

Sequenciamento de operações unitárias para processamento integral e sustentável do amendoim

Sequencing of unit operations for integral and sustainable peanut processing

Secuenciación de operaciones unitarias para el procesamiento integral y sostenible de maní

Recebido: 02/04/2020 | Revisado: 04/04/2020 | Aceito: 08/04/2020 | Publicado: 12/04/2020

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Resumo

As biomassas mais utilizadas são madeira, cana-de-açúcar, papel, casca de arroz e amendoim. Com base no conceito de sustentabilidade e redução de resíduos, a utilização plena da biomassa é uma alternativa capaz de proporcionar às pessoas uma melhor ingestão nutricional, melhorando a economia relacionada à biomassa e a relação ecológica entre o homem e o meio ambiente. Portanto, o objetivo deste trabalho foi o uso sustentável da biomassa, o amendoim, por meio de operações unitárias múltiplas. Assim, o material passou por vários processos, como o processo de moagem, onde foi possível obter dados de análise imediata (teor de umidade, teor de cinzas, teor volátil e carbono fixo) e tamanho das partículas. Em seguida, a secagem foi realizada utilizando energia solar e a eficiência de

secagem foi obtida. Testes de infravermelho e estufa foram realizados em laboratório para determinar as curvas de secagem e umidade. Após a secagem, também foi realizada a extração de óleo com etanol como solvente aquecido por energia solar. Depois disso, o bolo residual foi utilizado como material adsorvente na remoção do corante por adsorção em leito fixo. Assim, o adsorvente impregnado de corante passou por um processo de pirólise para formar carvão fino, bio-óleo e gases. Com produtos e subprodutos de cada processo, foi possível gerar o espectro infravermelho de cada um. Portanto, mostra-se como é possível o uso integral do amendoim como biomassa, utilizando processos sustentáveis de engenharia química, e como ele pode contribuir para a redução da poluição e da produção de resíduos.

Palavras-chave: Biomassa; Amendoim; Uso integral; Processos; Sustentabilidade.

Abstract

The most used biomass are wood, sugar cane, paper, rice husk, and peanut. Based on the concept of sustainability and waste reduction, the full utilization of biomass is an alternative capable of providing people with a better nutritional intake, improving the economy related to biomass and the ecological relationship between man and the environment. So, the aim of this work was the sustainable use the biomass, the peanut, through multiple unit operations. Thus, the material went through several processes as the grinding process, where it was possible to obtain immediate analysis data (moisture content, ash content, volatile content and fixed carbon) and particle size. Then, drying was performed employing solar energy and the drying efficiency was obtained. Infrared and greenhouse tests were performed in the laboratory to determine the drying and humidity curves. After drying, it was also carried out the oil extraction using ethanol as solvent heated by solar energy. After that, the residual cake was used as the adsorbent material in the dye removal by adsorption in a fixed bed. So, the dye-impregnated adsorbent has undergone a process of pyrolysis in order to form fine coal, bio-oil, and gases. With products and by-products of each process, it was possible to generate the infrared spectrum of each. Therefore, it is shown how the integral use of peanut as biomass is possible, using chemical engineering sustainable processes, and how it may contribute to the reduction of the pollution and to the reduction of waste production.

Keywords: Biomass; Peanuts; Full use; Processes; Sustainability.

Resumen

Las biomásas más utilizadas son madera, caña de azúcar, papel, cáscaras de arroz y maní. Basado en el concepto de sostenibilidad y reducción de desechos, el uso completo de la biomasa es una alternativa capaz de proporcionar a las personas una mejor ingesta nutricional, mejorando la economía relacionada con la biomasa y la relación ecológica entre el hombre y el medio ambiente. Por lo tanto, el objetivo de este trabajo fue el uso sostenible de biomasa, maní, a través de operaciones de unidades

múltiples. Por lo tanto, el material pasó por varios procesos, como el proceso de molienda, donde fue posible obtener datos de análisis inmediatos (contenido de humedad, contenido de cenizas, contenido volátil y carbono fijo) y el tamaño de las partículas. Luego, el secado se llevó a cabo utilizando energía solar y se obtuvo eficiencia de secado. Se realizaron pruebas de infrarrojos y de invernadero en el laboratorio para determinar las curvas de secado y humedad. Después del secado, el aceite también se extrajo con etanol como disolvente calentado por energía solar. Después de eso, la torta residual se usó como material adsorbente para eliminar el tinte por adsorción en un lecho fijo. Así, el adsorbente impregnado de colorante pasó por un proceso de pirólisis para formar carbón fino, bio-aceite y gases. Con los productos y subproductos de cada proceso, fue posible generar el espectro infrarrojo para cada uno. Luego, se muestra cómo es posible el uso completo del maní como biomasa, manejando procesos de ingeniería química sostenibles, y cómo puede contribuir a la reducción de la contaminación y la producción de residuos.

Palabras clave: Biomasa; Maní, Uso completo, Procesos, Sostenibilidad.

1. Introduction

The integral use of biomass is an alternative capable of providing people better nutritional consumption, improve the biomass-related economy and the ecological relation between man and the environment, since the use of this technology results in the reduction of waste (Silva et al., 2005). Thus, in the prospective scenario, it is important to outline technological options that allow such use and possible applications in the context of chemical engineering, since it has lignocellulosic biomass as the essential raw material in numerous processes, such as extraction, adsorption, and pyrolysis.

Due to the vast biodiversity found in its territory, Brazil has a wide variety of agricultural, agro-industrial and waste products whose processing would be of great economic and social interest. These include waste from activities such as pulp and paper mills, sawmills (sawdust), mills (sugarcane bagasse) and, in general, agricultural production units generating grains and crop residues such as straw cereals and corn, corn cob, peanut pods, rice and oat peels, among others (Assis, 1994).

The biomass consists of any material of plant origin derived from photosynthesis and includes the fraction previously processed by animals. Biomass sources include wood and processing residues (woody vegetables), non-woody vegetables, which are typically produced from annual crops, urban solid waste generated in the domestic and commercial environment, animal waste, food processing residue, aquatic plants, algae and biofluids (McKendry, 2002).

Numerous unit operations can be applied to biomass such as drying, extraction,

adsorption, pyrolysis, second-generation alcohol production, use of noble fractions for the production of cosmetics and biodiesel, beneficiation of by-products for the production of asphalt, that is, several process routes can be followed for the biomass to be fully utilized (Vieira, 2004).

Drying of agricultural products can be defined as a simultaneous process of heat and mass transfer between the product and the drying air. It consists of the removal of excessive moisture contained within the grain by means of evaporation, usually caused by forced convection of heated air, in order to allow the maintenance of its quality during storage, for long periods of time without the danger of deterioration (Souza, 2013). Since integral biomass processing also aims at environmental preservation, it is interesting to work with natural drying, a process widely used in tropical regions that require minimal investment, such as using solar dryers (Silva & Souza, 2020; Stoppe et al., 2020).

The data obtained during the drying process can be adjusted with the aid of empirical equations. The most used models (empirical equations) are the proposed by Lewis (1921), Brooker et al. (1974), Henderson & Henderson (1968), Page (1949), Overhults et al. (1973), Midilli et al. (2002), besides logarithmic and two terms exponential. After drying, the biomass can be submitted to the roasting process, extraction of oil if the biomass presents high contents or to go directly to human food, for example.

Solvent extraction is a mass transfer operation widely used in the food industry to remove oil from seeds and/or oilseed pulps. In relation to the extraction system, this can be in fixed or mobile bed, the latter being the most used for the production of vegetable oils of oilseeds, such as cotton, peanuts, and soybeans (Rydberg, 2004). In this process, the seeds are subjected to pre-treatments such as peeling, sometimes baking, partial dehydration and grinding. Therefore, the resulting extract, composed of solute and solvent, may contain some finely divided solid particles, which may be removed by filtration or centrifugation. For the removal of the solvent, additional evaporation is required (Geankoplis, 2003).

The extraction process has as products the pie or bran, the oil and the dregs. The pie can be followed for a new extraction to remove the remaining oil or for supplementation. The bran, because it contains lower oil content, can be used for animal feed, if it presents the nutritional values required by the market. In addition, both can be used in dye adsorption processes. The oil, in turn, can be used in the manufacture of soaps, be used in the production of biodiesel, in food, among other applications (Ramalho & Suarez, 2012).

The extraction processes can be sustainably performed using solar energy, such as demonstrated by Tavares & Santos (2016) for peanut oil extraction, Silva et al. (2017) for

vegetable oils, Tavares et al. (2019) to obtain *Jatropha* oil, Silva et al. (2019) for coconut oil and Silva et al. (2020) for Baru oil.

The contamination of natural waters is one of the great social problems that generate constant concern (Kunz & Peralta-Zamora, 2002; Nandi et al., 2008). Dyes are one of the most important classes of pollutants because they have a structure that makes them stable and harder to biodegrade, as they are designed to be resistant to environmental conditions such as sunlight, pH and microbial attack (Rai et al., 2004; Bestani et al., 2008).

Adsorption is a well-known separation process and an effective method for water decontamination applications. This process is considered a superior technique to the other techniques in terms of initial cost, flexibility and simplicity and also due to the ease of operation, and does not result in the formation of harmful substances (Dutta, 2004). In this context, due to the need to reduce costs with commercial adsorbents the use of plant residues as adsorbents for the removal of dyes from industrial effluents has been explored. For this, it is possible to use the pie obtained in extraction processes, which is a lignocellulosic material and a co-product generated by the oil extraction process as an adsorbent (Santos et al., 2007).

Among some waste materials that can be used as adsorbents can be mentioned, banana peel (Almeida & Santos, 2020), sugarcane bagasse (Jorge et al., 2015), sawdust (Tavares et al., 2017), coal of green coconut (Stoppe et al. 2019) and passion fruit seeds (Isquierdo et al., 2020). The use of peanuts as an adsorbent material also seems to be very promising for removing contaminants, as shown by some works in the literature (Xu & Liu, 2008; Ali et al., 2012; Hou et al. 2019).

In an adsorption process, the equilibrium of the system is reached when there are no changes in adsorbate concentrations in the solid phase and adsorbate in the solution. The equilibrium reflects the capacity and/or affinity of an adsorbent for an adsorbate, under a given set of conditions under which the system is subjected. Thermodynamically, the system is presented by adsorption isotherms (Rafatullah et al., 2010).

The adsorbent isotherms relate the amount of adsorbate adsorbed per gram of adsorbent, Q_e (mg g⁻¹), in equilibrium with the amount of adsorbate remaining in solution, C_e (g L⁻¹) at a given temperature (Özacar & Sengil, 2003). The isotherms often used are those developed by Langmuir (Al-Degs et al., 2000) and Freundlich (Choy et al., 1999). However, other equations are found, such as Brunauer, Emmett and Teller (BET) (Fu & Viraraghavan, 2002), Redlich-Peterson (Allen et al., 2003), Tempkin (Allen et al., 2003), Tóth (Allen et al., 2003) and Dubinin-Radushkevich (Choy et al., 1999).

After passing through the adsorption process, the biomass meal with adsorbed dye can

still be used in pyrolysis processes.

Pyrolysis can be defined as the thermal degradation of organic material in the partial or total absence of an oxidizing agent, or even in an environment with an oxygen concentration capable of preventing the intensive gasification of the organic material. Pyrolysis occurs generally at a temperature ranging from 400 °C to the start of the gasification system (Pedroza et al., 2010, 2011).

According to Gomes (2010) Raw materials (biomass), used in the pyrolysis process, can be punctuated with vegetable or agricultural residues (sugarcane bagasse, rice hulls, straw and corn cob, etc.), industrial waste (bark, wood chips, orange marc, cashew, pineapple, among others) and forest residues (leaves, branches, reforestation wood and materials resulting from distillation).

There are several works in the literature regarding biomass pyrolysis, using different biomasses, such as sugarcane bagasse (Lira et al. 2010; Santos et al. 2010, 2012a,b), spent coffee grounds (Batista Jr. et al., 2017), malt (Silvério et al., 2017), as well as other biomasses (Pimentel et al., 2019; Bernardes et al., 2020; Pereira, et al. 2020).

The pyrolysis is distinguished, when compared to the differentiated types of processes, to generate products (liquid, solid and gaseous), with aggregated values that can be used as a fuel source or among other uses (Pedroza, 2011).

The oil resulting from the pyrolysis of biomass, called generically bio-oil, besides being a renewable liquid fuel, which is the main advantage over petroleum derivatives, can be used for the production of numerous chemical substances. (Santos, 2011)

The modeling and optimization of the pyrolysis technique are essential for the improvement of processes and new large industrial applications. The knowledge of kinetics and transport phenomena is of great relevance for the design of pyrolysis gasifiers and reactors.

Therefore, the objective of this work was the integral and sustainable use of the biomass, the peanut, through the sequencing of these unit operations (drying, extraction, adsorption, and pyrolysis) and how it may contribute to the reduction of the pollution and to the reduction of waste production.

2. Material and Methods

In order to exemplify the full utilization of biomass, the peanut was chosen because of its low cost, availability in the region, high oil content and because all byproducts generated

in its processing have great application and quality. A laboratorial and quantitative research method was used in the present study as the following described, for the characterization and application of the chosen biomass were described (Pereira, 2018).

2.1. Characterization of biomass

To prepare the sample, all peanuts were peeled, crushed in an industrial blender and sieved in a Tyler type sieve with a 1.18mm opening (# 16). Subsequently, the classified peanut was used in the unit operations of drying, extraction, adsorption, and pyrolysis in order to demonstrate the possible operations of a process of integral use of biomass.

2.1.1. Immediate analysis

The peanut samples were submitted to moisture, volatile, ash and fixed carbon content analysis in order to determine the composition of the biomass.

Moisture Content: Two grams of peanuts were weighed and placed in a preheated laboratory kiln at 105 °C until the mass of the sample remained constant.

Volatile Content (Tv): The volatile content was quantified by measuring the mass fraction of the standardized and previously dried biomass that volatilizes during heating to temperatures of approximately 850 °C in a muffle furnace for seven minutes.

Ash Content: One gram of dry, volatiles-free biomass sample was placed in a crucible and brought into a muffle at 710 °C for one hour. After that one hour, only the ashes remained inside the crucible.

Fixed Carbon (Tcf): The fixed carbon content (Tcf) is obtained by subtracting the sum of volatile material and ash contents from the total sample (100).

2.1.2. Size distribution analysis

In order to know the size distribution of the material, the sample was homogenized and the sieving was carried out for 10 minutes. In this assay, sieves of # 10, # 12, # 16, # 20, # 25, # 30 and # 35 were used. It was then determined the mass fraction of particles retained in each sieve, which allowed the treatment of the data and adjustment in main size distribution models: the GGS, RRB and Sigmoid models, represented by equations (1) to (3), respectively.

$$\bar{X} = \left(\frac{D}{k}\right)^m \quad (1)$$

$$\bar{X} = 1 - e^{-\left(\frac{D}{D'}\right)^n} \quad (2)$$

$$\bar{X} = \left[1 + \left(\frac{D_{50}}{D}\right)^p\right]^{-1} \quad (3)$$

where D is the average diameter of each sieve and \bar{X} is the cumulative mass fraction.

2.2. Drying

2.2.1. Kinetics of drying

In order to determine the kinetics of peanut drying, an infrared moisture analyzer (model IV2500, Gehaka) was used to collect data on the percentage of moisture lost.

The experiments were carried out at temperatures of 50 and 60 °C, similar to those reached in the solar drying tests, which was also performed in this work. The moisture data were then annotated as a function of time until no variation was observed. The moisture ratio (MR) was calculated by Equation (4):

$$MR = \frac{X_{bsi} - X_e}{X_{bs0} - X_e} \quad (4)$$

where X_{bsi} is the dry basis moisture at time (i), X_e is the equilibrium moisture (90% of the final moisture on the dry basis - dynamic equilibrium) and X_{bs0} is the initial dry basis moisture.

The data were adjusted by means of empirical equations that represent the drying kinetics and the software *Statistica*® was used to calculate the kinetic parameters. The models (empirical equations) adjusted were Lewis (1921), Brooker et al. (1974), Handerson and Henderson (1968), Page (1949), Exponential of two terms, Midili et al (2002) and Logarithm, in which the parameter K is the drying constant, defined by the Arrhenius equation and used to determine the effective mass diffusivity as a function of temperature.

2.2.2. Solar drying

The solar drier used for this experiment was built by students from the Federal University of the Triângulo Mineiro (UFTM). In the dryer, a cooler was coupled to promote

forced convection of the drying air and thus, increase the moisture removal efficiency of the material. In each experiment, about 400 and 500g of peanut (peeled, crushed and sieved) were introduced into the dryer tray to form a thin layer. After lodging the peanut in the dryer, the tray was capped and the temperature of the peanut, air, and tray was measured every 30 minutes with a multimeter.

2.3. Oil extraction by leaching

2.3.1. Extraction of oil using Soxhlet

A sample of 6 g of peanuts was placed inside the cellulose Soxhlet extraction thimble and was introduced into the Soxhlet extractor (model MA491/6) in such a way that it was submerged in the solvent during extraction. About 80 ml of solvent (50% alcohol, 50% toluene in volume) was added, and then the water circulation was connected to condense the solvent and prevent significant loss thereof through evaporation in order to maintain the solvent volume constant.

The experiment was run for 4 hours (period considered to be optimal) and then the solvent was recovered by heating the mixture until the evaporation of the solvent ceased and there was no return to the sample. After completion of this process, the Soxhlet was switched off and the container containing the mixture (oil + solvent) and the extraction thimble containing the samples were taken to the kiln for 24 hours at a temperature of 105 °C for complete evaporation of the solvent.

After 24 hours, the vessel containing the extracted oil was transferred to the desiccator until it reached room temperature and afterward was weighed to determine the mass of extracted oil. By subtracting the weight of the container from the value obtained (container weight + oil), the weight of the oil was reached. The residual particulate material from the extraction (peanut) was stored for further analysis of the extractives. The yield of the extraction was calculated using the values of final moisture, dry mass of residual cake and mass of oil. This experiment was carried out in triplicate.

2.3.2. Extraction using solar energy

The experiment consisted of filling the entire absorber bed (295 cm³) with 120 g of dried peanut particles in a solar extractor and percolating the solvent (ethanol) with a flow rate

of 25 ml/ min, using a peristaltic pump. To avoid possible leaks, the sample was not packaged.

The whole system was exposed to solar radiation for about 2 hours, which is the average time required for peanut oil extraction (Tavares, 2015). Four extractions were carried out on different days considering favorable environmental conditions of operation, i.e. day with open sky, temperature above 25 °C and periods with solar peak incidence. At the end of the experiment, the cake in the absorber bed was dried in kiln at 105 °C for 24 hours, and the container containing the oil and ethanol mixture was taken to the freezer for further separation.

After the experiments of solar drying and solar extraction, it was possible to determine the moisture content and the oil content, respectively. To analyze the moisture, 20 grams of sample was dried in the kiln at 105 °C for 24 hours and reweighed in analytical balance. Through the difference between the masses, the moisture was obtained. For the oil content, the methodology is the same as for the moisture content. However, 20g of the sample was weighed before and after extraction.

2.4. Adsorption using biomass

The equilibrium study had as objective to determine the efficiency of the peanut pie (after extraction) as adsorbent, through the construction of the adsorption isotherm for the methylene blue.

First, the calibration curve was determined for methylene blue by solutions with concentrations equal to 1.25; 2.5; 5; 7.5; 10 and 12.5 mg/L, using a UV-VIS spectrophotometer (model 200-1000 NM, Bioespectro, Curitiba, Brazil) at a wavelength equal to 664 nm.

Using the Concentration vs. Absorbance (Abs) curve, it was possible to calculate the concentration of methylene blue in the following experiments by absorbance values.

The experiments were carried out in 6 beakers with 500 mL solutions at the concentrations of 25, 50, 100, 150, 200, 250 mg / L of dye and a fixed amount of adsorbent material of 2.5 g (Santos et al., 2007), under constant stirring for 24 hours, until the system was in equilibrium. After the end of the experiment, the solutions were filtered and the absorbance reading was performed on the UV-VIS spectrophotometer. With the data obtained, it was possible to calculate the amount of dye adsorbed by the peanut through Equation (5).

$$q_e = \frac{V(C_0 - C_e)}{m} \quad (5)$$

in which q_e is the amount of methylene blue adsorbed at equilibrium (mg/g); C_0 is the initial concentration of the dye solution (mg/L); C_e is the concentration of the solution at equilibrium (mg/L); V is the volume of the dye solution (L); and m is the mass of the adsorbent (g). The results were analyzed by fitting the Langmuir; Freundlich; Sips; Temkin; Tóth; Redlich and Peterson models, which represent the behavior of the isotherms in the adsorption process.

2.5. Pyrolysis: Thermogravimetric analysis

The thermogravimetric dynamic test was performed at the Energy Laboratory of the UFES, University Center of Espírito Santo, in a TGA/SDTA (model 851, Mettler Toledo), under conditions of kinetic control.

The experiment consisted of the continuous recording of the mass loss of the sample by means of a micro-scale, which was subjected to a heated inert atmosphere (N_2 inert gas with a flow rate of 50 ml/min). The analysis was performed in 70 μ L alumina crucible. The initial sample mass was about 9 mg. The dynamic test consists of subjecting the biomass sample to a drying step, followed by a linear increase in temperature from 298 K to a final temperature of 898 K at the heating rate of 50 K/min. In this work, the kinetic parameters of the slow pyrolysis of peanut meal impregnated with methylene blue dye were calculated using a single-stage global reaction model with a n reaction order. The biomass conversion into volatile material (X) is defined by Equation (6):

$$X = \frac{m_0 - m}{m_0 - m_\infty} \quad (6)$$

in which m is the mass analyzed at each determined time of the TGA experiment, m_0 is the initial mass and m_∞ is the final mass. Thus, the time conversion variation is given by a reaction of order n (Equation 7), in which the reaction constant is represented by the Arrhenius equation.

$$\frac{dX}{dt} = k_0 \exp\left(\frac{-E_a}{RT}\right) (1-X)^n \quad (7)$$

in which k_0 it is the pre-exponential factor; E_a is the activation energy; n is the order of the reaction, t is the time, T is the temperature and R is the universal gas constant.

The parameters, E_a and n were determined using the Differential Evolution technique as developed by Santos et al. (2012c). Equations 8 and 9 present the objective functions

defined for the TG (mass loss) and DTG (mass loss rate) curves respectively, and the objective function of the TG is supposed to be minimized by the least-squares method.

$$O.F._{TG} = \sum_{j=1}^N (m(t)_j^{obs} - m(t)_j^{calc})^2 \quad (8)$$

$$O.F._{DTG} = \sum_{j=1}^N ((dm/dt)_j^{obs} - (dm/dt)_j^{calc})^2 \quad (9)$$

To compare the results obtained in the literature, the deviation between the TG and DTG curves observed and the calculated for the optimal point was given as a percentage of the largest measure (Equations 10 and 11).

$$FIT_{TG} (\%) = 100 \sqrt{(O.F._{TG})/N} / \max(|m_j^{obs}|) \quad (10)$$

$$FIT_{DTG} (\%) = 100 \sqrt{(O.F._{DTG})/N} / \max(|(dm/dt)_j^{obs}|) \quad (11)$$

2.6. FT-IR analysis of products

The spectra were collected in a medium infrared spectrometer (FT-IR ALPHA model, Bruker). The sample module was QuickSnap Platinum ATR, equipped with ATR diamond module for the analysis of powders, solids, slurries, and liquids. It has a finger-operated clamping mechanism and pressure applicator with 360 ° rotation.

Thus, it was possible to obtain the infrared transmittance spectra (from 400 to 4000 cm⁻¹) for untreated samples of fresh peanut, dry peanut, bran, peanut oil obtained from the extraction and pie with adsorbed dye, through the OPUS / Mentor software.

3. Results and Discussion

3.1. Peanuts characterization

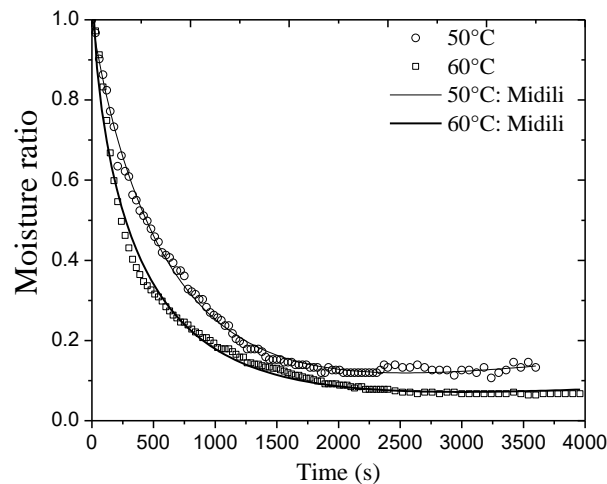
Samples of dried peanuts were subjected to several steps in order to quantify moisture content, ash content, volatile content, and fixed carbon. The immediate analyses showed about: 2.93% of moisture, 86.90% of volatiles, 1.19% of ash and 11.92% of carbon. It is worth remembering that the peanut used by the analyzes had already undergone a solar drying process and therefore had a moisture content of less than 5.4%, as predicted in the literature.

Regarding to the particle size distribution, after the sieving, the best model which fitted the data was the R.R.B. Model (R²=0.991), with parameters D¹=1.51 mm and n=3.83, showing that about 63.2% of particles were smaller than 1.51 mm.

3.2. Drying of peanuts

An infrared moisture analyzer (model IV2500, Gehaka) was used to collect data on the percentage of moisture lost. The moisture content curves of the peanuts as a function of the drying time at temperatures of 50 and 60 °C are shown in Figure 1.

Figure 1 – Curve of moisture ratio as a function of time for temperatures of 50 and 60 °C



Source: Authors (2020).

Figure 1 shows the Midili et al. (2002) model that a better fit the curves of moisture content of the peanuts as a function of the drying time at temperatures of 50 and 60 °C. It is observed that at both temperatures the drying time was close to 2500 seconds and that the final residual moisture at the temperature of 60 °C was lower (3.6%) when compared to the temperature of 50 °C (4.6% residual moisture on dry basis).

According to Agência Nacional de Vigilância Sanitária (2003), the moisture content of peeled raw peanuts should be less than or equal to 8% and the moisture content of raw peanuts with peel should be less than or equal to 11%. As the used peanut was bought in the supermarket and peeled, an initial moisture content of less than 8% was expected.

In order to determine the moisture content of the peanut as a function of drying time, the following kinetic model were tested: Lewis (1921), Brooker et al. (1974), Handerson and Henderson (1968), Page (1949), Exponential of two terms, Midili et al. (2002) and Logarithmic, according to Table 1.

Table 1 – Adjustment to Kinetic Models for drying at 50 °C and 60 °C.

Kinetic Models	Temperature		R ²	
	50°C	60°C	50°C	60°C
Lewis (1921) $MR = \exp[(-k \cdot t)]$	$k=0.0013$	$k=0.0019$	0.9142	0.8834
Brooker et al. (1974) $MR = c \cdot \exp[(-k \cdot t)]$	$c=0.8802$ $k=0.0011$	$c=0.8378$ $k=0.00152$	0.9326	0.9084
Handerson & Henderson (1968) $MR = \exp[(-k \cdot t) + \frac{1}{9} \exp(-9k \cdot t)]$	$k=0.0013$	$k=0.0019$	0.9142	0.8834
Page (1949) $MR = \exp[(-k \cdot t^n)]$	$k=0.0094$ $n=0.7081$	$k=0.0196$ $n=0.6385$	0.9683	0.9703
Exponencial de dois termos $MR = c \cdot \exp(-k \cdot t) + (1-c) \exp(-k \cdot c \cdot t)$	$c=0.2807$ $k=0.0034$	$c=0.2721$ $k=0.0052$	0.9519	0.9331
Midilli et al. (2002) $MR = c \cdot \exp[(-k \cdot t^n)] + b_1 \cdot t$	$c=1.0077$ $k=0.0029$ $n=0.9042$ $b_1=3.5756 \cdot 10^{-5}$	$c=1.0867$ $k=0.0170$ $n=0.6831$ $b_1=1.7541 \cdot 10^{-5}$	0.9965	0.9878
Logarithmic $MR = c \cdot \exp(-k \cdot t) + d$	$c=0.8759$ $k=0.0018$ $d=0.1136$	$c=0.8839$ $k=0.0026$ $d=0.0885$	0.9947	0.9799

Source: Authors (2020).

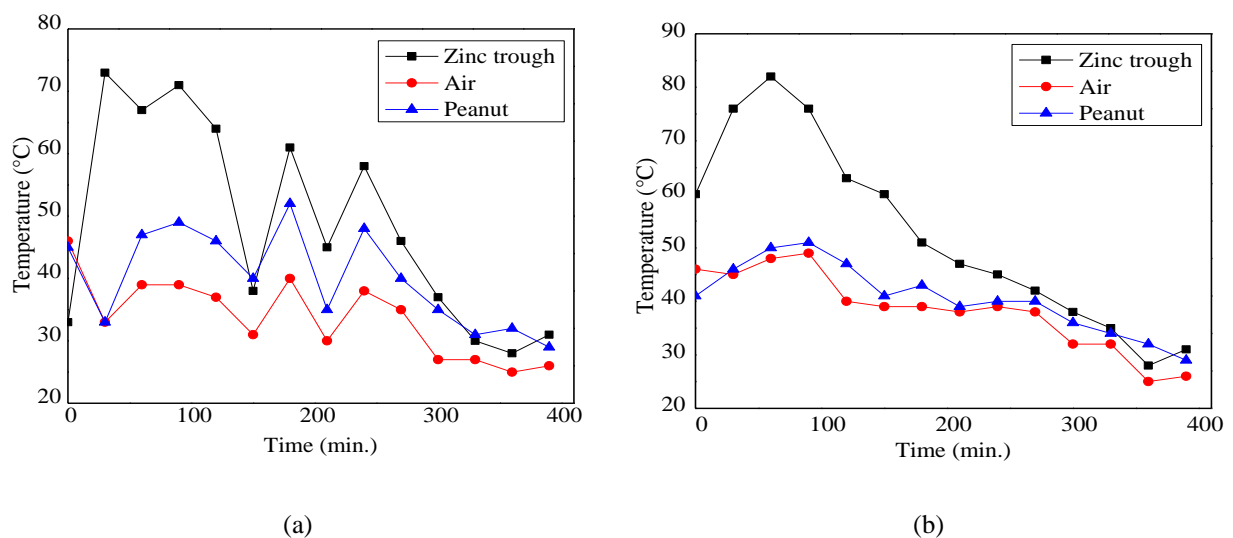
Table 1 shows that the model proposed by Midilli et al. (2002) presented better results in order to represent the experimental data of peanut drying kinetics at temperatures of 50 and 60 °C, with R² equal to 0.9965 and 0.9878 respectively. Table 1 also presents the adjustment values for the parameters of this model: c, k, n and b₁. A decreasing drying rate was observed in peanuts, which decreases continuously throughout the drying process, a behavior already expected in the grains. Thus, heat transfer is not always compensated by mass transfer in the same proportion. The limiting factor at this stage is the reduction of migration of moisture from the interior to the surface of the product.

However, as shown by Dalbello (1995) and Woodrof (1983) elevated temperatures lead to a reduction in grain quality, causing discoloration and rancidity, which affect the acceptance of the product at the marketing level.

In this work, the drying process was only a preservation step for the peanut to be used later, and the ground peanut at 60 °C presented acceptable characteristics of quality. In order to fully exploit the biomass in a sustainable manner, solar drying was also studied. In the experiments, it was possible to evaluate the effect of the mass variation (about 400 and 500 g) on the final moisture ratio, residual moisture on a dry basis and the influence of the trough, air and peanut temperature on the drying efficiency.

It is important to emphasize that the experiments were carried out on different days, but at similar times, from 11:00 am to 5:00 pm and with similar climatic conditions. The temperature data for each experiment were collected over time and are shown in Figure 2.

Figure 2 – Evaluation of the Zinc trough, air and peanut temperatures as a function of time for different masses: (a) Experiment 1: 400 g; (b) Experiment 2: 500 g.



Source: Authors (2020).

Figure 2 shows that a great variation in the trough temperature was observed in both experiments, evidenced by the standard deviation of the measurements. Such oscillations may be related to the low specific heat of the material of its composition, zinc (0.093 cal/g°C), which requires little energy to change its temperature by 1 °C, and is therefore very susceptible to climate change (sudden transition to a cloudy sky).

From the temperature data, it can be said that in experiment 1, an average temperature higher than experiment 2, was obtained. This fact is proved by meteorological data of the

experiment that showed higher solar incidence (maximum of 3500 kJ/m² against 2500 kg/m²) and lower wind speeds, which leads to higher temperatures and lower energy losses of the zinc trough by natural convection of air. In addition, experiment 1, performed under more favorable conditions presented a lower moisture ratio, as can be observed in Table 2.

Table 2 – Experimental data of solar drying.

Experiments	Variables			Results		
	Weight (g)	X _{bu0}	X _{bs0}	X _{bsf}	MR	Mean Temperatures [°C]
1	400	0.0245	0.0252	0.0082	0.046	T _{zinc} = 56.25 T _{air} =40.41 T _p = 42.33
2	500.35	0.0275	0.0283	0.0216	0.244	T _{zinc} = 48.71 T _{air} =33.78 T _p =40.14

Source: Authors (2020).

From the results of Figure 2 and Table 2 it can be noted for experiment 1, carried out with 400 g, the average temperature of the zinc trough was 56.25 °C with a standard deviation of 15.79 °C. The mean air temperature for the same experiment was 40.41 °C, with a standard deviation of 5.58 °C and the peanut temperature equal to 42.33 °C and a standard deviation of 5.26 °C. Experiment 2, carried out with 500 g, the average temperature of the zinc trough was 48.71 °C with a standard deviation of 16.41 °C. The mean air temperature for the same experiment was 33.78 °C, with a standard deviation of 6.35 °C and the peanut temperature equal to 40.14 °C and a standard deviation of 7.66 °C.

In the same way that the trough temperature was evaluated. It was observed in experiments 1 and 2 that the air and peanut temperature showed a little variation during the day, confirmed by the low standard deviation of the data. In addition, it was possible to verify that there was heat transfer from the trough to the peanut since it is perceived that T_p is greater than the T_{air}. Thus, by placing the peanut directly on the trough, the transfer by conduction was greater than the transfer by convection.

It was not possible to compare the drying kinetics of the infrared with the solar dryer, because the average temperature for experiments 1 and 2 was similar. It should also be noted that the moisture ratio was calculated in relation to the amount of moisture withdrawn using the kiln.

3.3. Extraction by conventional method (Soxhlet) and by means of solar concentrator

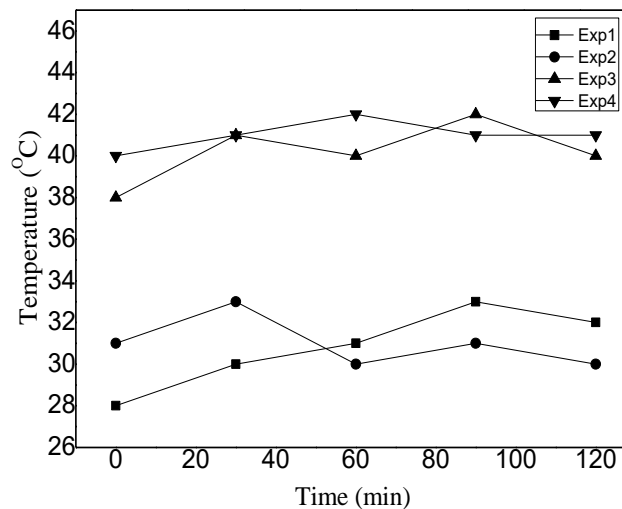
After the drying process, the oil was extracted by the conventional method, using Soxhlet and solar concentrator.

The experiments carried out in triplicate shown that the oil content extracted by the conventional is consistent with the literature since an average oil content of 53.70% was obtained, close to 52.37% obtained by Zullo et al. (1993). The value obtained experimentally was taken as efficiency equal to 100% for purposes of comparison with the method using the solar concentrator.

In order to extract the vegetable oil using the solar concentrator, the absorber bed was filled up with the particles and from the mass and the volume occupied by the sample; the bulk density (ρ_{bulk}) of the particles was calculated. Thus, the bulk density of the dried and ground peanut was equal to 406.78 kg/m³.

Figure 3 shows the temperature as a function of time of the oil and solvent mixture leaving the absorber bed.

Figure 3 – Temperature as a function of time for the process of peanut oil extraction.



Source: Authors (2020).

It is interesting to verify through Figure 3 that Experiments 3 and 4 presented the higher values of Temperature during the extraction. In order to minimize the heat loss of the absorber bed by convection due to the contact with the ambient air, a plastic wrap was used.

Table 3 shows the Oil content and leaching efficiency obtained by extraction with the solar concentrator.

Table 3 - Oil content and leaching efficiency obtained by extraction with solar concentrator.

Leaching Experiments	Oil content (g)	Efficiency (%)
1	37.89	75.78
2	38.58	77.16
3	41.45	82.90
4	49.91	99.82

Source: Authors (2020).

It can be noted in Table 3 that the efficiency of vegetable oil extraction compared to the literature is higher for higher temperatures. In addition, it is important to note by Table 3 that even at temperatures below those observed by Tavares (2015), solar extraction achieved high efficiencies, above 75% for all conditions

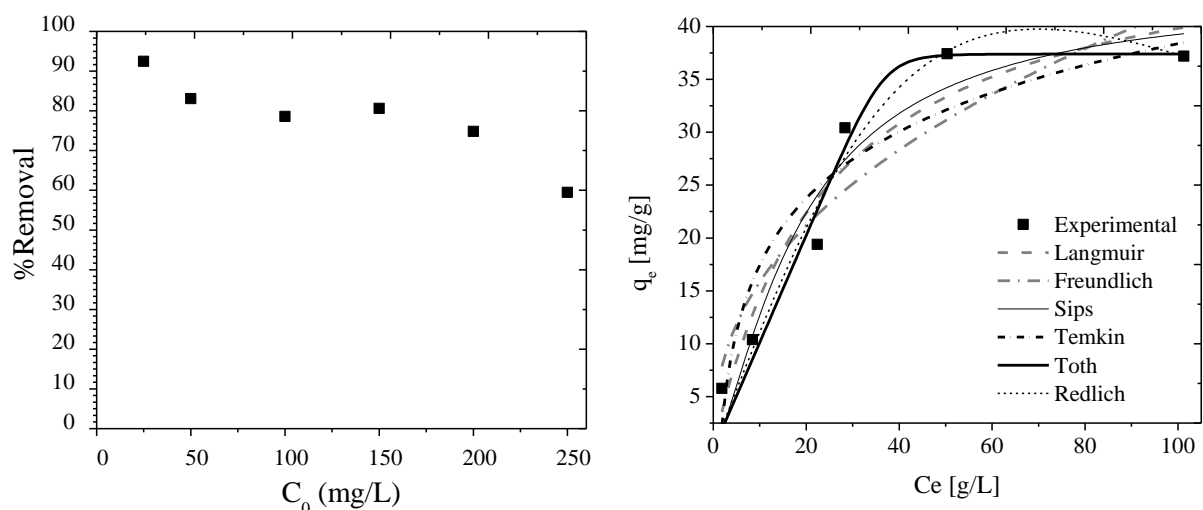
3.4. Adsorption

From the built-up calibration curve, a linear behavior was observed and the straight equation obtained is shown in Equation 12. The linear correlation coefficient was 0.9942.

$$\text{Absorbance} = 0.159 \cdot C_i \left(\frac{\text{mg}}{\text{L}} \right) + 0.005 \quad (12)$$

Thus, the equation of the line obtained can be used to calculate the remaining concentrations in the solutions after the adsorption process, guaranteeing reliable results. These results show the percentage of dye removal versus the initial concentration (Figure 4a).

Figure 4 – Adsorption calibration and fitting models: (a) amount of dye removal (%) vs. initial concentration (mg/g); (b) Models adjusted to experimental isotherm.



Source: Authors (2020).

It can be seen in Figure 4a that the removal of methylene blue was higher at lower concentrations, which was already expected, because at lower concentrations the available amount of active sites from the adsorbent to the dye is higher, whereas at higher concentrations the active sites are saturated more quickly. Although adsorption has been observed to decrease at higher concentrations, peanut can be considered a good adsorbent, since the lowest percentage of removal was 59.49% at a concentration equal to 250 mg/L of solution of methylene blue. The experimental data were adjusted to the isothermal models (Figure 4b), allowing a better understanding and prediction of the adsorption process.

Table 4 presents the fit results of adsorption isotherm models for the adsorption of dye in the peanuts residue

Table 4 - Adsorption Isotherm models for methylene blue adsorption in peanuts residue.

Modelos	Parâmetros	R ²
Langmuir: $q_e = \frac{q_{m\acute{a}x} b_L C_e}{1 + b_L C_e}$	$q_{m\acute{a}x} = 49.491 \text{ mg/g}$ $b_L = 0.041 \text{ L/mg}$	0.9096
Freundlich: $q_e = k_F C_e^n$	$k_F = 6.023 \text{ L/mg}$ $n = 0.412$	0.8304
Sips: $q_e = \frac{q_{m\acute{a}x} (k_s C_e)^{n_s}}{1 + (k_s C_e)^{n_s}}$	$q_{m\acute{a}x} = 43.36 \text{ mg/g}$ $k_s = 0.052 \text{ L/mg}$ $n_s = 1.363$	0.8901
Temkin: $q_e = \beta \ln(k_T C_e)$	$\beta = 9.079$ $k_T = 0.684 \text{ L/mg}$	0.8477
Tóth: $q_e = \frac{q_{m\acute{a}x} C_e}{\left(1/k_T + C_e^{n_T}\right)^{1/n_T}}$	$q_{m\acute{a}x} = 37.391 \text{ mg/g}$ $k_T = 7.401 \text{ L/mg}$ $n_T = 10.928$	0.9420
Redlich and Peterson: $q_e = \frac{k_{RP} C_e}{1 + \alpha_{RP} C_e^g}$	$k_{RP} = 1.133 \text{ L/g}$ $\alpha_{RP} = 1.992 \text{ mg/L}$ $g = 2.004$	0.937

Source: Authors (2020).

Analyzing the correlation coefficients for the models tested, in Table 4 and Figure 4b, it was observed that Freundlich presented the worst performance in the data correlation, with R² equal to 0.8304. The Tóth model presented the best fit, with R² equal to 0.9420 and parameters equal to $k_T = 7.401 \text{ L/mg}$ and $n_T = 10.928$.

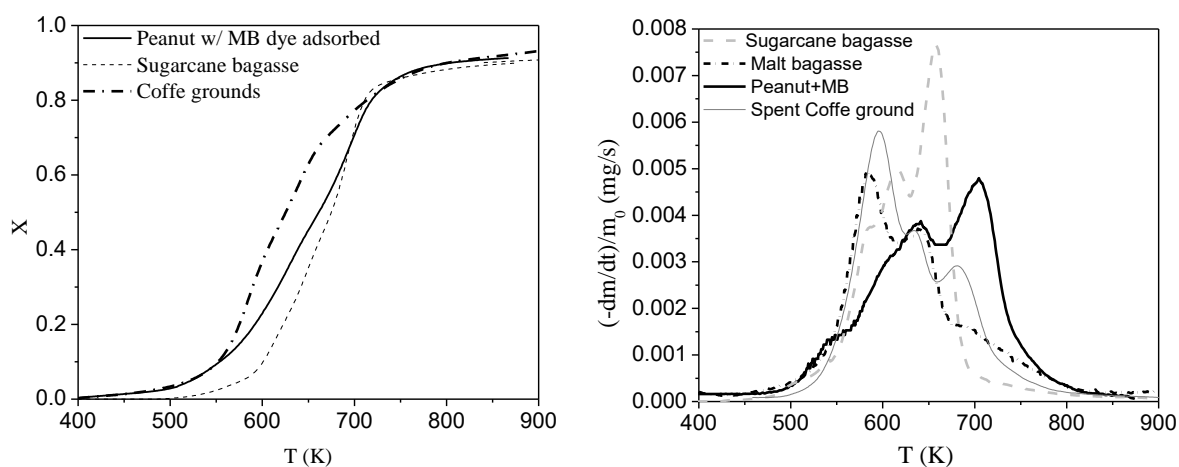
The results obtained for the adsorption capacity of the peanut as an alternative adsorbent show how much this material can be promising in such processes. In addition, the type of isotherm for this process makes it possible to adsorb large quantities of pollutants even at low concentrations of adsorbate in the fluid.

3.5. Pyrolysis (TGA)

From the experimental data of thermogravimetric, the kinetic parameters of the slow pyrolysis were estimated using the differential evolution method, with the following parameter values: $k_0 = 160.206$; $E_a = 50.782$ kJ/mol and $n = 1.116$. This is a low value of Activation Energy, compared with other biomasses, which is good as the process is endothermic, once little energy needs to be provided to start the reaction.

Figure 5 shows the conversion curves and mass loss rate of volatiles of the MB dye impregnated peanut meal, as well as the comparison with other biomasses.

Figure 5 – Experimental curve of pyrolysis kinetics as a function of temperature: (a) Conversion; (b) Derivative from mass loss (DTG).

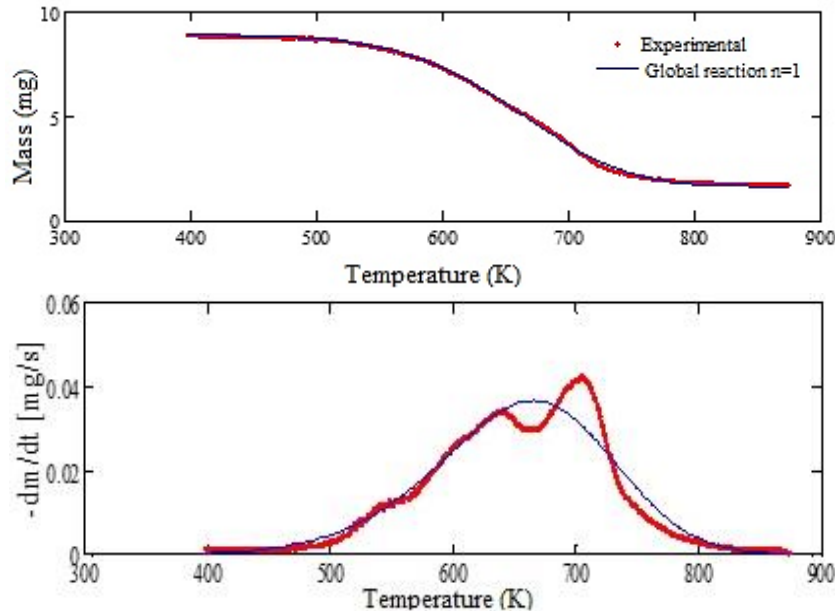


Source: Authors (2020).

Figure 5 also compares the conversion, DTG and the normalized curves for different materials, showing that the differences in the composition of the materials interfere with the kinetics of degradation and consequently the composition of the products formed. However, as the overall conversion behavior is different, it is believed that all biomasses presented have great potential for devolatilization, being good sources for bio-oil production.

Figure 6 shows the experimental and simulated data by the global reaction model for a heating rate equal to 50 K/min.

Figure 6 – Experimental and simulated data by the global reaction model for heating rate equal to 50 K/min.



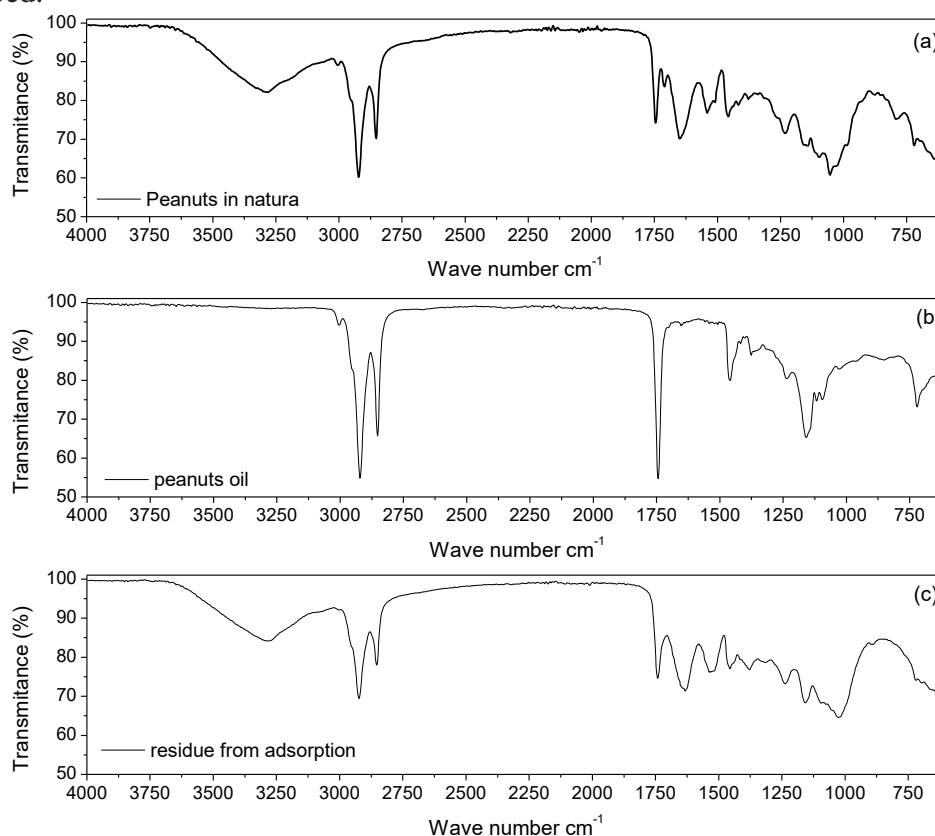
Source: Authors (2020).

Through the fit of the overall reaction model shown in Figure 6 can be noted that the mass loss curve showed R^2 equal to 0.999308 and FIT equal to 0.86% while the DTG curve presented an R^2 of 0.941454 and FIT equal to 7.6%. This model has presented a satisfactory result in the prediction of the mass loss curve and gives a general idea of the energy required to carry out the pyrolysis process, which is endothermic. Since about 93% of the volatile biomass material was released during the pyrolysis up to 600 °C, it is believed that this biomass, even if impregnated with dye, would produce a high proportion of oil, if a pyrolysis reactor was used to provide moderate temperatures, high heat, and mass transfer coefficients and low residence times of the gas phase.

3.6. Fourier transform infrared analysis (FT-IR)

Figure 7 (a) shows the Fourier transform infrared Analysis (FT-IR) of peanuts *in natura*. Figures 7(b) and 7(c) show these results for peanuts oil and peanuts residue with dye adsorbed, respectively.

Figure 7 – FT-IR spectra of: (a) peanuts *in natura*; (b) peanuts oil; (c) peanuts residue with dye adsorbed.



Source: Authors (2020).

The results in Figure 7a showed that at wavelengths close to 3300 cm^{-1} , a very pronounced band, probably due to the stretching of the O–H bond, can be attributed to the presence of water in the sample and to the carboxylic groups. Another important functional group that leads to the broadening of the band in such frequencies is the amine group, present in the proteins, that corresponds to 25% of the composition, in mass, of the peanut. It is interesting to note that the peak has a medium transmittance, which was already expected to take into account that peanut has only 5.4% water according to the literature and high concentration of carboxylic acids and amines. There were also three distinct peaks in the region between 2600 and 2900 cm^{-1} , similar to those obtained in the oil spectrum, which refers to the C–H bonds. The bands near 1700 cm^{-1} refer to the carboxylic acids and their derivatives, which belong to the structures of oil and proteins, the main components of peanuts. The pronounced peak observed near the 1200 cm^{-1} refers to axial deformation vibrations of the carbon-oxygen bond belonging to triacylglycerols and esters. The bands below 1000 cm^{-1} belong to the alkenes present in polyunsaturated carboxylic acids, such as linoleic and linolenic acids and structural components.

It was possible to observe, in Fig 7 (b), very distinct peaks due to the fatty acids present in the samples of vegetable oils. It was also observed the existence of 3 peaks in the region between 2600 and 2900 cm^{-1} . The first peak, at a lower wavelength, refers to the stretching of the C–H bond of the alkanes. It is important to note that the higher the number of C–H bonds, the lower is the transmittance intensity due to the overlap and accumulation of the peaks. These C–H bonds belong to the methyl, methylene and methane groups, which have very distinct peaks in this region of the spectrum and tend to 3000 cm^{-1} , as the concentration in the sample increases (Pereira, 2012). In addition, the simple aliphatic carboxylic acids presented a very strong band of the carbonyl group in the range of 1730-1700 cm^{-1} . In the peanut oil spectrum, the characteristic band of carboxylic acids occurring around 1740 cm^{-1} is due to the axially deformation occurring with the C=O bond. The observation of this band demonstrates the idea that it is a carbonyl that, together with the wide absorption bands in the region of 2900 cm^{-1} (characteristic of carboxylic acid) proves that the spectrum is relative to a sample of vegetable oil, composed of unsaturated and saturated fatty acids. The peaks between 1300 and 1500 cm^{-1} are due to the methyl and methylene groups, C–H bonds which were already expected due to the fatty acid chains of vegetable oils. In the wavenumbers between 1100 and 1250 cm^{-1} , the peaks are related to the axial deformation vibrations of the carbon-oxygen bond that belongs to triacylglycerols. The bands below 1000 cm^{-1} belong to the alkenes presented in polyunsaturated carboxylic acids, such as linoleic and linolenic acids.

For the peanut sample with adsorbed methylene blue, in Fig 7 (c), an increase in transmittance was observed for the peaks in the region of 2600 to 2900 cm^{-1} which are related to the C–H bond, due to the lower concentration of oil, which was extracted before being carried out the adsorption. In addition, there was a decrease in triacylglycerol peaks. It was not possible to observe the peak of the methylene blue since it absorbs in the visible region at 664 nm and the infrared begins at 780 nm.

4. Conclusion

In the present work, possible forms of integral use of the biomass were evidenced. The first one was for immediate analysis confirming that the peanut has a low content of moisture and ash and a high content of volatiles and carbon within the specified. This configuration is due to a large amount of oil present in this biomass.

By means of particle size distribution, it was possible to observe that the three models analyzed had a good fit and that among the three RRB was the one with the highest R^2 . It was possible also to conclude that 63.2% of the particles of the sample had a diameter smaller than 1.51 mm.

In both the infrared and solar drying operations it was possible to observe that the drying time is highly dependent on the operating temperature (drying curves), ie the higher the temperature the lower the drying time. Solar equipment showed that the drying efficiency could reach around 79%.

Solar extraction was one of the most efficient operations studied here. It was possible to achieve high efficiencies (99%), that is, practically all oil could be extracted and temperatures lower than those reached by Tavares (2015). The solar extractor proved to be a good sustainable option rather than Soxhlet extraction, due to the excellent extraction efficiency and energy saving.

Through the pyrolysis, it was possible to observe that the differences in the composition of the materials interfere in the kinetics of degradation and consequently in the composition of the products formed. However, as the overall conversion behavior is different, it is believed that both biomasses presented have great potential for devolatilization, being good sources for bio-oil production.

By the adsorption operation, it was observed that the removal of methylene blue was higher at lower concentrations and that the lowest removal percentage was 59.49% at a concentration of 250 mg/L. In addition, peanut meal is a good adsorbent and the Tóth model was the one that presented the best fit, with R^2 equal to 0.9420. Infrared spectrometry was used to evaluate the composition of peanut oil, fresh peanut, and peanut after adsorption. The water peaks in the samples, carboxylic acids, amines, triacylglycerols and esters could be observed.

From the experimental data of thermogravimetric (TGA), the kinetic parameters of the slow pyrolysis were estimated using the differential evolution method. The low value of Activation Energy ($E_a = 50.782$ kJ/mol) obtained was good as the process is endothermic, so little energy needs to be provided to start the reaction. Through the fit of the overall reaction model, it can be noted that the mass loss curve showed R^2 equal to 0.999308 and FIT equal to 0.86% while the DTG curve presented an R^2 of 0.941454 and FIT equal to 7.6%. In addition since about 93% of the volatile biomass material was released during the pyrolysis up to 600 °C, it is believed that this biomass, even if impregnated with dye, could produce a high

proportion of oil if a pyrolysis reactor was used to provide moderate temperatures, high heat, and mass transfer coefficients and low residence times of the gas phase.

Fourier transform infrared Analysis (FT-IR) was obtained for peanuts in nature, peanuts oil and peanuts residue with dye adsorbed, respectively, allowing to analyze the functional groups of each one and the differences between them.

Therefore, it can be said that the objectives proposed in the work were achieved since it was possible to show the full use of biomass, involving chemical engineering processes. For future work the analysis of the peanut extractives is proposed; the use of the oil removed in cakes and cosmetics, to perform the adsorption in bed and more tests in different conditions.

As suggestions for future work, we intend to add some other unit operations (flocculation, filtration, centrifugation, etc.) in the sequence for integral and sustainable use of biomass, as well as, to deepen with more detailed studies those processes that have already been used in this study (drying, extraction, adsorption, pyrolysis). In addition, it is intended to extend studies of full utilization to other biomasses, such as Baru, coconut, malt, sugarcane bagasse, spent coffee ground, etc.

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