CHEMICAL ENGINEERING TRANSACTIONS
VOL. 105, 2023

A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it

Guest Editors: David Bogle, Flavio Manenti, Piero Salatino

ISBN 979-12-81206-04-5; **ISSN** 2283-9216

DOI: 10.3303/CET23105052

Study of the Grinding of Coffee Waste Prior to its Valorization

Josefa Fernández-Ferreras*, Lucía Pérez Gandarillas, Yaiza Sisniega Maza

Department of Chemistry and Process & Resource Engineering, University of Cantabria, ETSIIyT, Avda. Los Castros 46, 39005-Santander, Spain
josefa.fernandez@unican.es

Coffee is one of the most important food products worldwide, but it also generates substantial amounts of waste, including spent coffee (SC), and coffee silver skin (CSS). Despite these organic wastes represent a major pollution hazard, they are also a potential source of compounds that can be used in different industrial applications such as pyrolysis, fermentation, solid-liquid extraction, or hydrolysis. To increase the efficiency of these operations, a reduced particle size of the wastes is beneficial, mostly below 400 µm. The use of a planetary ball wants to be an innovative and efficient method for reducing grinding times and costs for biomass materials such as residual coffee waste. This work is focused on the analysis of the influence of different grinding conditions of a planetary ball mill on the particle size distribution of SC and CSS wastes. A multiple linear regression analysis was applied to quantify the influence of variables. The results demonstrated that it was easier and higher the reduction in CSS (96% in 2 minutes) than in SC (max. 66% in 5 minutes), both at an optimal speed of rotation of 400 rpm. The speed of rotation and grinding time are the most influential variables, while pause time or reversal of direction do not favour the grinding process or in a less manner. This study highlights the potential of planetary ball mills for grinding organic materials, reducing grinding times and costs while improving the efficiency of waste utilization for subsequent industrial applications.

1. Introduction

Coffee is the most important food product worldwide and is the second most commercialized after crude oil (Polidoro et al., 2018). As a result of the high demand for coffee, large amounts of waste are generated (Mussatto et al., 2011). In fact, approximately 90% of the coffee produced becomes spent coffee (SC), which is obtained in the "instant coffee" elaboration process (Kourmentza et al., 2018). Another coffee residue is the coffee silver skin (CSS), which is obtained during the roasting process of the beans (Mussatto et al., 2011). These wastes contain significant amounts of caffeine and tannin, which could potentially harm the environment if improperly disposed (Getachew and Chun, 2017). Despite producing 6 million tons of these organic wastes annually (Tokimoto et al., 2005), they are also a potential source of compounds with functional properties similar to those of grains which can be used in different industrial applications, which are summarized in Table 1.

Table 1. Industrial applications of spent coffee and coffee silver skin

ORGANIC WASTE	APPLICATIONS
Spent coffee (SC)	Biosorbent and Extraction of its phenolics compounds (Solomakou et al., 2022)
	Fuel production (Silva et al., 1998)
	Fuel etanol production (Mussatto et al., 2012)
	Oil and water retention (Murthy and Naidu, 2012)
	Extraction of polysaccharides (Ballesteros et al., 2015)
Coffee Silver Skin (CSS)	Enzyme production (Mussatto et al., 2013)
	Dietary fiber (Borrelli et al., 2004)
	Soil fertilizer or fuel (Saenger et al., 2001)
	Antioxidant (Narita and Inouye, 2012)
	Water and oil retention (Murthy and Naidu, 2012)
	Pyrolysis (Polidoro et al., 2018)

Paper Received: 19 February 2023; Revised: 11 May 2023; Accepted: 21 April 2023
Please cite this article as: Fernandez-Ferreras J., Perez Gandarillas L., Sisniega Maza Y., 2023, Study of the Grinding of Coffee Waste Prior to Its Valorization, Chemical Engineering Transactions, 105, 307-312 DOI:10.3303/CET23105052

According to Murthy and Naidu, 2012, particle size plays a crucial role in these processes, being beneficial a size below 400 µm. For this purpose, a previous step of grinding the material is of great importance. In addition, in the case of CSS, milling is necessary to improve its workability, as unmilled CSS texture is very spongy, light, and unmanageable. Grinding is an important industrial operation that is mainly used for the size reduction of materials, but also for the production of large particle sizes (Ozkan et al., 2009). Ball mills can be used for various purposes, depending on their configuration, including mixing and blending, mechanical crushing and grinding and mechanical alloying of materials (Schilz et al., 1999). Generally, these mills have been used with inorganic materials (Liu et al. 2016), especially, hard and brittle materials for fine and ultrafine grinding down to the nanometer range (Rosenkranz et al., 2011). Some studies using organic materials show that planetary ball milling can produce fine particles when milling parameters are optimized (Rajkhowa et al., 2008).

Compared to other grinding devices, planetary ball mills provide one of the highest energy intensities due to the overlapping effect of two centrifugal forces produced by the rotation of the support disk and the rotation of the containers on their own axis and in the opposite direction (Burmeister and Kwade, 2013). This makes it an efficient method for producing nanopowders of a wide range of materials at industrial and laboratory scale (Wall et al., 2014).

The planetary ball mill can operate both dry and wet, but dry grinding is preferred to avoid separation of the solid and liquid phases and drying of the wet grinding particles. The processes within planetary ball mills are complex and highly dependent on the material processed and the synthesis, so the optimum grinding conditions must be evaluated for each individual system. (Burmeister and Kwade, 2013).

Although traditionally simple cutting or ball mills have been used for grinding residual biomass, the use of planetary ball mills for this type of material is innovative, due to their high energy input, that can reduce grinding times and costs.

Therefore, this study is focused on the grinding process of waste biomass in a planetary ball mill, specifically for two coffee wastes (SC and CSS), analyzing of the influence of operating variables and characteristics of feeding material on the particle size reduction. Four variables have been modified: grinding time, rotation speed, interval / pause time and reversal of direction

2. Materials and Methods

2.1 Waste Biomasses

SC and CSS were provided by “Cafetería Primos de Origen”, a specialty coffee shop in Santander (Spain) that imports green beans from El Salvador, Ethiopia, Rwanda, Brazil, and Indonesia.

Before performing the experiments, the residual biomasses were characterized. For this purpose, bulk and apparent densities, angle of repose and humidity were calculated, according to standard tests performed in solid operations (Table 2).

Table 2. Initial characterization of spent coffee and coffee silver skin.

	Spent Coffee (SC)	**Coffee Silver Skin (CSS)**
Bulk density (g/ml)	0.4	0.11
Apparent density (g/ml)	1.6	0.53
Angle of repose (º)	37.25	42.91
Humidity (%)	10.5	9.4

Both materials have similar moisture content, being a little more cohesive the CSS, but it can be remarked the lower bulk and apparent densities in the case of CSS according to its spongy and light texture.

2.2 Grinding process and design of experiments (DOE)

Grinding of SC and CSS was performed with a Retsch brand planetary ball mill, a high-energy mill with a single 500 ml container and 25 stainless steel 20 mm balls which, according to Danha et al., 2016, lead to highest yields in obtaining medium-sized particles.

To optimize the grinding conditions with respect to size reduction, grinding parameters such as grinding time, rotation speed, interval / pause time and reversal of rotation were varied for each of the materials.

The pause or rest time (PT) is considered to avoid possible phase changes or agglomerations due to the local increase in temperature of the sample (Kwon et al, 2002). Varying the interval (I) is very interesting because the shorter the interval time chosen, the greater the number of times the mill stops. In this case, PT was added to grinding time, being the actual PT chosen 30s in SC and 15s in CSS (Table 3).

Table 3 shows the design of experiments carried out on SC and CSS. The values indicated in the pause show the pause time after the one grinding interval (grinding interval/pause time).

Table 3. Design of Experiments for Spent Coffee (SC) and Coffee Silver Skin (CSS)

VARIABLES for Spent Coffee (SC)	VALUE				
Grinding time (min)	5	10	15		
Rotation speed (rpm)	350	400	450		
Pause time (I[s]/PT[s]) (PT added to grinding time)	No	Yes (120/30)			
Reversal of rotation	No	Yes			
VARIABLES for Coffee Silver Skin (CSS)	**VALUE**				
Grinding time (min)	2	3	5	10	15
Rotation speed (rpm)	300	400			
Pause time (I [s]/PT [s]) (PT added to grinding time)	No	Yes (15/15)	Yes (30/15)	Yes (60/15)	
Reversal of rotation	No	Yes			

2.3 Sieving process and particle size distribution

A CISA sieve shaker model RP.20 was used to calculate the particle size distribution. Depending on the initial particle size of the residual biomass, several sieves with different mesh sizes were chosen, taking ISO 3310/01 as a guiding reference. For SC, mesh sizes (μm) were 5000, 3550, 1600, 1000, 630, 400, 250, 150, 63 and 0. For CSS after grinding, 400, 300, 250, 200, 150, 112, 80, 63 and 0 μm.

To verify the efficiency of the planetary ball mill, a characterization of the samples was carried out before and after grinding. For this, the differential (fm) and cumulative (Fm) distributions of the mass fraction were made and from these, the parameters that refer to the representative size of the total distribution (diameters d_{10}, d_{50}, d_{90} and d_{mm}). The d_{10} is the diameter such that 10% of the mass of the sample is made up of particles with a diameter less than this value, d_{50} corresponds to 50% and is also the median of the sample and d_{90} represents 90%. These parameters were calculated by linear regression on the accumulated distributions of siftings. Sauter diameter was also calculated for optimal tests in SC and CSS.

Equation 1 shows the mass average diameter (d_{mm}) of the distribution.

$$d_{mm} = \frac{m_i \cdot d_i}{\sum m_i} \qquad (1)$$

2.4. Multiple Linear regression analysis (MLR)

To evaluate the effect of the operating conditions (factors) used on the values of d_{10}, d_{50}, d_{90} and d_{mm} (responses), the results were fitted to a multiple linear regression model (MLR), performed using MODDE Pro-13 software (Umetrics, Malmö, Sweeden). The MLR analysis allows to recognize the influence of variables on the material behavior under milling and to identify the most significant ones. This will also serve to evaluate the sensitivity of the grinding process to changes. The intervals considered are showed in Table 3.

The relative importance of the different factors on the responses was evaluated through the graphs of the coefficients of the MLR model. To make the regression coefficients comparable when the responses (d_{10}, d_{50}, d_{90} and d_{mm}) had different ranges, the coefficients were normalized, divided by the standard deviation of their respective response.The size of the terms in the model reflects the magnitude of the change in the response when a factor varies, with the magnitude of the change being directly proportional to the size of the terms. If a response increases when a factor increases, the sign of the coefficient will be positive.

3. Results

Experiments were done in duplicate to calculate errors in the reproducibility. Errors found were always lower than 2.4% for SC and 1.5% for CSS. Before conducting the experiments, the mass average particle diameter (d_{mm}) of the initial waste was calculated, being 922.2 µm for SC and 1829.7 µm for CSS.

3.1 Effect of grinding time and rotational speed

Grinding time is one of the most important variables to study, as it has a direct impact on the particle size and specific surface area. Actually, increasing the grinding time leads to a decrease in the particle size of the powder and an increase in the specific surface area. However, prolonged grinding times can cause excessive heating of the vessel, resulting in lower throughput, increased wear of the grinding media and larger agglomerations. (Lin et al. 2013). Figure 1 shows the influence of grinding time and rotational speed variables on the average particle size for both materials. Lower values are obtained in CSS despite its higher initial particle size, which indicates that this material grinds more easily. In SC mainly at 350 rpm and in CSS, a trend of decreasing average particle size during the first minutes of grinding is observed. However, as the grinding time increases, an inflection point is observed, followed by a progressive increase in the average particle size, due to

agglomeration. This is consistent with the findings of Liu et al., 2016, who claim that the grinding efficiency is high at the beginning and slows down as the grinding progresses. Furthermore, other authors suggest that there is a grinding limit in ball mills beyond which no further particle breakage occurs (Knieke et al., 2011).
In most cases, a minimum rotational speed is required to achieve adequate performance and greater size reduction. However, in some instances, rotational speed has no influence above a certain number of revolutions or no influence at all (Burmeister and Kwade, 2013). This can be clearly seen in the case of SC, where a greater particle size reduction is achieved when increasing the speed from 350 rpm to 400 rpm, the same as for the CSS. However, an increase in revolutions from 400 rpm to 450 rpm in the SC has the opposite effect, with a larger final particle size, not tested in the CSS.

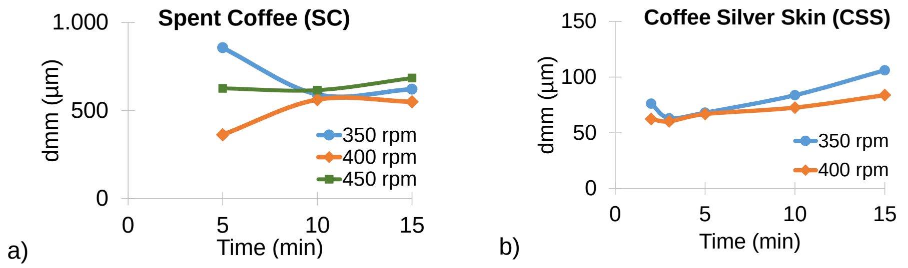

Figure 1. Influence of grinding time and rotational speed variables on the average particle size: a) SC, b) CSS

The results show that largest particle size reductions are obtained at speeds of 400 rpm, with a higher influence in the case of SC. On the other hand, higher throughput at lower speed leads to lower energy consumption and therefore lower cost and a more energy-efficient process.For both materials, the optimum rotation speed is 400 rpm and the optimum grinding times are 5 minutes for SC (d_{mm} 362.6 µm) and 3 minutes for CSS (d_{mm} 60.2 µm). However, similar results were achieved for the CSS at a time of 2 min and 400 rpm (d_{mm} 62.3 µm) and at 3 min at 350 rpm (d_{mm} 63 µm). Finally, the maximum percentage of reduction of the mass average diameter d_{mm}, was 60.68% for SC (362.6 µm) and 96.70% for CSS (60.2 µm).

3.2 Effect of Interval / pause time

The introduction of pause times during the grinding process is interesting to avoid overheating of the material and balls (Naghdi et al. 2017) and, consequently, prevent the material agglomeration.For SC, a pause time of 30 seconds was introduced every two minutes at 350 and 400 rpm. In CSS the pause time was 15 seconds every 15, 30 and 60 seconds in different experiments at 350 and 400 rpm.As it can be seen in Figure 2, for both SC and CSS, adding pause time does not contribute to smaller average particle diameters. The % of reduction of average particle size obtained were 56.97% for SC and 96.83% for CSS.

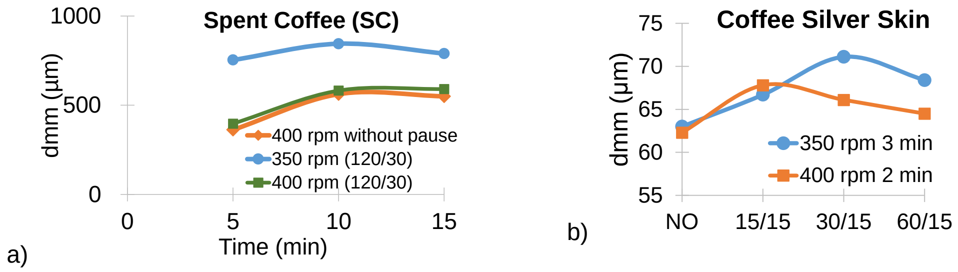

Figure 2. Optimization of the pause time variable: a) SC, b) CSS

3.3 Effect of reversal of direction

Table 4 shows a summary of the results of the optimal experiments for each material for the different variables studied, compared to initial values, that includes the influence of the reversal of direction.
For SC, the optimal values were obtained when grinding for 5 minutes at 400 rpm with a pause time of 30 seconds every 2 minutes and reversal of direction, with a percentage of particle size reduction of 66.12% (312.4 µm), very similar to that obtained at 10 minutes in similar conditions (66.75%,306.6 µm) but using half the time. For CSS the reduction was 96.87%, grinding for 3 minutes at 400 rpm with a pause time of 15 seconds every minute and reversal of direction.The reversal of direction decreases particle size, but its influence is very small compared to the rotation speed or grinding time.

Table 4. Summary of the results d_{10}, d_{50}, d_{90}, d_{mm}, d_{SAUTER} of the optimal tests for each organic material.

	t (min)	Speed (rpm)	Interval/Pause	Reversal	d_{10} (μm)	d_{50}(μm)	d_{90}(μm)	d_{mm} (μm)	d_{SAUTER} (μm)
Spent coffee (SC)	NO	NO	NO	NO	108.2	472.3	966.7	922.2	232.5
	5	400	NO	NO	36.5	221.8	769.4	362.6	108.1
	5	400	120/30	NO	52.4	309.1	813.0	396.9	137.1
	5	400	120/30	Yes	32.6	207.9	608.7	312.4	100.8
	10	400	120/30	Yes	22.6	166.6	763.7	306.6	81.78
Coffee Silver Skin (CSS)	NO	NO	NO	NO	-	-	-	1829.70	-
	3	400	NO	NO	10.4	51.9	108.5	60.2	42.5
	3	400	30/15	NO	8.9	44.5	111.1	58.0	39.3
	3	400	30/15	SI	9.2	46.2	120.3	59.8	40.2
	3	400	60/15	SI	9.0	44.9	108.1	57.3	39.2

3.4 MLR results

A MLR analysis was performed to quantitatively evaluate the influence of the operating variables (rotational speed, grinding time, pause time and reversal of direction) on the particle size distribution parameters (d_{10}, d_{50}, d_{90} and d_{mm}). Figure 3 shows the values of the coefficients obtained for SC and CSS for each operational variable, each one represented by a different color. The lower the coefficient, the lower the particle size achieved. Based on this statement, it can be observed that the rotational speed contributes the most to particle size reduction for CS, while grinding time has more influence in all the parameters determined in CCS.
For both materials high rotational speed results in smaller d_{10}, d_{50}, d_{90} and d_{mm}, and in general a higher grinding time produces higher particle size.
The pause time in general does not favor smaller particle size, mainly in the CSS. The reversal of the direction favors the obtention of smaller average particle diameters in SC but influences little the process in CSS.

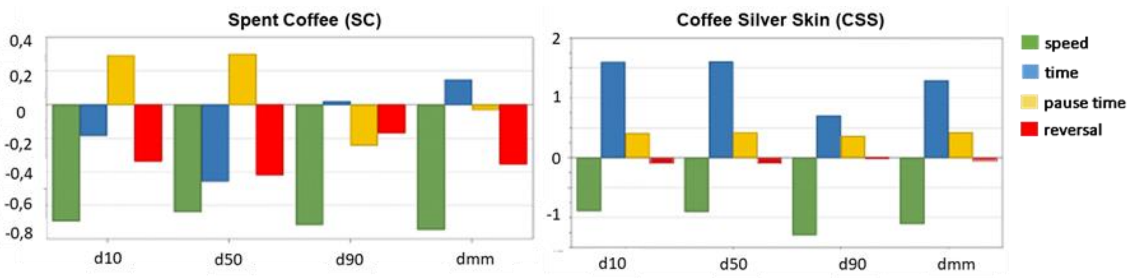

Figure 3. Graph of coefficients for d_{10}, d_{50}, d_{90} and d_{mm} for SC and CSS (rotational speed, grinding time, pause time, reversal of direction).

4. Conclusions

Throughout this work, the characterization and grinding of two organic materials as an example of waste biomass (coffee grounds and silver skin) has been carried out, with different operating conditions to analyse their influence on the particle size reduction in a planetary ball mill. The following conclusions can be reached:

- A particle size reduction in spent coffee of 66.12% (312,4 μm) and coffee silver skin of 96.87% (59,8 μm) were achieved for optimal grinding conditions.
- The optimum speed of rotation for both materials was found to be 400 rpm.
- The variables with the greatest influence for the two materials are the speed of rotation and the grinding time, sometimes having an influence 10 times greater than the interval/pause time and the reversal of direction.
- The pause time, regardless of the interval and the material, does not favour either the grinding process, probably because it increases the residence time causing heat to "cake" the product, in spite of the resting or turning time.
- The pause time with reversal of the direction of rotation has a minimal influence on the process, having a favourable influence when working at high rotational speeds.

The study shows that particle size reduction is more easy for CSS than form SC, achieving also lower values, but both get appropriate sizes for most industrial applications of the coffee wastes in a very short time with low revolutions.

References

Ballesteros L.F., Cerqueira M.A., Teixeira J.A., Mussatto, S.I., 2015, Characterization of polysaccharides extracted from spent coffee grounds by alkali pretreatment, Carbohydrate Polymers, 127, 347-354.

Borrelli R.C., Esposito F., Napolitano A., Ritieni A., Fogliano V., 2004, Characterization of a New Potential Functional Ingredient: Coffee Silverskin, Journal of Agricultural and Food Chemistry, 52, 1338-1343.

Burmeister C.F., Kwade A., 2013, Process engineering with planetary ball mills, Chemical Society Reviews, 42, 7660-7667.

Danha G., Legodi D., Hlabangana N., Bhondayi C., Hildebrandt D., 2016, A fundamental investigation on the breakage of a bed of silica sand particles: An attainable region approach, Powder Technol., 301, 1208-1212.

Getachew A.T., Chun B.S., 2017, Influence of pretreatment and modifiers on subcritical water liquefaction of spent coffee grounds: A green waste valorization approach, Journal of Cleaner Production, 142, 3719-3727.

Knieke C., Romeis S., Peukert W., 2011, Influence of Process Parameters on Breakage Kinetics and Grinding Limit at the Nanoscale, AIChE Journal, 57, 1751-1758.

Kourmentza C., Economou C.N., Tsafrakidou P., Kornaros M., 2018, Spent coffee grounds make much more than waste: Exploring recent advances and future exploitation strategies for the valorization of an emerging food waste stream, Journal of Cleaner Production 172, 980-992.

Kwon Y.S., Gerasimov K.B., Yoon S.K., 2002, Ball temperatures during mechanical alloying in planetary mills, Journal of Alloys and Compounds, 346, 276-281.

Lin J., Zhang X., Han W., Sun D., 2013, Influence of milling time on the synthesis, microstructure and mechanical properties of ZrB2SiCZrO2 ceramic composites, Ceramics International, 39, 4213-4219.

Liu S., Li Q., Xie G., Li L., Xiao H., 2016, Effect of grinding time on the particle characteristics of glass powder, Powder Technology, 295, 133-141.

Murthy P.S., Naidu M.M., 2012, Recovery of Phenolic Antioxidants and Functional Compounds from Coffee Industry By-Products, Food and Bioprocess Technology, 5, 897-903.

Mussatto S.I., Ballesteros L.F., Martins S., Maltos D.A.F., Aguilar C.N., Teixeira J.A., 2013, Maximization of Fructooligosaccharides and β-Fructofuranosidase Production by Aspergillus japonicus under Solid-State Fermentation Conditions, Food and Bioprocess Technology, 6, 2128-2134.

Mussatto S.I., Machado E.M.S., Carneiro L.M., Teixeira J.A., 2012, Sugars metabolism and ethanol production by different yeast strains from coffee industry wastes hydrolysates, Applied Energy, 92, 763-768.

Mussatto S.I., Machado E.M.S., Martins S., Teixeira J.A., 2011, Production, Composition, and Application of Coffee and Its Industrial Residues, Food and Bioprocess Technology, 4, 661-672.

NaghdI M., Taheran M., Brar S.K., Rouissi T., Verma M., Surampalli R.Y., Valero J.R., 2017, A green method for production of nanobiochar by ball milling- optimization and characterization, Journal of Cleaner Production, 164, 1394-1405.

Narita Y., Inouye K., 2012, High antioxidant activity of coffee silverskin extracts obtained by the treatment of coffee silverskin with subcritical water, Food Chemistry, 135, 943-949.

Ozkan A., Yekeler M., Calkaya M., 2009, Kinetics of fine wet grinding of zeolite in a steel ball mill in comparison to dry grinding, International Journal of Mineral Processing, 90, 67-73.

Polidoro A. dos S., Scapin E., Lazzari E., Silva A.N., Dos Santos A.L., Caramão E.B., Jacques R.A., 2018, Valorization of coffee silverskin industrial waste by pyrolysis: From optimization of bio-oil production to chemical characterization by GC × GC/MS, Journal of Analytical and Applied Pyrolysis, 129, 43-52.

Rajkhowa R., Wang L., Wang X., 2008, Ultra-fine silk powder preparation through rotary and ball milling, Powder Technology, 185, 87-95.

Rosenkranz S., Breitung-Faes S., Kwade A., 2011, Experimental investigations and modelling of the ball motion in planetary ball mills, Powder Technology, 212, 224-230.

Saenger M., Hartge E.U., Werther J., Ogada T., Siagi Z., 2001, Combustion of coffee husks, Renewable Energy, 23, 103-121.

Schilz J., Riffel M., Pixius K., Meyer H.J., 1999, Synthesis of thermoelectric materials by mechanical alloying in planetary ball mills, Powder Technology, 105, 149-154.

Silva M.A., Nebra S.A., Machado Silva M.J., Sanchez C.G., 1998, The use of biomass residues in the Brazilian soluble coffee industry, Biomass and Bioenergy, 14, 457-467.

Solomakou N., Tsafrakidou P., Goula A.M., 2022, Holistic Exploitation of Spent Coffee Ground: Use as Biosorbent for Olive Mill Wastewaters After Extraction of Its Phenolic Compounds, Water, Air, and Soil Pollution, 233, 173-179.

Tokimoto T., Kawasaki N., Nakamura T., Akutagawa J., Tanada S., 2005, Removal of lead ions in drinking water by coffee grounds as vegetable biomass, Journal of Colloid and Interface Science, 281, 56-61.