Biorefining of Waste Coffee Grounds: Turning an Environmental Problem into an Opportunity

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Abstract. The work presented herein proposes to turn the growing environmental threat posed by coffee waste disposal into an opportunity by proposing a number of valorization routes of waste coffee grounds (WCG) through biorefining. In this process, various steps were studied: drying of WCG on small and medium scales, coffee oil extraction from small to large scale, conversion of coffee oil into biodiesel, and suggestions of designs for the recycling of de-oiled WCG into briquettes. Drying was found to be best performed using an elevated hot air tunnel rather than an electric pan or a motorized stirrer. Solvent extraction of coffee oil from dried WCG on a 2000 g scale afforded a 9.2 wt% yield and a solvent recovery yield of 94 vol%. This oil was successfully converted into fatty acid methyl esters biodiesel with a 58% yield. Several original and cost-effective designs were tested, some of which being readily reproducible in most laboratory settings.

1. Introduction
With over 2.25 billion cups drunk every day, coffee is one of the most popular beverage and traded commodity in the world [1]. Unfortunately, waste coffee grounds (WCG) are rarely recycled, the common practice being to dispose of them in municipal landfills where they eventually degrade to release methane, a very potent greenhouse gas that contributes to climate change [2]. One effective solution to turn this problem into an array of opportunities is biorefining. Biorefining is defined as the sustainable processing of a biomass or a combination of different types of biomasses to produce a spectrum of marketable products and energy at a potentially better economic return [3-6]. Figure 1 depicts biorefining of WCG with a non-exhaustive list of value-added products. WCG are the waste resulting from coffee brewing. After solvent extraction of WCG, two new products are obtained: coffee oil and de-oiled WCG. Coffee oil can be converted into cosmetics, soap [7-8], or biodiesel [9-10]. As for de-oiled WCG, they have a large span of applications; they can be employed as soil conditioner [11], used for mushroom production [12], fermented into bioethanol [13] or biogas [14], converted into biochar and bio-oil [15], or compressed into briquettes or heating pellets [16-17]. Another important way to valorize coffee oil is its biorefining into bioactive molecules. Coffee contains more than 1000 chemicals [18-19], many of which have interesting physiological, biological and pharmacological effects; e.g. antioxidants like caffeine. One important part of coffee is its lipid fraction which contains a diverse set of chemicals [20]. Examples include triglycerides, which are useful to make soap or biodiesel, and diterpenes such as Cafestol and Kahweol that are the subject of extensive medical research because of their high anticarcinogenic activity [21].
2. Materials and methods

n-Hexane was 99% pure and supplied by Sigma Aldrich. Methanol was 99.5% pure and supplied by VWR. Phosphoric acid used as a 5.4 M (30 wt%) solution in methanol was supplied by Acros Organics, USA. Sodium methoxide as a 5.4 M (30 wt%) solution in methanol was supplied by Acros Organics, USA. Phosphoric acid used in the washings of biodiesel was made from 85% phosphoric acid supplied by Fluka, Switzerland. Oil and biodiesel samples were centrifuged in a 5500 rpm Hettich centrifuge, model EBA 30 or in a 2700 rpm Clay Adams/Becton Dickinson centrifuge, model 420227. Vacuum distillations and vacuum filtrations of coffee solutions were performed by means of piston-powered vacuum pump (0.85 atm, 38 L/min). Water content in waste coffee grounds was determined accurately with a VWR oven, model 1300U. Occasionally, samples were mixed with hexane by means of a VELP scientifica vortex shaker, model Zx3. Ultrasonication was performed with a Branson ultrasonic cleaner, model 1210-EMT (50W, 47 kHz). Gross Calorific Values were measured by means of a Parr 1344EE oxygen bomb calorimeter and medical grade oxygen supplied by Maghreb Oxygen, Morocco. The motorized stirrer made in-house was fitted with a 0.09kW DRdrives motor, model MS5624, coupled with a Motovario gear reducer, model NHRV/030, operating at a gear ratio of 30 and a gear output speed of 47 rpm.

3. Results and discussion

3.1 Drying methods

The very first step required prior to the extraction of coffee oil from WCG is drying. Indeed, experiments performed in our lab showed that typical water content in WCG is as high as 64 wt%. This substantial amount of water hinders solvent extraction of coffee oil, due to the immiscibility of water with the non-polar extraction solvent (n-hexane). Preliminary work in our lab showed that the oil extraction yield drops by 60% when performing the extraction on wet WCG instead of the dried form (Table 1, entries 1-2). Another significant advantage of drying WCG is the inhibition of microbial activity that leads to the formation of mold (Figure 2), which in turn enables the safe storage of WCG over a prolonged period of time [22].
Different drying methods were tested and compared in order to identify the most suitable drying method for wet WCG. Criteria selected in the comparison were drying time, drying temperature, labor intensiveness, energy consumption and scale.

**3.1.1 Electric pan.** A 30 cm 1500W round electric pan with nonstick coating and temperature control was tested (Figure 3). Though this pan’s primary purpose is the stir frying, braising and baking of food, we tested its potential to serve as a drying system of WCG. In drying experiments, its thermostat was adjustable from 200 to 450°C. A drying time of 15 min was found to be sufficient to get dry WCG, providing there is continuous stirring on the tray by the operator to prevent burning.

**3.1.2 Hot air tunnel.** The second drying system that was tested is a hot air tunnel, with WCG laid on a sheet of cardboard placed on the floor (Figures 4a-4b). This tunnel forms a rectangular cuboid that is 105 cm wide, 150 cm long and 90 cm high, thus providing a surface area of 1.58 m² for WCG. The flow of hot air in the system is created by a total of 5 electric fans, three of which are fan heaters and two are regular fans. This hot air is generated on both sides of the cuboid; one side is heated by means of 2 fan heaters, and the other using one fan heater and two regular fans. As one can see from Figures 4a-4b, the top of the cuboid, as well as two of its faces are closed with cardboard sheets, a material that has the advantage of being a good insulator that is inexpensive and readily available. Air temperature inside the tunnel and in the room were measured to be 29°C and 19°C respectively. Drying time was determined to be 24 h, with stirring of the WCG every 4 h.

![Figure 2. Mold generated on moist WCG.](image1)

![Figure 3. Dry WCG after 15 min of heating and stirring on an electric pan.](image2)

The hot air tunnel system was then enhanced by elevating the cardboard sheet on which WCG were laid (Figures 4c-4d). The rationale behind this modification was the fact that water condensation was found to form beneath the cardboard sheet laid on the cold floor, thus making the cardboard damp and the process unnecessary lengthy. To remedy this problem, WCG were raised by 45cm, several small perforations were made all over the sheet, a 20 cm hole was made at the center of the sheet, and
locations of the fans were rearranged. Two fan heaters were placed on one end of the cuboid to generate air flow from one side to the other. A third fan heater was placed underneath the sheet to blow hot air towards holes in the sheet and keep the cardboard warm and dry. Two additional regular fans were also employed in conjunction with fan heaters to increase air flow. Air temperature inside the tunnel, underneath the tunnel and in the room were measured to be 31°C, 27°C and 21°C respectively. Drying time was determined to be 8h, with stirring of the WCG after 4h.

3.1.3 Motorized stirrer. The last drying system to be tested was a motorized stirrer fitted atop a modified stainless-steel 12L pressure cooker vessel (Figure 5).

![Figure 5. Motorized stirrer utilized to dry WCG.](image)

The electrical motor rotates a shaft at the moderate speed of 47 rpm to ensure WCG inside the vessel are in constant motion, thus speeding up the loss of water vapor. To prevent burning of WCG sticking to the hot walls of the vessel, the stirrer’s dimensions are such that all the walls are always scraped. Heating of the system is gas powered and controlled with the valve of a portable stove.

3.1.4 Comparison of drying methods. The electric pan’s main advantage is that it can dry rapidly; however, its working temperature is too high which causes thermal degradation of WCG that is detectable by a burnt smell. Operating the pan requires a lot of labor because of the constant manual stirring required to prevent WCG from burning on the very hot pan. Finally, the pan is not suitable for large scale extraction since its output is about 600 g of WCG per hour. Benefits of the hot air tunnels is their capacity to dry large quantities of WCG with minimal labor. The elevated tunnel has the additional advantage of drying faster and at a higher temperature than its regular counterpart thanks to its better insulation and hot air flow. Finally, the motorized stirrer’s appealing feature is its lower energy consumption due to its gas-powered heat generation. This being said, its long drying time and high temperature which is hard to control, does not make it the best option. Based on the pros and cons summarized above, the most suitable drying system for WCG is the elevated hot air tunnel.

3.2 Solvent extraction of coffee oil

3.2.1 Preliminary small-scale coffee oil extraction. Extraction of the lipid fraction of dried WCG was done by means of an organic solvent in a batch system mode. Typical setups employed in lipid extraction from biomass at the laboratory scale consist in using a Soxhlet apparatus [10,23] in which a solvent such as n-hexane, isopropanol, ethanol or methanol is refluxed [10,24]. However, because the Soxhlet apparatus is inherently limited by its relatively small size, it was decided to perform coffee oil extraction on a larger vessel, with n-hexane as a solvent. Literature reports that oil content in coffee beans is in the range of 10-15 wt% depending on the coffee variety [19,25]. Determination of the oil content in our own WCG collected from the University cafeteria was done by running experiments with variable drying methods, sonication time and extraction time (Table 1). These preliminary experiments were run on a small scale, with 100 g of WCG.
Table 1. Conditions tested in coffee oil extraction from WCG (solvent: hexane, T=69°C).

| Entry | Type of WCG | Drying method | Sonication time (min) | Extraction time (min) | Extraction yield (wt%) |
|-------|-------------|---------------|-----------------------|-----------------------|------------------------|
| 1     | wet         | none          | 5                     | 30                    | 4.4                    |
| 2     | dry         | oven, 100°C, 4h | 10                    | 30                    | 10.9                   |
| 3     | dry         | oven, 100°C, 24h | 10                   | 90                    | 7.9                    |
| 4     | dry         | oven, 150°C, 2h | 0                     | 30                    | 10.3                   |

The poor extraction yield in Table 1, entry 1 (4.4 wt%) shows the importance of performing coffee oil extraction on dry WCG rather than wet WCG, thereby highlighting the importance of the drying process in coffee recycling. The best conditions identified to get an optimal extraction yield (10.9 wt%) are given in Table 1, entry 2: drying at 100°C for 24h, sonication time of 10 min, and an extraction time of 30 min with n-hexane at 69°C.

3.2.2 Medium-scale coffee oil extraction. Once extraction conditions of coffee oil were determined on a 100 g scale, a new setup was designed to perform the extraction on the scale of 2000 g of WCG, i.e. a scale up by a factor of 20. To this end, large extraction vessels fitted with water condensers were constructed and tested (Figures 6a-6b). It was found that using a copper Liebig condenser was not appropriate as large amounts of hexane vapor could reach the top of the condenser, even when increasing the cold-water flow. This drawback needed to be addressed since the loss of hexane makes the extraction costly and environmentally harmful. Replacing the Liebig condenser by a glass Dimroth condenser, which has a greater cooling capacity due to the higher surface area, solved this problem. Once the reflux in the extraction unit was complete, the oil solution in hexane was transferred to a desolventation unit to get solvent-free coffee oil (Figures 6c-6d). The goal of this unit is also to recover the expensive solvent which will be used again in the next batch.

Figure 6. Medium-scale coffee oil extraction and desolventation units. (a) extraction unit with 12L stainless steel vessel, electrical heating and copper Liebig condenser. (b) extraction unit with 18L aluminium vessel, gas-powered heating and glass Dimroth condenser. (c) desolventation unit heat at 60°C under vacuum. (d) desolventation unit connected to a rotary evaporator for a rapid and efficient distillation.

The desolventation vessel was built in-house by perforating the lid of a 12L stainless steel pressure cooker and welding into it a copper pipe which was insulated at a later stage. To take advantage of the rotary evaporator available in our facilities, the desolventation vessel was heated and connected to vacuum via the rotary evaporator. This combination allowed a quick distillation of the solvent, while at the same time preserving the quality of the coffee oil extracted by avoiding thermal degradation.
throughout the process. The desolventation unit was tested on a 2000 g scale. Coffee oil was extracted in a multi-step extraction, resulting in a total of four oil/hexane fractions. Each fraction was obtained by refluxing the WCG-hexane suspension in the extraction unit (Figures 6a-6b) at 69°C for 1 h. Transfer of the resulting oil solution and its subsequent distillation in the desolventation unit was done one fraction at a time. At the end of each distillation, the amounts of recovered hexane and oil were recorded. In this system, the overall coffee oil extraction yield was found to be 9.2 wt%, which is lower than the 10.9 wt% found in the small extraction system (Table 1, entry 2). This is most likely due to the fact that, in the medium-scale extraction, ultra-sonication was not utilized. Moreover, it is well possible that more than four fractions should have been collected in the medium-scale extraction to increase the extraction yield. A solvent recovery yield of 94 vol% was attained, which proves that the desolventation unit we developed loses very little solvent. Reaching a higher percentage in the solvent recovery should be feasible by using a dry ice trap placed before the vacuum pump of the rotary evaporator.

3.3 Conversion of coffee oil into fatty acid methyl esters biodiesel

As shown in Figure 1, coffee oil can be valorized by blending with other oils to become a cosmetic oil, saponified to become soap, or transesterified to become biodiesel. The latter route was done using methanol as alcohol (30 equivalents) and sodium methoxide as catalyst (2.5 wt% solution in methanol) at 60°C for 1 h. The reaction afforded coffee fatty acid methyl esters biodiesel with a yield of 58%. Characterization of the oil and the biodiesel obtained is given in Table 2.

| Table 2. Characterization of coffee oil and its resulting biodiesel. |
|---------------------------------------------------------------|
|                 | Free fatty acid (%) | Density at 15°C (kg/m³) | Gross calorific value (kJ/g) |
| Coffee oil      | 2.01               | 842                      | 39.62                       |
| Coffee biodiesel| 0.23*              | 832b                     | 38.78                       |

* EN14214 limit: 1%  
  b EN14214 limit: 860-900

Results obtained show that the coffee biodiesel made meets European fuel specifications EN14214 in terms of free fatty acid content, but not in terms of density. This problem can be easily addressed by blending the coffee oil extracted with a denser vegetable oil in a small proportion before turning it into biodiesel.

4. Designs

4.1 Design of a large-scale coffee oil extraction unit

Based on the medium-scale coffee oil extraction system successfully tested that is capable of processing 2000 g of dry WCG, a large-scale system was designed (Figure 7). This larger system is designed to process 100 kg of material, thus making it suitable for a small recycling plant.

Tank A is first opened, loaded with 100 kg of dried WCG and closed. Next, a pre-determined volume of hexane is added by opening valve V1 and the mechanical agitator in tank A is turned on. The tank is heated by means of a stove and a built-in ultrasonic probe on the side of the tank is activated to accelerate the diffusion of the oil in the solvent. As the solvent is refluxed, vapors of hexane go through valve V2a, reach the condenser and return to tank A. Once the first extraction cycle is complete and heating is paused, valves V3a, V3b and V3c are opened to transfer the coffee oil solution in hexane from tank A to tank B which serves as desolventation unit. Two fine woven wire meshes are placed inside tank A. One at the bottom to prevent WCG from reaching valve V3b, one on the side of the tank to prevent WCG from reaching valve V3a. In case these meshes get both clogged, V3b is closed, V6 is opened, V3a semi-opened, and compressed air is injected into the pipes. To start the next extraction cycle, valves V3a and V3b are closed, and V1 is opened to introduce more hexane in tank A. Once all extraction cycles are finished, V2a and V2b are closed, V4a, V4b and V4c are opened, a high vacuum pump is turned on and mild heating is applied on tank B to start vacuum
distillation of the solvent. During this distillation, reclaimed hexane is stored in tank D. Once condensation of the recovered hexane is no longer detectable, distillation can be stopped, and pure coffee oil can be transferred out of tank B via V5. This extraction system is designed to be operated safely as proven by temperature and pressure sensors placed in tanks A and B. In the event WCG loaded in tank A require additional drying before starting oil extraction, the system is devised to heat WCG in tank A and collect water vapor in tank C via valve V4b.

Figure 7. 2D sketch of the coffee extraction unit.

4.2 Design of a medium-scale coffee log unit
As shown in Figure 1, de-oiled WCG can be valorized into a large number of products, the simplest of which is coffee briquettes which result from the compression of the grounds in a press. We propose here two designs for the press; a manual press and a hydraulic press. Figures 8a-8b shows our designed manual press. It is composed of a perforated cage meant to be filled with de-oiled WCG mixed with a binding agent. Once filled, a perforated mobile plate attached to a press arm is moved by the operator to squeeze the grounds and compact them to form a briquette.
Testing of this press in the lab showed that, because of the low pressure applied, de-oiled WCG must be mixed with a binding material such as glycerin prior to briquetting to improve mechanical properties. Another suggestion of improvement is to weld fine woven wire mesh on the walls of the cage to minimise loss of material through the perforations.

Figure 8c shows an improved design of the press that uses a 2-ton hydraulic bottle jack to generate a much greater compressive force. In this new design, the cage is not perforated on all sides, but has only one opening at its bottom to allow liquid removal.

5. Conclusion
This study has highlighted the importance and benefits of processing waste coffee grounds which are currently largely unexploited at the global scale. Through biorefining, a key pillar of circular economy, a large array of value-added products can be marketed: coffee oil, biodiesel, heating briquettes, etc. This work also gives to researchers in the field of waste valorization, practical, efficient and affordable methods to dry WCG and extract coffee oil for further research. These methods are of course not limited to coffee only but can be extended to many other oil-containing biomass wastes.

6. References
[1] Ray K 2013 Liver: Caffeine is a potent stimulator of autophagy to reduce hepatic lipid content--a coffee for NAFLD? vol 10, Gastroenterol Hepatol. p 563
[2] Mussatto S I, Machado E M S, Martins S and Teixeira J A 2011 Production, composition, and application of coffee and its industrial residues vol 4, Food Bioprocess Tech. p 661–672
[3] René v R and Bert A 2007 Status Report Biorefinery NL-6700 AA Wageningen: Agrotechnology and Food Sciences Group
[4] Cherubini F 2010 The biorefinery concept: Using biomass instead of oil for producing energy and chemicals vol 51, Energy Conversion and Management p 1412–1421
[5] Cherubini F, Jungmeier G, Mandl M, Philips C, Wellisch M, Jørgensen H, et al. 2007 IEA bioenergy Task 42 on biorefineries: co-production of fuels, chemicals, power and materials from biomass IEA
[6] Parajuli R, Dalgaarda T, Jørgensen U, Adamsen A P S, Knudsen M T, Birkved M, Gyling M and Schjørring J K 2015 Biorefining in the prevailing energy and materials crisis: a review of sustainable pathways for biorefinery value chains and sustainability assessment methodologies vol 43, Renew. Sustain. En. Rev. p 244-263
[7] Cavitch S M 1994 The Natural Soap Book Storey Publishing ISBN 0-88266-888-9
[8] Anneken D J, Both S, Christoph R, Fieg G, Steinberner U and Westfechtel A 2006 Fatty Acids Ullmann’s Encyclopedia of Industrial Chemistry
[9] Uddin M N, Techato K, Rasul M G, Hassan N M S and Mofijur M 2019 Waste coffee oil: A promising source for biodiesel production vol 160, Energy Procedia p 677–682
[10] Caetano N S, Silva V F M and Mata T M 2012 Valorization of coffee grounds for biodiesel production vol 26, Chem. Eng. Trans. p 267–272
[11] Gathuo B, Rantala P and Määtä R 1991 Coffee Industry Wastes vol 24, Water Sci. Technol. p 53–60
[12] Murthy P S and Madhava Naidu M 2012 Sustainable management of coffee industry by-products and value addition-A review vol 66, Resour. Conserv. Recy. p 45–58
[13] Sampaio A R M 2010 Desenvolvimento de tecnologias para produção de etanol a partir do hidrolisado da borra de café MSc thesis, Braga, Portugal: Department of Biological Engineering, University of Minho
[14] Kim J, Kim H, Baek G and Lee C 2017 Anaerobic co-digestion of spent coffee grounds with different waste feedstocks for biogas production vol 60, Waste Management p 322–328
[15] Vardon D R, Moser B R, Zheng W, Witkin K, Evangelista R L, Strathmann T J, Rajagopalan K and Sharma B K 2013 Complete utilization of spent coffee grounds to produce biodiesel, bio-oil, and biochar vol 1, ACS Sustain. Chem. Eng. p 1286–1294
[16] Ciesiereczuk T, Karwaczyńska U and Sporek M 2015 The possibility of disposing of spent coffee ground with energy recycling vol 16, J. Ecol. Eng. p 133–138
[17] Haile M 2014 Integrated valorization of spent coffee grounds to biofuels vol 2, Biofuel Res. J. p 65-69
[18] Illy A, Viani R 1998 Espresso Coffee: The Chemistry of Quality, Academic Press, London
[19] Speer K and Kölling-Speer I 2006 The lipid fraction of the coffee bean vol 18, Braz. J. Plant Physiol. p 201-216
[20] Maier HG 1981 Kaffee. Paul Parey Verlag, Berlin and Hamburg
[21] Yaqi R, Chunlan W, Jiakun X and Shuaiyu W 2019 Cafestol and Kahweol: A review on their bioactivities and pharmacological properties vol 20, Int. J. Mol. Sci. p 4238
[22] Rocha M V P, de Matos L J B L, Lima L P D, Figueiredo P M S, Lucena I L, Fernandes F A N and Gonçalves L R B 2014 Ultrasound-assisted production of biodiesel and ethanol from spent coffee grounds vol 167, Bioresour. Technol. p 343–348
[23] Al-Hamamre Z, Foerster S, Hartmann F, Krüger M and Kaltschmitt M 2012 Oil extracted from spent coffee grounds as a renewable source of fatty acid methyl ester manufacturing vol 96, Fuel p 70–76
[24] Somnuk K, Eawlex P and Prateepchaikul G 2017 Optimization of coffee oil extraction from spent coffee grounds using four solvents and prototype scale extraction using circulation process vol 51 p 181-189
[25] Jenkins R W, Stageman N E, Fortune C M and Chuck C J 2014 Effect of the type of bean processing, and geographical location on the biodiesel produced from waste coffee grounds vol 28 Energy Fuels p 1166–1174

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