tensile strength. 	Reinforcement in a Natural Rubber Composite	[155]
Spent Coffee Grounds.	<ul style="list-style-type: none"> SCGs were defatted using hexane and treated using sodium chlorite at 1% (<i>w/w</i>) and acetic acid at 1% (<i>v/w</i> biomass), at a solid/liquid ratio of 1:10 (<i>w/v</i>) at 80 °C, for 3 h for the SCG-derived polysaccharides. A two-stage enzymatic hydrolysis (short- and long-term) was performed to produce short-chain Manno-oligosaccharides (MOSs) and monosaccharides (MSs), 	<ul style="list-style-type: none"> Amounts of 77% delignified SCGs and 61% SCG-derived polysaccharides, amounts of 15.9 g of first bio sugars (mostly MOSs), 25.6 g of second bio sugars (mostly MSs), and 3.1 g of bioethanol, were recovered from 100 g dry weight (DW) amounts of SCGs. 	Oligosaccharides (OSs), manno-oligosaccharides (MOSs), mannose, and bioethanol	[156]

The following schematic demonstrates the potential of utilising spent coffee grounds in a refining scheme of chemical activation to activated carbon, and retrieval of bio-oil [157].

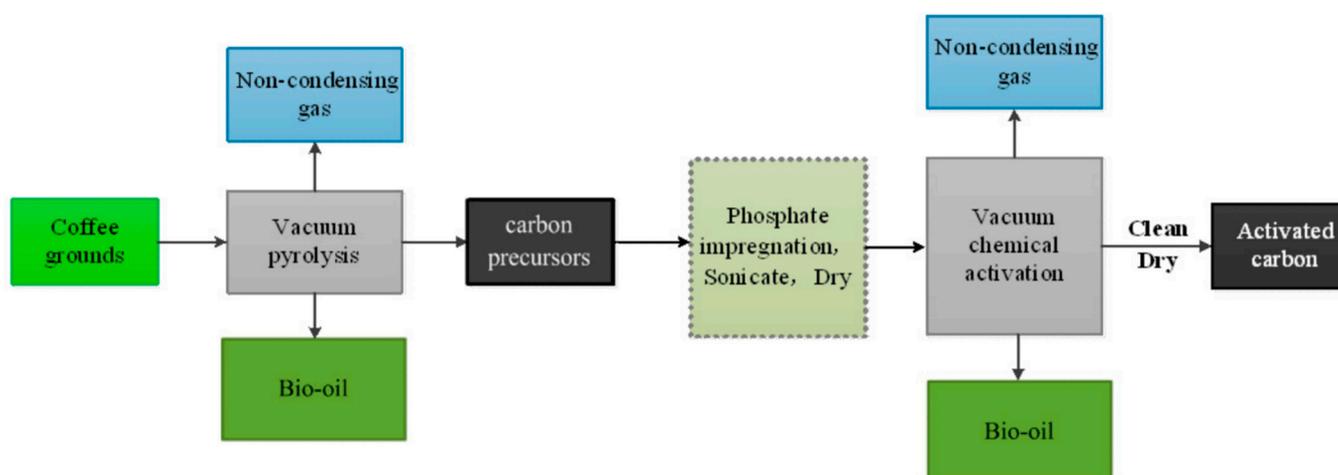


Figure 3. Refining scheme of the chemical activation of SCGs to activated carbon [157].

2.6. Other Applications of Spent Coffee Grounds

2.6.1. Soil Amendment and Agriculture

Studies have consistently highlighted the potential of SCGs as an effective soil amendment [158]. SCGs are rich in essential nutrients and organic matter, making them valuable inputs for enhancing soil quality [159]. When incorporated into agricultural practices, SCGs can improve the soil structure, water retention, and nutrient availability [160]. This not only enhances crop yields but also reduces the need for synthetic fertilisers, thereby diminishing the environmental footprint of conventional agriculture [161]. The circular economy potential here lies in the repurposing of a waste product to support sustainable food production.

2.6.2. Carbon Sequestration

The organic nature of SCGs contributes to their carbon-rich composition, making them an important resource for carbon sequestration [162]. Incorporating SCGs into soils can aid in the capture and storage of carbon dioxide (CO₂) from the atmosphere [163]. This carbon sequestration potential aligns with the sustainability goal of mitigating climate change by reducing CO₂ emissions. Utilizing SCGs in this manner exemplifies the circular economy principle of reusing waste materials for environmental benefits.

2.6.3. Energy Production

In addition to soil amendments, SCGs can be converted into biofuels through various processes such as pyrolysis, anaerobic digestion, or direct combustion [164]. This bioenergy generation not only reduces the dependence on fossil fuels but also provides a renewable and sustainable energy source. The circular economy potential is evident in the repurposing of SCGs as a source of energy, turning a waste product into a valuable resource. However, it is important to consider the efficiency of these biofuel processes and their overall environmental impact.

2.6.4. Biodiversity and Ecosystem Impacts

While the focus of SCG utilisation is on the direct benefits it offers, it is essential to consider the broader environmental impacts of coffee production itself. Studies on the environmental feasibilities of individual processes of utilization of SCG for its compounds are necessary, to fully assess the biodiversity and ecosystem impacts of the processes. One study, for example, assesses the environmental feasibility of the biofuel produced from

SCGs via transesterification with pretreatment and pyrolysis via process simulation with 500–4000 kg of SCGs, which found promising results using recycling systems which improved carbon dioxide emissions in one of the two recycling systems [165]. This highlights the importance of environmentally assessing the impacts of the processes used to utilize SCGs, to fully understand their impacts.

3. Role of the Valorisation of Spent Coffee Grounds in the Circular Economy

The utilisation of SCGs aligns with key circular economy principles, including reducing waste, reusing materials, and recycling resources. By repurposing SCGs, we extend their lifecycle and reduce the demand for virgin materials, thereby promoting circularity in resource use. This approach is essential for addressing environmental challenges and achieving long-term sustainability.

3.1. Economic and Social Implications

The sustainability and circular economy potential of SCG utilisation are closely linked to economic and social factors. The economic viability of SCG utilisation can influence its widespread adoption [166]. Additionally, the social aspects of SCG collection and processing, including job creation and community engagement, can have positive implications for local economies and sustainable practices. The utilisation of SCGs as a biobased resource holds substantial promise in contributing to sustainability and embracing circular economy principles [167]. By diverting waste from landfills, enhancing soil quality, sequestering carbon, generating renewable energy, and aligning with circular economy principles, SCG utilisation presents an opportunity to address pressing environmental challenges. Collaborative efforts among stakeholders in the coffee industry, policymakers, and researchers are essential to unlock the full potential of SCGs as a valuable biobased resource, fostering a more sustainable and circular economy. Figure 4 portrays the potential circular bioeconomy of spent coffee grounds [168].

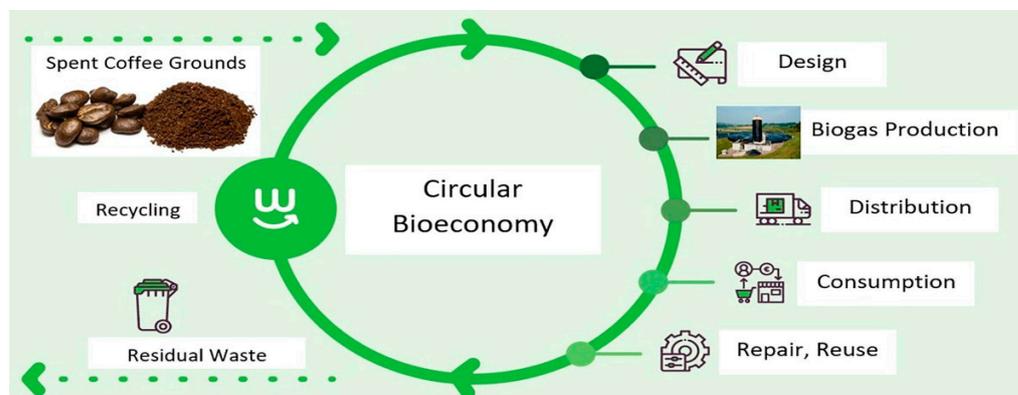


Figure 4. The potential of SCGs in the circular economy [168].

3.2. Challenges and Future Perspectives

The utilisation of SCGs as a resource for biobased chemicals and materials has gained significant attention in recent years due to their sustainability and abundance. However, the utilisation of SCGs as a resource for biobased chemicals and materials presents certain challenges that need to be addressed for successful industrial applications. One of the primary challenges is the heterogeneity of SCGs from their various sources and the logistics of collections from their various sources especially the coffee shops and household consumers. Regarding the variability in SCG composition, factors such as the coffee type, brewing method, and processing conditions are the main influencing parameters. This variability can make it difficult to standardize extraction processes and optimize the production of biobased chemicals and materials. Researchers must develop efficient methods to sort and preprocess SCGs to ensure their consistent quality and composition. Table 4 gives

insight into recent initiatives that are currently converting large quantities of SCGs into valuable products.

Since a huge amount of SCGs is obtained from coffee shops and household consumers, the logistical challenges faced in concentrating large volumes of spent coffee grounds to processing levels by bioprocessing plants requires innovative approaches and technological advancements to overcome these challenges. Various initiatives have been explored to improve the collection and processing of SCGs at a commercial scale. For example, there are initiatives such as ANDRITZ, a company in Austria, Finland, and Germany who are working closely with businesses to transform their coffee waste into biofuels [169]. This would suggest that with proper infrastructure and processes, the concentration of coffee grounds on a large scale is feasible. Similarly, the Kafsimo project in Greece is a community-led approach that involves collecting spent coffee ground waste from local coffee shops, drying it under natural processes, and then converting it into solid fuels such as briquettes and pellets. This project demonstrates the potential for similar projects in other regions to successfully collect and process coffee waste on a larger scale [170]. Matrapazi and Zabaniotou [171] conducted an experimental study into the feasibility of SCGs via their pyrolysis into biochar and energy with a capacity of 2566 tons of SCGs per year.

The authors demonstrated that SCGs, via their pyrolysis into biochar and energy, are economically profitable at a net profit of 47 euros per ton of treated SCGs [171]. The study demonstrates the feasibility of concentrating SCGs to a scale necessary for bioprocessing. Statistics on the mass of coffee grounds collected in Ireland and the EU are limited. However, based on the advancements and recent innovations in other regions, it would demonstrate that for countries such as Ireland. Further research on the mass of SCGs generated by various coffee processing institutions such as Insomnia as well as the concentration of spent coffee grounds is needed to fully harness the potential of the material.

Despite these challenges, the prospects for implementing SCGs for biobased chemicals and materials are promising. As sustainability becomes a driving force in various industries, the demand for alternative, renewable resources is expected to rise [172]. SCGs offer an eco-friendly solution that can contribute to reducing waste and greenhouse gas emissions. Potential research directions in this area include the development of innovative extraction techniques to efficiently recover valuable compounds from SCGs, such as antioxidants, polyphenols, and cellulose [173]. Additionally, exploring novel applications for SCGs in bioplastics, biofuels, and speciality chemicals could open up new avenues for sustainable product development [174]. Collaboration between academia, industry, and government entities is crucial to overcome certain obstacles and unlock the full potential of SCGs as a valuable resource for biobased chemicals and materials. Also obtaining data on the amount of SCGs generated in EU countries will be beneficial. By addressing these challenges and pursuing innovative research, SCGs can play a significant role in the transition towards a more sustainable and circular economy.

Table 4. SCG collection statistics that could hold potential feasibility in systems at bio-processing plant levels.

Institute/Organisation/Company/Body	Findings	City/Country	References
Imbibe—Coffee Roasters	<ul style="list-style-type: none"> • 6 kg Beans Delivery and SCGs 'Zero Waste' Return Scheme. • Currently this is utilised for composting purposes only. 	Ireland	[175]
Coffee 4 Planet Ark—The Council of the City of Sydney	<ul style="list-style-type: none"> • 921 café and coffee shops produce 3000 tonnes of coffee grounds annually (93% end up in landfill). • Shops' sending of coffee grounds via a postage service—Sendle (distribute where necessary in country) • \$8.75 to collect and receive 25 kg of SCG. • Additional cost of 19c-50 for sustainable transport liners. 	Sydney, Australia	[176]
ANDRITZ	<ul style="list-style-type: none"> • Helping large processing plants and coffee companies dispose of tonnes of spent coffee grounds each day. • Turning this waste into biofuel (sustainably) • Coffee companies using biofuel from coffee grounds to fuel their steam heaters, saving on energy. • Mechanical dewatering of wet grounds (Paddle dryer, fluid bed dryer, pelletizing, or steam generation) • Providing coffee producers with a complete automated production line of proven technology to transform their exhausted coffee waste. • Circular Coffee Production 	Operating in: Austria, Brazil, Canada, China, Finland, Germany, United States. - 280 Cities Worldwide	[169]
Bio-Bean Ltd., Nafigate, and WaysTUP!	<ul style="list-style-type: none"> • First coffee recycling company • Transforming SCGs into biofuels and biomass pellets (chemical and plastics industry). • Uses in plastics, brake pads, and more. • Research in areas such as utilising SCGs into natural flavour extracts. • Collection system from hundreds of coffee shops, bars, and factories across England. • Initial factory setup in 2015 could process up to 50,000 tonnes annually. • Navigate produces biopolymers from SCGs that are sourced through Biobean. • Low industrial-scale cost of polymers. 	- Alconbury and Cambridgeshire, England. - Prague, Czech Republic	[177–179]
Agricultores De La Vega Valencia and WaysTUP! Project	<ul style="list-style-type: none"> • Utilising spent coffee grounds to extract flavours, polyphenols, oils, and carotenoids 	Valencia, Spain.	[180,181]

Table 4. Cont.

Institute/Organisation/Company/Body	Findings	City/Country	References
Coffeefrom	<ul style="list-style-type: none">• Create products by the injection and moulded 3D Printing of exhausted coffee grounds that are sold through numerous avenues.• Coffee espresso cup and saucer• Coffee mugs.• 100% Italy-based supply chain.• Collaborates with local enterprises.	Italy	[182]

4. Conclusions

In conclusion, the utilization of SCGs has the potential to contribute to the production of biobased chemicals and materials. SCGs can be harnessed to create a wide range of sustainable products, including bioplastics, biofuels, and adsorbents. This could reduce the environmental burden associated with coffee waste disposal and also contribute to the development of more sustainable alternatives to conventional petroleum-based materials. Effectively repurposing SCGs may reduce the environmental footprint of both the coffee industry and the materials sector. Additionally, this innovation aligns with the principles of the circular economy, promoting resource efficiency and waste minimisation. The utilisation of SCGs for biobased materials represents a step towards a more sustainable and environmentally conscious future. It exemplifies the transformative potential of waste-to-value approaches, offering a solution to the challenges of resource depletion and environmental degradation. Continued research and development in this field will undoubtedly play a crucial role in advancing the circular bioeconomy and mitigating the environmental impacts of traditional materials production.

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