Comparison of Energy Properties of Pellets from Shells of Different Nut Species

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Bio-waste is a source of energy-rich material. Therefore, it can be used in further processes and efficiently utilized. Further processes of waste utilization include compressing them to pellets. However, this type of pellet often has a low bulk density, high ash content, low ash melting temperature, and low calorific value. This research dealt with the energy properties of peanut and pistachio nut shells that were pure or mixed with spruce sawdust in half proportion and compressed into pellets. As a continuation of previous research, the properties of these pellets prepared were measured and compared with pure spruce pellets and with pellets from walnut shells. The tested shell pellets had calorific values similar to wood pellets from spruce. However, mixing nut shells with spruce solved the problems of high ash content and low ash melting temperatures. The amount of wood present in the pellets resulted in increased ash melting temperatures and decreased the ash content.

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INTRODUCTION

Residues from logging in forestry and crop harvesting are a source of large amounts of bio-waste every year that can be used in the energy sector. Otherwise, they are landfilled. These energy materials can help meet the European Union's carbon neutrality targets by 2050 (Cardozo *et al.* 2014). The current energy crisis has reopened Europe's energy dependence on third-country fossil fuels and highlighted the need for energy independence and self-sustainability.

Nevertheless, the high degree of deforestation in Europe and the enormous logging in many European countries also prove that this problem cannot be solved unless wood is harvested in a sustainable manner. Because of poor timber management, Europe is also facing a shortage of wood materials for construction purposes as well as wood pellets for heating in small heat sources. Therefore, the price of wood in the markets is continuously rising and wood is becoming a valuable commodity requiring careful treatment. In addition, it is necessary to replace some of the wood demand by utilization of waste biomass, especially in the production of energy materials.

Many studies have already focused on the production of pellets from alternative biomass or its admixture into wood sawdust. An example is an Italian study that assessed the energy performance of pellets made from hazelnuts and olives obtained by pruning on local farms. The research showed a slight deterioration in the quality of the produced pellets, and further research was proposed to determine the appropriate ratio of the given materials with clean sawdust (Acampora *et al.* 2021). It is also possible to press waste shells into larger forms, which was effectively demonstrated in one Indonesian study using local cashew nut shells waste to make briquettes (Ifa *et al.* 2020).

Much current research has gone even further and investigated various types of thermal decompositions, such as pyrolysis and hydrothermal carbonization, or liquefaction of different types of shells. The complex results of the pyrolysis of the utilization of the local source of Brazilian nut shells and peels are also discussed in a study by Colpani *et al.* (2022).

Alternative sources of waste biomass from forestry can be divided into basic groups such as thin branches, roots, bark, seed shells, leaves, and needles. For agricultural residues, these are mainly stems and shells. Shells are one of the waste raw materials common to agricultural and forestry biomass and can be obtained after separation of the seed at harvest in large quantities in one place. Sometimes they form a hard woody seed shell, but for agricultural biomass, they form rather soft paper-like tissues. In addition, the seed shells have optimal properties in terms of bulk density and moisture content. Often their production is linked to a local agricultural resource, such as camellia plantations, in China, which produce energy-rich waste camelia nut shells (Zhang *et al.* 2015).

Two types of nut shells were chosen for this study. Pistachios were selected as a representative of hard nut shells and peanuts as a representative of soft agricultural shells. World production of pistachios last year was 0.8 million metric tons, mainly in countries such as Iran, USA, and Turkey, and also in Southern European countries such as Greece, Spain, and Italy (Apaidin-Varol *et al.* 2007; Acikalin *et al.* 2012). For peanuts, last year's world production was even at the level of 51 million metric tons, and the largest producers this time were China, India, and Nigeria and some South Europe countries produce small amounts too (Perea-Moreno *et al.* 2018; Kumar *et al.* 2021). Despite the most suitable conditions for growing these crops, they still form part of the European diet and are imported into the EU from other countries. Thus, nut processing factories generate tons of biodegradable waste every year, and these have potential to be used in pelletizing lines to create an ecological type of fuel. However, before the technical treatment of this waste, it is necessary to consider its energy and technical properties related to the quality of incineration.

The problems arising in using of some types of alternative biomass pellets can be connected to a deterioration of properties such as calorific value, durability, or chemical content. Combustion of such unsuitable pellets can lead to a decrease in the performance of the combustion plant or problems with maintaining the combustion process. The right ratio of raw materials for pellet production is one of the possible solutions to mitigate the negative effects of some types of alternative biomass pellets.

One possibility is even mixing less calorific biomass with energy-richer materials such as coal. This was confirmed by research in which various types of waste biomass were mixed with lignite and bituminous coal (Guo *et al.* 2020). However, to determine the suitability of certain alternative biomass and its correct ratio in the pellet, it is necessary to experimentally determine the basic properties of the pellets before using them for commercial purposes.

This research dealt with the analysis of pellets of peanut and pistachio nut shells that were either pure or mixed with spruce sawdust in half proportion. The aim was to determine the energy properties of the input raw materials of alternative waste biomass from pistachios and peanuts and their improvement by mixing with spruce sawdust. Through mixing input raw materials of lower quality with higher quality raw materials, the authors were able to improve energy properties and increase the quality of combustion.

MATERIAL AND METHODS

As a raw material to produce pellets, spruce sawdust, pistachio shells, and peanut shells were used in this work, as shown in Fig. 1. Spruce sawdust was obtained from a local wood processor (Zilina, Slovakia) in the optimal size without the need for further treatment. Pistachio and peanut shells were obtained by shucking the seeds and were ground using a 750 W industrial crusher (CS-700; Yongkang Tianqi Shengshi Industry and Trade Co., Ltd., China) before pressing. The ground feed material did not require further drying. The moisture of spruce sawdust ranged from 15% to 25%.

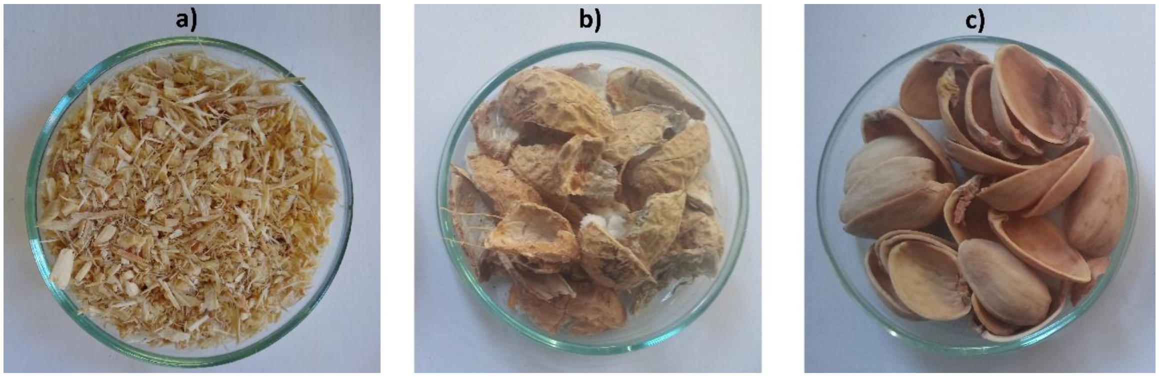


Fig. 1. Input materials: a) Spruce sawdust; b) Peanut shells; c) Pistachio nut shells

A small pellet press (M-200; Cronimo Ltd., Ludgerovice, Czech Republic) with a power of 7.5 kW was used for the pelletization. In total, three pure samples were created with 100% content of spruce sawdust, pistachios, and peanuts. The remaining two samples with 50% peanut and pistachio content were mixed with spruce sawdust. The input materials were pressed with roughly the same amount of pressure. The formed pellets were then air-dried in the laboratory. The air-drying process took approximately one week, and the temperature range for the pellets was between 23 and 25 °C with a relative humidity of 40% to 50%. The formed pellets are shown in Fig. 2.

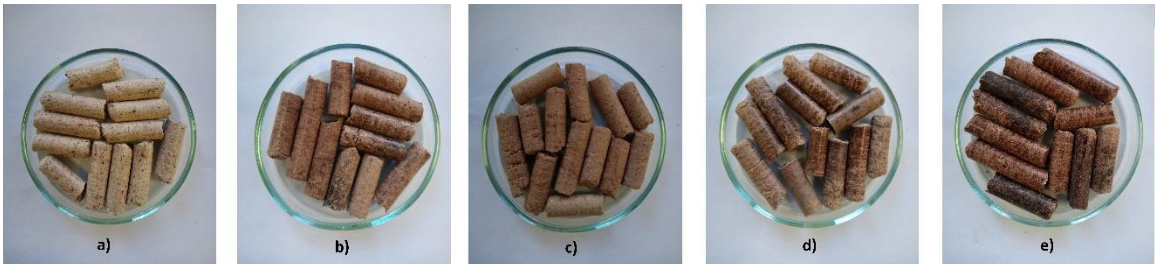


Fig. 2. The formed pellets: a) 100 Spruce pellets; b) 50 Pistachio; c) 100 Pistachio; d) 50 Peanut; and e) 100 Peanut

After the pelletizing process, the elemental, thermogravimetric (TGA), and calorific analyses, including the measurements of ash melting temperature of formed pellets were performed. An elemental analyzer CHN628 (Leco Corporation, St. Joseph, MI, USA) was used for elemental analysis. Through the burning of pre-weighed individual samples (~ 0.1 g), this elemental analyzer could detect the contents of carbon, hydrogen, and nitrogen. The TGA was completed using a TGA 701 thermogravimetric analyzer (Leco Corporation, St. Joseph, MI, USA). The amount of moisture, volatile matter, fixed carbon, and ash were obtained from TGA. Each pre-weighed sample of 1.2 g was used. Individual samples were heated at 107 °C in air atmosphere, then the temperature was slowly increased to 900 °C in nitrogen atmosphere, and finally cooled at 550 °C in air atmosphere following standards for solid biofuels ISO 18134 (2015), ISO 18122 (2015), and ISO 18123 (2015).

The calorific analysis was realized using a Leco AC 500 calorimeter (Leco Corporation, St. Joseph, MI, USA). The calorimeter can carry out the high calorific values (HCV) of pre-weighted individual samples of 1 g. The usable potential of HCV must be determined based on low calorific value (LCV), the value of which depends on the heat absorbed by evaporation of the moisture of the burned sample. The LCV then can be calculated using Eqs. 1 and 2.

$$\mathrm{LCV} = \mathrm{HCV} - r_{H2O}(W_p + 8.94x_H) \quad (1)$$

$$x_H = 0.01 \cdot H_h \cdot B_p \quad (2)$$

In Eqs. 1 and 2, r_{H2O} represents the water heat of vaporization (MJ/kg); W_{p} represents the water content (kg/kg); 8.94 is the hydrogen to water conversion coefficient; H_{h} represents the hydrogen content (kg/kg); and B_{p} represents the volatile contents (kg/kg) of the tested samples.

The Ash Fusion Determinator LECO AF 700 was used to determine the ash melting temperatures. Four standardized ash melting temperatures were measured by this analyzer: (1) the deformation temperature (DT); (2) the shrinkage temperature (ST); (3) the hemisphere temperature (HT); and (4) the flow temperature (FT). Oxidizing conditions were used during measurements. Individual samples were prepared from the fuel by ashing at 550 °C according to ISO 21404 (2020) for solid biofuels.

All the measurements were repeated at least three times. The resulting values were mean measurement values with standard deviations calculated in Microsoft Excel (Microsoft Corp., version 2209, Redmond, WA, USA) program.

EXPERIMENTAL RESULTS AND DISCUSSION

The following section describes the results from the fundamental energy analysis from thermogravimetry, elemental analysis, calorific value, and fusibility of ash.

The results of the elementary analysis of pressed pellets with different contents of spruce sawdust, peanut shells, and pistachio nut shells are shown in Table 1. The samples showed approximately the same proportion of carbon, but the highest carbon content was shown by the peanut samples and the least amount was found in pistachios. Bigger differences were noticeable especially in the nitrogen and hydrogen contents. Spruce sawdust contained noticeably less nitrogen and higher hydrogen content, while pistachios had higher nitrogen and lower hydrogen contents. Even larger values of nitrogen and smaller values of hydrogen were measured in the peanut sample. Both samples of peanuts and pistachios showed a slight increase in sulfur content at the level of twice the content in spruce sawdust, while a lower sulfur content was recorded in the sample of peanuts.

Based on the results from the study of Čajová Kantová *et al.* (2022), carbon content in walnut shells is between spruce and pistachio, and nitrogen content in walnut shells is similar to pistachio. The sulfur content was not measured for walnut shells. Therefore, pistachio shells present a prospective energy potential with regard to the emerging emissions of nitrogen or sulfur oxides. Peanut shells have a higher content of nitrogen, which could cause the higher formation of nitrogen oxides during their combustion.

Table 1. Elemental Analysis of Tested Pellets

Sample	C Content (%)	H Content (%)	N Content (%)	S Content (%)
100% Spruce	48.15 ± 0.25	6.05 ± 0.04	0.06 ± 0.003	0.03 ± 0.001
50% Pistachio	46.58 ± 0.03	5.45 ± 0.03	0.10 ± 0.004	0.06 ± 0.002
100% Pistachio	47.27 ± 0.07	5.35 ± 0.05	0.14 ± 0.007	0.07 ± 0.001
50% Peanut	47.97 ± 0.13	5.40 ± 0.05	0.16 ± 0.005	0.05 ± 0.003
100% Peanut	49.12 ± 0.02	5.22 ± 0.04	0.49 ± 0.009	0.06 ± 0.002

The calorific value results are shown in Fig. 3. The measurements showed that the high calorific value of alternative fuels based on peanuts and pistachios was comparable to standard spruce wood. The calorific value of pistachios was found to be slightly lower than that of spruce wood, whereas peanuts showed a slightly higher calorific value. Based on the previous study (Čajová Kantová *et al.* 2022) the higher calorific value of walnut shells is significantly higher than of peanut and pistachio shells. The stated value of HCV of walnut shells was 20.01 MJ/kg.

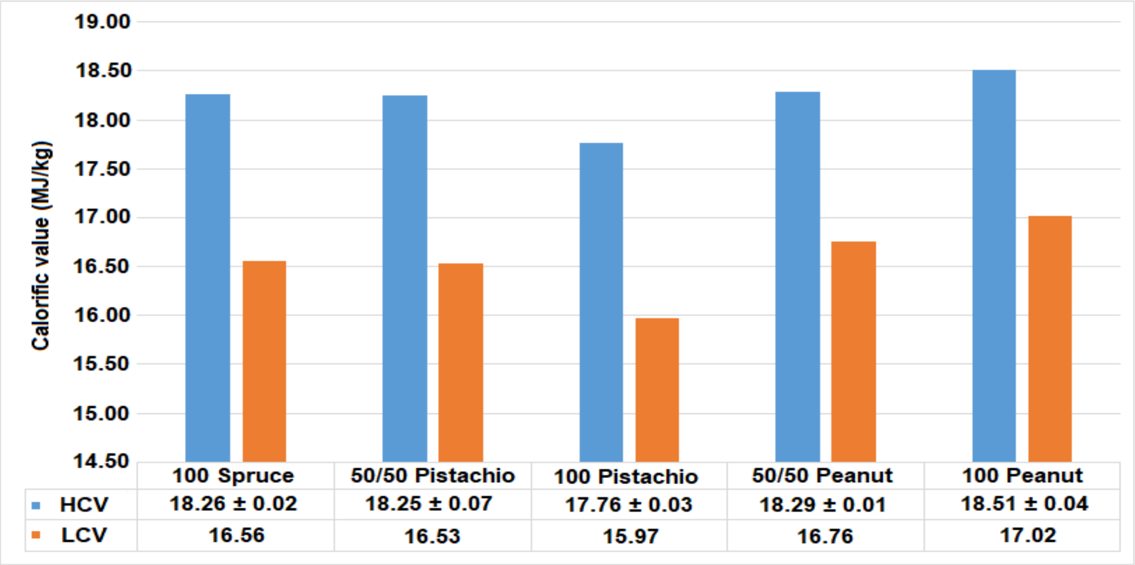


Fig. 3. Calorific values of tested pellets

The lowest decrease in calorific value from HCV to LCV was calculated on the peanut sample, which, according to Table 2, also had the lowest moisture value. On the other hand, the lowest results for LCV were exhibited by the pistachio sample with highest moisture. Similar results were obtained in Swedish research on both types of shells (Noszczyk *et al.* 2021).

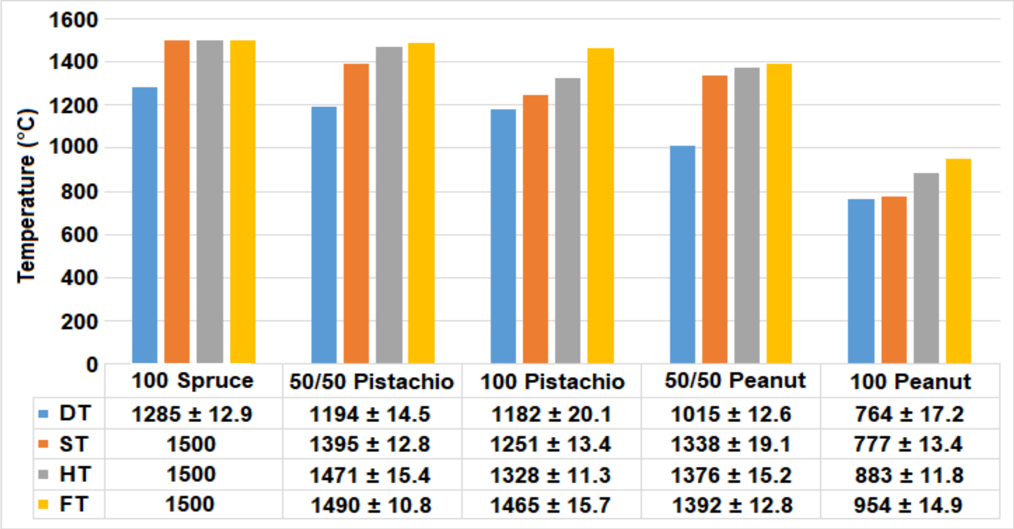
Table 2 shows the results of thermogravimetric measurements determining the content of moisture, volatile combustibles, fixed carbon, and ash in the samples. From the obtained data, a higher content of fixed carbon and a lower content of volatile combustibles are evident in pistachio samples, but especially in peanut samples compared to spruce wood. It is also possible to see a proportional increase in the ash content in the sample of pistachios and peanuts. The pistachio sample contained the highest moisture values, while a significant decrease in moisture was found in the peanut sample, which was even drier than the sawdust sample.

Based on the previous study of Čajová Kantová *et al.* (2022), the volatile matter in walnut shells is between peanut and pistachio shells and the ash content in walnut shells is similar to that from peanut shells. Therefore, pistachio shells present prospective energy potential with regard to the volatile matter and ash content.

Table 2. Proximate Analysis of Tested Pellets

Sample	Moisture (%)	Volatile Matter (%)	Fixed Carbon (%)	Ash (%)
100% Spruce	6.59 ± 0.09	74.99 ± 0.12	17.89 ± 0.15	0.49 ± 0.01
50% Pistachio	6.68 ± 0.38	73.47 ± 0.76	18.56 ± 0.63	1.29 ± 0.18
100% Pistachio	7.12 ± 0.07	72.63 ± 0.22	18.66 ± 0.02	1.59 ± 0.08
50% Peanut	4.97 ± 0.13	73.29 ± 0.79	20.34 ± 0.69	1.41 ± 0.01
100% Peanut	4.24 ± 0.18	70.56 ± 0.06	23.13 ± 0.21	2.07 ± 0.05

Figure 4 presents the results of ash melting temperature measurements. As expected, there was a decrease in the melting temperatures of alternative materials based on pistachios and especially peanuts compared to spruce wood, on which only the deformation temperature (DT) was measured. DT of pistachios remained at a high level even in the pure sample, and its mixing with spruce sawdust increased it only negligibly.



	100 Spruce	50/50 Pistachio	100 Pistachio	50/50 Peanut	100 Peanut
■ DT	1285 ± 12.9	1194 ± 14.5	1182 ± 20.1	1015 ± 12.6	764 ± 17.2
■ ST	1500	1395 ± 12.8	1251 ± 13.4	1338 ± 19.1	777 ± 13.4
■ HT	1500	1471 ± 15.4	1328 ± 11.3	1376 ± 15.2	883 ± 11.8
■ FT	1500	1490 ± 10.8	1465 ± 15.7	1392 ± 12.8	954 ± 14.9

Fig. 4. Ash melting temperatures of tested pellets

However, the addition of spruce sawdust helped to increase shrinkage temperature (ST) and hemisphere temperature (HT), while flow temperature (FT) did not increase much. Unlike pistachios, significantly lower deformation temperatures were recorded for the pure peanut sample, and the addition of spruce sawdust resulted in a substantial increase in DT and a larger jump in ST, while growth at the HT and FT levels slowed down.

The DT of walnut shells is significantly higher than of peanut shells but still significantly lower than of spruce or pistachio shells (Čajová Kantová *et al.* 2022).

CONCLUSIONS

1. The problems of agricultural pellets are their high ash content and low ash melting temperatures, which cause sintering problems during their combustion. Tested shell pellets from this study had calorific values similar to wood pellets from spruce. However, the mixing of nut shells with spruce solves the problems with the high ash content and low ash melting temperatures, because the amount of wood in waste pellets raises ash melting temperatures and decreases the ash content.
2. Pistachio shells present prospective energy potential with regard to the emerging emissions of nitrogen or sulfur oxides, also with their volatile matter, ash content, and relatively prospective value of ash melting temperatures or calorific values. Their ash content was between spruce and peanut shells. Peanut and pistachio shells had similar calorific values than spruce.
3. Peanut shells, despite the increased content of ash and potentially emitting elements of nitrogen and sulfur, according to the findings, had a significantly lower moisture than the spruce sawdust used for standardized pellets. This may be caused by the oiliness of the peanuts, which may also show up in the soft structure of the shell, which may also be a consequence of the higher calorific value. In comparison, the hard shell of pistachios behaves like standard wood and absorbs even higher moisture values.

4. A lot of the agricultural biomass including shells, such as peanut or pistachio nuts shells, still ends up in landfills without further usage. However, the findings from the measurements in the article prove their suitability for use in the energy industry. Furthermore, it is necessary to look for the possibility of using these resources in accordance with the requirements and the minimum burden on the environment.

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REFERENCES CITED

Acampora, A., Civitarese, V., Sperandio, G., and Rezaei, N. (2021). “Qualitative characterization of the pellet obtained from hazelnut and olive tree pruning,” *Energies* 14(14), article 4083. DOI: 10.3390/en14144083

Acikalin, K., Karaca, F., and Bolat, E. (2012). “Pyrolysis of pistachio shell: Effects of pyrolysis conditions and analysis of products,” *Fuel* 95, 169-177. DOI: 10.1016/j.fuel.2011.09.037

Apaidin-Varol, E., Putun, E., and Putun, A. E. (2007). “Slow pyrolysis of pistachio shell,” *Fuel* 86(12-13), 1892-1899. DOI: 10.1016/j.fuel.2006.11.041

Čajová Kantová, N., Čaja, A., Belány, P., Kolková, Z., Hrabovský, P., Hečko, D., and Mičko, P. (2022). “Mechanical and energy properties of pellets formed form walnut shells bledned with spruce sawdust,” *BioResources* 17(1), 1881-1891. DOI: 10.15376/biores.17.1.1881-1891

Cardozo, E., Erlich, C., Alejo, L., and Fransson, H. T. (2014). “Combustion of agricultural residues: An experimental study for small-scale applications,” *Fuel* 115, 778-787. DOI: 10.1016/j.fuel.2013.07.054

Colpani, D., Santos, O. V., Araujo, O. R., Lima, R. M. V., Tenório, S. A. J., Coleti, J., Chaar, S. J., and Souza, C. K. L. (2022). “Bioenergy potential analysis of Brazil nut biomass residues through pyrolysis: Gas emission, kinetics, and thermodynamic parameters,” *Cleaner Chemical Engineering* 1, article ID 100002. DOI: 10.1016/j.clce.2022.100002

Guo, F., He, Y., Hassanpour, A., Gardy, J., and Zhong, Z. (2020). “Thermogravimetric analysis on the co-combustion of biomass pellets with lignite and bituminous coal,” *Energy* 197, article ID 117147. DOI: 10.1016/j.energy.2020.117147

Ifa, L., Yani, S., Nurjannah, N., Darnengsih, D., Rusnaenah, A., Mel, M., Mahfud, M., and Kusuma, S. H. (2020). “Techno-economic analysis of bio-briquette from cashew

nut shell waste," *Heliyon* 6, article ID e05009. DOI: 10.1016/j.heliyon.2020.e05009
ISO 18122 (2015). "Solid biofuels – Determination of ash content," European Committee for Standardization, Brussels, Belgium.
ISO 18123 (2015). "Solid biofuels – Determination of the content of volatile matter," European Committee for Standardization, Brussels, Belgium.
ISO 18134 (2015). "Solid biofuels – Determination of moisture content," European Committee for Standardization, Brussels, Belgium.
ISO 21404 (2020). "Solid biofuels – Determination of ash melting behaviour," European Committee for Standardization, Brussels, Belgium.
Kumar, M., Rai, D., Bhardwaj, G., Upadhyay, S. N., and Mishra, P. K. (2021). "Pyrolysis of peanut shell: Kinetic analysis and optimization of thermal degradation process," *Industrial Crops and Products* 174, article ID 114128. DOI: 10.1016/j.indcrop.2021.114128
Noszczyk, T., Dyjakon, A., and Koziel, A. J. (2021). "Kinetic parameters of nut shell pyrolysis," *Energies* 14(3), article 682. DOI: 10.3390/en14030682
Perea-Moreno, A. M., Manzano-Agugliaro, F., Hernandez-Escobedo, Q., and Perea-Moreno, A. J. (2018). "Peanut shell for energy: Properties and its potential to respect the environment," *Sustainability* 10(9), article ID 3254. DOI: 10.3390/su10093254
Zhang, J., Wang, Z., Du, M., Yao, X., and Hu, L. (2015). "Physicochemical properties of camellia nut shell and its thermal degradation characteristics," *BioResources* 10(1), 647-659. DOI: 10.15376/biores.10.1.647-659

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