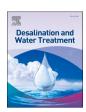
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Strategies toward Green Deal implementation in the context of SCG reuse and recovery in the circular economy model

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ABSTRACT

The article explores the future outlook on integrating and using a resource database for the recovery and reuse of coffee industry residues, focusing on economic and environmental perspectives within the Circular Economy Model (CEM). With the continuous rise in global coffee consumption, the production of Spent Coffee Grounds (SCG) has also surged worldwide, presenting significant opportunities for resource recovery and recycling. The reuse of SCG, a solid byproduct of coffee brewing, whether at home or on an industrial scale for soluble coffee production, is the subject of the article. This review aimed to consolidate existing knowledge on SCG output, management, characterization, treatment, and various methods for resource recovery and recycling to enhance understanding of SCG. Recent approaches include biodiesel production, biochar conversion, composting, codigestion, extraction, as well as utilization in water treatment or construction. Despite these advancements, the SCG research community remains relatively small and disconnected, lacking timely exchange of information. Therefore, the development of high value-added products within the framework of Green Deal Implementation and CEM is strongly encouraged.

1. Introduction

Coffee, a beverage that is widely consumed across the globe, is produced in numerous countries spanning Africa, Asia, Central America, and South America [1,2] The International Coffee Organization (ICO) has reported that the global coffee consumption for the 2022/23 season has reached approximately 170 million bags, each weighing 60 kg [3]. Coffee grounds are the common beverage consumed and lead to high amounts of spent coffee grounds (SCG) [4,5]. The instant coffee sector makes up around half of the overall coffee consumption [6,7]. On average, 1 ton of green coffee can produce roughly 650 kg of SCG, and the production of 1 kg of soluble coffee results in about 2 kg of wet SCG [8,9]. Consequently, it is estimated that approximately 6 million tons of SCG are generated globally on an annual basis [10,11].

Implementing the circular economy principles could solve the issue of spent coffee grounds. Circular economy model considers the issues of

mitigating the impact of production and consumption on the environment and emerged as a reaction to the existing unsustainable linear "take, make, consume, and waste" economic model [12,13]. The most commonly cited definition of circular economy is the one in the European Commission's Communication "Closing the loop - an EU action plan for a closed loop economy (COM(2015) 614) " [14], which states that "a closed loop economy is one where the value of products, materials and resources in the economy is maintained for as long as possible and the generation of waste is kept to a minimum." Circular economy can be effectively utilized in the management of waste feedstocks [15, 16], such as coffee grounds[17], to produce a variety of value-added materials and generate economic advantages.

At present, the majority of spent coffee grounds is either burned as waste or dumped in landfills, although a small portion is utilized for composting [18–21]. Improper handling of SCG can lead to significant pollution due to the high oxygen consumption during the breakdown of

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organic matter and the potential release of residual caffeine, polyphenols, and tannin [22,23]. As a result, managing SCG has emerged as a growingly complex issue. Furthermore, the focus on global environmental consciousness and stringent laws has prompted research into sustainable practices. As SCG is rich in carbon and lipids, more and more people are exploring how to produce clean biofuels or other high value-added products from SCG. It has also promoted the emergence of research papers and application of environmental technologies for more efficient approaches. Therefore, it has great significance to integrate relevant literature and find the most promising research direction through economic analysis.

SCG has been previously explored for value added products, such as biodiesel, extracted valuable chemicals, thermally or pyrolysis treatment strategies and biodiesel production. Few review paper discussed the resourceful utilization of SCG from different aspects. In 2012. Murthy and Naidu emphasized the exploration of value addition from SCG through the implementation of a valorization strategy. They focused on integrating various techniques and applying bioengineering principles to recover fine chemicals such as natural pigments, proteins, lipids, starch, cellulose, enzymes, vitamins, and antioxidants [24]. In 2015, Campos-Vega et al. aimed to establish a biorefinery platform that would enhance the value of SCGs. They summarized the characteristics of SCGs and the components they contain, including fatty acids, amino acids, polyphenols, minerals, and polysaccharides, which justify their valorization [25]. In 2022, Bomfim et al. indicate that spent coffee grounds can be used as biopolymer precursors and fillers for polymer composite production, benefiting various industries from food packaging to automotive. [26] In addition, they explored pre-processing techniques to recover antioxidants and sugars, while bioprocessing methods were applied to the production of enzymes, bioethanol, polyhydroxyalkanoates and biogas.[27,28] SCGs can be considered as valuable sources for the recovery of high-value compounds within the context of a circular economy [29,30].

The above reviews mainly illustrated the useful components of SCG, and resource recovery through direct use, pre-treatment and biochemical processing based on various ingredients such as the bio-refinery of SCG. Recently, SCGs were investigated as environmental functional materials for heavy metal and organic pollutants adsorption, and application as filter Medias, which were considered to be indispensable

in high value-added products. However, this part has not been reported yet. Therefore, in this review, emphasis has been given in the application and pretreatment techniques of SCGs for pollutants control as environmental functional materials. Meanwhile, the various treatment methods of SCG according to the value-added and application prospects were also discussed

Moreover there is an urgent need for practical and new ideas and innovative technological solutions to use this low cost SCG resources and exploit its full potential increasing the overall sustainability of the coffee agro-industry and zero-waste in the contest of Green Deal as well as Circular Economy Model in EU.

2. SCG generation and amounts

Global coffee production reached 168.2 million bags during the 2022/2023 season [3]. The leading coffee producers worldwide were Brazil, Vietnam, Colombia, and Indonesia, collectively accounting for over half of the global coffee supply. The consumption of coffee has been steadily rising, with significant increases in global consumption in the fiscal years 1990/1991, 2000/2001, and 2010/2011, showing growth rates of 83.35 %, 50 %, and 20 % respectively [21]. Data for the 2022/23 coffee year published by ICO indicate that the world's coffee consumption was over 10 million tonnes [3]. As coffee production increases, the amount of SCG generated increases, as illustrated by the data shown in Fig. 1. Hence, the effective utilization of SCG has garnered significant interest due to the brewing of millions of coffee posts and the disposal of millions of pounds of SCG on a daily basis worldwide.

3. Characterization and functional properties of SCG

3.1. The composition of SCG and potential valorization

Table 1 summarizes the composition of SCG obtained from different studies in the literature. The carbon content of SCG was about 61.13~% and the nitrogen content was about 2.91~%, thus C/N ratio was 21~[32]. Carbohydrates correspond to the primary chemical constituent of the carbon content. As far as the composition of sugars is concerned, SCG consists of 37.03~% mannose, 31.90~% galactose, 24.08~% glucose, and 6.99~% arabinose. Additionally, it contains 23.90~% lignin [33]. SCG

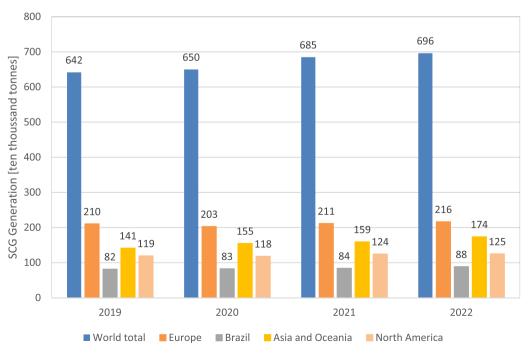


Fig. 1. Generation of SCG (Data from ICO, FAO and calculated according to the research from Pfluger.1975 and Ramalakshmi et al., 2009) [8,31].



Table 1Characterization of spent coffee grounds.

Chemical components	Value	Reference
Total carbon (%, dwt)	61.13, 47.8– 69.5	[32,35]
Total nitrogen (%, dwt)	2.91, 1.9-2.3	[4,32,35]
Carbon/nitrogen (C/N ratio) (wt)	21.00: 1, 19.80: 1	[32,36]
Cellulose (Glucose) (g/100 g)	$12.40 \pm 0.79, 8.6 15.3$	[4,24,33]
Hemicellulose (g/100 g)	$39.10 \pm 1.94, 36.7 \pm 5.0$	[1,33]
Arabinose (g/100 g)	$3.60 \pm 0.52, 1.7$	[1,33]
Mannose (g/100 g)	$19.07 \pm 0.85, 21.2$	[1,33]
Galactose (g/100 g)	$16.43 \pm 1.66, 13.8$	[1,33]
Lignin (g/100 g)	23.90 ± 1.70	[33]
Insoluble (g/100 g)	17.59 ± 1.56	[33]
Soluble (g/100 g)	$6.31 \pm 0.37, 1.6 1.7$	[4,33]
Klason lignin (%, dwt)	30.9-31.9	[4]
Total dietary fiber (g/100 g)	60.46 ± 2.19	[33]
Insoluble (g/100 g)	50.78 ± 1.58	[33]
Soluble (g/100 g)	9.68 ± 2.70	[33]
Lipids (wt%)	10–15	[36-38]
Protein (w/w)	13.60	[25]
Non-protein nitrogeneous compounds(%)	3.70	[25]
Total sugars (mg/kg)	8.50 ± 1.20	[24]
Total polyphenols (%, dwt)	1.50 ± 1.00	[24]
Caffeine (%, dwt)	0.02-0.4526	[24]
Minerals (mg/kg)	-	
Potassium (mg/kg)	$11{,}700 \pm 0.01, 3549.00$	[1,33]
Calcium (mg/kg)	$1200 \pm 0.00^{\circ}$ 777.40	[1,33]
Magnesium (mg/kg)	$1900 \pm 0.00, 1293.30$	[1,33]
Ashes (%, dwt)	0.43-2.20	[1,4]

contains a total fiber content of 43 %, with 35 % being soluble and 8 % insoluble [24]. Moreover, the coffee fibers derived from SCG possess antioxidant properties, with a trolox content of 2.4 mmol per 100 g of dry weight [34]. This is comparable to well-known food antioxidants like red wine products (43 %) and peaches (36 %). Consequently, the dietary fiber obtained from SCG can be classified as an antioxidant dietary supplement with potential benefits.

Proteins and non-protein nitrogenous compounds, such as free amino acids, peptides, and alkaloids, constitute the major chemical components of the nitrogen content. The protein content in SCG is measured at 13.60 % [25]. Utilizing SCG protein, food products can be formulated to provide various health benefits for individuals suffering from liver diseases, oxidative stress, and hypertension [25]. Additionally, SCG contains 3.7 % nitrogenous compounds. Due to the presence of non-protein nitrogenous compounds, SCG can be beneficial in agriculture and positively impact soil amendment [35].

SCG comprises lipids in the range of 10-15 percent by weight (wt %), although certain studies have indicated lipids reaching up to 20 wt% [37]. The lipid composition of SCG is suitable for conversion into biodiesel [39]. It is estimated that approximately 1 billion liters of biodiesel can be produced from SCG annually [36].

In essence, SCG primarily consists of cellulose (glucose), hemicellulose, lignin, protein, and lipids, with a minor presence of caffeine (0.02–0.4526 %), minerals, and ashes.

3.2. The functional and structural characterization of SCG and potential valorization

Table 2 presents a summary of the structural and functional analysis of SCG derived from various studies in the literature. The water holding capacity (WHC) and oil holding capacity (OHC) refer to the ability of a material to retain water or oil after the application of external centrifugal gravity force or compression. The WHC of SCG is reported to be 5.73 ± 0.10 (g water/g dry sample), while the OHC of SCG is 5.20 ± 0.30 (g water/g dry sample) [33]. OHC and WHC both play a crucial role in the preparation, processing, and storage of food products.

WHC water holding capacity; OHC oil holding capacity; DPPH antioxidant activity by the 2,2-diphenyl-1-picrylhydrazyl assay; FRAP antioxidant activity by the ferric reducing antioxidant power assay.

Table 2 Functional and structural characterization.

Functional and physiological properties	Value	Reference
WHC (g water/g dry sample)	5.73 ± 0.10	[40]
OHC (g oil/g dry sample)	5.20 ± 0.30	[40]
Emulsifying activity (%)	54.72 ± 0.90	[40]
Emulsion stability (%)	92.38 ± 0.90	[40]
Antioxidant potential	-	
DPPH (µmol TE/g dry material)	20.04 ± 0.05	[40]
FRAP (mmol Fe(II)/g dry material)	0.102 ± 0.01	[40]
Higher heating value, HHV (MJ/kg)	19.00-26.90	[41]
Lower heating value (MJ/kg)	18.8-21.8	[42]
Structural Characterization		
Morphology and Porosity	-	
Particle size (µm)	100-500	[42]
BET surface area (m ² /g)	4.03	[40]
Specific surface areas (m ² /g)	2.00	[40]
Specific pore volumes (cm ³ /g)	0.003	[40]
Thermal Behavior	-	
Melting point (°C)	76.89	[40]

DPPH (antioxidant activity by the 2,2-diphenyl-1-picrylhydrazyl assay) of SCG is $20.04\pm0.05~\mu mol~/g$ and FRAP (antioxidant activity by the ferric reducing antioxidant power assay) of SCG is $0.102\pm0.01~mmol/g$ [33]. This suggests that the reuse of SCG to obtain such compounds is possible.

Particle size of SCG is 100–500 μm [42]. BET (Brunauer–Emmett–Teller) surface area of SCG is 4.03 m^2/g and Specific surface areas of SCG is 2.00 m^2/g . By enhancing the porosity of SCG, it becomes suitable as a biomass precursor in the manufacturing of activated carbon through chemical and physical activation methods [18,43–47].

4. SCG valorization within circular economy

The utilization of SCGs, that leads to the production of bio-based chemicals and bio-materials, is a very promising step towards sustainable development and circular economy policy. SCGs can be successfully used to produce bioplastics, biofuels, and adsorbents etc, what reduces environmental footprint associated with coffee waste disposal and contribute to the development of more sustainable alternatives to conventional petroleum-based materials. SCG can undergo various treatment methods to minimize its environmental footprint, given its composition of organic residues such as cellulose, hemicellulose, lignin, fatty acids, and other polysaccharides.

A wide range of alternative options is available for use in recycling SCGs as a valuable resource. Some of the current valorization techniques for SCG involve its utilization in animal feeding [48], the creation of organic compost [26], or its use as a fertilizer to enhance soil fertility by boosting N, P, and K levels, reusing for construction and building material [49], anaerobic digestion [50,51], extraction of high-value compounds [52-56] biodiesel production [57-61] Other applications are currently under consideration, including being evaluated as a potential feedstock and an alternative for coal used in preparing biochar fuel in the industrial sector [62,63]. In addition, there are a few studies about using SCG for water treatment [64]. The removal of heavy metals from water using biochar derived from SCG has been demonstrated by authors [65]. There has been a growing interest in the utilization of specific volatile compounds found in SCG, which could potentially serve as anti-inflammatory, antimicrobial, and antioxidant agents in the food industry [66]. These compounds can be used to formulate innovative functional foods that offer health benefits [67]. Additionally, SCG have also been utilized in the textiles and home furnishings industries [68].

Fig. 2 summarizes the various value-added products of spent coffee grounds and the components used. High value-added products are mainly environmental functional materials, biofuels and extracts. Other products, such as construction materials and fertilizers, have relatively low added value. The basis for this classification depends on the number





Fig. 2. Various value-added products and the components used.

of relevant literatures, the degree of technological maturity and the prospect of industrial and commercial applications.

4.1. Environmental functional materials for water and wastewater treatment

Different natural or discarded substances have been examined and experimented with for the purpose of adsorbing heavy metals and organic compounds from water-based solutions. Examples of these natural substances include coal, wood, lignite, and bauxite, while discarded materials such as wheat and rice waste, coffee and tea waste, as well as fruit peels have also been investigated. The main objective of these studies was to find ways to reduce the production cost of adsorbents [69,70]. Despite having a lower surface area compared to activated carbon, the SCG (presumably referring to a specific material) contains a wide range of valuable organic components that contribute to its sorption capacity [71–73]. The research papers on application of SCG are categorized according to the SCG pretreated or not. Table 3 summarizes the SCG utilization in heavy metals and dyes removal including the preparation steps of the SCG, the target pollutants, and the adsorption capacity of target pollutants. The SCG was directly used as adsorbents for heavy metals removal, and the removal efficiency of Cd⁺ and Pb^{+2} by the raw SCG was 15.65 and 2.46 mg g^{-1} , respectively [74,75]. Meanwhile, the removal efficiency of Hg⁺² and Pb⁺² by the SCG combined with silicone elastomer via sugar leaching technique was 17.1 and 13.5 mg g⁻¹ [76]. Furthermore, acid-activated SCG can remove $61.6~\text{mg g}^{-1}$ and $9.05~\text{mg g}^{-1}$ for lead and fluoride. This finding provides robust evidence for the effectiveness of SCG in eliminating lead from both industrial and potable water sources [60]. The activated form of the pretreated SCG can create a significantly larger active-surface area compared to the non-pretreated SCG, resulting in the presence of functional groups that are capable of capturing lead and fluoride contaminants.

Biomass pyrolysis is a process that entails heating biomass in an inert environment at temperatures ranging from 400 to 600° C. This results in the formation of liquid bio-oil, combustible gas, and solid char. Biochar, a stable carbon-rich residue, is produced through the pyrolysis/ carbonization of plant- and animal-based biomass. The potential applications of biochar include carbon sequestration, enhancement of soil fertility, pollution mitigation, and recycling of agricultural by-products and waste. SCG, due to its relatively high content of organic compounds like lignin, cellulose, hemicellulose, and fatty acids, has the potential to be utilized in biochar production compared to typical solid waste [85]. Therefore, some studies have already explored approaches to produce biochar from SCG [82,84,86]. In addition, the produced biochar from SCG was tested for aqueous contaminants removal. Kim et al. [84] conducted a controlled experiment where SCG was subjected to slow pyrolysis in an electric furnace. The temperature was carefully regulated, and the SCG was heated gradually at a rate of around 20 °C per minute until it reached 400 °C. It was then maintained at this temperature for a duration of 30 minutes, resulting in the production of SCG-char. The performance of SCG and SCG-char on acid mine drainage

Table 3Metal ions, anions, and dyes adsorption on adsorbents derived from SCG.

Preparation of SCG	Target Pollutant	Adsorption capacity(mg/g)	Reference
Untreated SCG at 25 °C	Pb^{2+}	2.46 mg g ⁻¹	[74]
Dried and then sieved	Cd^+	15.65 mg g ⁻¹	[75]
Combined SCG with	Pb ²⁺ and Hg ²⁺	13.5 and	[76]
silicone elastomer using sugar leaching technique	from water	$17.1 \; \mathrm{mg \; g^{-1}}$	
Activated with HCl and	Lead and fluoride	61.6 and	[77]
heated for 2 h		9.05 mg g^{-1}	
Dried at 105 °C for 5 h	Methylene blue	23.0 mg g^{-1}	[78]
Dried at 100 °C for 10 h	Acid Red 44	27.8 mg g^{-1}	[79]
Washed and dried	Rhodamine dyes	2.33 and	[80]
		$8.33~\mathrm{mg~g}^{-1}~\mathrm{forRh}$	
		B and Rh 6 G	
Dried in oven at 120 °C	H_2S	127 mg g^{-1}	[81]
for 48 h and ZnCl ₂			
powder as an activating agent			
Magnetically modified	Acridine orange	73.4 mg g^{-1}	[20]
by contact with water-			
based magnetic fluid			
Engineered biochar with	As(V)	9.20 mg g^{-1}	[82]
paper mill sludge in CO ₂			
Engineered biochar with	As(V)	13.1 mg g^{-1}	[83]
FeCl ₃ in N ₂ and CO ₂			
atmosphere			
Heated at 20 °C /min	Cd, Cu, Pb and	SCG (91 %, 58 %,	[84]
up to 400 °C and held	Zn in acid mine	>99 % and 82 %)	
for 30 min	drainage	SCG-char (99 %,	
		88 %, >99 % and	
		99 %)	

treatment was investigated and compared. SCG was effective in removal Cd (91 %), Cu (58 %), Pb (>99 %) and Zn (82 %) in the acid mine drainage. Moreover, SCG-char enhanced the removing efficiency of Cd (99 %), Cu (88 %), and Zn (99 %) effectively. In the Cho et. al. study, untreated SCG yielded the biochar that presented very low adsorption capability for AS(V). However, in the case of SCG co-pyrolysis with paper mill sludge, higher adsorption affinity towards AS(V) – reaching removal efficiency of 84.4 % – was observed with AS(V) adsorption capacity of 9.20 mg/g [82]. Furthermore, magnetite coating on the above biochar obtained higher adsorption capacity on As(V) of 13.1 mg/g. The cost of pyrolysis must be considered although the pyrolysis of SCG into biochar presented effective removal capacity on heavy metals.

Furthermore, SCG has the capacity to effectively eliminate heavy metals from wastewater. Additionally, it can serve as an adsorbent for hydrogen sulfide. Additionally, by subjecting it to a drying process in an oven at $120\,^{\circ}\text{C}$ for 48 hours using ZnCl_2 powder as an activating agent, it can be transformed into efficient desulfurization adsorbents. This transformation allows for the preparation of activated carbons that can effectively adsorb H_2S [81]. Although the activation process can be optimized or modified, the presence of nitrogen in the precursor (caffeine) proves to be a valuable asset of SCG. The nitrogen functional groups within SCG play a catalytic role in the oxidation of hydrogen sulfide.

The efficiency of eliminating methylene blue (MB) [78], Acid Red [79], and Rhodamine dyes [80] by SCGs from a water-based solution was analyzed, as detailed in Table 3. The findings suggest that SCGs exhibit significant promise as a cost-effective and readily accessible substitute adsorbent for eliminating organic dyes in wastewater treatment. Additionally, spent coffee grounds can be conveniently converted into a magnetic form through magnetic fluid treatment and utilized as an affordable magnetic adsorbent for eliminating water-soluble dyes [20].

At present, there is a growing interest in utilizing SCG as a natural filler. The research conducted by Kruszelnicka et al. [87] validates that incorporating spent coffee grounds as a filler in polymer composite

moldings efficiently enhances the growth of microorganisms in the bioreactor.

4.2. Biofuels

In recent times, the growing consumption of fossil fuels has led to a increased awareness regarding alternative resources. Numerous solid biomass resources hold significance in this context, including straw, miscanthus, paper fiber residues, bagasse, sunflower shells, palm kernel, olive dry pomace, and more [88]. Among these resources, SCG stands out due to its distinctive characteristics. With a high heating value ranging from 19.0 to 26.9 MJ/kg [4,89] and a lipid content of up to 15 wt%, SCG presents the potential for direct combustion or utilization as a fuel through transesterification and other purification processes.

The study examined the combustion properties of SCG in a small boiler system with an input of 6.5 kW based on lower heating value. The lower heating value of spent coffee grounds used as fuel is approximately 18.8 MJ/kg (equivalent to 4500 kcal/kg at 10 % water content), which surpasses that of wood pellets at a similar water content. Blended logs consisting of 20 wt% SCG and 80 wt% sawdust, when compressed, show potential as a biofuel for energy generation in residential settings. However, it is important to note that increasing the proportion of SCG in the blend leads to higher CO and particle emissions [90].

Untreated SCG used as a burning fuel can lead to further environmental issues, and there is a significant risk of spontaneous combustion when it is stored in a dried state [38]. Given these risks, extracting lipids and oil from SCG can be utilized for valuable purposes in biodiesel production. Additionally, the transesterification process for biodiesel production can be carried out either after the extraction of oil [41,91] or via in situ or direct transesterification of the SCG biomass [57,92,93].

Döhlert et al. [94] devised a procedure for converting coffee oil (triglycerides) from spent coffee grounds into hydrocarbons suitable for use as diesel fuel. For example, approximately 77 g of hydrocarbons can be generated from 1 kg of spent coffee grounds. Park et al. [57] discussed the in-situ transesterification of wet SCG to produce biodiesel, with a maximum yield of fatty acid methyl esters (FAME) at 16.75 wt% based on the weight of dry SCG at 95°C. Liu et al. [93] introduced a direct transesterification method for producing biodiesel from SCG without the need for oil extraction and esterification steps. The yield of coffee biodiesel was 17.1 $\pm\,0.7$ wt% with an acid value of 0.31 mg KOH/g after alkaline washing, comparable in quality to the conventional solvent extraction method. Additionally, Tuntiwiwattanapuna et al. [95] devised and scaled up an in-situ transesterification (TE) process for industrial spent coffee grounds (IND-SCG) biodiesel production, highlighting that increased reaction temperature and size reduction enhanced the IND-SCG biodiesel yield. A biodiesel yield of > 80 % was achieved within 3 hours at 50°C using whole deacidified IND-SCG. It was also noted that producing SCG biodiesel through conventional methods consumed 43 % less energy compared to on-site in-situ TE, although the in-situ TE process scored better in terms of respiratory health and land use.

The physical fuel properties of the biodiesel produced by SCG were found to be comparable to standard sunflower, palm, and rapeseed biodiesels in terms of energy densities. A study conducted on the recycling of SCG as a cost-effective feedstock for biofuels production in Turkey revealed that the quality of the biodiesel met ASTM D6751 standards, with all properties meeting the required specifications [96].

As we all know, biofuels contribute to lower carbon dioxide and sulfur emissions, making them more environmentally friendly than fossil fuels. However, the two most widely used biofuel feedstock currently is corn and soybeans, and the main drawback is that they must meet the food, feed and other economic needs first. In order to save the non-renewable resources of oil and coal, the large-scale conversion of different crops to corn and soybeans for producing biofuels will not only cause food shortages, but also consume a huge amount of water resources and electricity, and even destroy the ecological environment.



The solid waste such as SCG, if it can be efficiently converted to biofuels with low cost, will improve this dilemma and reduce the wide range of social and environmental issues associated with the large-scale production of biofuels.

4.3. Extraction chemicals

Spent coffee grounds are frequently subjected to extraction operations as a preliminary step in fuel preparation. By employing four different solvents and a prototype-scale extraction with circulation process, the experimental yields obtained were as follows: 14.7 wt% using hexane, 13.1 wt% using anhydrous ethanol, 11.8 wt% using hydrous ethanol, and 7.5 wt% using methanol. These optimization techniques were employed to extract coffee oil from the spent coffee grounds [54].

The extraction of spent coffee grounds can yield food additives, as well as cosmetic, and pharmaceutical substances, such as antioxidant phenolic compounds and polysaccharides. The crude ethanolic extract obtained from SCG was found to possess valuable properties, including a favorable volatile profile, antioxidant activity, and sun protection factor when evaluated in capsules [52].

The optimal condition for extracting phenolic compounds from spent coffee grounds was found to be using 60 % methanol in a solvent/solid ratio of 40 mL/g SCG for a duration of 90 minutes. This extraction method resulted in an extract with a high content of phenolic compounds (16 mg gallic acid equivalents/g SCG) and a high antioxidant activity (FRAP of 0.10 mM Fe(II)/g) [1]. In terms of encapsulation techniques, both freeze-drying and spray-drying were evaluated for phenolic compound encapsulation. Among these methods, freeze-drying with maltodextrin as the coating material proved to be a favorable option for encapsulating antioxidant phenolic compounds extracted from spent coffee grounds [97].

The research focused on enhancing the extraction of polysaccharides from spent coffee grounds through autohydrolysis to increase their antioxidant properties. Additionally, polysaccharide-rich extracts obtained from alkali pretreatment or autohydrolysis of spent coffee grounds were utilized in creating carboxymethyl cellulose based films to introduce new functionalities. Another study explored eight diverse pretreatment methods, including chemical, physical, and physicochemical approaches, followed by a sequential, combinatorial strategy to maximize sugar yield from spent coffee waste [98].

In addition, the use of coffee ground extracts in water can serve as a potent and environmentally friendly corrosion inhibitor for C-steel in a 1 mol/L HCl solution [99].

It is worth noting that the extraction process also yields valuable products such as natural pigments, proteins, lipids, starch, cellulose, enzymes, vitamins, and antioxidants, which hold great importance for the food, cosmetic, and pharmaceutical sectors [24]. Compared to the energy consumed by biorefinery to obtain biofuels, the energy used to obtain SCG extracts is much less. Therefore, it is also a good choice to obtain high value-added products of SCG through extraction operations. Additionally, this innovation aligns with the principles of the circular economy, promoting resource efficiency and waste minimisation according to Ahmed et al. [17]

4.4. Other ways and perspectives for the reuse of SCG

In addition to water treatment and burning, spent coffee grounds have many other potential applications such as co-digestion, compost for fertilizer and reusing for construction. However, their prospects may be limited due to the complex process, high cost, low product value and output rate, and substitutability of other biomass.

To achieve a satisfactory methane yield, it is essential to effectively optimize the quantity of substrates utilized during the fermentation process of SCG. This is primarily accomplished by co-digesting SCG with other organic wastes like food waste or manure [21].

Kim et al. investigated the anaerobic co-digestion of spent coffee grounds with different organic wastes, including whey, waste activated sludge, food waste, and Ulva for biomethanation. It was found that co-digestion with waste activated sludge had a negative impact on both methane production rate and yield. On the other hand, the other co-substrates improved the reaction rate and maintained methane production at a comparable or higher level compared to the mono-digestion of spent coffee grounds [100]. Additionally, the potential of Ulva biomass as a co-substrate for anaerobic digestion of spent coffee grounds was investigated. The results showed that co-digestion with Ulva (25 % on a COD basis) was beneficial for SCG biomethanation in terms of process stability and performance [101].

Orfanoudaki et al. [102] conducted research on the co-digestion of pig manure with SCG. Their findings revealed a significant improvement in methane production, with an increase ranging from 0.12 to 1.4 L methane per liter of reactor per day.

In a separate study, Atelge et al. [103] investigated the impact of oil extraction from spent coffee grounds as a pre-treatment method. They conducted Mesophilic Biomethanation Potential tests using defatted spent coffee grounds in combination with co-substrates such as macroalgae, glycerin, and spent tea waste. The ratios used were 25 %, 50 %, and 75 %, respectively. The highest methane yield was observed when defatted spent coffee grounds were digested alone, resulting in 336 \pm 7 mL CH₄ per gram of volatile solids. Furthermore, the methane yield increased along with the mass ratio of defatted spent coffee grounds in co-digestion.

Kampioti and Komilis [104] conducted a study to investigate the feasibility of anaerobic co-digestion of coffee waste (CFW) with other organic wastes, namely anaerobic sludge (AS), food waste (FW), and cow manure (CM). The researchers measured the production of biogas from different combinations of these substrates. The findings of the study revealed that the co-digestion of CFW with AS exhibited a synergistic effect, resulting in the generation of 201 mL g $^{-1}$ VS_{mixture}, which was 12 % higher than the amount produced from the mono-digestion of AS. Conversely, when CFW was co-digested with CM and FW, it had an antagonistic effect on biogas production, showing that CFW contributes to inhibition of biogas generation as a result of mixing with CM and FW.

Furthermore, by subjecting hydrolyzed spent coffee grounds (HSCG) to a simulated digestion fermentation process, the antioxidant properties of the resulting digestion and fermentation products were demonstrated in the human hepatocellular carcinoma HepG2 cell line, indicating the potential use of HSCG as a dietary supplement [105].

SCG have the ability to be combined with other substances for use as a fertilizer. Research by Ronga et al. [106] showed that when SCG and biochar are enriched with 30 % weight of glass fertilizer, grapevine production can be increased. These repurposed materials align well with the principles of a circular economy. The process of composting, aided by microbial augmentation and biochar amendment, can accelerate composting time and improve the quality of functional compost. By utilizing SCG, poultry manure, and biochar derived from agricultural waste to create functional composts through microbial bioaugmentation, the degradation and humification process can be enhanced. For instance, the addition of cow dung and SCG together proved to be particularly effective in promoting degradation and humification during the two-stage co-composting of green waste [107].

On the basis of the impact of varying rates of SCG on the composting process, gaseous emissions, and the quality of the final product, it is possible to successfully compost SCG in different proportions. This can result in reduced greenhouse gas emissions and improved quality of the end product [108].

SCG possess a remarkably high calorific value and consist of a significant quantity of water, rendering them highly compatible as a supplement to clay in the manufacturing of porous ceramic materials that exhibit thermal insulation properties. In their research, Fonseca et al. [90] examined the impact of various mixtures of clay and coffee waste on structural ceramics by introducing varying proportions of spent



coffee grounds as an additive. They discovered that SCG could serve as a secondary clay raw material, enabling the production of well-formed bricks with exceptional thermal insulation characteristics.

Utilization of SCG as a burnout additive in ceramic materials has been extensively documented in the literature [109,110]. Researchers have highlighted that the organic composition of SCGs leads to their combustion during the ceramic firing process, resulting in the formation of pores (gas-filled voids) in their initial position. These pores contribute to a decrease in the thermal conductivity of ceramic materials, which is advantageous for structural building materials [111].

Arulrajah et al. [112] discovered that utilizing spent coffee grounds-bagasse ash in geopolymer production, along with slag supplements, can result in an innovative eco-friendly construction material that satisfies the necessary strength criteria for pavement subgrade applications.

Mayson and Williams [113] introduced a practical plan for combining energy and waste management in a coffee company to establish a circular economy model. By recycling and reusing SCG for roasting, they managed to decrease carbon emissions by 5.06 kg $\rm CO_2e/kg^{-1}$ fuel per batch, in contrast to traditional methods. Additionally, they highlighted the significance of embracing the principles of a circular economy, such as "resource sharing" and "enhanced collaboration," in the relationship between coffee roasters and stores to achieve zero waste goals.

Another instance of the retrieval of different SCG components through the zero waste method can be observed in the extraction of water-soluble polyphenols, caffeine, and antioxidants. These extracted substances possess numerous applications in the pharmaceutical and cosmetic industries [21].

The utilization of SCGs for bio-based 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. Further research and development in this field will undoubtedly play an important role in the circular economy approach and mitigating the environmental impacts of traditional materials production.

5. Conclusions

As shown in the article, SCG is a valuable raw material that can be processed and used in many sectors: construction, wastewater treatment, agriculture, biofuel production, etc. The indicated examples of applications fit perfectly into the model of a circular economy.

Judging from the growth trend of SCG around the world, resource utilization is conducive to the sustainable development of this industry. Some substances contained in SCG, such as natural proteins, lipids, starch, cellulose, enzymes, vitamins, and antioxidants, can provide a basis. Various proposals for SCG biorefineries for producing different high-value products are presented. The typical procedure commences with the extraction phase in order to acquire the intended compound or initial material. The leftover substance is subsequently subjected to additional processing to yield alternative products, or to produce energy for the operation. Extraction yields certain premium products of considerable importance to the pharmaceutical, cosmetic and food sectors. The development of separation and purification technology to improve the extraction efficiency of target substances in SCG is a prerequisite for large-scale application of extraction process.

Currently, the technology for manufacturing biofuels from used coffee grounds is quite advanced, resulting in stable and reliable products. The process of using SCG as a potential source for biodiesel typically involves extracting coffee oil through solvent extraction, followed by converting it into coffee biodiesel through acid esterification and alkaline transesterification. Nevertheless, in order to implement SCG biorefinery on a larger scale, additional research and development endeavors are necessary to minimize expenses and enhance profitability.

SCG can effectively remove heavy metal ions, especially Pb⁺² and Hg⁺² ions from water or adsorb organic dyes to reduce the chroma of water after certain low-cost physical and chemical treatment. Therefore, SCG is highly prospective in the field of water treatment, for example, the drinking water treatment facility can be filled with a SCG filter to intercept certain heavy metals and organic pigments. Compared to the cost of traditional incineration and landfill disposal of SCG and the environmental capacity consumed, the use of SCG in the water treatment field may improve environmental quality while reducing the cost of such solid waste treatment and even obtaining profits. Nevertheless, the application of spent coffee grounds must be based on a sufficient quantity of spent coffee grounds recovered. It is imperative to conduct a thorough study and develop a comprehensive plan for a large-scale, efficient coffee-residue recovery system in order to maximize the utilization of spent coffee grounds from both environmental and economic perspectives within the framework of circular economy principles.

CRediT authorship contribution statement

Lurui Chen: Writing – original draft, Visualization, Resources, Formal analysis. Jacek Mąkinia: Writing – review & editing, Validation, Supervision, Conceptualization. Joanna Szulzyk-Cieplak: Writing – review & editing, Visualization, Validation. Li Xie: Writing – review & editing, Formal analysis. Hongyu Mao: Writing – original draft, Visualization, Software, Data curation. Jakub Drewnowski: Writing – original draft, Supervision, Methodology, Conceptualization. Jun Xu: Writing – original draft, Methodology, Formal analysis, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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