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Emerging potential of spent coffee ground valorization for fuel pellet production in a biorefinery

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Abstract
The global market for fuel pellets (FPs) has been steadily growing because of a shift to coal substitutes. However, sustainability and the availability of biomass are the main issues. Various kinds of bio-wastes can be valorized through cutting-edge technologies. In the coffee industry, a valuable organic waste called spent coffee grounds (SCGs) is generated in bulk. SCG can be divided into two components, namely spent coffee ground oil and defatted spent coffee grounds (DSCG). SCG and DSCG can be used to produce FPs with excellent higher heating values. This review highlights that burning FPs composed of 100% SCG is not feasible due to the high emission of NO_x. Moreover, the combustion is accompanied by a rapid temperature drop due to incomplete combustion which leads to lower boiler combustion efficiencies and increased carbon monoxide emissions. This was because of the low pellet strength and bulk density of the FP. Mixing SCG with

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other biomass offers improved boiler efficiency and emissions. Some of the reported optimized FPs include 75% SCG + 20% coffee silverskin, 30% SCG + 70% pine sawdust, 90% SCG + 10% crude glycerol, 32% SCG + 23% coal fines + 11% sawdust + 18% mielie husks + 10% waste paper + 6% paper pulp, and 50% SCG + 50% pine sawdust. This review noted the absence of combustion and emissions analyses of DSCG and the need for their future assessment. Valorization of DSCG offers a good pathway to improve the economics of an SCG-based biorefinery where the extracted SCGO can be valorized in other applications. The combustion and emissions of DSCG were not previously reported in detail. Therefore, future investigation of DSCG in boilers is essential to assess the potential of this industry and improve its economics.

Graphical abstract



Keywords Spent coffee grounds · Defatted spent coffee grounds · Fuel pellets · Calorific value · Valorization · Biorefinery · Circular bioeconomy · United Nations Sustainable Development Goals.

Abbreviations

ABE	Average bias error
b	Combustible constituent in the residue
BET	Brunauer–Emmett–Teller
BE%	Boiler efficiency
BD	Bulk density
C	Carbon
CB	Coffee bean
CBE	Circular bioeconomy
CGA	Chlorogenic acid
CH	Coffee husk
CO	Carbon monoxide
CO_2	Carbon dioxide
CP	Coffee pulp
C_{pmd}	Specific heats of dry flue gases

C_{pmH2O}	Specific heats of water vapor
C_{pw}	Water calorific capacity
CS	Coffee silverskin
CV	Calorific value
C_r	Carbon content of the residue passing through the grate
DSCG	Defatted spent coffee grounds
DRDCB	Defatted roasted defective coffee beans
ED	Energy density
EU	European Union
FC	Fixed carbon
FPs	Fuel pellets
GCV	Gross calorific value
GHGs	Greenhouse gases
H	Hydrogen
H_2O	Water
H_2S	Hydrogen sulfide
HHV	Higher heating value
ICTs	Information and Communication Technologies
IWB	Integrated waste biorefineries
L	Lignin
LHV	Lower heating value
CH_4	Methane
N	Nitrogen
NH_3	Ammonia
NHV	Net heating value
NO	Nitric oxides
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides
NPV	Net present value
O	Oxygen
OSHA	Occupational Safety and Health Agency
P_c	Calorific mass flow
PM	Particulate matter
P_n	Nominal useful power heat of the boiler
$q\%$	Combustion efficiency
q_a	Thermal heat loses in the flue gas
q_b	Chemical heat loses in the flue gas
Q_{comb}	Biomass mass flow
q_r	Heat losses due to combustible constituents in the residue
Q_w	Water mass flow
R	Residue mass passing through the grate
RDCB	Roasted defective coffee beans
RHI	Renewable heat incentives
ROI	Return on Investment
S	Sulfur
S_{BET}	BET surface area
SCG	Spent coffee grounds
SCGO	Spent coffee ground oil
SDGs	Sustainable Development Goals

SEM	Scanning electron microscopy
SFR	Sustainable Fuel Register
SO_2	Sulfur dioxide
SO_x	Sulfur oxides
T_a	Air temperature
T_g	Exhaust gas temperature
T_{in}	Inlet temperature of water from the boiler heat exchanger (K)
T_{out}	Outlet temperature of water from the boiler heat exchanger (K)
VM	Volatile matter
VOC_S	Volatile organic compounds (VOC_S)

1 Overview of global fuel pellets (FPs) production and consumption and current challenges

Biomass is the 4th largest energy source around the world, providing around 10% of the energy used worldwide. Rising environmental concerns have led the efforts to reduce the emission of greenhouse gases (GHGs) such as carbon dioxide (CO_2), methane (CH_4), nitrogen oxides (NO_x), and sulfur oxides (SO_x) and bolstered the use of biomass as a substitute for fossil fuels (Chawla et al., 2018; Kim et al., 2014). Unlike other biofuel feedstocks, biomass residues or wastes do not compete with food supplies and are widely available at reasonable prices (Abbas et al., 2020; Ahmed et al., 2020). Direct combustion is a thermochemical technique used to convert biomass to heat and work (Shahbaz et al., 2020; Sheng & Azevdo, 2005). Densified solids known as fuel pellets (FPs) are sustainable and good-quality, biomass-based materials that are granular and come in shapes of spheres or cylinders with sizes of around a few centimeters (Smaga, 2016). FPs are consumed for electricity and power generation as well as for residential and district heating (Thrän et al., 2017). In 2018, 52% of FP consumption was in the residential sector and 48% of FP consumption was in the industrial sector (European Pellet Council (EPC), 2019). There are different FPs available that are produced from different sources such as wood, agro, and bio-wastes by different processing conditions including torrefaction (Thrän et al., 2017). FPs and their components are not hazardous because they consist of organic matter and do not contain any harmful chemicals. Furthermore, they serve as an organic matter stream to dispose of biogenic waste from the environment. FPs have calorific values comparable to those of wood, low-sulfur content, and humidity, and produce small amounts of ash (Smaga, 2016). Combustion of FPs does not affect the overall balance of CO_2 in the atmosphere because FPs are obtained from renewable resources.

Driven by rising global and environmental demand, wood FPs production increased over the past 15 years from 6–7 million tonnes in 2006 to 14.3 million tonnes in 2010 (Thrän et al., 2017). The increase in the worldwide FPs production continued in 2013 with 24.93 million tonnes produced and reached 55.70 million tonnes in 2018. This is an increase of 14.13 and 36.67% compared to 2017 and 2016, respectively. With a production volume of 20.25 million tonnes in 2018, China was the largest producer of the FPs globally (36%) followed by EU 28 (16.87 million tonnes, 30%) and North America (10.90 million tonnes, 20%) (European Pellet Council (EPC), 2019). In 2018, the largest FP consumer was EU 28 with 26.05 million tonnes (50%) followed by China with 17.6 million tonnes (33%) (European Pellet Council (EPC), 2019). Asia is continuing to grow its production and consumption, providing a driving force for pellet market development alongside

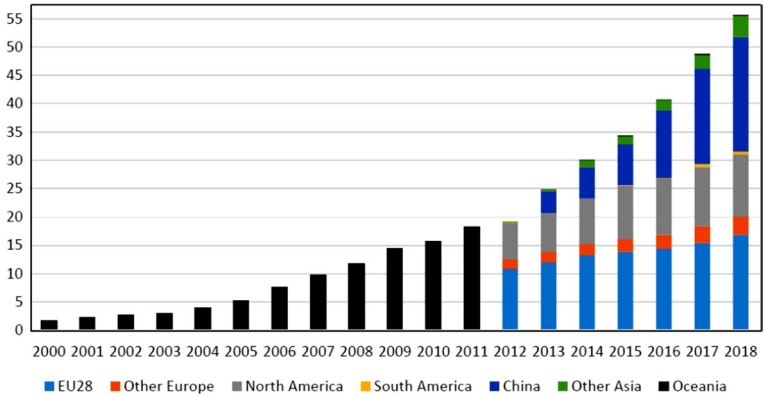


Fig. 1 Evolution of global pellet production (million tonnes) between 2000 and 2018 (European Pellet Council (EPC), 2019)

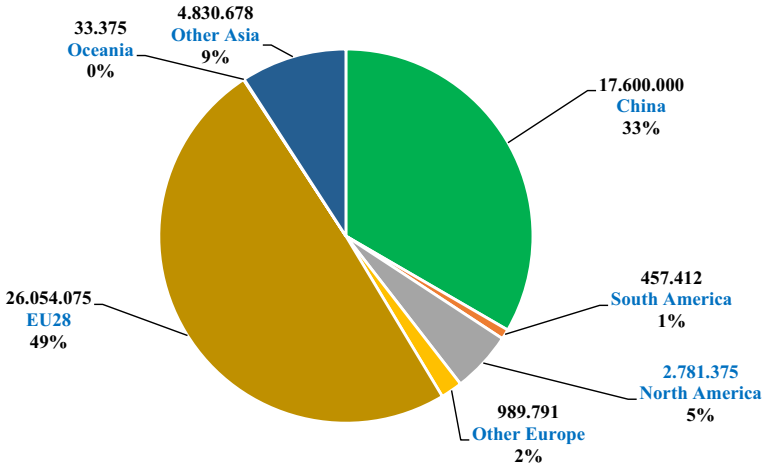


Fig. 2 Global wood pellet consumption in 2018 (tonnes and %) (European Pellet Council (EPC), 2019)

Europe (European Pellet Council (EPC), 2018). South Korea and Japan will continue to be the largest consumers in Asia. In 2016, they imported 2 million tonnes of FPs from Asia (Vietnam, Malaysia, Indonesia, Thailand), Canada, and Russia (Gauthier et al., 2017). Figure 1 presents global pellet production between 2000 and 2018, and Fig. 2 depicts global wood pellet consumption in 2018.

The global market for FPs is steadily growing due to the increasing demand to replace coal in larger power plants. This growth has promoted the industry to look for raw materials like wastes collected from agriculture, forestry, or a combination of both (Miranda et al., 2015). Since 2011, this market has been growing 14% a year on average (Thrän et al., 2017). One of the strongest reasons for this is the harmful emission released when burning coal. In 2020, China set a goal of using 30 million tonnes of non-hazardous FPs from biomass to replace 15 million tonnes of coal (Thrän et al., 2017). However, such plans are highly affected by sustainability and biomass availability to meet electricity and heating

demand. Additionally, the uncertainty of bioenergy support from many countries remains a serious challenge. On the other hand, various kinds of bio-waste, such as those from the food industry, can be valorized through cutting-edge recycling technologies into agro-pellets. This will significantly reduce the amount of waste to be returned to the environment and solve the problem of organic waste disposal. The United Nations Sustainable Development Goals (SDGs) set 17 goals to transform our world into a more sustainable world. Waste recycling into biofuels and value-added products is under these goals to help transform our world into a greener world with affordable and cleaner energy, sustainable cities and communities, responsible production and consumption of resources and fighting climate change (United Nation (UN), 2021). Such an approach can be realized through circular bioeconomy (CBE) in which integrated waste biorefineries (IWB) are being developed to turn these wastes into useful products and clean energy resources (Leong et al., 2021). Spent coffee grounds (SCGs) are the primary by-product from coffee production that is currently treated as worthless waste and have yet to be explored for recycling (Scully et al., 2016). Additionally, SCG pellets have gained attention because of their viability as an alternative fuel (best lower calorific value = 21.08 MJ/kg) (Nosek et al., 2020). Using SCG in the form of briquette as an energy resource can contribute to the saving of 10 to 25% of the post-exploitation waste during the infusion process (Ciesielczuk et al., 2015). The available literature about the production of FPs from different raw biomass residues is abundant (Miranda et al., 2015). However, no previous work has highlighted the potential of the SCG for FPs production. This review is the first review paper to highlight the potential of SCGs as promising raw materials for FPs production. The article gathers some important information that discusses the potential of the SCG for FPs production including the characteristics, potential, feasibility, advantages, and current business opportunities of FPs from SCG.

2 FPs quality standards

The quality of the produced FPs is assessed based on their physical and chemical properties. Density, abrasion, calorific value, and strength (durability) are some of the most important physical properties of the FPs. In addition, pellet shape, size, durability, and density and some chemical qualities including heavy metal type, elemental composition, and energy content can be used to assess the FPs quality. High strength and low abrasion indicate a minimum dust amount during pellet handling. This characteristic affects the combustion quality, emissions, and effect on human health which includes respiratory diseases (Miranda et al., 2015; Narra et al., 2012). Therefore, they play a crucial role in determining whether the FPs will be suitable for the global market or not.

The quality of the manufactured FPs can be controlled and monitored using different international standards and to assure the proper use of FPs in combustion equipment (Miranda et al., 2015). In Europe, some FP standards such as DIN 51,731 (Germany), ÖNORM M 7135 (Austria), and DIN EN 14,961 have been set to regulate the quality of FPs in the market (European Commission (EC), 2009; Peksa-Blanchard et al., 2007;). In the USA, the Pellet Fuels Institute (PFI) provided a quality control standard (Thrän et al., 2017). Additionally, France set two standards (Agro + and Agro) for agro-pellets (Narra et al., 2012). Table S1 (supplementary section) summarizes these standards. In recent years, EN ISO 17225 replaced the former EN14961 specification (European Commission (EC), 2009; Peksa-Blanchard et al., 2007;). Under this standard, EN ISO 17225-2 was

applied to FPs produced from woody biomass (groups: A1, A2, and B used for household and commercial applications and I1, I2, and I3 for industrial applications) while EN ISO 17225-6 was used for FPs produced from non-woody biomass (herbaceous and biomass of fruit classified as A and B) (Miranda et al., 2015). Tables S2 and S3 (supplementary section) present EN ISO 17225-2 and EN ISO 17225-6 standards for woody biomass and non-woody biomass. FPs quality can be optimized based on the combination of its physical and chemical characteristics (Narra et al., 2012).

3 Overview of the coffee industry and spent coffee grounds (SCGs) as a valuable food waste

Coffee is grown in around 80 countries with Brazil, Vietnam and Columbia being the primary producers and exporters of coffee beans (CB) (Al-Hamamre et al., 2012; Karmee, 2018). Coffee is one of the world's most popular beverages and the second largest traded commodity after petroleum (Campos-Vega et al., 2015). In 2018–2019, global coffee production and consumption were estimated to be 168.87 and 164.82 million bags. This represents an increase of 3.7 and 2.1% compared to 2017–2018. In 2019–2020, global coffee consumption has been projected to increase to 166.06 million bags, which represents an increase of 0.75% compared to 2018–2019. However, exports in the first six months of the year 2019–2020 (October 2019–March 2020) decreased by 3.9% to 61.96 million bags compared to 64.5 million bags in the same period in 2018–19 due to the COVID-19 pandemic (International Coffee Organization (ICO), 2020). Europe has the highest coffee consumption per capita in the world (Caetano et al., 2017). This means that people in Europe drink more coffee on average.

However, this industry generates a huge amount of organic waste such as coffee pulp (CP), coffee silverskin (CS), coffee husk (CH), defective beans, SCG, and roasted defective coffee beans (RDCB) (Atabani, Al-Muhtaseb, et al., 2019; Ballesteros et al., 2014; Efthymiopoulos et al., 2018). SCG represents the most important and valuable waste generated by this industry (Mussatto et al., 2011). It is a solid residue of fine particles with a dark-brownish color and high moisture content, organic content and acidity that is produced during the brewing process of roasted CB with hot water or steam (Barbero & Fiore, 2015). SCG is a highly distributed source, being a kind of waste that can be produced by end users, public bars, hotels, restaurants, catering operators, and vending machine companies (Bottani et al., 2019). SCG and RDCB amount to approximately 40% and 20% of the total CB mass (Efthymiopoulos et al., 2018; Koh & Hong, 2017). The average annual production of SCG is 8 million tonnes (Efthymiopoulos et al., 2018). On average, 650 kg of SCG is generated from 1 ton of CB. It is believed that 93% of SCG is directly sent to landfills without proper recycling. In the best cases, SCG has been only reutilized for fertilizer or compost production. Direct dumping of SCG to the landfills represents an environmental threat as the emission of GHGs such as CO_2 and CH_4 may contribute to global warming. Furthermore, SCG as an organic waste demands large quantities of oxygen to degrade. Additionally, dumping has an economic cost that is associated with the collection, transportation, and treatment of such waste. There are great political and social pressures to reduce the pollution arising from the disposal of organic waste without proper recycling (Ballesteros et al., 2014; Mussatto et al., 2011). Considering the generation of 103,000 tons of SCG in 2014 in South Korea alone, the total amount of energy (heating value) of unused SCG is more than 940 TJ (46.3 Gtoe, ton of oil equivalent) per year (Kang et al., 2017).

The large amount of energy available in SCG makes the utilization of this waste stream lucrative.

SCG is a valuable waste that contains many organic components (i.e., lipids and wax, amino acids, lignin, carbohydrates, polyphenols such as chlorogenic acid (CGA), residual caffeine, melanoidins, tannins, diterpenes, xanthines, vitamin precursors, minerals, ashes, and polysaccharides) that have good physical characteristics, but may be toxic at high concentration and are not yet exploited (Fig. 3) (Barbero & Fiore, 2015). Most of the recent studies concluded that SCG should be split into two main components: the lipid/extracts from SCG called spent coffee ground oil (SCGO) and the exhausted coffee grounds that remain after the extraction process known as defatted spent coffee grounds (DSCG) (Atabani, Al-Muhtaseb, et al., 2019; Go et al., 2016). The average oil content of SCG has been found to be between 10 and 20 wt% (Atabani, Al-Muhtaseb, et al., 2019). This strategy will improve the economics of the industry as SCGO and DSCG can be utilized in many

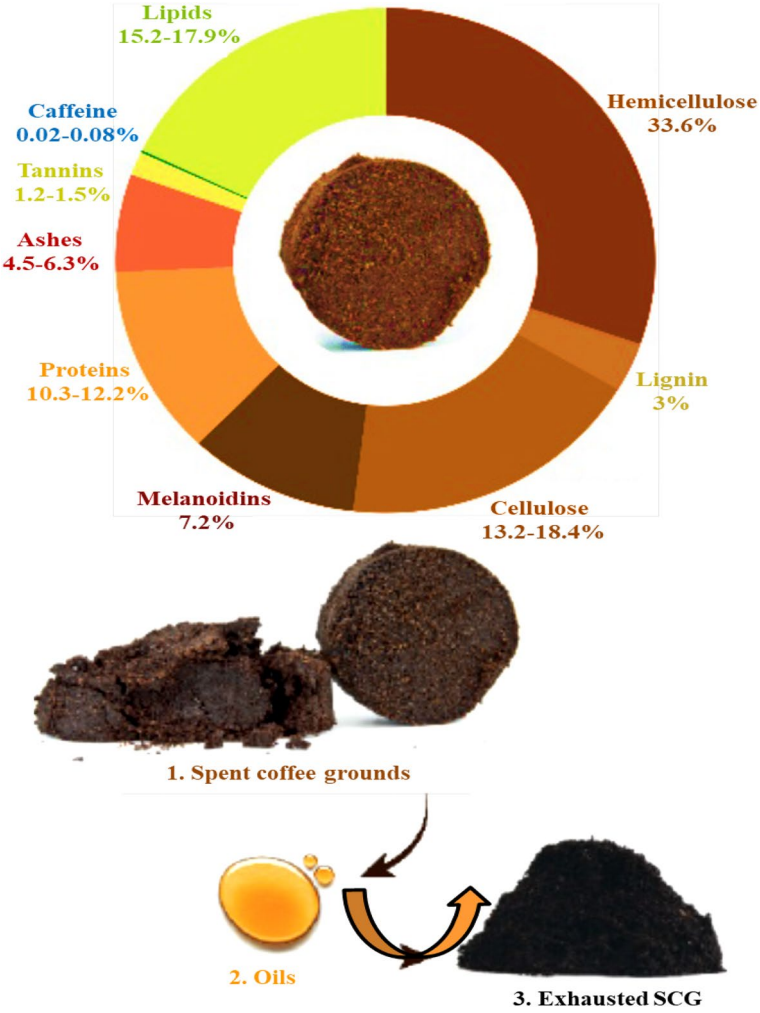


Fig. 3 Composition of anhydrous SCG and images of SCG, SCGO, and DSCG (Barbero & Fiore, 2015)

applications (Atabani, Al-Muhtaseb, et al., 2019). Moreover, SCGO has been reported to be a very stable oil (does not decompose quickly) as it has a high content of antioxidants (Al-Hamamre et al., 2012).

This makes SCG an excellent raw material for IB aiming to generate biofuels and value-added products to enhance the CBE of many nations. The body of literature is packed with many articles highlighting the potential of SCG recycling. Some valorization options include transesterification to produce biodiesel (Al-Hamamre et al., 2012; Atabani, Shobana, et al., 2019), anaerobic digestion to produce biogas (Girotto et al., 2017; Kim et al., 2017a, 2017b), and fermentation to produce bioethanol (Kwon et al., 2013; Rocha et al., 2014). SCG have also been classified as excellent raw materials used for the production of adsorbents (Shen & Gondal, 2017), sugar (Mussatto et al., 2011), cream-scrubs, detergents, and soaps (health-promoting/care applications) (Barbero & Fiore, 2015), biopolymers (Obruca et al., 2014), protein (Somnuk et al., 2017), antioxidants (Boyadzhiev et al., 2010), compost (Cruz et al., 2015), vermicomposting (Barbero & Fiore, 2015), activated carbon (Kante et al., 2012), textiles (Koh & Hong, 2017), and many other products (Atabani, Al-Muhtaseb, et al., 2019). These recycling opportunities raise the potential of the SCG biorefinery to produce both sustainable biofuels and value-added products (Fig. S1) (Barbero & Fiore, 2015).

4 Characterization and potential of SCG as FPs

Valorization of SCG into FPs is one of the most promising recycling options. Compared to bioethanol, biodiesel, pyrolysis oil, and biogas, FPs have the greatest obtainable energy per kg from the direct combustion of SCG FPs (Girotto et al., 2018).

As stated in Sect. 2, FPs characteristics significantly influence the design and operation of biomass combustion systems. These characteristics include pellet size, strength and durability, moisture content, calorific value, energy density (ED), bulk density (BD), morphology, and porosity (Jeguirim et al., 2014; Sheng & Azevedo, 2005). The available literature about the production of FPs from different raw biomass residues is abundant (Miranda et al., 2015) while from the SCGs has been reported in (Kang et al., 2017; Kristanto & Wijaya, 2018; Limousy et al., 2013; Nosek et al., 2020; Potip & Wongwuttanasatian, 2018) and DSCG in (Deligiannis et al., 2011; Haile, 2014a, 2014b). This is attributed to their excellent chemical properties compared to other crops and biomass residues (Atabani, Al-Muhtaseb, et al., 2019). Moreover, the short growing period of CB provides another advantage compared to other biomasses, which require a longer growing time (Nosek et al., 2020). This makes both SCG and DSCG potential fuels in boilers (electricity generation) and stoves (heat generation) (Bio-bean, 2021a; Limousy et al., 2013). The following sections highlight the most important characteristics of SCG and DSCG and compare them with the literature results for other biomass fuels.

4.1 Pellet size

Pellet size is one of the most important FP characteristics (Kang et al., 2017; Narra et al., 2012). According to (Kang, 2017), 0.9% of dried, SCG particles have a size between 850 and 500 µm, 68.2% of dried, SCG particles have a size between 500 and 250 µm, 28% of dried, SCG particles have a size between 250 and 100 µm and 2.6% of dried, SCG particles have a size of ~100 µm (Fig. 4). However, this size distribution may vary depending on

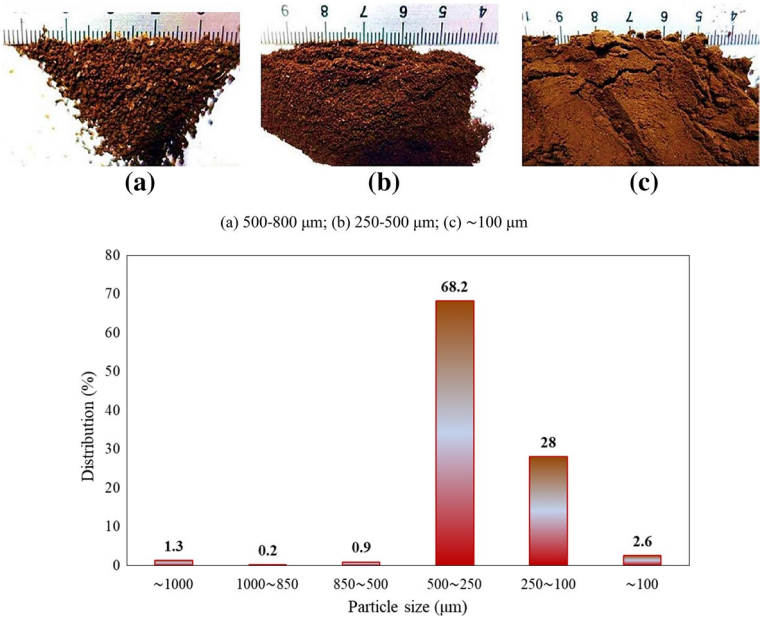


Fig. 4 SCG particle size distribution (Kang et al., 2017)

the coffee grinding machine. These results are in good agreement with (Go et al., 2016) who tested two samples of SCG. Their results showed that the average particle sizes are 430.5 ± 32.3 and 374.7 ± 8.3 µm, respectively. The authors also measured the average particle size of DSCG. The obtained results were 291.1 and 314.2 µm, respectively.

4.2 Calorific value (CV)

Calorific value (CV) is defined as the amount of energy generated from the combustion of a unit mass of a substance (Telmo & Lousada & Lousada, 2011). It is usually reported in units of kJ/kg or MJ/kg. It is one of the most important properties for the design, control, and numerical simulation of thermal systems (Friedl et al., 2005; Sheng & Azevedo, 2005). CV indicates the combustion quality of FPs. It is also important because it is used to determine the price of FPs. CV can be expressed either as a higher heating value (HHV), also known as gross calorific value (GCV), or lower heating value (LHV), also known as net heating value (NHV). HHV is defined as the enthalpy of complete combustion of a fuel when all carbon in the fuel is completely converted to CO_2 and all hydrogen is completely converted to water (H_2O) in the liquid phase. The HHV also includes the condensation enthalpy of water. HHVs are measured at standard conditions (101.3 kPa and 25 °C). The LHV is obtained by excluding the enthalpy of condensation of water and all of the water produced by the reaction remains as water vapor (Sheng &Azevedo, 2005; Telmo & Lousada, 2011). The HHV is generally used in the USA, while the LHV is more commonly used in European countries (Friedl et al., 2005). The calculation of HHV of biomass from the heat of formation of CO_2 and H_2O and other combustion reaction products is difficult. This is due to the complex and varying mixture of compounds (Friedl et al., 2005).

Therefore, HHV can be either determined experimentally using bomb calorimetry which quantifies the enthalpy change of reactants and products or it can be predicted from basic analyses such as ultimate (C, H, N, and O contents), proximate (fixed carbon, volatile matter, and ash), and chemical (cellulose, hemicellulose, lignin, and extractives) analysis using empirical equations (regression models) that are developed statistically based on a larger database of samples. Bomb calorimetry is a time-consuming process and involves manual laboratory work as well as calculations. However, this has been made easier now through the development of fully automatic equipment. Nevertheless, it is relatively expensive. On the other hand, proximate and ultimate analyses are cheaper, easier, and quicker methods that can be determined by modern laboratory equipment (Friedl et al., 2005; Sheng & Azevedo, 2005). The available literature is packed with many formulae proposed to empirically estimate the HHV of biomass with different accuracies (Demirbaş, 1997, 2001; Friedl, 2005; Sheng & Azevedo, 2005).

Demirbaş (1997) proposed the following three formulae to predict the HHV (MJ/kg) from ultimate and proximate analyses as follows:

$$\begin{aligned} \mathrm{HHV} &= [33.5(\mathrm{C}) + 142.3(\mathrm{H}) + 15.4(\mathrm{O}) - 14.5(\mathrm{N})] \times 10^{-2} \\ \mathrm{ABE} &= -1.67\%\ \text{(understimates)} \quad R^2 = 0.081 \end{aligned} \tag{1}$$

$$\begin{aligned} \mathrm{HHV} &= [0.196(\mathrm{FC}) + 14.119] \\ \mathrm{ABE} &= -5.60\%\text{(significantly understimates)} \quad R^2 = -0.647 \end{aligned} \tag{2}$$

$$\begin{aligned} \mathrm{HHV} &= [0.312(\mathrm{FC}) + 0.1534(\mathrm{VM})] \\ \mathrm{ABE} &= -6.97\%\text{(significantly understimates)} \quad R^2 = -0.30 \end{aligned} \tag{3}$$

where C, H, O and N ≡ Carbon, hydrogen, oxygen and nitrogen content (ultimate analysis) (% mass, dry basis), FC ≡ Fixed carbon (% mass, dry basis), VM ≡ Volatile matter (% mass, dry basis), ABE ≡ Average bias error.

Friedl et al (2005) proposed the following formula to predict the HHV (kJ/kg) of biomass samples consisting of plant material from their ultimate analyses as:

$$\begin{aligned} \mathrm{HHV} &= \left[3.55\left(\mathrm{C}^2\right) - 232(\mathrm{C}) - 2230(\mathrm{H}) + 51.2(\mathrm{C}) \times \mathrm{H} - 131(\mathrm{N}) + 20,600\right] \\ R^2 &= 0.943 \end{aligned} \tag{4}$$

Sheng and Azevedo (2005) proposed the following formulae to predict the HHV (MJ/kg) from ultimate and proximate analyses:

$$\begin{aligned} \mathrm{HHV} &= [-1.3675 + 0.3137(\mathrm{C}) + 0.7009(\mathrm{H}) + 0.0318(\mathrm{O})] \\ \mathrm{ABE} &= 0.07\% \quad R^2 = 0.834 \quad \text{Accuracy} > 90\% \end{aligned} \tag{5}$$

$$\begin{aligned} \mathrm{HHV} &= [19.914 - 0.2324(\mathrm{Ash})] \\ \mathrm{ABE} &= 0.3\% \quad R^2 = 0.625 \end{aligned} \tag{6}$$

$$\begin{aligned} \mathrm{HHV} &= [-3.0368 + 0.2218(\mathrm{VM}) + 0.2601(\mathrm{FC})] \\ \mathrm{ABE} &= 0.26\% \quad R^2 = 0.617 \end{aligned} \tag{7}$$

Parikh et al (2005) proposed the following formulae to predict the HHV (MJ/kg) using proximate analyses:

$$\begin{aligned} \text{HHV} &= [0.3536(\text{FC}) + 0.1559(\text{VM}) - 0.0078(\text{Ash})] \\ \text{ABE} &= 0.12\% \end{aligned} \tag{8}$$

Demirbaş (2001) proposed the following formula to predict HHV (MJ/kg) based on chemical composition:

$$\begin{aligned} \text{HHV} &= [0.0889(\text{L}) + 16.8218] \\ \text{ABE} &= -8.5\%(\text{significantly understimate}) \quad R^2 = -0.875 \end{aligned} \tag{9}$$

where, $L \equiv$ Lignin content (wt% and extractive-free sample basis).

The lower heating value (LHV) (MJ/kg) and ED (MJ/m^3) can be calculated from (Miranda et al., 2015):

$$\text{LHV} = \text{HHV} - 2.447 \times \left[\frac{\text{H}}{100}\right] \times 9.011 \tag{10}$$

$$\text{ED} = \text{BD} \times \text{LHV} \tag{11}$$

where H ≡ Hydrogen content (% db), BD ≡ Bulk density (kg/m^3).

Therefore, FPs with high bulk density and LHV possess large energy per unit volumes (ED). FPs with such characteristics reduce both logistic and storage needs, especially for the large-scale supply (Miranda et al., 2015). This makes these pellets attractive in many countries from both the demand and supply side of the market (Peksa-Blanchard et al., 2007).

Compared to proximate and chemical analyses, correlations based on ultimate analyses have been reported to provide the most accurate models to predict the HHV of biomass (Eqs. 4 and 5). It was also concluded that HHV models based on chemical composition are the least accurate as compared to others (Eq. 9). This is because chemical components, except cellulose, have different structures, such as lignin (Demirbaş, 2001; Sheng & Azevedo, 2005). It is important to note that the chemical composition determined depends on the analysis procedure used (Sheng & Azevedo, 2005).

Several papers have reported the energy content of various biomass and bio-waste such as SCG, rice husk, and bagasse. This is because it is one of the most critical characteristics of FPs. However, the moisture (water) content of biomass radically influences this property and must be low (< 15%) (Kang, 2017; Narra et al., 2012). This is because if wet FPs are directly burned, moisture requires energy to evaporate (Kang et al., 2017; Miranda et al., 2015). Additionally, high moisture contents negatively influence the flame temperatures, combustion, and residence completion times in the combustion chamber (Miranda et al., 2015). According to Kang et al., 2017, wood pellets with a water content of 7–10% have an LHV of 16.3–17.6 MJ/kg, while wood chips with a water content range between 18 and 35% have LHVs that range between 6.3 and 14.6 MJ/kg. CB with less than 5% water content has been reported to have an LHV between 20.9 and 21.8 MJ/kg. Wet SCG prepared through the brewing process has been reported to have an LHV of 8.4 MJ/kg. This is attributed to the high water content of SCG. Wet SCG has been reported to have an average water content of ~55% (Atabani et al., 2017; Kang et al., 2017). Therefore, drying SCG immediately after brewing is important to increase the LHV of SCG to more than 18.8 MJ/kg and to maintain the quality of SCG. Drying SCG to remove water prevents fungi and

mold from growing, which deteriorate its quality. This makes the LHV of SCG higher than wood pellets (Kang et al., 2017). Drying of SCG may be performed by an oven with heating at 105 °C for several hours (Atabani et al., 2017) or under sunlight or open-air using a cuboid tray with proper dimensions for several days (Ciesielczuk et al., 2015).

Table 1 conducts a comprehensive comparison between the HHVs of SCG and DSCG with other biomass residues. Based on the survey from literature, it was found that the HHVs of SCG and DSCG range between 19.55(min)–24.39(max) MJ/kg and 17.86(min)–21.54(max) MJ/kg. Therefore, the average reduction in the HHV of DSCG compared to SCG is between 10 and 20%. HHV values of SCG and DSCG were found to be greater than most of the HHV of other crop and biomass residues such as rice husks (14–15.91 MJ/kg), bagasse (7.7–19.22 MJ/kg), corn cob (17.36–17.48 MJ/kg), grape pomace (19.54 MJ/kg), pyrenean oak (19.3 MJ/kg), cassava stalk (18.1 MJ/kg), palm empty brunch (17.5 MJ/kg), rice straw (15.2 MJ/kg), coffee husk (16 MJ/kg), sugarcane leaves (18.4 MJ/kg), wheat straw (17.0–18.55 MJ/kg), barley straw (17.43 MJ/kg), corn stover (17.8 MJ/kg), tobacco stalk (17.7–18.43 MJ/kg), beechwood (18.77–19.23 MJ/kg), coconut fiber (19.678 MJ/kg), maize residue (17.51 MJ/kg), hard wood (18.66–18.97 MJ/kg), and some coals types such as peat (11–16 MJ/kg), and lignite (brown) coal (8.5–16.6 MJ/kg). These results were also compared to those of oil cake (20.57–21.23 MJ/kg), pine sawdust (20 MJ/kg), softwood (19.55–20 MJ/kg), bituminous (black) coal (19.43 MJ/kg), anthracite (hard) coal (16.7–32.7 MJ/kg), and hazelnut shell (19.3 MJ/kg). It was also found that SCG has higher LHV than some bio-wastes such as green and red tea waste (Smaga, 2016). Additionally, it has been observed that CVs of SCG and DSCG do not differ significantly from the CVs of wood pellets that are currently available in the market. Results were found to be quite comparable to different wood species such as *Eucalyptus globulus* (17.631 MJ/kg), *Castanea sativa* (18.75 MJ/kg), *Hymenaea courbaril* (19.3 MJ/kg), and *Bowdichia nitida* (20.81 MJ/kg) (Telmo & Lousada & Lousada, 2011). However, charcoal (34.388 MJ/kg) and coke (32.428 MJ/kg) were found to have greater CVs than both SCG and DSCG (Parikh et al., 2005). The CVs of SCG and DSCG are lower than that of LPG (49.3 MJ/kg), paraffin (25.92 MJ/kg) and coal (42 MJ/kg) (Pilusa et al., 2013). Overall, the results proved the potential of SCG as a raw material to produce FPs. A study conducted by Zuorro & Lavecchia, 2011 suggested that phenolic-extracted SCG has a comparable HHV (23.4 ± 0.5 MJ/kg) to SCG (24.3 ± 0.8 MJ/kg). This method supports another pathway, in which the extraction of some rich ingredients from SCG can be valorised to produce raw materials used in cosmetic, pharmaceutical, and food industries while what remains of SCG can be valorised to produce FPs.

Alternatively, SCG can be mixed with other crops and biomass residues to increase its HHV (Table 1). For instance, the addition of 10 and 25% of SCG to beechwood improved the calorific values from 18.77 to 19.12 MJ/kg (~1.86% increase) and 20.32 MJ/kg (~8.26% increase), respectively (Ciesielczuk et al., 2015). Kristanto and Wijaya (2018) supported that the addition of SCG to coffee silverskin (CS) resulted in the increase of its HHV from 17.77 MJ/kg for FP made from 55%SCG + 40%CS + 5% starch to 19.71 MJ/kg for FP made from 95%SCG + 5%CS + 5% starch representing an increase of ~11%. Nosek et al., 2020 reported the same improvement when SCG is mixed with sawdust to form 50SCG/50sawdust which has an LHV of 20.06 MJ/kg compared to the 17.15 MJ/kg obtained when 100% sawdust is used. On the other hand, the positive effect of adding crude glycerol (by-product of transesterification reaction) to increase the HHV of SCG has been demonstrated by (Potip & Wongwuttanasatian, 2018). FPs of 90%SCG + 10% glycerol and 95%SCG + 5% glycerol have HHV of 21.55 and 21.21 MJ/kg, respectively, compared to 21.14 MJ/kg of SCG.

Table 1 Comparison of HHVs of dried SCG, DSCG, RDCB, DRDCB, and other crops and biomass residues (MJ/kg)

Type	Ref	HHV
SCG	(Obruca et al., 2014)	19.61
DSCG		17.86
SCG	(Somnuk et al., 2017)	23.10
DSCG		20.4
SCG	(Li et al., 2014)	23.20
SCG	(Caetano et al., 2017)	19.30
SCG	(Atabani et al., 2017)	24.00
DSCG		21.54
SCG	(Vardon et al., 2013)	23.40
DSCG		20.10
SCG	(Yang et al., 2016)	20.20
DSCG	(Deligiannis et al., 2011)	21.16
DSCG	(Kondamudi et al., 2008)	20.21
SCG	(Sakuragi et al., 2016)	21.1
SCG (1)	(Go et al., 2016)	24.39 ± 1.16
DSCG (1)		20.27 ± 1.46
SCG (2)		22.83 ± 0.86
DSCG (2)		20.03 ± 0.77
SCG	(Efthymiopoulos et al., 2018)	22.37 ± 0.07
DSCG		19.85 ± 0.11
RDCB		21.34 ± 0.18
DRDCB		19.60 ± 0.54
SCG	(Limousy et al., 2013)	19.55
Pine sawdust pellets		19.23
50% SCG + 50% pine sawdust pellets		19.63
SCG	(Potip & Wongwuttanasatian, 2018)	21.14
95%SCG + 5% glycerol		21.21
90%SCG + 10% glycerol		21.55
Lignite (brown) coal		16.38
Bituminous (black) coal		19.43
Corn cob		17.36
Hard wood		18.66
Sugarcane leaves		18.40
Cassava stalk		18.10
Palm empty brunch		17.50
Rice straw		15.20
SCG	(Silva et al., 1998)	24.91
Rice husks		15.91
Wheat straw		17.49
Sugar cane bagasse		18.69
Sugar cane bagasse		19.23
Coconut fiber		19.68

Table 1 (continued)

Type		Ref	HHV
DSCG after bioethanol production		(Haile et al., 2013)	20.80
Coffee husk			16.00
Bagasse			7.70–8.00
Rice husk			14.00
Pure beechwood		(Ciesielczuk et al., 2015)	18.77
10%SCG + 90% pure beechwood			19.12
25%SCG + 75% pure beechwood			20.32
Anthracite (hard) coal			16.7–32.7
Lignite (brown) coal			8.5–16.6
Peat			11–16
SCG before phenolic extraction		(Zuorro & Lavecchia, 2011)	24.3 ± 0.80
SCG after phenolic extraction			23.4 ± 0.50
Pyrenean oak		(Miranda et al., 2015)	19.30
Pyrenean sylvestris			19.70
Pine sawdust			20.80
Vine shoots			18.70
Olive branches			18.82
Barley straw			17.43
Wheat straw			18.25
Olive pomace			22.03
Grape pomace			19.54
Oil cake		(Jóvér et al., 2018)	20.57–21.23
Maize residue			17.51
Wheat residue			16.545
Sunflower residue			14.39
Bagasse			16.80
Wheat straw		(Demirbaş, 1997)	17.00
Olive husk			19.00
Tea waste			17.10
Hard wood			18.80
Soft wood			20.00
Hazelnut shell			19.30
Corn stover			17.80
Tobacco stalk			17.70
Eucalyptus globulus	Wood species	(Telmo & Lousada, 2011)	17.63
Castanea sativa			18.75
Hymenaea courbaril			19.30
Bowdichia nitida			20.81
SCG		(Bok et al., 2012)	22.74
Mallee			17.32
Charcoal		(Parikh et al., 2005)	34.39
Coke			32.43

Table 1 (continued)

Type	Ref	HHV
95%SCG + 5%CS + 5% starch composition	(Kristanto & Wijaya, 2018)	19.71
85%SCG + 10%CS + 5% starch composition		19.58
75%SCG + 20%CS + 5% starch composition		19.32
65%SCG + 30%CS + 5% starch composition		18.12
55%SCG + 40%CS + 5% starch composition		17.77
80%DSCG + 20% glycerin	(Haile, 2014a, b)	19.30
75%DSCG + 25% glycerin		19.70
70%DSCG + 30% glycerin		19.80
65%DSCG + 35% glycerin		20.20
60%DSCG + 40% glycerin		21.60
Eco-fuel briquette	(Pilusa et al., 2013)	18.90
LPG		49.3
Paraffin		25.92
Coal		42.00
Wood		16.00
Tobacco leaf	(Demirbaş, 2001)	17.7
Hazelnut shell		20.05
Olive cake		21.53
Wheat straw		18.55
Corn cob		17.48
Corn straw		18.27
Hard wood		18.97
Tobacco stalk		18.43
Soft wood		19.55
Beechwood		19.23
100%SCG	(Nosek et al., 2020)[a]	21.8
50SCG/50sawdust		20.06
40SCG/60sawdust		19.34
30SCG/70sawdust		18.19
100%sawdust		17.15
SCG	(Smaga, 2016)[a]	21.15
Black tea		19.50
Green tea		19.39
White tea		18.20
Red tea		18.76

[a]Results are based on LHV

[b]32% SCG + 23% coal fines + 11% sawdust + 18% mielie husks + 10% waste paper + 6% paper pulp

Results of HHVs of DSCG indicated its potential as FPs. Some of the results were: 21.54 (Atabani et al., 2017), 21.16 (Deligiannis et al., 2011), 20.21 (Kondamudi et al., 2008), 20.1 (Vardon et al., 2013), 20.03 (Go et al., 2016), 20.17 (Efthymiopoulos et al., 2018), and 20.8 MJ/kg (Haile et al., 2013). These findings satisfy DIN 5173, Ö NORM M 7135, EN14961, Agro+, and Agro standards (Table S1). This supports the concept of removing/extracting lipids from SCG that can be valorized into other products while valorizing DSCG to produce FPs. SCGO can be valorized to produce biodiesel. Glycerin collected as a by-product of the transesterification can be added to DSCG to produce FPs with higher HHV. Haile (2014b) indicated that FPs prepared by mixing 60%DSCG with 40% glycerin produce a heating value of 21.6 MJ/kg in comparison with 19.3 MJ/kg (80%DSCG + 20% glycerin).

Furthermore, it has been found that hydrolyzed DSCG used for bioethanol production has a comparable HHV to DSCG (20.80 MJ/kg) (Haile et al., 2013). Therefore, the authors suggested another pathway to recycle SCG in which the extracted oil (SCGO) can be transesterified into biodiesel. DSCG can be then hydrolyzed into bioethanol and the remains of DSCG besides glycerin (by-product of transesterification reaction) can be valorized as FPs. The same approach was proposed by (Kondamudi et al., 2008) in which hydrolyzed DSCG can be finally utilized as FPs. Another approach was reported by (Go et al., 2016) in which SCG is firstly hydrolyzed so the hydrolysate can be processed into bioethanol while the SCGO can be then extracted from hydrolyzed SCG and the solid remains can be used as FP.

4.3 Ultimate and proximate analysis and chemical composition of SCG and DSCG

Several papers have reported the ultimate and proximate analysis and chemical composition of SCG and DSCG. The main findings from the literature are tabulated in Table 2. The following Sects. (4.3.1–4.3.2) provide a detailed discussion of these findings obtained from the literature.

4.3.1 Ultimate analysis

The ultimate analysis gives the weight percent of the elements in biomass. C, H, and O are the major biomass elements representing 97–99% of the organic biomass. Other elements include N, sulfur (S), chlorine (Cl), etc. (Sheng & Azevedo, 2005). As per the previous section's discussion, correlations between HHV and elemental composition are given in Eqs. (1, 4, and 5). The HHV is affected by H and C content in the sample. An increase in C and H contents increases the HHV. However, O content was reported to offer a slightly negative effect on HHV (Sheng & Azevedo, 2005). Ultimate analysis is a useful tool to predict the harmful emissions caused by the combustion of FPs (Miranda et al., 2015).

The most interesting finding is that SCG and DSCG have more C and H and less O content than many other biomasses. This indicates that SCG and DSCG are potential sources to produce FPs as their HHVs are high. H content in SCG has been found in the range between 6.04–8.99% for SCG and 6.17–6.74% for DSCG. C content in SCG has been found in the range between 46.42–71.6% for SCG and 48.34–52.03% for DSCG. N content in SCG has been observed to range between 1.41–17.78% for SCG and 0.49–16.7% for DSCG. However, both SCG and DSCG contain more N than other biomasses and fail to satisfy EN ISO 17225, EN 14,961, Agro+, and Agro standards (Tables S1, S2, and S3).

Table 2 Comparison between ultimate analysis (elemental composition) of SCG, DSCG and other crops and biomass residues

Ultimate analysis						
	Ref	C%	H%	N%	O%	S%
SCG	(Silva et al., 1998)	59.50	7.30	2.50	30.70	–
SCG (1)	(Pujol et al., 2013)	57.16	7.17	1.18	–	–
SCG (2)		59.77	7.57	1.32	–	–
CB	(Kang et al., 2017)	53.25	6.94	1.62	34.25	0.06
SCG		53.05	7.19	1.45	36.20	0.05
Wood pellets		48.48	6.33	0.42	44.21	0.01
SCG	(Vardon et al., 2013)	56.10	7.20	2.40	34.00	0.14
DSCG		51.80	6.30	2.80	38.80	0.17
SCG	(Yang et al., 2016)	50.40	7.20	2.10	40.30	–
SCG	(Li et al., 2014)	54.50	7.10	2.40	34.20	0.10
SCG	(Kan et al., 2014)	51.80	6.40	2.08	39.67	0.15
SCG	(Kelkar et al., 2015)	52.37	7.31	2.42	36.50	0.14
SCG	(Kristanto & Wijaya, 2018)	43.00	8.65	1.41	45.44	1.60
CS		49.00	6.46	3.21	38.83	2.80
SCG	(Jeguirim et al., 2014)	61.13	8.99	2.91	26.60	0.37
Pine sawdust		47.10	6.10	< 0.1	46.27	< 0.01
SCG	(Bok et al., 2012)	54.61	6.59	3.97	34.83	–
Mallee		48.40	6.30	0.10	45.20	–
SCG	(Atabani et al., 2017)	46.42	6.04	15.50	–	0.561
DSCG		49.59	6.74	16.70	–	0.851
DSCG	(Deligiannis et al., 2011)	52.03	6.31	0.49	–	–
SCG	(Somnuk et al., 2017)	52.95	6.76	2.10	38.07	0.12
DSCG		48.34	6.17	2.39	43.01	0.09
SCG	(Kim et al., 2018)	54.3 ± 0.1	7.4 ± 0.1	2.1 ± 0.1	36.9 ± 0.4	0.00
SCG	(Luz et al., 2017)	60.20	6.80	2.12	30.80	0.02
Cow manure		41.65	5.81	1.91	49.46	0.27
SCG	(Qiao et al., 2013)	55.23 ± 3.71	7.07 ± 0.39	2.33 ± 0.24	34.44 ± 4.54	0.30 ± 0.19
Sludge		34.04 ± 0.22	5.47 ± 0.07	5.93 ± 0.09	25.69 ± 0.42	0.70 ± 0.02
SCG	(Nosek et al., 2020)	54.56	7.44	17.78	–	–
50SCG/50sawdust		52.69	6.99	46.87	–	–
40SCG/60sawdust		52.13	6.89	50.64	–	–
30SCG/70sawdust		51.29	6.74	61.66	–	–
Anthracite (hard) coal	(Ciesielczuk et al., 2015)	65–85	4.7–5.9	1.4–3	–	0.8–2
Lignite (brown) coal		66–73	5–7	0.7–1.5	–	0.7–7
Peat		58.00	5.50	1.80	–	0.20

Table 2 (continued)

Ultimate analysis						
	Ref	C%	H%	N%	O%	S%
SCG	(Kim et al., 2017a)	51.4 ± 0.2	7.0 ± 0.1	2.2 ± 0	38.4 ± 0.1	–
Food waste		48.4 ± 0.7	7.1 ± 0.1	4.4 ± 0.1	40.4 ± 0.5	0.2 ± 0
Ulva		37.8 ± 0.3	7.2 ± 0.3	5.4 ± 0	35.7 ± 0.3	1.9 ± 0.2
WAS		37.1 ± 0.4	5.7 ± 0	6.6 ± 0.1	27.2 ± 0.1	0.5 ± 0.1
Whey		35.3 ± 0.3	7.3 ± 0.1	1.5 ± 0	45.8 ± 0.5	0.2 ± 0
SCG	(Sakuragi et al., 2016)	51.50	6.97	2.43	37.10	0.15
SCG	(Girotto et al., 2018)	58.80	8.90	3.40	28.70	0.20
SCG	(Yang et al., 2017)	51.40	7.70	2.10	38.80	–
Paper filter		50.10	6.20	0.20	43.50	–
Corn stalk		44.40	6.30	0.50	48.90	–
White pine		42.70	6.80	0.00	50.50	–
SCG	(Limousy et al., 2013)	61.13	8.99	2.91	26.60	0.37
Pine sawdust		47.10	6.10	< 0.1	46.27	< 0.01
Millet	(Friedl et al., 2005)	45.90	5.30	0.9	41.10	0.10
Eco-fuel briquette	(Pilusa et al., 2013)	36.65	4.60	0.75	36.3	0.34
Coal		61.2	4.30	1.2	7.4	3.9
Pyrenean oak	(Miranda et al., 2015)	50.94	6.34	1.81	~40.81[a]	0.10
Pyrenean sylvestris		51.80	5.70	0.37	~41.10[a]	0.03
Pine sawdust		50.50	6.10	0.48	~42.89[a]	0.03
Vine shoots		46.90	5.70	0.58	~46.77[a]	0.05
Olive branches		47.02	7.62	0.34	~45.02[a]	0.00
Barley straw		43.85	5.50	0.77	~49.78[a]	0.10
Wheat straw		45.10	6.00	0.91	~47.99[a]	0.00
Olive pomace		51.42	6.56	1.98	~39.94[a]	0.10
Grape pomace		42.97	9.28	2.05	~45.53[a]	0.17
Char	(Kelkar et al., 2015)	75.30	3.52	4.40	0.11	0.12
Charcoal	(Parikh et al., 2005)	92.04	2.45	0.53	2.96	1
Coke		89.13	0.43	0.85	0.98	1

Proximate analyses					
	Ref	VM (wt%)	FC (wt%)	Ash (wt%)	Moisture (wt%)
SCG	(Sakuragi et al., 2016)	81.1	17.00	1.90	–
SCG	(Li et al., 2014)	82.0	16.30	1.70	8.1
SCG	(Kan et al., 2014)	76.16	15.25	1.78	6.81
CB	(Kang et al., 2017)	76.75	17.27	3.88	2.10
SCG		70.03	16.22	2.06	11.69
Wood pellets		74.85	17.21	0.55	7.39
SCG	(Bok et al., 2012)	77.51	19.83	1.35	1.31
Mallee		81.90	17.60	0.50	

Table 2 (continued)

Proximate analyses					
	Ref	VM (wt%)	FC (wt%)	Ash (wt%)	Moisture (wt%)
SCG	(Jeguirim et al., 2014; Limousy et al., 2013)	68.94	17.46	1.82	11.78
Pine sawdust pellets		77.61	14.19	0.30	7.90
50% SCG + 50% Pine sawdust pellets		78.23	13.94	1.18	6.65
Charcoal	(Parikh et al., 2005)	9.88	89.10	1.02	–
Coke		0.92	91.47	7.61	–
Pistachio shell		82.03	16.84	1.13	–
Coffee chaff		75.80	19.60	4.60	–
Wheat straw		80.60	11.70	7.70	–
Wheat straw		63.00	23.50	13.50	–
Bagasse		7.90	7.00	22.10	–
Bagasse		86.30	11.90	1.80	–
Tea waste		85.00	13.60	1.40	–
Castor seed cake		67.90	25.20	6.90	–
Rice husk char		5.90	41.20	52.90	–
CS	(Kristanto & Wijaya, 2018)	91.00	Nd	1.30	–
95%SCG/5%CS/5% starch composition		85.50	0.50	0.10	–
75%SCG/20%CS/5% starch composition		86.00	1.50	0.50	–
55%SCG/40%CS/5% starch composition		86.80	2.40	0.60	–
SCG	(Yang et al., 2017)	75.20	–	1.10	–
Paper filter		79.40	–	0.50	–
Corn stalk		68.90	–	2.40	–
White pine		70.50	–	1.10	–
80%DSCG + 20% glycerin	(Haile, 2014a, 2014b)	73.40	16.40	5.08	5.12
75%DSCG + 25% glycerin		76.80	13.81	4.40	4.99
70%DSCG + 30% glycerin		78.00	13.97	3.70	4.33
65%DSCG + 35% glycerin		79.10	14.58	3.20	3.12
60%DSCG + 40% glycerin		77.80	14.37	2.80	5.03

Chemical composition				
Property	References	Biomass		Result
Lignin	(Bok et al., 2012)		%	33.32 ± 0.91
Lignin	(Caetano et al., 2017)		%	31.02

Table 2 (continued)

Chemical composition					
Property		References	Biomass		Result
Holocellulose		(Yang et al., 2017)	SCG	%	61.40
Protein				%	13.00
Extractives				%	10.50
Lignin				%	27.70
Holocellulose			Paper filter	%	91.50
Protein				%	1.20
Extractives				%	7.30
Lignin				%	0.70
Holocellulose			Corn stalk	%	67.90
Protein				%	3.10
Extractives				%	1.30
Lignin				%	18.60
Holocellulose			White pine	%	44.10
Protein				%	–
Extractives				%	5.10
Lignin				%	49.70
Cellulose		(Ballesteros et al., 2014)	SCG	%	12.4 ± 0.8
Arabinose	Hemicellulose			%	3.60 ± 0.52
Mannose				%	19.70 ± 9.85
Galattose				%	16.43 ± 1.66
Lignin				%	23.90 ± 1.7
Fat				%	2.29 ± 0.3
Ashes				%	1.30 ± 0.1
Protein				%	17.44 ± 0.1
Total dietary fiber				%	60.46 ± 2.29
Cellulose			CS	%	23.77 ± 0.09
Arabinose	Hemicellulose			%	3.54 ± 0.29
Mannose				%	1.77 ± 0.06
Galattose				%	3.76 ± 1.27
Lignin				%	28.58 ± 0.46
Fat				%	3.78 ± 0.40
Ashes				%	5.36 ± 0.20
Protein				%	18.69 ± 0.10
Total dietary fiber				%	54.11 ± 0.10

Table 2 (continued)

Chemical composition				
Property	References	Biomass		Result
Hemicelluloses	(Demirbaş, 2001)	Tobacco leaf	%	41.54
Cellulose			%	43.45
Lignin			%	15.01
Hemicelluloses		Wheat straw	%	45.20
Cellulose			%	33.82
Lignin			%	20.98
Hemicelluloses		Hazelnut shell	%	30.29
Cellulose			%	26.70
Lignin			%	43.01
Hemicelluloses		Hardwood	%	32.26
Cellulose			%	45.85
Lignin			%	21.89
Hemicelluloses		Beechwood	%	31.86
Cellulose			%	46.27
Lignin			%	21.87
Hemicelluloses		Softwood	%	24.82
Cellulose			%	42.68
Lignin			%	32.50
Hemicelluloses		Olive cake	%	21.63
Cellulose			%	23.08
Lignin			%	55.29
Hemicelluloses		Tobacco stalk	%	28.89
Cellulose			%	44.32
Lignin			%	26.79

[a]Calculated by differences, Approximately = (100—C–H–N–S)

This is attributed to the existence of protein and caffeine (Bok et al., 2012). High N content may contribute to the increased oxides of nitrogen (NO_x) emitted during the combustion of FPs (Kang et al., 2017). S content in SCG has been reported to range between 0.02–1.6% for SCG and 0.09–0.851% for DSCG. It can be noticed that some results of S contents of SCG and DSCG do not match the requirement of DIN 51,731, ÖNORM M 7135, DIN EN 14,961, Agro+, and Agro standards (Tables S1, S2, and S3).

4.3.2 Proximate analysis

The proximate analysis provides the weight percent of moisture, FC, VM, and ash in the biomass material (Sheng & Azevedo, 2005). It is one of the easiest and most widely used methods to characterize the quality of fuel, such as HHV [Eqs. (2, 3, 6, 7 and 8)] (Demirbaş, 1997; Parikh et al., 2005; Sheng & Azevedo, 2005). Equation 8 indicated the lowest ABE among others (0.12%). From these empirical formulae, it can be concluded that HHV increases with an increase in FC and VM content and decreases with an increase in ash content. The wide range of proximate analysis results of SCG, DSCG, and other

crops as well as biomass residues are provided in Table 2. It has been observed that the ash content of SCG is higher than some biomass-derived residues such as pine sawdust (Limousy et al., 2013), wood pellets (Kang et al., 2017), and lower than others such as rice husk char and castor seed cake (Parikh et al., 2005). It can be seen that FPS from agro-pellet produced more ashes than wood pellets. Besides producing FP with lower HHV, high ash content is not desirable as it forces operators to clean and service their boilers and stoves more frequently. Results from Table 2 indicate that SCG has an ash content between 1.35 and 2.06%. This was reported to be lower than CB (3.88%) (Kang et al., 2017). These results are acceptable according to EN 14,961 ($\leq$3–10%) and EN ISO 17225-6 ($\leq$6%) standards but do not satisfy the Ö NORM M 7135 ($<$0.5%) and DIN 51,731 ($<$1.5%) standards. Generally, high contents of ash are associated with low durability (Miranda et al., 2015). Durability plays an important role as it indicates the ability of FPs to withstand handling stresses (Narra et al., 2012). The durability of FPs is affected by the density and lignin and water content (Kristanto & Wijaya, 2018). According to Kang et al., 2017, FPs with an ash content of less than 0.7% are classified as 1st grade. Therefore, it can be considered that both CB and SCG can be categorized as either 2nd or 3rd grade FP in this aspect. Miranda et al., 2015 indicated that high ash content is no longer a problem that cannot be solved. This is due to new automatic systems dedicated to ash removal. The removal of ash may also increase the HHV and durability of FPs.

4.3.3 Chemical composition

The chemical analysis of SCG, other crops, and biomass residues is provided in Table 2. SCG features a high amount of fiber content. The average content of hemicellulose was (30–40%), cellulose (8–13%), and lignin (25–33%), respectively. In general, the HHVs of lignocellulosic fuels increase with an increase in their lignin contents (Eq. 9) (Demirbaş, 2001). As it can be seen, SCG has higher lignin content than CS.

4.4 Bulk density, porosity, and morphological analyses

The density of FPs is determined by the ratio of the mass of pellets to a predefined volume. The higher the BD, the higher is the ED, and the lower the transport and storage costs (Narra et al., 2012). Compared to fossil fuels, FPs from biomass have two main drawbacks: low BD and high porosity. These key bottlenecks are a hindrance to their use as energy carriers. The low density of biomass affects combustion efficiency. Densification, a process that involves compressing raw material to obtain denser fuel (high BD), may solve these drawbacks as it increases the density and homogeneity of raw materials 2–4 times and therefore the energy content 1–4 times. It was reported that the BD of FP of ~ 0.7 t/m^3 drastically reduces the transport, storage, and logistics costs. BD is influenced by certain factors including moisture content, particle size, natural adhesive content, and raw material morphology. Pelletizing is the most widely used technique to densify biomass and produce high-density FPs and thus, more ED (Miranda et al., 2015; Peksa-Blanchard et al., 2007). Table 3 compares the bulk density of SCG blends with other biomass residues. SCG has a lower bulk density than other biomass residues. Similar results have also been reported by (Atabani et al., 2017; Ballesteros et al., 2014). Therefore, it can be inferred that mixing SCG at controlled percentages with other biomass that have high HHV and BD such as olive pomace (22.03 MJ/kg and 0.780 gm/cm^3), pine sawdust (20.8 MJ/kg and 0.650 gm/cm^3), grape pomace (19.54 MJ/kg and 0.824 gm/cm^3), pyrenean sylvestris (19.7 MJ/kg

Table 3 Comparison of the BD of SCG, blends, and other crops and biomass residues

Sample	Bulk density	EN 14,961/ EN ISO 17225 g/cm^3	Agro + /Agro	Ref
SCG	0.3131 ± 0.0036	≥ 0.6	≥ 0.65	(Ciesielczuk et al., 2015)
10%SCG + 90% beechwood	0.83 ± 0.2			
25%SCG + 75% beechwood	0.73 ± 0.01			
Beechwood	0.95 ± 0.02			
95%SCG/5%CS/5% starch composition	0.806			(Kristanto & Wijaya, 2018)
85%SCG/10%CS/5% starch composition	1.026			
75%SCG/20%CS/5% starch composition	1.051			
65%SCG/30%CS/5% starch composition	1.154			
55%SCG/40%CS/5% starch composition	1.167			
80%DSCG + 20% glycerin	0.9966			(Haile, 2014b)
75%DSCG + 25% glycerin	0.9957			
70%DSCG + 30% glycerin	0.996			
65%DSCG + 35% glycerin	0.9978			
60%DSCG + 40% glycerin	0.9955			
Pyrenean oak	0.678			(Miranda et al., 2015)
Pyrenean sylvestris	0.675			
Pine sawdust	0.650			
Vine shoots	0.700			
Olive branches	0.582			
Barley straw	0.644			
Wheat straw	0.620			
Olive pomace	0.780			
Grape pomace	0.824			
Eco-fuel briquette[a]	0.721			(Haile et al., 2013)

[a]32% SCG + 23% coal fines + 11% saw dust + 18% mielie husks + 10% waste paper + 6% paper pulp

and 0.675 gm/cm^3) (Miranda et al., 2015), and beechwood (18.77 MJ/kg and 0.95 gm/cm^3) (Ciesielczuk et al., 2015) helps to produce FPs with a high HHV and BD. This strategy can also be adopted with other biomass residues with relatively lower HHV than SCG but higher BD such as rice husk, rice straw, and bagasse (Table 1). From Tables 1 and 5, it can also be seen that mixing 75% of beechwood with 25% SCG yields an FP with a BD of 0.73 (0.01) g/cm^3 and HHV of 20.32 MJ/kg. Both properties satisfy the standards of EN 14,961, EN ISO 17225 (≥ 0.6 g/cm^3), Agro +, and Agro (≥ 0.65 g/cm^3), respectively, for FPs from biomass. Similar observations can be drawn with DSCG when mixed with glycerin (Haile, 2014b). For instance, mixing 40% of glycerin with 60% DSCG yields an FP with a BD of 0.9955 g/cm^3 and HHV of 21.60 MJ/kg. Table 2. Comparison between ultimate analysis (elemental composition) of SCG, DSCG, and other crops and biomass residues.

It has been reported that the high bulk density of the final FPs does not always imply proper compaction. Therefore, the relationship between bulk density and durability of the final FPs must be tested experimentally (Miranda et al., 2015). This is because the durability of the FP is as important a quality parameter as HHV and BD (Table S2–S3). For instance, Kristanto & Wijaya, 2018 reported that FP prepared from 75%SCG/20%CS/5% starch composition has the highest durability and optimum water content. Moreover, it had a constant flame and a decrease in temperature, although its HHV is lower than other samples (Table S1) (19.32 MJ/kg). Additionally, its BD was also quite excellent (1.051 g/cm^3) which satisfied DIN 5173, Ö NORM M 7135, EN14961, Agro+, and Agro standards.

Ballesteros et al., 2014 indicated that both SCG and CS have low porosities and possess mesopores. Brunauer–Emmett–Teller (BET) analyses indicated that the BET surface area (S_{BET}) of CS and SCG was 2.1 and 4.3 m^2/g, respectively. The total volumes of pores were 0.003 (CS) and 0.004 cm^3/g (SCG). These results were validated by BJH. Therefore, SCG has a higher surface area than CS. These results have been further confirmed by (Kristanto & Wijaya, 2018) who indicated that CS has a denser fiber structure as compared to SCG, implicating that CS has a lower porosity than SCG. The higher porosity of SCG leads to lower density because of the many voids in the particles. This can be seen clearly in the images obtained by scanning electron microscopy (SEM) in Fig. S2 and Fig. S3, respectively, showing the significant morphological differences between CS and SCG. Therefore, adding CS to SCG can increase the density of FP and thus the combustion efficiency.

Atabani et al. (2017) compared the porosity of SCG (CWBE) and DSCG (CWAE) using SEM and BET/BJH analyses. SEM analysis revealed that SCG possesses a smooth, monolith surface deprived of noticeable pores, whereas the DSCG sample contains large lumps with a rough surface and high porous structure. The DSCG sample presents a granular morphology connected by many carbon nanospheres and has a loose porous structure. These results were in agreement with those of (Somnuk et al., 2017), which indicated that DSCG has smoother surfaces than SCG due to oil extraction. Additionally, S_{BET} and BJH pore assembly, viz., pore size and pore volume distribution, characterization of SCG and DSCG are shown in Fig. 5. The structural characteristics of SCG and DSCG are significantly dissimilar. SCG is non-porous and has a S_{BET} of 4.4526 m^2/g (similar to Ballesteros et al., 2014), whereas DSCG sample provides S_{BET} specific surface area value of 0.4056 m^2/g, suggesting that it consists of a highly porous structure. These outcomes are consistent with SEM results. The morphology differences are affected by the activation ratio and the SCGO extraction process. According to the SEM, BET/BJH analyses presented in (Atabani et al., 2017; Ballesteros et al., 2014), the order of porosity is DSCG > SCG > CS. Therefore, CS has the highest density, followed by SCG and DSCG. This indicates that a densification process is required to increase the ED of both SCG and DSCG. On the other hand, the higher energy content of SCG compared to CS gives another indication that mixing both wastes can also be another technique to increase the energy content of FP (Kristanto & Wijaya, 2018). Figure 6 shows the isotherm linear plots of SCG and DSCG.

4.5 Thermogravimetric analysis (TGA) and differential thermogravimetry (DTG)

Results of thermogravimetric analysis (TGA) and differential thermogravimetry (DTG) curves of SCG were reported in (Jeguirim et al., 2014; Kan et al., 2014). Figure 7 shows the results of TGA and DTG of SCG, DSCG and mallee. Bok et al., 2012 compared the TGA/DTA curves of SCG and mallee. Two peaks after moisture loss occurred at 550 K (degradation of hemicelluloses and cellulose) and 673 K (degradation of lignin).

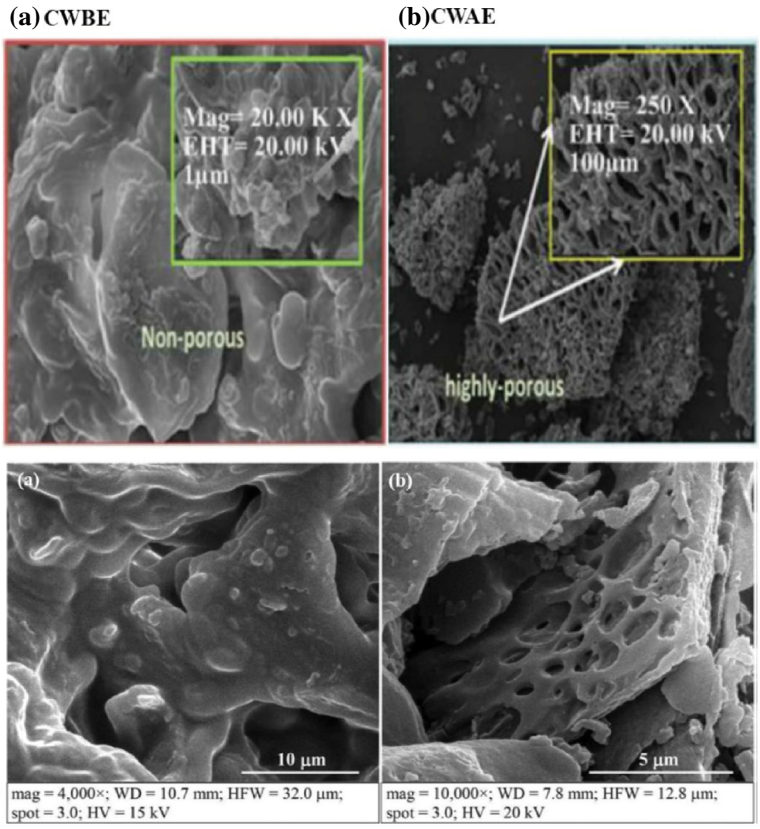


Fig. 5 SEM analysis **a** SCG **b** DSCG (Atabani et al., 2017). CWBE ≡ Coffee waste before extraction (SCG). CWAE ≡ Coffee waste after extraction (DSCG)

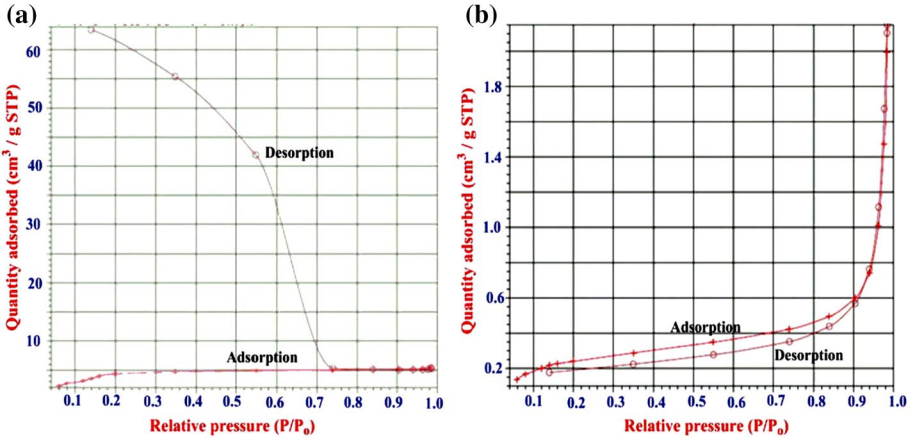
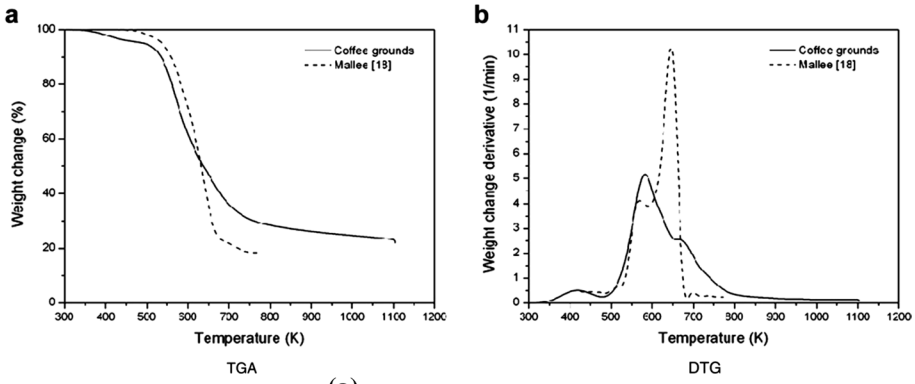
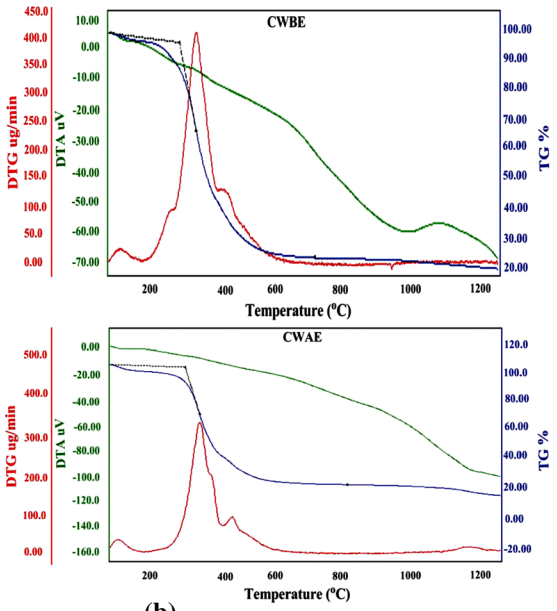


Fig. 6 Isotherm linear plots of **a** SCG and **b** DSCG (Atabani et al., 2017)



(a) SCG and mallee (Bok et al., 2012)



(b) SCG and DSCG (Atabani et al., 2017)

Fig. 7 TGA and DTA of SCG, DSCG, and mallee **a** SCG and mallee (Bok et al., 2012), **b** SCG and DSCG (Atabani et al., 2017)

While for SCG, the maximum peak was recorded at 581 K, which is lower than mallee (651 K). These results indicate that the devitalization step of SCG occurred earlier than mallee. During thermal degradation, a short shoulder appears around 500 K. The mass loss and weight change derivative at this shoulder is 10% and 0.55/min^{-1} approximately.

Atabani et al. (2017) indicated both SCG and DSCG samples showed a similar trend of decomposition curves with a three-step decomposition up to 1200 °C. The rresults showed three main decomposition stages for both samples as follow:

(1) At the 1st stage, a mass loss of (4.05–4.53%) in the temperature range of 60.38–77.03 °C was detected. This loss is associated with crystalline water molecule removal from both SCG and DSCG.
(2) At the 2nd stage, a mass loss of 44.32 and 49.03% was recorded between 247–376 and 245–382 °C for SCG and DSCG, respectively. This loss represents the removal of polysaccharides and fatty oils from both samples.
(3) At the 3rd stage, a mass loss of 34.78 and 38.51% occurred at 509 and 498 °C for both samples.

4.6 Combustion and emissions analyses

The stability of temperature during the combustion process is a crucial parameter in the supply of fuel to a furnace or boiler. Irregular changes or sudden drops in the temperature can reduce heating efficiency, whereas the expected boiler temperature is comparatively constant. Besides, high VM enhances the combustion efficiency of the FPs. However, the extreme increase in VM leads to abridged duration (Kristanto & Wijaya, 2018). On the other hand, ultimate analysis is also important to predict the level of emission of FPs. The efficient combustion of FPs is primarily affected by the quality of the FPs (physical and chemical qualities) and the design of the combustion system. Many studies dealt with the combustion analyses of SCG and other biomass residues (Kang et al., 2017; Limousy et al., 2013; Nosek et al., 2020; Pilusa et al., 2013; Potip & Wongwuttanasatian, 2018). However, it has been observed that no studies dealing with DSCG were found. This necessitates the investigation of DSCG in boilers in future studies.

Kang et al. (2017) investigated the combustion and emission characteristics of a small boiler system (6.5 kW) fed with 100% dried SCG. The combustion chamber type was crucible with primary and secondary air supply and heat exchanger. A primary and secondary air supply chamber was selected for the combustion of SCG using this prototype and was stable over a longer time. SCG was supplied to the burner at a rate of 1.167 kg/h. The total combustion duration was 146 min. At this time, the temperature of the water in the storage tank reached more than 75 °C. Main findings showed that the concentration of O_2, CO, and NO_x in the flue gas was about 17.8%, 643 ppm, and 163 ppm, respectively. It was observed that the emission of NO_x from SCG was higher than the emission of NO_x from pine, citrus pectin waste, and peach stones. This is attributed to the higher N content in SCG than other biomass (Table 2). The high N content is attributed to the existence of protein and caffeine in SCG (Bok et al., 2012).

Limousy et al. (2013) experimentally measured the combustion and boiler efficiency, emissions of gaseous yields, and particulate matter of biomass boiler (residential) fired with 100% SCG, 100% pine sawdust, and (50% SCG and 50% pine sawdust) pellet.

The combustion efficiency (q %) of FPs was been calculated using the following formula:

$$q = 100 - \left(\frac{q_a + q_b + q_r}{\mathrm{LHV}}\right) \times 100 \tag{12}$$

where, $q_a \equiv$ Thermal heat loses in the flue gas (kJ/kg), $q_b \equiv$ Chemical heat loses in the flue gas (kJ/kg), $q_r \equiv$ Heat losses due to combustible constituents in the residue (kJ/kg)

$$q_{\mathrm{a}} = \left(T_{\mathrm{g}} - T_{\mathrm{a}}\right) \times \left[\left(\frac{\mathrm{C_{pmd}} \times \left(\mathrm{C} - \mathrm{C}_r\right)}{0.536 \times \left(\mathrm{CO} + \mathrm{CO_2}\right)}\right) + \left(\frac{\mathrm{C_{pmH2O}} \times 1.244 \times \left(9\mathrm{H} + W\right)}{100}\right)\right] \tag{13}$$

$$q_{\mathrm{b}} = \left(\frac{12644 \times \mathrm{CO} \times \left(\mathrm{C} - \mathrm{C_r}\right)}{0.536 \times \left(\mathrm{CO} + \mathrm{CO_2}\right) \times 100}\right) \tag{14}$$

$$q_{\mathrm{r}} = \left(\frac{335 \times b \times R}{100}\right) \tag{15}$$

$$\mathrm{C_r} = \left(\frac{b \times R}{100}\right) \tag{16}$$

where $T_{\mathrm{g}} \equiv$ Exhaust gas temperature (K), $T_{\mathrm{a}} \equiv$ Air temperature (K), $C_{\mathrm{pmd}} \equiv$ Specific heats of dry flue gases (kJ/K m^3), $C_{\mathrm{pmH2O}} \equiv$ Specific heats of water vapor (kJ/K m^3), CO ≡ Content in dry flue gas (% volume), $\mathrm{CO_2} \equiv$ Content in dry flue gas (% volume), $b \equiv$ Combustible constituent in the residue (% mass), $R \equiv$ Residue mass passing through the grate (% mass), $\mathrm{C_r} \equiv$ Carbon content of the residue passing through the grate, C ≡ Carbon content of the fuel.

The boiler efficiency (BE %) was calculated using the following formula:

$$\mathrm{BE}\ \% = \left(\frac{P_{\mathrm{n}}}{P_{\mathrm{c}}} \times 100\right) \tag{17}$$

$$P_{\mathrm{n}} = \left[Q_{\mathrm{w}} \times C_{\mathrm{pw}} \times \left(T_{\mathrm{out}} - T_{\mathrm{in}}\right)\right] \tag{18}$$

$$P_{\mathrm{c}} = \left[Q_{\mathrm{comb}} \times \mathrm{LHV}\right] \tag{19}$$

where, BE% ≡ Boiler efficiency, $P_{\mathrm{n}} \equiv$ Nominal useful power heat of the boiler (kW), $P_{\mathrm{c}} \equiv$ Calorific mass flow (kW), $Q_{\mathrm{w}} \equiv$ Water mass flow (kg/s), $C_{\mathrm{pw}} \equiv$ Water calorific capacity (kJ/kg K), $T_{\mathrm{out}} \equiv$ Outlet temperature of water from the boiler heat exchanger (K), $T_{\mathrm{in}} \equiv$ Inlet temperature of water from the boiler heat exchanger (K), $Q_{\mathrm{comb}} \equiv$ Biomass mass flow (kg/s).

The summary of the results is tabulated in Table 4. It has been observed that the highest emissions of CO, NO, $\mathrm{NO_2}$, volatile organic compounds ($\mathrm{VOC_S}$), and particulate matter (PM) were recorded for SCG. However, the combustion of a blended sample (50% SCG/50% pine sawdust) resulted in a remarkable reduction in all emissions. These reported results were close to those obtained for wood pellets. Similarly, both boiler and combustion efficiencies increased remarkably with the blend. The study concluded that 50% SCG/50% pine sawdust blends meet French Standards (NF agro-pellets).

Kristanto and Wijaya (2018) indicated that the peak temperature of combustion for SCG and CS (stoichiometric) ranges between 500 and 600 °C. The experiment indicated that the resulting temperature resulting from the combustion of 100% SCG remains constant for a few minutes, and drops dramatically in around the seventh minute. In comparison, the addition of CS to SCG results in a slight decrease in temperature and tends to be constant. This phenomenon is attributed to the density as it directly affects combustion efficiency.

Table 4 Combustion and boiler efficiencies and particle emissions of SCG, pine sawdust, and their 50/50% blend (Limousy et al., 2013)

Sample	Combustion efficiency (%)	Boiler efficiency (%)	CO	NO	NO_2	VOCs	PM
SCG	86.3	64.1	1785[a]	178[a]	28[a]	539[a]	1071[a]
			3069[b]	328[b]	79[b]	530[b]	1472[b]
Pine sawdust	90.8	84.3	153[a]	45[a]	0[a]	330[a]	104[a]
			263[b]	84[b]	0[b]	324[b]	143[b]
Blend[c]	91.9	83.5	353[a]	193[a]	8[a]	209[a]	310[a]
			606[b]	355[b]	22[b]	205[b]	426[b]

[a]Expressed in ppm (v) for gases and in mg/Nm^3 for particles at 13% O_2

[b]Expressed in mg/Nm^3 at 10% O for gases and particles)

[c]50% SCG/50% pine sawdust

VOCs Volatile organic compounds, *PM* Particulate matter

As per the previous section's discussion, CS is denser than SCG in part because SCG has a higher porosity than CS. It was concluded that FPs with 20% CS, 75% SCG, and 5% artificial adhesive are the best FP among other samples as they are the most durable FPs and exhibit the most constant flame and reduction in temperature. These results conformed to German pellet standard DIN 51,731. Moreover, the addition of CS could enhance the energy generated because of the pellet density increase. Therefore, adding SCG and CS together does not meaningfully disturb the value of energy. The reduction in temperature by mixing SCG with CS is comparatively constant due to pellet density effects as well as combustion efficiency.

The authors concluded that the addition of CS beyond 20% is not recommended due to the following limitations:

(1) An increase in mixing CS with SCG beyond 20% results in an increase in NO_x emission. This is because of the higher nitrogen content in CS as compared to SCG (Table 3).
(2) An increase in mixing CS with SCG beyond 20% results in an increase in the ash content in the FPs (Table 2).

Nosek et al., 2020 investigated the potential of using SCG as an FP along with wood sawdust. The authors reported the same observation as Kristanto & Wijaya, 2018 that the boiler efficiency decreased with the combustion of 100% SCG. The boiler heat power with 100% SCG was 3.98 kW, four times lower than that associated with wood pellets (15.18 kw). Authors attributed this to incomplete combustion due to the low pellet strength of SCG. This has remarkably increased carbon monoxide (CO) emission as compared to 100% sawdust. However, CO emission with 100% SCG combustion (2248 mg/m^3) still satisfies the STN EN 303-5 2012 standard which specifies a maximum concentration of CO of about 3000 mg/m^3. However, both CO_2 and NO_x emissions were the lowest with 100% SCG. It was found that FPs consisting of 50% SCG and 50% pine sawdust (wt%) yield similar parameters of combustion (boiler efficiency and emissions) for sawdust pellets and meet the NF agro-pellets standard. However, a sample consisting of 30% SCG and 70% pine sawdust (wt%) seems to provide the best

FP as compared to other samples when considering the measured emission and boiler performance.

Potip and Wongwuttanasatian (2018) tested FPs prepared by mixing SCG with crude glycerol which acts as a binding agent at two different compositions: 95% SCG + 5% crude glycerol and 90% SCG + 10% crude glycerol. Results were compared with a sample that consists of 100% SCG. Experiments were conducted at four theoretical air supplies of 550, 700, 850, and 1000%. The equations for the combustion of 100% theoretical air and 100 g SCG and 1 mol crude glycerol are:

$$\begin{aligned}\left(4.551\mathrm{C} + 3.295\mathrm{H_2} + 1.088\mathrm{O_2} + 0.142\mathrm{N_2}\right) + 5.111\left(\mathrm{O_2} + 3.76\mathrm{N_2}\right) \\ + \mathrm{Ash} \rightarrow 4.551\mathrm{CO_2} + 3.295\mathrm{H_2O} + 19.359\mathrm{N_2} + \mathrm{Ash}\end{aligned} \tag{20}$$

$$\mathrm{C_3H_8O_3} + 3.5\left(\mathrm{O_2} + 3.76\mathrm{N_2}\right) \rightarrow 3\mathrm{CO_2} + 4\mathrm{H_2O} + 13.16\mathrm{N_2} \tag{21}$$

Results revealed that FPs prepared from 90% SCG + 10% crude glycerol and 850% theoretical air provided the maximum peak temperature of 533.4 °C along with a combustion rate of0.20 g/s, respectively. Emission characteristics of 90% SCG + 10% crude glycerol and 850% theoretical air were 1262.3 mg/m^3 (CO), 19% (CO_2), 38.1 mg/m^3 (NO_x), 270.3 mg/m^3 (HC), 0 mg/m^3 sulfur dioxide (SO_2), and 20.6% (O_2). These results were in accordance with the standards specified by the Irish BioEnergy Association (IRBEA) for CO (1500), NO_x (200), and SO_2 (200), respectively. It was also found that 800–1000% theoretical air resulted in the best combustion for densified FPs (Kristanto & Wijaya, 2018).

Pilusa et al. (2013) investigated the emission characteristics of eco-fuel FP consisting of 32% SCG, 23% coal fines, 11% sawdust in a ceramic-lined stove at a burning rate of 2 g/min. The emission results of CO, CO_2, hydrogen sulfide (H_2S), nitric oxides (NO), nitrogen dioxide (NO_2), ammonia (NH_3), and SO_2 emissions are tabulated in Table 5 and compared against the standard of the *Occupational Safety and Health Agency (OSHA)*. The results indicated the emissions characteristics of the developed of eco-fuel FP conform to the OSHA. The CO_2 produced is 2 892 times the CO. The high CO_2 concentration indicates the complete combustion resulting in CO been oxidized to less toxic CO_2. This is advantageous since the CO_2 is less toxic than CO emissions.

Table 5 Emission characteristics of eco-fuel FP and comparison with OSHA (Pilusa et al., 2013)

Emission	FP	OSHA max-limit (ppm)
CO_2	21,332	5000
CO	73.78 ppm	50–200
NO_2	2.73 ppm	5
NO	1.34 ppm	25
NH_3	–	50
SO_2	3.67 ppm	5
H_2S	4.32 ppm	20
Ash	28.4%	–

5 Feasibility and advantages of SCG as FPs

Given the presented fuel properties and combustion qualities of the SCG in the previous chapter, SCG and DSCG are considered as potential feedstocks for FPs production for the following important reasons:

(1) High C content and HHV: It has been proven that SCG and DSCG have high C content (> 50%) (Table 3). This means for each 100 kg of SCG and DSCG, there will be at least 50 kg carbon. Thus, their HHVs are higher compared to other biomass residues (Table S1). This supports the combustion feature of SCG and its advantages over various kinds of feedstocks for biomass wood pellets.
(2) Lower ash content: Additionally, SCG and DSCG have also lower ash contents (Table 2) than many forestry residues and agricultural wastes.
(3) High lignin content: SCG features a high amount of fiber content. The average content of hemicellulose (30–40%), cellulose (8–13%), and lignin (25–33%), respectively. Lignin plays the role of additive during the pelletizing process. Without lignin, pelleting might be hard to mold and easy to break.
(4) No need for crushing: Due to the small pellet size of both SCG and DSCG (Sect. 4.1), there is no need for crushing.
(5) Short growing period of raw materials: The growing period of the coffee tree is about 4 years which is much shorter than the growing period of many forestry and agricultural residues.
(6) Clean property of the feedstock: Unlike forestry residues and agricultural wastes, SCGs as by-products are clean and have no harm to the human body.
(7) Save the environment: Production of FPs from recycled SCG and DSCG can contribute to saving the environment as the direct disposal of SCG can add a decontamination burden.
(8) FPs can be produced locally from the SCG and thus reduce reliance on virgin timber or imported pellets.

6 A business model of the SCG as FPs

Bio-bean company, a UK-based company, is one of the leading global companies that achieved great progress in valorizing the SCG into various types of biofuels and value-added products. The company is a pioneer in producing FPs (Bio-bean, 2020) and coffee logs (Bio-bean, 2021b) from the SCG. The net calorific value of the produced FPs from the SCG was found ≥ 15% higher than standard timber pellets along with low moisture content, high bulk density, high ash melting point, and good durability making SCG a high-performing alternative FPs. Additionally, the company highlighted that their efficient burn profile saves businesses money, requiring reduced volume and therefore fewer deliveries to achieve the same energy output as wood pellets. The company indicated SCG FPs are Sustainable Fuel Register (SFR) accredited, and therefore meet the criteria for renewable heat incentives (RHI) approval (Bio-bean, 2020). Figure S4 shows the images of the pelletizer machine and FP samples from bio-bean.

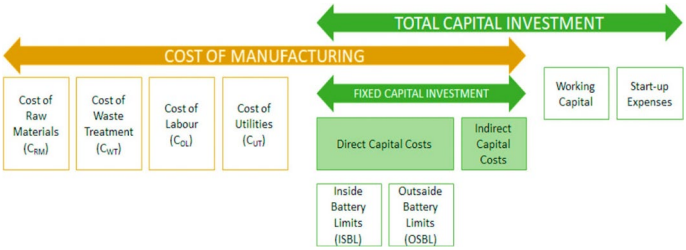


Fig. 8 Calculation of the total capital investment and cost of manufacturing (Cristóbal et al., 2018)

7 The techno-economic and profitability assessment of the SCG as FPs for the biorefinery

The techno-economic and profitability assessment of the SCG as FPs for the biorefinery can be divided into the calculation of total costs (including total capital investment and costs of manufacturing (Fig. 8) and the calculation of revenues and profitability ratios (Cristóbal et al., 2018).

To evaluate the feasibility of the proposed FPs production from the SCG, two profitability ratios must be taken into consideration; Return on Investment (ROI) and payback period. ROI and the payback period can be calculated as follows (Cristóbal et al., 2018):

$$\text{ROI}(\%) = \frac{\text{Annual net profit}}{\text{Total capital investment}} \tag{22}$$

$$\text{Payback period(years)} = \frac{\text{Total capital investment}}{\text{Annual net profit}} \tag{23}$$

The ROI is the ratio of gains to cost and it is measured in %, per period. ROI measures the rate of return on money invested in the biorefinery. A positive ROI means that the investment gains compare favorably to the costs; hence, the larger the ROI, the better the biorefinery economic performance. On the other hand, the payback period is the length of time that it takes for the gains from the investment to equal the costs. Namely, it measures the time it takes for an investment to pay for itself. The smaller the payback period, the better the biorefinery economic performance.

On the other hand, the net present value (NPV) assessment allows to point out how the biorefinery can be profitable and, on the other side, to stress that the high sales incomes, due to the retail model assumed, should be better investigated in terms of its acceptance by the customers and real feasibility. NPV can be calculated by the following formula (Bottani et al., 2019):

$$\text{NPV} = \sum_{t=1}^{n} \frac{R_t}{(1+i)^t} \tag{24}$$

where, R_t = Net cash inflow–outflows during a single period t, i = Discount rate or return that could be earned in alternative investments, T = Number of timer periods.

Although many studies reported the potential applications of SCG as FPs, however, no studies considered the integration of these ideas with a comprehensive feasibility study. It has been demonstrated that the cost of the raw materials (SCG) is almost neglectable when

compared to the costs of the production including transportation, manpower, energy, and facility costs. In particular, the manpower and transportation (for SCG collection) costs are the most impacting costs to be deeply considered and analyzed (≈ 35–40% and 20%, respectively, on average) when developing an FP biorefinery from the SCG. Manpower is the greatest cost, approximately doubling transportation costs;

A very recent study conducted by Bottani et al., 2019 stated that the adoption of Information and Communication Technologies (ICTs) can dramatically improve the efficiency of the whole SCG FPs system, reducing the production costs, and thus keeping the proposed thermal valorization economically feasible. To confirm this point, a case study evaluating four different scenarios (two including ICT system and two without an ICT system, considering two different pellet compositions, 50% SCG/50% pine sawdust and 98% SCG), has been conducted in 4 different plants in Italy to demonstrate that better results can be achieved with the deployment of ICT tools. A detailed logistics model adopting an ICT tool has been developed to evaluate the total cost associated with the collection and valorization of SCG as FPs. The results are tabulated in the supplementary section (Tables S4 and S5) considering two different pellet compositions, 50% SCG/50% pine sawdust and 98% SCG, respectively.

The profitability of the investment is strictly depending on the selling price of the SCG FPs. The bigger the quantity of FPs directly sold to end users, the more profitable the investment. Accordingly, the choice of spreading the production capability over FPs plants located in different regions is useful. On one hand, it minimizes the overall transportation costs by reducing the length of the routes, and on the other side, it enables the possibility to sell the FPs to the end users in a shop corner in the same facility. As it can be seen from Tables S6 and S7, manpower shares the greatest cost amongst all the analyzed scenarios, approximately doubling transportation costs. The adoption of the ICT technologies may lead to some savings in which it would make a "2 operators over 3 shifts" scenario feasible. Through this approach, operators working during the daytime shifts can manage the plant and prepare it for the night shift, which will be unattended but remotely supervised; in another hypothesis, only one plant operator could be involved for both daytime shifts thanks to the integration of his work with the truck driver, who may supervise the production and check the plant every time the vehicle is unloaded. Looking at the results of this study, the model demonstrates the economic advantage of collecting SCGs and using them for producing FPs that can be sold on the market.

8 Conclusions and future prospects

The environmental concerns of reducing harmful emissions such as carbon dioxide (CO_2) and oxides of nitrogen (NO_x) have promoted the world to search for environmentally friendly fuels. Biomass residues and bio-waste are promising resources to substitute fossil fuels since they do not compete with food, and they are renewable, besides being widely available at low prices. Furthermore, they can be reasonable solutions to dispose of biogenic waste from the environment.

FPs are solid, densified, sustainable, and good-quality biofuels having a form of granules in the shape of spheres or cylinders and a size of a few centimeters. FPs are consumed in two main different market sectors: the electricity and power generation sector and the residential and district heating sector. The qualities of FPs are assessed against some international standards such as DIN 51,731, ÖNORM M 7135, DIN EN

14,961, Pellet Fuels Institute (PFI), Agro +, and Agro. The physical and chemical qualities of FPs play an important role in ensuring a high combustion efficiency.

FP production increased over the past 15 years from 6–7 million tonnes in 2006 to 14.3 million tonnes in 2010, 24.93 million tonnes in 2013, 34.39 million tonnes in 2015 and amounting to 55.70 million tonnes in 2018. This represents an increase of 14.13 and 36.67% compared to 2017 and 2016, respectively. The global market for FPs is steadily growing due to the increasing demand to replace coal. It is believed that this market is growing 14% a year on average since 2011. However, such plans are highly affected by sustainability and the availability of biomass. Therefore, various kinds of bio-waste, such as from the food industry, can be valorized through cutting-edge recycling technologies to produce agro-pellets. This will significantly reduce the amount of waste returned to the environment and solve the problem of organic waste disposal. The global production and consumption of coffee are increasing annually. Therefore, a valuable organic waste that contains large amounts of organic compounds known as SCG is generated in huge amounts daily. SCG amounts to approximately 40% of the total CB mass. This huge potential makes SCG an excellent raw material for biorefineries aiming to generate biofuels and added-value products, thus contributing to enhancing the circular bioeconomy of many nations. Valorization of SCG into FPs is one of these potential recycling options. Most of the recent studies concluded that SCG should be split into two main components with the lipid/extracts from SCG known as SCGO and the exhausted coffee grounds remain after the extraction process known as DSCG. FP characteristics significantly influence the design and operation of biomass combustion systems. These characteristics include pellet size, strength, and durability, moisture content, calorific value, energy density, bulk density, morphology, and porosity. SCG and DSCG are promising resources to produce FPs. This is attributed to their excellent properties compared to other crops and biomass residues, such as higher heating value (HHV), etc.

However, a literature survey indicates that the burning of 100% SCG FPs in boilers is not feasible. This is because SCG emits higher NO_x compared to many other biomasses. This is attributed to the higher N content in SCG. Moreover, a rapid drop of temperature caused by the incomplete combustion of SCG was reported leading to lower boiler combustion efficiency, boiler heat power, which is four times lesser than wood pellets and remarkably increased carbon monoxide emissions. One of the main causes of these phenomena has been attributed to the low pellet strength and bulk density of SCG.

Therefore, it has been concluded that mixing SCG with other biomass offers optimum boiler efficiency and emissions. Some of the reported FPs prepared by blending SCG with other biomasses at different percentages are:

(1) 75% SCG, 20% coffee silverskin.
(2) 30% SCG and 70% pine sawdust (wt%).
(3) 90% SCG + 10% crude glycerol.
(4) 32% SCG, 23% coal fines, 11% saw.
(5) 50% SCG + 50% pine sawdust.

It shall be noted that no studies dealing with the combustion and emissions of DSCG were reported in this review. Therefore, investigation of DSCG in boilers in future studies is essential to assess the potential of this industry. This is because valorization of DSC offers a particularly good pathway toward improving the economics of this industry.

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