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Review of potential and prospective strategies for the valorization of coffee grounds within the framework of a sustainable and circular bioeconomy

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ABSTRACT

Moving from the linear production model to the circular economy approach is the main concern of the EU Circular Economy Action Plan. Population growth coupled with high demand for goods has led to a significant increase in solid waste, especially food waste, triggering the depletion of natural resources and the environmental burdens associated to their disposal. However, their physical, chemical and biological characteristics make them by-products with a high potential to be valorized and therefore used as resources for other industrial production models. This is the case of spent coffee grounds (SCGs), which are produced in large quantities on a daily basis. Therefore, valorization models under the approach of the biorefinery concept can be envisaged with the aims of recovering potential bioactive compounds and renewable energy. This has been the approach developed in this critical review, in which SCG recovery alternatives have been studied to obtain lignin, lipids, biofuels and phenolic compounds. In addition, a critical analysis of the outcomes of techno-economic and environmental evaluations available in the literature is included, in order to identify those indicators that provide information on the feasibility of their valorization.

1. Introduction

The search for an adequate management of all by-products produced in industrial facilities has become one of the main drivers of research and development in the context of the circular economy. However, a huge percentage of the total waste generated daily is managed in landfills or, depending on its composition, its energetic valorization is considered. However, it should be taken into account that, in general terms, the waste produced is suitable for its chemical composition, according to various routes to recover products with a higher added value in the market, as is the case of antioxidants, antimicrobials or bioactive compounds. Therefore, a biorefinery approach should be proposed, with a cascade strategy to produce several products, which greatly increases the value of this waste stream, which could now be defined as a "feedstock" rather than a waste product.

It is in this context that spent coffee grounds (SCG) could be included. This is the main waste associated with the coffee industry, as it is obtained by mixing coffee powder with water to obtain soluble coffee. Although it is possible to manage them as waste in composting and biomethanization stages, it is possible to valorize this type of waste in a biorefinery strategy, which is considered the objective of this critical review. This manuscript identifies the different valorization routes for SCG, focusing mainly on the extraction of lignin, lipids and phenolics. For this purpose, an in-depth evaluation of the research reports that have been published on this topic has been addressed. In addition, since economic and environmental issues must be considered as feasibility indicators for the processes under development, techno-economic assessment (TEA) and life cycle assessment (LCA) reports have also been taken into account. In addition, software tools for reference management have been used: Mendeley Desktop® and VosViewer®, mainly, which allows the compilation of research reports and the visualization of the most important data. Critical analysis of this information can provide researchers and stakeholders with the key operational conditions and potential for biorefinery development using SCGs as raw material.

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2. Preliminary analysis of available literature on SCGs valorization

In order to develop the literature review of published reports based on the valorization of SCGs, the time frame of the last 12 years, from 2011 to September 2023, was used as a search filter. In addition, the logical operators AND, to include the SCGs together with the words “lignin”/“lipids”/“phenolics”/“oil”/“LCA”/“TEA”/“polymer”, and also * to consider both singular and plural forms (i.e., “lipid*”) were used.

As a database for the research, Scopus® has been used, as it is considered one of the largest peer-reviewed scientific literature databases in the world, so that its use guarantees an in-depth overview on the current state of the art of SCG valorization. A large amount of research articles has been found, with a wide range of keywords, most of them referring to the same topic, but with other denominations (Figure 1SM). For example, this is the case for the keyword SCGs, as it has been found to vary as: “Spent Coffee Ground”, “spent coffee grounds”, “Coffee Grounds”, “SCG”, “SCGs”, but all these terms refer to the same material. To have a better overview of the evaluated topics, the selected keywords of the authors have been standardized according to the topics developed in this review article (Figures 2SM-5SM).

Four main groups can be distinguished: spent coffee grounds, coffee, phenolic compounds and fermentation. In addition, the keyword "extraction" is one of the most used by the authors, which is presented as the main focus of the research developed for spent coffee grounds, the recovery of active compounds, mostly phenolics with antioxidant properties, with high added value in the market. On the other hand, the keyword "biorefinery" is not as widespread as expected, which is a signal of the early stage of development of valorization routes for SCGs.

Moreover, considering the year of publication of the research articles based on SCGs recovery, the interest on this topic has been gaining weight over the last year, with the year 2021 presenting the highest number of research manuscripts on the topic (Fig. 1). However, it is believed that in the present year, the number of manuscripts will be even higher, since the number of research papers published until mid-2023 is more than half compared to last year. To this end, it can be concluded that the interest of researchers in the valorization of SCGs is growing and it is very likely that SCG biorefinery approaches will be considered at an industrial level in the near future, given their compositional potential, their content in bioactive compounds, their possible use as a source of fermentable sugars for biofuels and their heat capacity as an alternative to fossil fuels.

The most relevant journals that have published articles and reviews evaluating the recovery of SCG for the extraction of lipids, lignin, phenolics or others, based on the TEA and LCA analysis, are Bioresource Technology (with 28 manuscripts), followed by Waste and Biomass Valorization (with 16 research articles), Chemical Engineering Transactions (with 12 reports), Fuels (with 10 manuscripts) and Journal of

Table 1
Top-authors on the scientific productivity, according to Scopus database, on SCGs recovery strategies and valorization topics.

Author	Reports on SCGs	Institution	h-Index	Citations	References
Coimbra, M.A.	9	University of Aveiro	56	10763	(Oliveira et al., 2021; Cláudia P. Passos et al., 2019; Passos et al., 2015; Simões et al., 2013)
Mussatto, S.I.	9	Technical University of Denmark	53	11579	(Ballesteros et al., 2015; Lina F. Ballesteros et al., 2017a, 2017b;Conde and Mussatto, 2016;Machado et al., 2018)
Chuck, C. J.	8	University of Bath	25	2100	(Jenkins et al., 2017; Massaya et al., 2021b, 2021a, 2019; Pereira et al., 2021)
Márovà, I.	8	Brno University of Technology	29	2158	(Hudeckova et al., 2018; Kovalcik et al., 2018; Obruca et al., 2015; S. Obruca et al., 2014; Stanislav Obruca et al., 2014; Petrik et al., 2014)
Teixeira, J.A.	8	University of Minho	83	30787	(Lina F. Ballesteros et al., 2017a, 2017b; Mussatto et al., 2012; Sampaio et al., 2013)

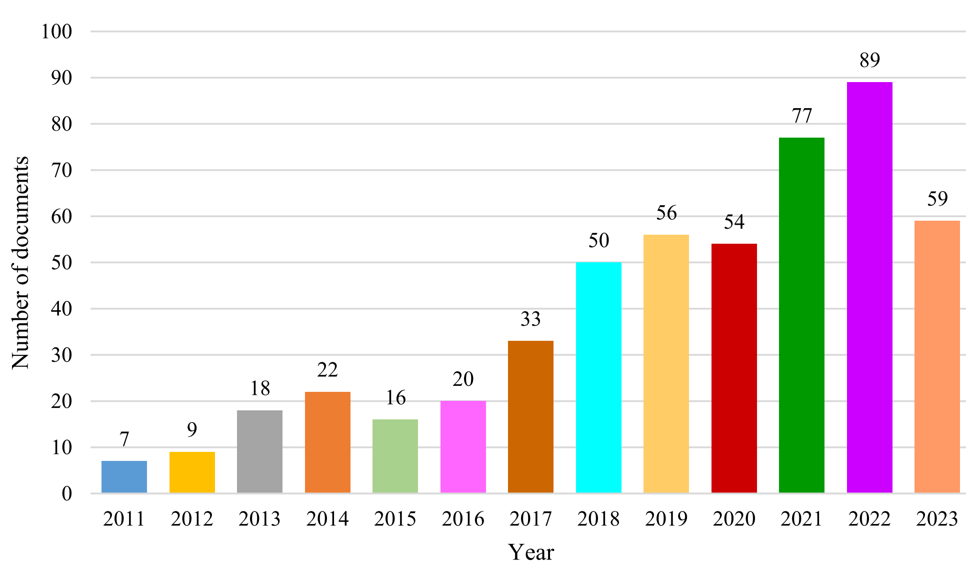

Fig. 1. Column chart including the number of references published per year in the period 2011–2023.

Cleaner Production (with 9 research reports). The top-authors in terms of scientific productivity, according to the Scopus database, are shown in Table 1.

3. Composition of spent coffee grounds

When thinking about the valorization of SCGs and the recovery of the high value-added components available in their molecular structure, the first step is to consider their composition. However, it should be borne in mind that, depending on the coffee variety, the composition of certain components, especially in terms of phenolic, antioxidant and flavonoid content, can vary significantly. Therefore, before considering a cascade valorization of SCG, it is important to analyze the composition of the SCG in terms on lignin, phenols and lipids in order to decide which cascade biorefinery process is the most convenient, considering economic, environmental and technological aspects. Bearing this in mind, Table 2 and Table 3 are depicted below, including the chemical composition of SCGs in a range percentages according to dry matter content, as can be found on literature (Table 2) and the content on phenolics, flavonoids and antioxidant compounds in μg/g, also in dry matter basis (Table 3). On the other hand, it is worth mentioning that the ranged values included in both Tables 2 and 3 are the result of the analysis of different research reports that have taken into account how the composition of SCGs could be influenced by external factors (i.e. cultivation conditions, geographical area, climate conditions, etc.).

4. Valorization routes: paving the way for circular economy

The different techniques of valorization of SCGs are described in the following sections, focusing on the recovery of lignin, lipids, phenolics and oil. The huge variety of applications of these products in different sectors, such as the food, pharmaceutical, medical and energy, makes them potential products of great interest.

4.1. Lignin extraction alternatives for valorizing SCGs

Lignocellulosic biomass could represent an important renewable resource due to the different alternative routes that could be developed in the search for the substitution of fossil resources. In particular, the recovery and valorization of lignin is one of the most interesting options, due to the various products that could be obtained, both energetic and bio-chemicals. However, due to its molecular structure and physico-chemical properties, it is also considered as a challenging feedstock, as it is necessary to break strong molecular bonds for its useful valorization.

Table 2
Chemical composition of SCGs (*Unit: % dry matter).

Component	Amount*	Reference
Cellulose	8.6-12.4	(Arya et al., 2022; Ballesteros et al., 2014; Kwon et al., 2013; Lavecchia et al., 2016; López-Barrera et al., 2016; Mussatto et al., 2011)
Hemicellulose	19.0-39.1	(Ballesteros et al., 2014; Kelkar et al., 2015; Mussatto et al., 2011)
Arabinose	1.9-3.6	(Ballesteros et al., 2014; Kwon et al., 2013; López-Barrera et al., 2016)
Mannose	13.2-19.1	(Ballesteros et al., 2014; Kwon et al., 2013; López-Barrera et al., 2016)
Galactose	16.4-26.0	(López-Barrera et al., 2016; Passos et al., 2019)
Lignin	23.9-33.6	(Ballesteros et al., 2014; Caetano et al., 2014; Kelkar et al., 2015)
Glucan	8.6-13.8	(Caetano et al., 2014; Mussatto et al., 2011)
Ashes	1.2-2.3	(Hernandez-Arriaga et al., 2017; Kelkar et al., 2015)
Protein	10.0-17.4	(Caetano et al., 2014; Lerda, 2016)
Carbohydrates	45.0-68.4	(Hernandez-Arriaga et al., 2017; Lerda, 2016; Martinez-Saez et al., 2017)
Lipids	15.1-27-0	(Hernandez-Arriaga et al., 2017; Lerda, 2016)
Total fiber	57.1-60.5	(Ballesteros et al., 2014; De Cosio-Barron et al., 2020; Martinez-Saez et al., 2017)
Insoluble fiber	50.8-57.1	(Ballesteros et al., 2014; De Cosio-Barron et al., 2020; Martinez-Saez et al., 2017)
Soluble fiber	1.6-9.7	(Ballesteros et al., 2014; De Cosio-Barron et al., 2020; Martinez-Saez et al., 2017)

Table 3
Main composition on phenolics/flavonoids/antioxidant compounds on SCGs (*Unit: μg/g dry matter).

Component	Amount*	Reference
Caffeine	209-439	(Badr et al., 2022; Ho et al., 2020)
Caffeic aid	7.2-41.4	(Badr et al., 2022; Ho et al., 2020)
Catechin	16.9-24.0	(Badr et al., 2022; Ho et al., 2020)
Chlorogenic acid	7.4-24.0	(Badr et al., 2022; Okur et al., 2021)
Gallic acid	3.1-18.2	(Badr et al., 2022; Ho et al., 2020)
Ferulic acid	21.1-119	(Angeloni et al., 2021; Ho et al., 2020)
p-cumaric acid	0.2-18.3	(Badr et al., 2022; Ho et al., 2020)
Quercetine	1.4-3.96	(Angeloni et al., 2021; Ramón-Gonçalves et al., 2019)
Rutin	4.9-10.11	(Angeloni et al., 2021; Badr et al., 2022)
Syringic acid	64.1-78.63	(Angeloni et al., 2021; Badr et al., 2022)
Vanillic acid	0.5-54.3	(Badr et al., 2022; Ho et al., 2020)
Sinapic acid	10.1-17.1	(Badr et al., 2022; Hussein et al., 2022)
Salicylic acid	7.61-12.7	(Badr et al., 2022; Hussein et al., 2022)
Epicatechin	37.2-53.8	(Badr et al., 2022; Ho et al., 2020)
Naringin	0.40-0.62	(Angeloni et al., 2021)
Kaempferol	0.4-3.2	(Badr et al., 2022; Król et al., 2020)

It is true that there are not many research articles analyzing lignin removal as a main objective, but it is a pretreatment step aiming at higher productivity for the production of fermentable sugars from lignocellulosic biomass (Sugebo, 2022). To this end, this section of the manuscript discusses the different techniques used for lignin removal.

There are novel techniques that have been used for the recovery/removal of lignin from SCGs, such as the use of ionic liquids. Tolesa et al. (2018) have considered the use of ammonium-based ionic liquid for the extraction of lignin using mild conditions, up to 71.2% after 4 h of reaction time at 120 ºC (Tolesa et al., 2018).

Nevertheless, conventional procedures are the most common, due to their simplicity, mostly based on physical and chemical treatments. Those are based on the chemical modification of the lignin content of SCGs, by phenolation and acetylation, which not only implies low costs and shorter reaction time, but also allows the reuse of lignin for other applications, since it is not degraded (Taleb et al., 2020). One of the most innovative and recent research for lignin applications is its use in the production of aluminum-air batteries, where it is used as an electrolyte additive. The addition of lignin implies enhanced corrosion inhibition and improved battery performance, given the chemisorption properties of lignin molecules, which involves an electrostatic-based interaction between the battery surface and the hydroxyl groups of lignin (Lee et al., 2021).

To achieve high-purity and high-quality lignin from SCG, organosolv pretreatment has proven to be a viable alternative. In this case, an organic solvent and catalyst in combination of high temperatures are required. (Ravindran et al., 2018) have studied the optimization in the organosolv pretreatment of SCG, considering both the maximum amount of lignin removal, the largest phenolic extraction yield and the highest amount of reduced sugars extracted. To this end, the optimized process was obtained when using ethanol (68%) as organic solvent, 1.5% H_2SO_4 as catalyst at 51ºC during 45 min. In addition, the authors also concluded that the requirements of this pretreatment process can be scaled up in a biorefinery approach, given the ease, cost-effectiveness and yield values obtained (Ravindran et al., 2018).

On the other hand, it is also important to consider the downstream process required to obtain lignin with the highest purity possible. A sequential separation procedure based on centrifugation for the liquid separation, and a precipitation stage with methanol was considered (Lee et al., 2019). Another way to recover lignin with a higher level of purity is to perform precipitation stages, using 60% ethanol with the addition of HCl to maintain the pH at a value of 2. Within the precipitation stages, centrifugation is required, as the supernatant is used for precipitation, while the insoluble part requires additional dissolution stages, at high temperature (150 ºC) during 70 min with ethanol/water, to allow the subsequent precipitation stage (Du et al., 2021).

Alkaline methods have also proven to be an effective procedure for lignin recovery/removal, as they lead to an increase in internal surface area and the breaking of lignin-carbohydrates bonds. However, the highest yields of this pretreatment method are obtained when the lignin content of the lignocellulosic biomass is low (Amin et al., 2017). When medium to high lignin content is available, as it is the case of SCGs, the organosolv pretreatment method is more adequate, as it has a higher capacity to break the internal bonds of hemicelluloses and lignin (Loh et al., 2019).

Thermal pretreatments, such as steam explosion, have been used for many lignocellulose-based industries as it leads to higher yields when the breakdown of lignin structures is desired. But, when developing a biorefinery process, this thermal pretreatment could lead to degradation of the available sugars in the lignocellulosic feedstock, if the temperature requirement of the process is too high (Xia et al., 2020). In addition, the high energy demand leads to significant environmental damage due to the use of fossil fuels for the production of energy requirements (Prasad et al., 2016).

Currently, there is a strong tendency to try to use biological pretreatment methods, as they are considered low demanding in chemical and energetic terms, more environmentally friendly and with reduced costs (Khir and Pan, 2019). In this case, the degradation of the bonds and recalcitrant cell wall structures of the lignocellulosic feedstock is performed by microorganisms. But, despite the high yields and efficiency of the procedure, biological treatments are time demanding and require enormous control over microbial growth, leading to more difficulties in applying this method in industrial facilities considering the economic profitability of the process (Joshi et al., 2021).

But what about the emerging uses of lignin for the extraction of high value-added compounds or for the production of biocomposites? The high molecular weight of lignin makes it a potential source for biocomposites production, as it could be used as a coupling agent, resulting in a high-strength matrix that could be used as an adhesive, i.e., in wood panels. Then, to take advantage of this, a pretreatment stage of the lignin is required, to increase the availability of the phenolic hydroxyl groups that are available in its structures. There are different ways to "activate" lignin, which implies a breakdown of its structure to turn it into a more accessible molecule, the most common ones are based on phenolation, demethylation (Zhao et al., 2022), oxidation (Azadfar et al., 2015), depolymerization (Gao et al., 2021) and/or glyoxalation (El Mansouri et al., 2007; Younesi-Kordkheili et al., 2016).

Looking for more sustainable process, Arias et al. (2022) have carried out an environmental assessment on the use of lignin as a renewable resource for the production of wood bioadhesives (Arias et al., 2022). The production of bioadhesives has been based on a first lignin functionalization step based on a carbonation reaction with dimethyl carbonate followed by a crosslinking step with hexamethylene diamine. The feasibility of the process and the environmental profile have been found to be adequate to consider the use of lignin as an alternative resource to formaldehyde in the production of wood adhesives (Arias et al., 2022). But, on the other hand, a previous study by the same authors has shown that the pretreatment of lignin for activation is a crucial step, since if a glyoxalation reaction occurs, the amount of glyoxal needed as a crosslinking agent, together with the raw amount of energy needs towards the process, turns the lignin-based bioadhesive into a not so good process alternative (Arias et al., 2020).

In addition, another option that is gaining attention in the valorization of lignin is the recovery of high value products such as phenolic compounds, vanillin, aromatic diacids and quinones, among others. Faustino et al. (2010) have used ethyl acetate as an extraction agent for the recovery of 17 phenolic compounds from lignin representing 1009 mg GAE/g and an antioxidant index of 11.4 (Faustino et al., 2010).

Seeking to reduce the amount of chemicals used, Vázquez-Olivo et al. (2019) have performed an acid hydrolysis, giving a total amount of 1421 mg GAE/100 g of lignin residues and an antioxidant capacity of 11.75 mmol ET/g (Vazquez-Olivo et al., 2019). In addition, renewable aromatic chemicals could also be obtained using lignin as a renewable feedstock. Mycroft et al. (2015) have studied the microbial degradation of lignin into aromatic chemicals, using Rhodococcus jostii as a strain (Mycroft et al., 2015). This procedure has also been recently evaluated for Pseudomonas putida to obtain pyridine dicarboxylic acid, as a precursor of bioplastics (Gómez-Álvarez et al., 2022).

Fenton oxidation has also been used for lignin depolymerization. Cronin et al. (2017) have used sodium percarbonate to enable depolymerization and subsequent extraction of dicarboxylic acids from lignin (Cronin et al., 2017). The alkaline reaction medium provided by the use of this chemical agent allows to reduce the amount of waste produced, as thermal degradation is prevented, which has been the main drawback detected by other authors when using Fenton oxidation procedure (Ma et al., 2014).

This opens the scope of potential uses of lignin. Its valorization beyond energy production has been gaining importance and is one of the main current research topics. In fact, some industries have also focused on marketable products from lignin, as is the case of Stora Enso ("Stora Enso," n.d.), which has developed different lignin-based products: Lineo®, lignin to replace phenol in resins and to manufacture biodegradable polymers, Lignode®, a lignin hard carbon battery to replace lithium-ion batteries, Neoligno®, a bio-based binder with uses in the manufacture of particleboard and insulation products and Neofiber®, a renewable carbon fiber composed of cellulose and lignin. METGEN is another industry that has developed METNINTH, which is a lignin refining technology based on converting lignin into a more accessible molecule to take advantage of its molecular compounds. And, lignin biopolymers are also produced by Borregaard, located in Norway and one of the most advanced sustainable biorefineries based on wood materials ("Borregaard," n.d.).

4.2. Lipid extraction alternatives for valorizing SCGs

The lipid composition of SCGs (around 16% w/w) makes this bio-based by-product a potential feedstock for biodiesel production (Go et al., 2020; Loyao et al., 2018), given its availability, its affordable price and the high-quality of the biodiesel produced (Muharam and Ramadhany, 2021).

To obtain them, both mechanical extraction and solvent extraction could be developed, the latter being the most efficient in terms of yield, due to the fact that it is more convenient when the lipid content is lower than 20% w/w, as is the case of SCGs (Koubaa et al., 2016). Different solvents has been used by authors: hydrocarbons, such as n-octane (Caetano et al., 2013), n-hexane (Efthymiopoulos et al., 2019) or toluene (Al-Hamamre et al., 2012), alcohols, such as ethanol and isopropanol (Battista et al., 2020; Son et al., 2018), esters, with ethyl acetate being the most common (Go et al., 2020; Loyao et al., 2018; Supang et al., 2022), and mixed solvents, based on the combination of hexane (Ahangari and Sargolzaei, 2013) with isopropanol or methanol (Cholakov et al., 2013; Efthymiopoulos, 2018). The co-occurrence of keywords used by authors in the research articles based on the recovery of lipids from SCG is depicted in Figure 6SM.

Regarding n-hexane, Efthymiopoulos et al. (2019) has investigated which are the most convenient extraction conditions to allow higher extraction yield, leading to optimized productivity of the recovery process (Efthymiopoulos et al., 2019). Large scale extraction has been evaluated, to have more accurate values closer to those of an industrial level approach. According to the report, the main findings were the extraction duration, which should be less than 10 h to avoid the decrease of the extraction yield, being 8 h the optimal value, for the laboratory level case. This time is reduced to 2 h when pilot scale approaches are developed, reaching results similar to those of the laboratory level. On the other hand, it has been concluded that the moisture content of SCGs directly affects the overall productivity of the recovery process, with a value of 10% moisture leading to the highest yield (Efthymiopoulos et al., 2019).

In search of more environmentally friendly options for lipid extraction from SCGs, solvent alternatives have been evaluated. This is the case of aqueous 2-methyloxolane, a bio-based and safe solvent (Claux et al., 2021; Gharby et al., 2020), used in a conventional Soxhlet system, requiring a solid-to-solvent ratio of 1:10 and a process time of 6 h. This alternative has been compared to hexane, and a higher yield has been achieved using 2-methyloxolane, probably due to the higher polarity and solubilization for polar lipids, as is the case for phospholipids. Among them, it has been identified that triacyl glycerides enabled the highest extraction yield, reaching 94% (Chemat et al., 2022). The solvent 2-methyltetrahydrofuran produced from renewable lignocellulosic resources has also been used for lipid extraction (Pace et al., 2012). Its use leads to a higher lipid extraction yield, when compared to hexane, almost 10 points higher, making it a potential alternative in lipid recovery from SCGs (Mkhonto and Chetty, 2021).

Not only greener solvent extractants were used, but also emerging extraction technologies, such as supercritical CO_2 extraction (Muharam and Ramadhany, 2021). It has been reported that, as expected, extraction conditions directly affect lipid yield, with increased pressure, at constant pressure, reduced particle size and higher solvent flowrate (Couto et al., 2009; Muangrat and Pongsirikul, 2019; Muharam and Ramadhany, 2021).

4.3. Phenolics extraction alternatives for valorizing SCGs

The polyphenol content of SCGs makes them a potential source of bioactive compounds, although there is not much research on this topic, at least on a large scale, both from an economic and technological point of view (Gąsecka et al., 2020). However, recent research articles have been focused on the recovery of phenolic compounds from SCGs, as these products could give significant and raw value for biorefinery development. The market for phenols has a high added value, with a multitude of applications in sectors such as cosmetics and medicine (Badr et al., 2022; Bondam et al., 2022).

Chlorogenic acids (CGAs) are the main components of the phenolic fraction of SCGs, and usually the total content of phenolic compounds is expressed as milligrams of gallic acid equivalent (mg GAE). (Panusa et al., 2013) characterized the SCG extracts in terms of their composition in total phenolic content and antioxidant activity, and also evaluate the possible changes that could occur in terms of composition and antioxidant activity by replacing aqueous ethanol with pure water as extraction solvent. These authors reported that SCG is a rich source of natural phenolic antioxidants, as it contains a high percentage of residual CGAs. Furthermore, although the use of ethanol and water can dissolve a wider range of phenolic compounds and flavonoids than the case where only pure water is used as solvent, not all types of CGAs are affected by the type of solvent. Thus, the alternative of using pure water to produce extracts rich in specific CGAs is preferable and desirable, since the reduction in the amount of chemicals leads to both lower costs and environmental impacts.

The evaluation of different parameters for the extraction of phenolic compounds using solvents, as well as the use of alternative techniques that are considered environmentally friendly for the efficient extraction of phenolic compounds, are common topics in the literature. Solid-liquid extraction with organic solvents, ultrasound-assisted extraction, microwave-assisted extraction, supercritical fluid extraction and high-pressure processes are some of the alternative techniques. (Solomakou et al., 2022) presented the conventional methodology for the extraction of phenolic compounds, as well as three different alternatives, namely ultrasound-assisted extraction, microwave-assisted extraction, and solvent extraction using β-cyclodextrin as solvent. The factors evaluated were temperature, solvent concentration, liquid/solid ratio and power. The optimum extraction yield (31.79 ± 0.25 mg GAE/g SCG) was achieved using microwave-assisted extraction, whereas the lowest yields were obtained with β-cyclodextrin as solvent. (Solange I. SolangeI. Solange I. SolangeI. Mussatto et al., 2011; S.I. Mussatto et al., 2011) also reported that the extraction of phenolic compounds was affected by the methanol concentration, solvent/solid ratio and extraction time used. The maximum value of phenolic compounds extracted from SCG was 18 mg GAE/g SCG, which was obtained using 50% methanol at a ratio of 25 ml per g SCG, for 90 min

Further processing of the extracted phenolic compounds has been evaluated to preserve their properties for a longer time. Encapsulation of these compounds is an important strategy as the phenolic compounds could be protected from oxidation as the coating material acts as a barrier against oxygen and water. Typical encapsulation techniques are usually based on spray drying, fluidized bed drying, fluid bed coating and freeze drying, due to the liquid nature of the extracts containing the bioactive compounds (Ballesteros et al., 2017).

The bioactive and phenolic content of SCGs, with anti-inflammatory, neuroprotective, antimicrobial, and anticancer properties (Dorsey & Jones, 2017), suggest their use as food supplements and ingredients, as well as in cosmetic products. There is a wide variety of products on the market that contain formulations based on coffee extracts or its by-products, as shown in Table 4. For example, the company Pectcof developed a product called Dutch Gum that has emulsifying and stabilizing properties and is formulated from coffee pulp; this is sold as an ingredient to the food and beverage industry (Pectcof, 2020). In addition, the company Aqia Nutrition has developed a line of products called AQIA coffee (AQIA, 2020), based on green coffee and cherry coffee. Among the products marketed are green coffee and cherry coffee oils, extracted by cold pressing coffee seeds (Bondam et al., 2022).

4.4. Fermentative production of chemicals and polymers by valorizing SCGs

Butanediol is a chemical compound with many applications in industry, as it is used in polyesters, cosmetics, pharmaceuticals, food additives and fertilizers, among others. Its usual production route is based on acid fermentation, but various co-products are obtained, such as ethanol, lactate, acetone, etc., depending on the type of microorganism and the fermentation conditions. The microbial production of

Table 4
Some examples of commercialization and application of SCGs in the cosmetic, pharmaceutical and food products.

Commercial product	Bioactive compound	Application	Reference
Cosmetic	Caffeine and chlorogenic acid	Anti-photoaging agent	(Choi and Koh, 2017)
Sunscreen	Coffee oil	Skin treatment and protection	(Kanlayavattanakul et al., 2021)
Pharmaceutical	5-caffeoylquinic acid	Anti-inflammatory	(Marto et al., 2016)
Cosmetic	Chlorogenic acid	Anti-wrinkle effects	(Cho et al., 2017)
Food ingredient	5-caffeoylquinic acid	Higher nutraceutical value	(Bertolino et al., 2019)
Cosmetic	Phenolics	Skin antiaging and lightening effect	(Ribeiro et al., 2018)
Food ingredient	Rosmarinic and syringic acids	Antifungal, anti-mycotoxigenic and anti-cytotoxic effect	(Badr et al., 2022)
Cosmetic	Coffee silver skin	Phyto cosmetic effects	(Rodrigues et al., 2016)

butanediol from *Cellulosimicrobium cellulans* (Ribeiro et al., 2020) could be developed by considering a sumerged coffee fermentation in a controlled pH medium and addition of pectinolytic enzymes, but also using *Rhizopus oligosporus* (Lee et al., 2016) with a solid-state fermentation. This butanediol could also be used to produce polymers, as it is a precursor of polyurethanes, and other products, such as methyl ethyl ketone, a fuel additive with many applications to produce additives, resins or other solvents (Hazeena et al., 2020; Tinôco et al., 2021).

Another microorganism used for the valorization of SCGs within a solid-state fermentation (SSF) process is *Aspergillus* sp. for the biotechnological production of polyphenols such as chlorogenic, quinic and caffeic acids, compounds recognized for their antioxidant and neuroprotective properties. After a SSF process with hydroalcoholic extraction, an increase of 2.3 times g GAE/kg is obtained compared to the use of the simple and conventional hydroalcoholic extraction with ethanol (Arancibia-Díaz et al., 2022), in addition to the lower amount of solvent associated with the SSF process.

In the nutraceuticals section, SCGs could be used as a source of prebiotics, after a process of acid hydrolysis and incubation with lactic acid bacteria (Prasanna and Rastall, 2017; Varzakas et al., 2018). The results obtained showed that SCG extracts have a more effective prebiotic capacity compared to that of inulin, an established and commonly used commercial prebiotic (Sarghini et al., 2021).

Another alternative for the valorization of SCGs is the production of biopolymers, potential substitutes for petroleum-derived polymers such as plastics. Biopolymers are biocompatible and degradable with physicochemical, thermal and mechanical properties very analogous to those of petro-based origin (Saratale et al., 2020; Saratale and Oh, 2015). Stanislav et al. (2014) explored that the production of PHBs [poly (3–hydroxybutyrates)] by *Cupriavidus necator H16* in culture media containing SCGs (Stanislav et al., 2014).

Aerobic fed/discontinuous aerobic fermentation was performed with an initial coffee oil concentration of 30 g/L, at a controlled neutral pH medium and at room temperature. In the case of a fed-batch fermentation, in the feed stage, an additional amount of 20 g/L coffee oil and 3 g/L ammonium sulfate (as a nitrogen source) need to be added to increase the yield and productivity of the process (Stanislav et al., 2014). Furthermore, one of the advantages of using oil from SCGs as a bioresource for PHB production is based on the fact that the weight percentage of the polymer in the biomass produced is around 90%, which facilitates its isolation and the subsequent steps required. *C. necator* has also been used to produce PHA [polyhydroxyalkanoate], in a mineral medium supplemented with 20 g/L of SCG oil, previously extracted by a semi-continuous supercritical extraction. After 48-h incubation time, the polymer must be extracted, using lyophilized cells and chloroform as solvent, followed by a filtration step under vacuum conditions. With this biotechnological route and operating conditions, a polymer content of 78% w/w and a yield of 0.77 kg PHA/kg SCG oil was obtained, with a molecular weight of $2.34 \cdot 10^5$ and a low polydispersity index (Cruz et al., 2014).

5. Assessing the environmental loads and economic profitability of SCGs valorization

5.1. Life cycle assessment for the environmental evaluation of SCGs

When it comes to assessing the sustainability of a new production process under development, it is important to establish what environmental benefits it brings and what the advantages are compared to existing production schemes at the industrial production level. The new biorefinery concept is usually developed on the basis of the utilization of unusable waste resources, the direct management of which is mostly based on landfill disposal. This type of management, although one of the most widespread, is not the most interesting from an environmental point of view, since the emission of particles, as well as the production of gases, mainly methane and carbon dioxide, and the damage to the landscape, give rise to significant environmental impacts. For this reason, more and more efforts are being made to valorize these by-products as feedstock for the development of biorefineries, i.e. various cascading products. In most cases, energy valorization appears as the most developed option, but given the richness of the biochemical composition of SCGs, a wide variety of high value-added products can be obtained, as introduced in the previous sections of this critical review. But, in order to select which of them is the most suitable, or to determine which one contributes more positively to the concepts of circular economy and sustainability, the use of the Life Cycle Assessment methodology is essential. This methodology is based on the evaluation of the potential environmental burdens that may result from the development of a given production process or the manufacture of one or more products from SCGs. However, given the lack of development of biorefinery processes based on SCGs on a large scale, life cycle analysis studies are not extensive in the literature, given the lack of necessary inventory data.

Several scenarios were evaluated in which different SCG management strategies were combined, including biodiesel production, anaerobic digestion, composting, direct application to cropland, incineration and thermal energy generation, and landfilling with recovery of the generated biogas for electricity generation (Schmidt Rivera et al., 2020). Among all the options evaluated, it was found that anaerobic digestion and direct application of the SCGs as fertilizer were the most environmentally sustainable options (Schmidt Rivera et al., 2020). On the other hand, different routes for biodiesel production have also been evaluated, although the conventional one is based on a first solvent extraction, to continue with a 2-step transesterification with an acid pretreatment and using NaOH as catalyst, an attempt has been made to develop a new process based on transesterification (Tuntiwiwattanapun et al., 2017). In this case, the aim would be to reduce the number of process steps, as well as the use of different types of solvents, such as the catalyst or the first extractive solvent. However, it was concluded that the energy consumption of the conventional process was significantly lower than that of the transesterification one, resulting in a lower environmental impact, as the energy requirement contributes to the consumption of fossil resources, which implies a high environmental burden. (Yang et al., 2021) have compared two thermochemical valorization routes, one focused on a first biodiesel extraction followed by a hydrothermal liquefaction of the defatted SCGs to produce biocrude, and the other based on the production of biocrude directly from the HTL of the raw SCGs. The lower yield of the route 1, compared to that of direct HTL, led to a greenhouse gas emission value of almost 3 times higher, being 297.6 g CO_2eq /MJ for route 1 and 103.3 g CO_2eq/MJ for the direct conversion (Yang et al., 2021).

On the other hand, it is also important to evaluate which stages of the SCGs valorization process for the production of biodiesel have the greatest environmental impact and, therefore, required a greater degree of improvement or optimization in order to reduce the environmental damage of the process. Although the drying of the SCGs gives rise to a large energy requirement, this is not the stage with the greatest environmental contribution, the oil extraction stage being more than 10 times higher, according to the (Bui et al., 2021) research report, given the enormous amount of solvent required for extraction, in addition to the energy requirements, both electrical and calorific.

5.2. Techno-economic assessments for considering the profitability of SCGs valorization

Usually, the development of an environmental analysis using the LCA methodology is combined with a techno-economic evaluation (TEA), as it is also important to consider the profitability of the process. No matter how environmentally friendly a process may be, if it is not adequate in economic terms, i.e., if it does not generate sufficient benefits to offset all costs and, in addition, generate income, the biorefinery approach developed will not be implemented at an industrial production level. This has been the concern in the studied developed by (I. K. Kookos, 2018), as in his research it has been evaluated the environmental and techno-economic analysis of the production of biodiesel, glycerol and electricity using SCGs as raw materials. It has been confirmed the fact that the environmental loads of the process are comparable to the BATs (Best Available Techniques) in the production of biodiesel, but the large-scale production of this biorefinery is only economically affordable when a centralized manufacture is developed.

(Banu et al., 2021) have considered different biorefinery pathways on the valorization of SCGs under a cascade approach. Given the chemical and physical properties of SCGs, there is a wide range of possibilities in its recovery, from the production of biofuels, bioplastics and biopolymers to the manufacture of polyurethane foams and bioactive compounds, a carotenoids and antioxidants. According to the most recent research articles, the most profitable SCGs biorefineries scenarios are the ones based on the production of biomolecules, derived from the saponification and neutralization of the free fatty acids (FFA) obtained from the extracted oil of SCGs (De Melo et al., 2014).

Another common route of valorization relies on the use of SCGs for bioethanol production, following an ABE fermentation procedure. It also requires a pretreatment stage, based on a milling procedure, to increase the productivity of the following stages (as it leads to an increase on the surface area allowing a higher available contact surface) and an acid hydrolyzation with sulfuric acid. Afterwards, an enzymatic saccharification is carried on, using a cellulase enzyme, with the aim of obtaining the fermentable sugars for the ABE fermentation, in a free-form, as the solid residue containing the lignin is separated by filtration (Carmona-Garcia et al., 2019). This valorization route also gives economic profitability, with revenues that amounts to $ 6.84 M/y.

Electricity production has also been considered as an alternative valorization route for SCGs, the extraction of oil is also required, but afterwards it is needed to develop a transesterification reaction using methanol and an acid catalyst, for obtaining the bio-glycerol, which will be used for the power production by its combustion. Even though this valorization route is not taking advantage of the biomolecules and polyphenols of the composition of SCGs, the profitability of this alternative, which revenues could amount to $ 1.461 M/y (I. K. Kookos, 2018).

In the field of biodiesel production (Thoppil and Zein, 2021) and despite the fact that it could be considered as a sustainable approach, since a renewable biofuel is produced from waste, it does not reach economic profitability (Thoppil and Zein, 2021). The main reason is the low selling price, which leads to a revenue value that is not sufficient to offset the cost associated with the purchase of equipment, utilities, operation, labor and other direct/indirect costs. A similar conclusion was raised from a TEA analysis (I. K. Kookos, 2018), which reported that the economic viability of SCGs for biodiesel production is difficult to achieve when considering production capacities below 42 t/year. The availability of SCGs could be considered for this purpose as a bottleneck in their valorization pathways. Likewise, the market price of the product and feedstock transportation could be listed as key aspects to ensure the profitability of biorefineries (Cristóbal et al., 2018).

6. Sustainability and circular economy with SCGs

The EU Circular Economy Action Plan aims to move from linear to circular production. To this end, the utilization of waste streams, and co-products, as resources for other facilities has turned out into a real possibility. The increasing population and high demand for coffee beverage has led to the production of large quantities of coffee waste, specifically SCGs, making them potential sources for developing biorefinery processes. But, seeking to gain an advantage in using these biomass-based renewable resources, it has been reported that such biorefineries should be developed on a large scale and centralized, meaning that they should be placed next to the coffee factory (Yeoh and Ng, 2022). Furthermore, SCGs biorefineries have been shown to be suitable for meeting biodiesel demand, with lower GHG emissions and similar production costs, making them a potential alternative to fossil fuels (Mayson and Williams, 2021; Yeoh and Ng, 2022). This conclusion is based on the fact that, although the energy potential of SCGs is lower compared to fossil fuels, their energy content is higher than that of other conventional biomasses used as biological resources for energy production. In fact, SCGs have a disadvantage, when assessing their environmental suitability, as there is a high concentration of nitrogen in their composition, leading to the production of NOx emissions (Mayson and Williams, 2021).

Furthermore, taking into account circular economy and cascading production approaches, it has been assessed that the use of SCG only for energy production is not cost-effective. The need for recovery of bioactive compounds, with many applications in pharmaceuticals, cosmetics and medical products, has become a necessity to ensure both environmental and economic suitability of SCGs biorefineries (Bijla et al., 2022; Massaya et al., 2019). To this end, the development of novel technologies for the extraction and recovery of bioactive compounds should be the main focus of future research, seeking to achieve new biorefinery approaches that can be categorized within the 12 Principles of Green Chemistry.

7. Conclusions

The availability of spent coffee grounds, and their suitability for use as a raw material in biorefinery processes, have made them potential candidates for obtaining bio-based and bioactive compounds. The different recovery routes that could be developed are beneficial for circular economy approaches and the preservation of fossil resources, as waste becomes a raw material input, also favoring the industrial symbiosis strategy: "your waste, my feed". This review has focused on the evaluation of SCG valorization alternatives to obtain lignin, lipids, polymers and phenolics, key products for the development of other production routes. From lignin, production of bioadhesives for wood, from lipids, bio-oil, bioplastics obtained from SCG polymers and phenolics, with a multitude of applications in cosmetics, food and pharmaceutical industries. The LCA and techno-economic literature reports available have also been evaluated, as environmental awareness and economic profitability are essential aspects to develop large-scale production strategies. To this end, it is hoped that this review article can be used as a reference for researchers, decision-makers and stakeholders to decide where to focus on SCG valorization under a biorefinery approach.

CRediT authorship contribution statement

A.A.: Methodology, Formal analysis, Investigation, Writing – original draft. **S.M.I.:** Investigation, Writing – original draft. **N.G.:** Formal analysis, Investigation. **G.F.:** Supervision, Writing – review & editing. **M.T.M:** Supervision, Writing – review & editing. **A.K.:** Conceptualization, Formal analysis, Supervision, Writing – review & editing.

Declaration of Competing Interest

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

Data Availability

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

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.indcrop.2023.117504.

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