TRENDS IN VALORISATION OF SPENT COFFEE GROUNDS: A REVIEW

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Abstract: *Coffee is nowadays one of the most appreciated and also consumed beverage. Its consumption presents many benefits for humans, but has also a great inconvenience because of the major quantities of waste resulted in the brewing process, which is not further exploited and reaches the environment with a significant negative impact. This residue is represented by spent coffee grounds (SCG). Due to its chemical composition, SCG represents a good source of some useful active compounds, which can be exploited for obtaining high-value products. This paper overviews aspects regarding the coffee world production, consumption and total quantities of residues resulted from coffee industry, pointing out the possibilities of SCG valorisation. Thus, it reviews the most important researches regarding SCG potential use in different domains, such as energy, agriculture, food and health, material construction and wastewaters treatment.*

Keywords: coffee, spent coffee grounds, biofuel, value-added product.

INTRODUCTION

Nowadays, coffee plants are cultivated in more than 70 countries. Coffee berries are obtained from two usually grown species: *Coffea canephora* (Robusta), the most widely cultivated variety, especially in Central Africa, Southeast Asia and Brazil and *Coffea arabica* (Arabica), cultivated in Latin America, Eastern Africa and Asia. About 60% of coffee beans worldwide production is arabica and the rest of 40% is Robusta (Batista et al., 2016).

Coffee berries are picked when ripped, then are processed and dried, becoming coffee beans. Roasting of coffee beans constitutes a very important stage in coffee obtaining process, because it influences physically and chemically properties of beans and determine their sensorial quality, especially flavour and colour. Roasted beans are grinded and brewed with near-boiling water, in order to obtain the coffee beverage.

Coffee is one of the most commercialized commodities worldwide, after petroleum. In the same time, it is the second most popular beverage, next after water (Mussatto et al., 2011b; Girotto et al., 2018). Thus, there is a great worldwide interest for its production and commercialization.

COFFEE WORLD PRODUCTION AND CONSUMPTION

According to latest statistical data provided by International Coffee Organization (ICO), total coffee production in 2018 was about 10.2 million tonnes, 6 million tonnes being represented by Arabicas and 4.2 million tonnes by Robustas. Leading coffee production country was Brazil, with an amount of about 3.7 million tonnes, meaning 36% of total. Compared to world production registered in 2015, in 2018 can be observed a major increase of about 10% (http://www.ico.org/prices/po-production.pdf).

Regarding coffee world consumption for a 2018/19 period, a total of about 10 million tonnes were consumed. Leading continent was Europe, with a total amount of about 3.3 million tonnes, followed by Asia & Oceania with 2.1 and North America with 1.9 million tonnes. European Union was the consumption's leader in Europe, with a total amount of 2.7 million tonnes, which represents 27% of total, followed by USA with 1.6 million tonnes. In 2018/19 it was registered an important increase of 6% in world coffee consumption, relative to 2016/17 period (http://www.ico.org/prices/new-consumption-table.pdf).

In 2017, the largest coffee-consuming country worldwide was Finland, with 10.35 kg per capita, followed by Netherlands with 9.58 kg per capita. USA was only 18th place, with an amount of 4.43 kg coffee consumed per capita (https://www.statista.com/chart/8602/top-coffee-drinking-nations/).

COFFEE RESIDUES

As world total coffee consumption increases every year, an important problem becomes to be the major quantities of organic waste resulted from coffee industry: by-products from beans processing and roasting (>50% of the fruit mass) and spent coffee grounds (SCG), from beverage preparation (Campos-Vega et al., 2015a). SCG is the solid residue which remains after roasted coffee beans are grinded and brewed, both in coffee shops chains and in industry, for obtaining instant coffee and represents the most abundant coffee by-product (45%) (Murthy and Naidu, 2012).

SCG chemical composition

Spent coffee grounds contain important quantities of organic compounds (phenolics, lipids, proteins, lignin, cellulose, hemicellulose and other polysaccharides), which determine its importance as a real source of valuable products (Kourmentza et al., 2018). Thus, Table 1 presents values reported by literature for most important components of spent coffee grounds (Massaya et al., 2019).

Table 1. Values reported by literature for components of spent coffee grounds (wt%) (Massaya et al., 2019)

Component	SCG
Hemicellulose	32-42
Cellulose	7-13
Lignin	0-26
Lipids	2-24
Proteins	10-18
Ash	1-2
Caffeine	0-0.4
Chlorogenic Acids	1-3
Moisture	50-60
Pectins	0
Total sugars	7-14
Total dietary fibre	21-59

Polysaccharides fraction covers about 50% of SCG total mass, of which about 50% are galactomannans, 25% arabinogalactans and 25% cellulose (Oosterveld et al., 2003). Presence of mannose, galactose, glucose and arabinose, polymerized into hemicellulose and cellulose (Ballesteros et al., 2014; Mussatto et al., 2011a) and high content of galactomannans (Simões et al., 2013) are highlighted in SCG, lignin being also present in a significant amount (Pujol et al., 2013). Dietary fiber represent about 43% of total SCG dry weight (42% insoluble, 1% soluble fibre respectively), which are approved to be used as raw material to develop functional foods. The fibre from SCG includes, among others, resistant starch, oligosaccharides and manno-oligosaccharides (Campos-Vega et al., 2015b; Vázquez-Sánchez et al., 2018; Tian et al., 2017).

There are also present some bioactive secondary metabolites, such as diterpenes, sterols, chlorogenic acids, flavonoids and caffeine (Massaya et al., 2019). Caffeine is a major biological active compound of spent coffee grounds (Cruz et al., 2012b).

SCG amounts

Brewing of 1 kg of soluble coffee generates 2 kg of wet SCG (Mussatto et al., 2011b). At the same time, 1 kg of green coffee beans produces 0.65 kg of dried SCG (Kookos, 2018). Thus, usually accepted mass ratio of generated SCG to produced coffee beans is 0.65 and the mass ratio of SCG to roasted beans is 0.91 (Eshetu, M. 2018). As a consequence, a high amount of more than 6 million tonnes of dried SCG is generated worldwide each year (Hardgrove and Livesley, 2016).

SCG impact on the environment

Kamil et al. (2019) achieved an estimation of total amount of wet SCG produced per year and its impact on the environment, by the CO_2 emissions from landfills point of view. Thus, considering the total number of coffee cups daily drunk of 1.4 billion and every cup waste of 30 g (15 g ground coffee beans and 15 g water), total amount of SCG landfilled can be estimated at 15.33 million tonnes per year. Considering that one tone of SCG produces 682 kg CO_2, annually there are generated 28.644 million tonnes of CO_2 from landfilling, a quantity equivalent with that produced by 10.6 million liters of burned diesel fuel.

Even if SCG contain polyphenols and tannins, which are compounds with established ecotoxicity, this residue is either disposed of in sanitary landfills or incinerated (Mata et al., 2018). This practice is dangerous both for humans and ecosystems, especially the soil one (Mussatto et al., 2011b; Cruz et al., 2012b). Thus, an interesting method for decreasing SCG toxicity by elimination the phenolic compounds from its structure was proposed, meaning a biological treatment with fungal strains from genus *Penicillium*, *Neurospora* and *Mucor* applied before releasing this residue to the environment (Machado, 2009).

Because of its content in caffeine, fatty acids and metals, SCG can be toxic for aquatic organisms on a long-term exposure (Fernandes et al., 2017). Its high humidity, which may exceed 65% by mass, acidic pH and listed toxic constituents, are responsible for high environment damage (Leifa et al., 2000; Páscoa et al., 2013). Due its organic nature, SCG may form leachate, a major pollutant of waters, when the final destination are waste landfills (Renou et al., 2008; Foo and Hameed, 2009).

Interest in SCG exploitation

Valorisation of spent coffee grounds presents nowadays a major interest, both for researches and industry sector. Some reasons conduct to these tendencies: the large number of organic compounds contained, such as phenolics, lipids, carbohydrates, proteins, which constitutes potential functional food ingredients (Campos-Vega et al., 2015a); some bioactive secondary metabolites, such as diterpenes, sterols, caffeine, because of their physiological effects (Bonita et al., 2007); SCG physical structure enables fast removal of these substances, by using the suitable solvent; SCG is a residue available in large amounts, because of the immense worldwide coffee consuming (Peshev et al., 2018).

To reduce the negative impact of SCG disposed of in landfills, there is a real interest for its utilization as a significant substrate for obtaining some useful value-added energy and non-energy by-products, as functional additives or antioxidants (Zuorro and Lavecchia, 2012; Esquivel and Jiménez, 2012; Li et al. 2014; Campos-Vega et al. 2015a; Kourmentza et al., 2018; Kovalcik et al., 2018; Karmee, 2018; Janissen and Huynh, 2018; Atabani et al., 2019; Iriondo-DeHond et al., 2019). Many important food companies take seriously into account utilization of coffee residue as a source for obtaining chemicals or energy (Kookos, 2018).

POSSIBILITIES OF SPENT COFFEE GROUNDS VALORIZATION

Till present, several important applications have been experimental developed for spent coffee grounds valorisation, especially as biofuels, composts and animal feed, a functional ingredient for food products with real health benefits, bio-composite materials, decontaminants of wastewaters.

Energy

A very remarkable utilization of SCG is energy production. Due to its high organic content, SCG is very attractive for being use as biomass. It represents an alternative source of energy and biofuels production (Li et al., 2014), namely fuel pellets, biochar, bio-oil, biogas, biodiesel and bioethanol, besides the other value-added products (Kondamudi et al., 2008; Atabani et al., 2019).

Direct combustion

SCG can be directly burned, in order to produce heat energy. Calorific value measurements demonstrated that coffee residues possess high energy content, proving a higher heating value than woody biomass (Bok et al., 2012). SCG was tested as combustible in some various ways, by direct boiler burning both alone, as pellets or woodchip logs and also mixed with other biomasses, such as sawdust pellets or pine sawdust pellets (Limousy et al., 2013; Jeguirim et al., 2014; Limousy et al., 2015; Jeguirim et al., 2016; Kang et al., 2017). Results of these combustion studies were summarized by McNutt and He (2019). Thus, boiler-style combustion of SCG produced higher heating value than wood pellets, at the same water content, but it is a need to reduce emissions of O_2, CO and NO_x in flue gas. When burning SCG pellets, heating value level was closed to standard agro-industrial pellets, but slightly lower. By combining 20% SCG and 80% pine sawdust and burning as woodchip logs, combustion yields were comparable to regular wood logs, while pelletization of SCG and pine sawdust conducted to higher particle emissions in flue gas than pellets obtained with pure sawdust.

Biochar, bio-oil and biogas

SCG pyrolisis leads to obtaining of biochar, bio-oil and biogas (Ktori et al., 2018), depending on technique and reaction parameters (slow or fast pyrolisis). Biochar is suitable for being used as a solid fuel in industry because of its high calorific value (Tsai et al., 2012), as a fertilizer in arid fields (Ktori et al., 2018) or for activated carbon (Safarik et al., 2012). Bio-oil was studied as a precursor for biodiesel production (Li et al., 2014) or as a chemical, due to its insecticidal and bactericidal properties (Bedmutha et al., 2011). Biogas can be used as a power source in engines, turbines and boilers (Ktori et al., 2018).

Vardon et al. (2013) proposed a complete utilization of SCG. Thus, coffee lipids extracted from SCG were subjected to transesterification, for producing biodiesel, while biochar and bio-oil were obtained from defatted SCG, through slow pyrolisis (Figure 1). Generated biochar showed a calorific value of about 31 MJ/kg.

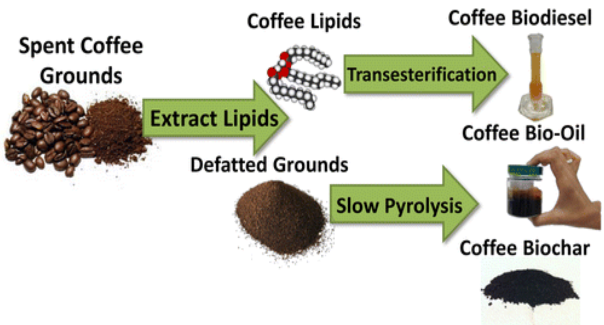

Figure 1. Complete utilization of spent coffee grounds to produce biodiesel, bio-oil and biochar (image from Vardon et al., 2013)

Regarding pyrolisis process applied for bio-oil production, Bok et al. (2012) obtained a maximum yield of bio-oil (55%) at 550°C, Li et al. (2014) recorded highest yield (66%) at 630°C, while Ktori et al. (2018) achieved maximum amount (36%) at 540°C. Because of SCG high moisture content, of approximately 50-60%, a pre-drying phase need to be applied prior to pyrolisis (Girotto et al., 2018).

Biodiesel

SCG chemical composition is characterized by large amounts of oil, with values around 10-15% (Al-Hamamre et al., 2012). Thus, it can be exploit with notable results in producing biodiesel. Nowadays, there are many researches which demonstrate its potential in this sense (Kondamudi et al., 2008; Vardon et al., 2013; Girotto et al., 2018; Kookos et al., 2018; Mata et al., 2018; Atabani et al., 2019; Kamil et al., 2019; Masaya et al., 2019; Tongcumpou et al., 2019). One possibility for biodiesel production process is the method which implies the extraction of lipids from spent coffee grounds first, by using solvents, then the transesterification reaction in two steps: acid esterification, followed by alkaline transesterification (Caetano et al., 2014). There is one more possibility for obtaining biodiesel, when transesterification is performed directly or *in situ* on SCG biomass, without prior lipids extraction, in only one step, by acid transesterification (Park et al., 2016; Liu et al., 2017). Conversion in two steps is suitable when coffee oil is highly acid (Al-Hamamre et al., 2012; Caetano et al., 2014).

However, some studies demonstrated that it is difficult to support an economically feasible process for biodiesel production from SCG (Kokoos, 2018). Kamil et al. (2019) concluded that profitability appears only when the production capacity of a biodiesel factory is in order of 10.000 tonnes SCG/year, smaller capacities demanding high initial investment per unit produced.

Bioethanol

Because of its high content in cellulose and hemicellulose, SCG represents a potential source for biotechnological production of bioethanol, further used as a fuel. This procedure needs an initial conversion of hemicelluloses and partially cellulose into fermentable sugars, by hydrolysis (Obruca et al., 2015). For converting SCG sugars to ethanol, a fermentation needs to be applied, by using *Saccharomyces cerevisiae* yeast (Mussatto et al., 2012). Bioethanol production proved efficiency, but due to fatty acids and triglycerides content from SCG, the hydrolysis process of polysaccharides was restricted. Thus, it is more advantageous obtaining biodiesel than conversion to bioethanol (Kwon et al., 2013).

Agriculture

Due to its content in polysaccharides and minerals, SCG has been studied as potential organic fertiliser. In order to stimulate plants growing, by improving their mineral nutrition, some researches regarding utilization of SCG as organic amendments have been realised, its application being either as such or as compost (both fresh and initially composted or composted directly on the soil) (Murthy and Naidu, 2012; Cruz et al., 2014b; Cruz et al., 2015; Campos-Vega et al., 2015a; Cervera-Mata et al., 2019). Different studies noticed that low amounts of SCG increased soil mineral content, bioactive compounds and antioxidant activity (Cruz et al., 2012a; Cruz et al., 2014a). However, because of its high C/N ratio, acidity and total content of phenols and caffeine, its direct application on soils might be harmful, both for plants and soil microorganisms (Hardgrove and Livesley, 2016). Thus, Gomes et al. (2013) reported that composting of SCG, before its application, may reduce toxicity to plants, due to decreasing of total phytotoxic constituents' amount. Added at the beginning of composting, SCG supplies compounds such as mannose, galactose, arabinose, glucose, proteins, calcium and phosphorus, enhancing degradation of waste, by aiding microbial activity and enzymes generation (Murthy and Naidu, 2012; Wu, 2014). SCG can also be considered a natural herbicide, being able to eliminate weed seeds in composting (Low et al., 2015). Zhang and Sun (2017) proposed the combination, in different proportions, of cow dung and SCG, as amendments in two-stage co-composting of green waste. The most qualitative, mature and rapid compost was obtained when 20% cow dung and 45% SCG were combined, in only 21 days. As well, nonphytotoxic compost products were caused, improving compost properties and nutrient content.

Because of SCG physical properties, such as small size particles and high surface area, this residue might help water management in acidic soils, which possess low cohesion and are sensitive to erosion (Kasongo et al., 2013). Turek et al. (2019) examined physical-hydraulic soil properties modifications, in case of SCG administration. It was observed an improvement of soil water retention and aeration, its application proving efficiency in soils with poor retention capacity. However, larger than 10% SCG contents are not recommended to be applied in sandy loam soils.

Considering its high nutritional content, SCG was investigated also as a supplement in animal feed, for pigs, chickens, rabbits and ruminants (Claude, 1979; Givens and Barber, 1986). However, because of its high content in lignin (~25%), tannins and caffeine, SCG proved a limited utilization in this regard (Cruz, 1983). Fuller (2004) concluded that a higher concentration than 2.5% of the last two compounds make SCG inedible, because they produce a diminution of protein digestibility. Seo et al. (2015) evaluated the effect of SCG inclusion, in concentration of up to 100 g/kg in the concentrate of ruminants, as a functional feed ingredient, on the yield and quality of milk. Results showed that milk production and composition were improved, with no side effects regarding feeding behaviour or apparent digestibility at ewes.

Food and health

In addition to SCG application in animal feed, currently there are studies which support the usage of this by-product as a nutraceutical or food functional additive, improving both nutrition and health (del Castillo et al., 2014).

SCG constitutes a valuable source of phenolic compounds and melanoidins, which can be further included as functional ingredients in human diet (Borelli et al., 2004; Mussatto et al., 2011c; Zuorro and Lavecchia, 2012; Xu et al., 2015). Thus, it was demonstrated the SCG antioxidant, antihypertensive and antimicrobial activities in intestine microbiota (Rufián-Henares and Morales, 2007; Campos-Vega et al., 2015b), with an important role in preventing diseases related to free radicals (Wang et al., 2011). Phytochemicals from SCG can be digested, absorbed and fermented in colon, exerting healthy effects by influencing the metabolic activity of the microbiota (López-Barrera et al., 2016).

SCG phenolic extracts can also be used as anti-inflammatory additives (Lopez-Barrera et al., 2016) and dermatological anti-melanogenesis agents (Huang et al., 2016). Regarding extracting of natural antioxidants and caffeine from SCG, there were proposed different methods, such as solid-liquid extraction using aqueous alcohol solution (methanol, ethanol and isopropanol) (Mata et al., 2018) or by pressurized liquid extraction (PLE) method with water and ethanol (Shang et al., 2017).

Dietary fibre can be fermented by colonic microbiota, releasing short chain fatty acids, with anti-inflammatory properties. Thereby, it can protect the onset or progression of inflammatory diseases, such as inflammatory bowel, colon cancer and rheumatoid arthritis (López-Barrera et al., 2016; García-Gutiérrez et al., 2017; Hernández-Arriaga et al., 2017). Due to its rich content in dietary fibre, SCG might be a good source of these compounds in the food industry (Martinez-Saez et al., 2017a). Vázquez-Sánchez et al. (2018) evaluated antioxidant dietary fibre extracted from SCG as a functional food ingredient, by their adding in biscuits. Antioxidant capacity after in vitro digestion, bioaccessability of phenolics and aminoacids and also total dietary fibre content were improved. Same study seemed to indicate that anti-diabetic compounds, with inhibitory effects on α-glucosidase activity, might be released in small intestine during these enriched biscuits digestion, making them capable to regulate sugar metabolism. Thus, dietary fibre extract from SCG might help production of foods diabetics friendly (Martinez-Saez et al., 2017b).

Other potential use of SCG is obtaining of valuable bio-sugars, such as oligosaccharides, manno-oligosaccharides and mannose, after its delignification and defatting, process which proved large-scale feasibility (Nguyen et al., 2019).

Peshev et al. (2018) revealed utilization of SCG for obtaining water extracts with sufficiently high caffeine concentration. Appliance of nanofiltration to these extracts, by using a suitable membrane, conducted to valuable products, as permeate and retentate fractions. Permeate can be further used for soft and energy drinks, while retentate for coffee drink or as functional food ingredient.

Materials construction

Because SCG contain high amounts of cellulose and hemicellulose, it has the potential to be part of bio-composite materials, being applied as filler and additive for polymer composites (Baek et al., 2013; García-García et al., 2015; Wu et al., 2016; Moustafa et al., 2017a; Moustafa et al., 2017b).

Vilela et al. (2016) successfully obtained a material which is rich in insoluble fibres by using the polysaccharide-rich fraction from SCG, obtained by alkaline hydrogen peroxide treatment of the coffee residue. Ballesteros et al. (2018) enhanced light barrier of carboxymethyl cellulose films for food packaging, by incorporating polysaccharide-rich extracts from SCG. These films proved almost similar properties as those noticed in literature, but their usage must be limited at applications where aesthetics is not important.

SCG was also studied as a subgrade material for construction industry, mixed with another adequate waste materials with proved compression resistance (e.g. recycled glass) (Arulrajah et al., 2017).

Wastewaters treatment

Considering its high lignocellulosic content, SCG proved to be an efficient adsorbent (Ballesteros et al., 2014), more exactly a precursor of activated carbon used for removing of basic dyes from dye-contaminated industrial waters (Pagalan et al., 2019). Due its unique microporous structure with a high surface area of about 300–1000 m^2/g and metal-chelating activity, it proved efficiency in adsorption of phenols and also heavy metals (Castro et al., 2011; Zuorro and Lavecchia, 2012; Davila-Guzman et al., 2013; Liu et al., 2015).

Besides its proven efficiency in wastewaters treatment (Franca and Oliveira, 2009), activated carbon from SCG can also be used as a removal of the organic matter from landfills leachate (Chávez et al., 2019).

Complete utilization of wet SCG

Tongcumpou et al. (2019) proposed a complete method for wet SCG utilization, by its continuous subjection to three processes: the extraction of antioxidants, direct biodiesel production from the remanent SCG and bio-char obtaining from defatted SCG. By antioxidants extraction with methanol, free fatty acids and water were also extracted, making possible direct *in situ* transesterification for obtaining biodiesel, with no previous drying and conditioning of SCG waste. In third process, defatted SCG was transformed in bio-char briquette, by a slow pyrolisis. Applying this chain of processes for SCG valorisation, energy and time consumption was reduced. In the same time, the energy content in bio-char briquette proved to be sufficient to support all processes.

CONCLUSIONS

Although many current studies indicate, as a result of extensive researches, some achievable and environmentally sustainable methods regarding valorisation of spent coffee grounds, this residue is nowadays still disposed of in landfills or directly burned. The main reason is that the majority studies have been realised at a laboratory scale, with no proved economic profitability and reliability at industry level. Thus, it is imperative that future researches to be accomplished together with industry sector, in order to demonstrate their viability for large scale implementation and economic feasibility.

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