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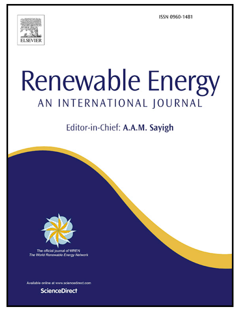

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PHYSICAL AND COMBUSTION PROPERTIES OF AGRICULTURAL RESIDUE BIO-CHAR BIO-COMPOSITE BRIQUETTES AS SUSTAINABLE DOMESTIC ENERGY SOURCES

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Abstract

Domestic energy sources in sub-Saharan Africa are still mainly comprised of firewood and charcoal. One of the main challenges affecting the uptake of carbonized briquettes is their inefficiency in transferring heat. In this study bio-composite briquettes were developed from rice husks, coffee husks and groundnut shells, in varying proportions. The briquettes were developed under low pressure (≤ 7 MPa) after carbonization and application of starch binder. Thermal properties of the developed bio-composite briquettes were determined by using a bomb calorimeter and thermogravimetric analysis to determine calorific values and physical properties, respectively. Drop strength tests and particle density determinations were performed to study the mechanical strength and integrity of the developed briquettes. The water-boiling test was used to determine time taken to boil 1 liter of water. Fourier's Law of heat conduction was used to investigate heat transfer rates across the briquettes for conditions of binder/binder-less bonding conditions. Calorific values for the developed briquettes ranged between 16.6 MJ/kg and 22 MJ/kg. Results for drop strength for the developed composite briquettes were all above 86 %, indicating satisfactory characteristics. Bio-composite briquettes developed using coffee and rice husks bio-chars took less time to boil water compared to all the other bio-composite briquette combinations. Particle densities ranged between 430 kg/m^3 and 580 kg/m^3. Heat transfer was enhanced when no binder was present and coffee and rice husks were sequentially placed in the briquette composition. This study showed the advantages of producing bio-char bio-composite briquettes over single constituent briquettes. Bio-composite carbonized briquettes produced from rice husks, coffee husks and groundnut shells are a suitable and sustainable alternative to firewood and charcoal use in sub-Saharan Africa.

Keywords: Bio-composite briquettes; Burning rates; Carbonization; Heat flux; Thermogravimetric analysis.

1. Introduction

Commercial agriculture is a major component in the utilization of land resources in Africa [1]. However, processing of agricultural produce generates tremendous amounts of agricultural wastes [2]. This has resulted in significant challenges in waste management of these agricultural wastes, particularly in Sub-Saharan African countries. One potentially sustainable way of utilizing these wastes is by application of thermo-chemical/ biological/ mechanical techniques to increase their energy potential when converted into briquettes [3].

Briquettes have gained traction because of their advantages over traditional cooking means of firewood and charcoal [4,5]. Development of briquettes and briquetting technology, are not new concepts with recent reviews in literature of briquetting mechanisms [6]. Briquettes have been developed using both low and high pressure techniques with the former yielding more advantages. An underlying factor in both processes is the moisture content of the biomass material. It should be below 13 % for successful briquetting [4,5]. Studies on the development and properties of briquettes developed from agricultural wastes are substantial in the literature. Often times, binders/adhesives are used in the development of briquettes so as to enhance cohesion of the bio-char [6]. This is in addition to the naturally existing lignin in agricultural wastes that provides binding ability during briquetting. However, most of these studies focused on a single monotonous and homogeneous agricultural waste. Whereas previous efforts on briquette development are commendable, one major significant drawback is the inadequacy of information on physical properties, which is extremely important for large-scale utilization of briquettes in domestic cooking applications in sub-Saharan Africa. Bio-composite briquettes like all composite materials enable strengths of each constituent raw material to become amplified. For example, rice husks have high ash content, but coffee husks have good ignition properties. Bio-composite briquettes developed from rice and coffee husks enable the developed briquette to have reduced ash content from a quantitative viewpoint, but also enhances its ignitability due to the presence of coffee husks [4,5]. Additionally, in order for briquettes to make a significant dent in charcoal and fire wood utilization in sub-Saharan Africa, combinations of different agricultural wastes must be considered in their totality. This is because energy demand for fuel for domestic cooking application continues to increase as population growth in sub-Saharan Africa increases.

Composite briquettes blend different properties from different materials hence counter-balancing the strengths and weaknesses of the developed briquettes [7-10]. However, very few studies have investigated the effect of the combinations of various agricultural raw materials on the properties of the bio-composite briquette developed. Research by Muazu and Stegemann, (2015) demonstrated that blending rice husks and corn cobs improved durability of the developed briquettes and reduced their water absorption [9]. Other researchers have used combinations of rice husks with other agricultural materials to develop briquettes. These briquettes yielded energy values, strength and combustion qualities that are sufficient enough to produce required heat for domestic applications [10-18]. Sawdust is another material that has been combined with other agricultural materials to form and characterize briquettes. Such briquettes contain substantial combustion properties for domestic energy [15,19,20]. Additionally, Gil et al., (2010), Mitchual et al., (2013) and Obi, (2015) cited increasing strengths in such sawdust composite briquettes [21-23]. Combinations of several materials to form briquettes led to increasing briquettes strengths compared to single material briquettes [24,25].

Oladeji, (2010) developed briquettes from combinations of rice husks and corncob. The developed briquettes have sufficient calorific values and strength properties [26]. Rice bran and palm kernel shells have been used to form briquettes with increased calorific values compared to single material briquettes [27]. In their study, Rezania et al., (2016) developed briquettes from combinations of water hyacinth and empty fruit bunch. Increasing empty fruit bunch led to an increase in calorific value and volatile matter of the developed briquettes [28]. Somerville, (2016) and Sotannde et al., (2010) used wood species to form briquettes [29,30]. Composite agricultural materials of different nut shells were used to form briquettes by Tembe et al., (2018). Developed briquettes contained high calorific values and densities [31]. Wamukonya and Jenkins, (1995) studied the durability and relaxation of composite sawdust and wheat-straw briquettes as possible fuels for Kenya. Wheat straw briquettes were less durable and expanded most but a mixture of sawdust and wheat-straw greatly improved the durability of the developed briquettes because sawdust reduces the total length increases in briquettes during their production. Wheat straw has lower binding capacity compared to sawdust because the former contains lower amounts of native components, such as lignin and extractives. Binding capacity of raw materials enhances durability of developed briquettes [32]. Additionally, sawdust has a higher bulk density and higher drop resistance compared to wheat straw. Raw material bulk density and drop resistance positively enhances the durability of the developed briquettes. Other composite briquettes have been developed with compositions of coal to enhance their properties [33,34].

Nurhayati et al., (2016) developed composite briquettes from corn cob and rice husks as a way of endeavoring to attain sustainable food security and fuel energy for small scale industries and household communities. They realized that the calorific value of the developed briquettes ranged between 2.7 MJ/kg and 4.4 MJ/kg noting increase in starch binder levels as a possible solution to increasing the calorific value [35]. Rodzkin et al., (2017) undertook a life cycle assessment of biomass production from drained wetlands areas for composite briquettes fabrication. They discussed the possibility to produce energy in form of composite briquettes from grass as well as the need to make briquettes from drier biomass [36]. Hydrochar composite briquettes were developed for food wastes using molasses as binder. The developed briquettes had excellent compressive strength and impact resistance indices. Briquettes made with molasses binder had lower ignition temperature and higher combustion intervals compared to those made with molasses and lime binders [37]. Ndindeng et al., (2015) optimized the mechanical and thermal performance of rice husk, bran, and clay composite briquettes. The briquetting process led to an increase in density of rice husks from 120 kg/m^3 to 600 kg/m^3. The developed briquettes had lower start-up time (< 5mins) compared to charcoal at 10 minutes. The developed briquettes yielded acceptable quality in terms of hardness, burning rate and ignition time [38]. Cao et al., (2018) developed high-strength charcoal composite briquettes using waste cotton stalk and wood sawdust biomass samples pretreated by two different thermal methods, namely, dry torrefaction and hydrothermal treatment. Their realization was that density and compressive strength of briquettes increased with increasing blending temperature [39]. Effects of particle size, pressure and mold diameter on physical characteristics of rice straw briquettes with sawdust as a binder have been studied. It was concluded that sawdust binding material increased the stable density of briquettes, shatter index and calorific values [40]. Boumanchar et al., (2017) studied the effects of material mixtures on the heating values of composites from biochar, different biomass, plastics, synthetic rubber and cardboard [41]. Yank et al., (2016) investigated the physical properties of rice husk and bran composite briquettes under low-pressure densification for rural applications. Rice husks briquettes had a 16.08 MJ/kg calorific value, representing an interesting alternative to fuelwood. Additionally, the type of binder used greatly affected the density and strength properties of the developed briquettes [42]. Jittabut, (2015) determined physical and thermal properties of briquette fuels from rice straw and sugarcane leaves by mixing molasses. Fixed carbon was obtained between 9.06 % and 13.63 %, ash content was between 7.84 % and 12.85 %, volatile matter was 68.14 % - 74.67 while moisture content was in the range of 4.2 % and 6.2 %. Heating values ranged between 16.3-17.83 MJ/kg, density was in the range of 0.53-0.58 kg/m^3 while the compressive strength was in the range of 32.44-44.7 kg/cm^2 [15]. Husain et al., (2002) developed composite briquettes from palm fiber and shell from the processing of palm nuts to palm oil. The briquettes' density ranged between 1100-1200 kg/m^3. The higher heating value, ash content and moisture content were 16.4 %, 6 % and 12 % respectively. [43]. Kaliyan and Morey, (2010) studied the impact of natural binders and solid bridge type binding mechanisms in composite briquettes from corn stover and switchgrass [44]. Amaya et al., (2007) developed activated carbon composite briquettes from rice husks and eucalyptus wood [45]. Biomass associated products have been noted to improve the nitrification process through their interaction of ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) which reduces the amount of greenhouse gases in the atmosphere [46].

The combination of two or more raw agricultural wastes in the developed bio-composite briquette affects the thermal conductivity and heat conduction behavior of the developed briquette. Fibers may be assumed to be continuous, and in some cases longitudinally set within a layer. In addition to matrix formation, heat propagation in composite materials along the direction of fiber layers depends on thermal conductivity of the fibers. In composite materials, weak bonds between layers are along transverse direction to the fiber axis [47]. Interfacial thermal resistance in the transverse fiber direction significantly reduces the thermal conductivity of the composite materials. It is therefore general understood that, thermo-conductivity in woods is direction dependent i.e. anisotropic as opposed to isotropic conductance is metals [48,49]. Therefore, much more work is needed to understand the heat conduction behavior of bio-composite briquettes developed from two or more agricultural wastes. In this study, carbonized briquettes from different combinations of rice husks, coffee husks and groundnut shells were developed as bio-composite briquettes.

2 Experimental

2.1 Materials

Agricultural waste raw materials used in this work were obtained from specific agricultural zones where each crop is grown in Uganda. Groundnut shells were obtained from Ibanda district in Uganda. Coffee husks were obtained from Kanyiginya Coffee factory. Rice husks were obtained from Eastern Uganda. The agricultural waste samples were first cleaned thoroughly to remove dirt. They were then sun-dried for 8 hours to reduce their moisture content to about 13 %. The agricultural waste raw materials were then carbonized into bio-char and crushed for bio-composite briquette development. To form bio-char, the dried/cleaned agricultural waste raw materials were fed in a carbonizer made of a 200 L volume capacity steel drum (2:1 height: diameter ratio) with openings of 0.02 m diameter for air regulation. Ignition of the agricultural waste raw material took place from the top of the carbonizer drum after which the top of the drum was covered. During the carbonization process the holes were covered with mud in order to limit the amount of air available for complete combustion in the carbonizer as the agricultural waste raw material reduced due to pyrolytic processes. [4,5] (see Figure 1).

Figure 1: Agricultural residues before (Top) and after carbonization (bottom) to form bio-char: coffee husks (a), rice husks (b) and groundnut shells (c).

2.2 Briquette development

Bio-chars obtained from the carbonization process were ground to particles sizes of less than 15mm particles and sieved in order to increase their surface area for binding with starch. This was done to enhance the binding ability and eventual drop strength of the developed bio-composite briquette. The particles were then mixed with cassava starch binder in different proportionality from 10 % - 40 %. The combination of the bio-char and binder were then poured in a metallic mold of inner diameter 5cm and a height of 8cm. The mixtures were compressed in the mold to take up the shape of the mold. Specific details of the briquette development process have been provided elsewhere [4,5]. Four categories of bio-composite briquettes were developed namely: (1) rice and coffee husk bio-composite briquettes; (2) groundnut shell and coffee husk bio-composite briquettes; (3) rice and groundnut shell bio-composite briquettes; and (4) Rice, coffee and groundnut shell bio-composite briquettes. In development of low cost carbonized briquettes, human energy dominates as contributions from electrical, chemical and thermal energy are negligible [50]. There was no particle size reduction of raw material prior to carbonization. In the carbonization process, no electrical energy was used and no petrochemical fuels were used. Additionally, no thermal energy was needed to dry the raw materials because they were sufficiently dry (< 13 %) using solar radiation. Typical applied pressure of ≤ 7 MPa is sufficient for producing low-cost briquettes [5, 43].

2.3 Physical properties and thermal analysis

Physical properties including moisture content, ash content, fixed carbon and volatile matter of the developed bio-composite briquettes, were determined using an Eltra Thermostep non-isothermal Thermo gravimetric analyzer (TGA), Haan, Germany. TGA experiments were carried out from room temperature to 920 ^oC with a heating rate of 16 $^oC/min$. High-purity compressed air (oxygen: nitrogen = 21:79, > 99.5 %) was used for cleaning the crucibles and chamber prior to TGA experimentation. Nitrogen gas was used as the purge gas for pyrolysis experimentation. The flow rate was maintained at 1 L/min and the sample masses averaged 1.1 g. Thermogravimetric analysis was also used to determine weight loss of the developed bio-composite briquettes with increase in temperature. An analysis of the burning rates with time increase for the developed bio-composite briquettes was done. The burning rates were measured through Equation 1. Calorific values for the briquettes developed were determined using an IKA C 2000 oxygen bomb calorimeter. Approximately 1 g of developed briquette was placed in a nickel crucible and fired inside the bomb calorimeter using an ignition wire in the presence of oxygen. The experimental values of calorific value were compared to models obtained by Garcia et al., (2014) (See Equation 2) [51]. Standard water boiling tests were used to determine how long it took for 200 grams of briquettes to boil half a liter of water in an improved domestic cook stove typically used in sub-Saharan Africa.

$$B_R = \frac{(W_i - W_0)}{t_i} \quad (\%/\text{min}) \text{ ……………………………………………….……..….Equation 1}$$

Where B_R is Burning rate, W_i is Weight at time i, W_0 is initial weight and t_i is the time it takes for the initial weight to reach W_i.

$$HHV\ \left(\frac{MJ}{kg}\right) = 17300 - 117.51AC + 165.55FC - 232.69MC \text{ …..……………..Equation 2}$$

Where AC is Ash content, FC is Fixed Carbon and MC is Moisture content of the developed briquettes.

For steady-state conditions with no distributed source or sink of energy within the wall, the appropriate form of heat conduction is given by Fourier's heat conduction equation (see Equation 3 and Equation 4) [52]. However, an additional component of thermal resistance associated with heat transfer by convection at the developed bio-composite briquette must be considered (see Equation 5) [52]. Considering the schematic representation for heat flow through particular agricultural wastes that constitute the developed bio-composite briquette with and without starch binder as shown in Figure 2 for a composite consisting of rice husks, coffee husks and groundnut shells.

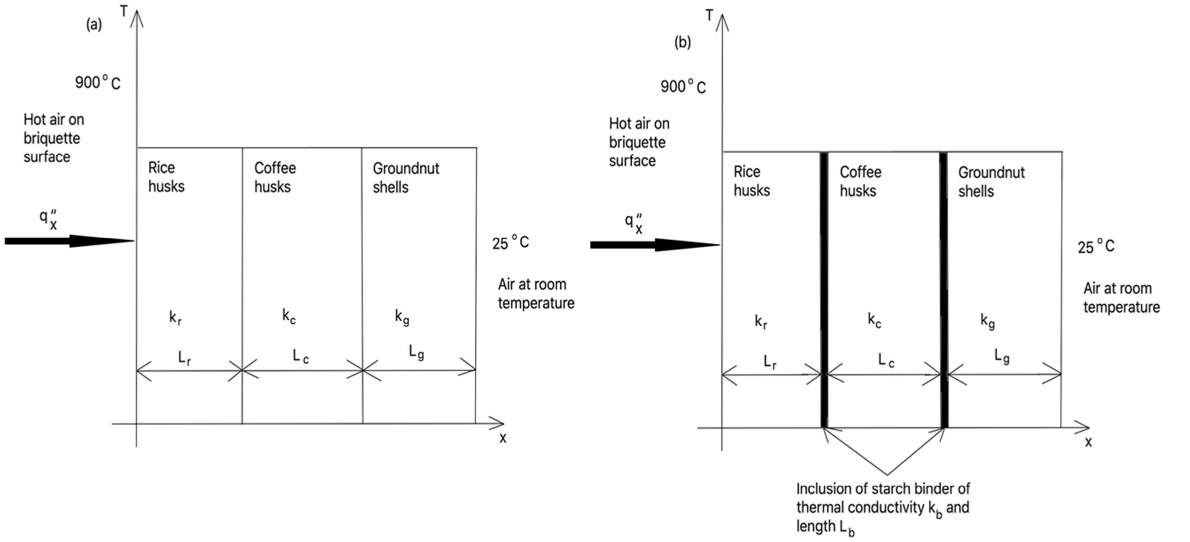

Figure 2: Schematic representation of heat flow through component materials constituting bio-composite briquettes: (a) without starch binder; and, (b) with starch binder

Therefore, for heat conduction:

$$q_x = kA\left(\frac{-dT}{dx}\right) \text{...} \text{Equation 3}$$

$$q'' = \frac{q_x}{A} = k\left(\frac{T_{hot\ air\ surface} - T_{air\ at\ room\ temp}}{L}\right) \text{.....................................} \text{Equation 4}$$

The additional heat transfer by convection is determined from Newton's law of cooling as:

$$q = hA(dT) \text{...} \text{Equation 5}$$

2.4 Mechanical properties

Mechanical integrity of the briquettes under impact loading was determined using drop strength tests. Developed bio-composite briquettes were elevated up to 2 m and then were dropped onto a thick steel metal plate. The ratio of weight after dropping to weight before dropping was recorded as the drop strength. Particle densities of the developed bio-composite briquettes were obtained by standard procedures involving ratios of mass and volume for each bio-composite briquette developed [5].

3 Results and discussions

3.1 Physical properties

Samples of the resultant agricultural waste raw material after carbonization are shown in Figure 1. It was observed that after carbonization, the agricultural wastes all retain their inherent morphological structure. The moisture content, volatile matter, ash content and fixed carbon of the rice husks, coffee husks and groundnut shell agricultural wastes are shown in Figure 3. Rice husks had higher moisture contents at 12.7 %, followed by 9.3 % for groundnut shells and 8.1 % for coffee husks. Rice husks also had the highest ash contents of 19 % compared to 5.3 % and 3.8 % for coffee husks and groundnut shells, respectively. Coffee husks have lower ash contents compared to rice husks because of their lower SiO_2 levels [4]. The relatively high volatile matter content in all the raw materials (> 54 %) make them more readily devolatilized because of increased ignitability potential [4,5]. Moisture contents for all of the agricultural wastes raw materials were all lower than 10 % and 18 % reported by Križan et al., (2009) as being necessary for successful briquetting [53]. Low moisture content values are preferred because higher levels of moisture content in biomass-based fuels result significant amounts of energy being used to evaporate water, which lowers the combustion efficiency [54]. Additionally, combustion of biomass fuels with high moisture contents leads to increasing production of greenhouse gases [55]. However, in other fields like gasification, water content in biomass is advantageous as it increases the Integrated Gasification Combined Cycle (IGCC) efficiency [56].

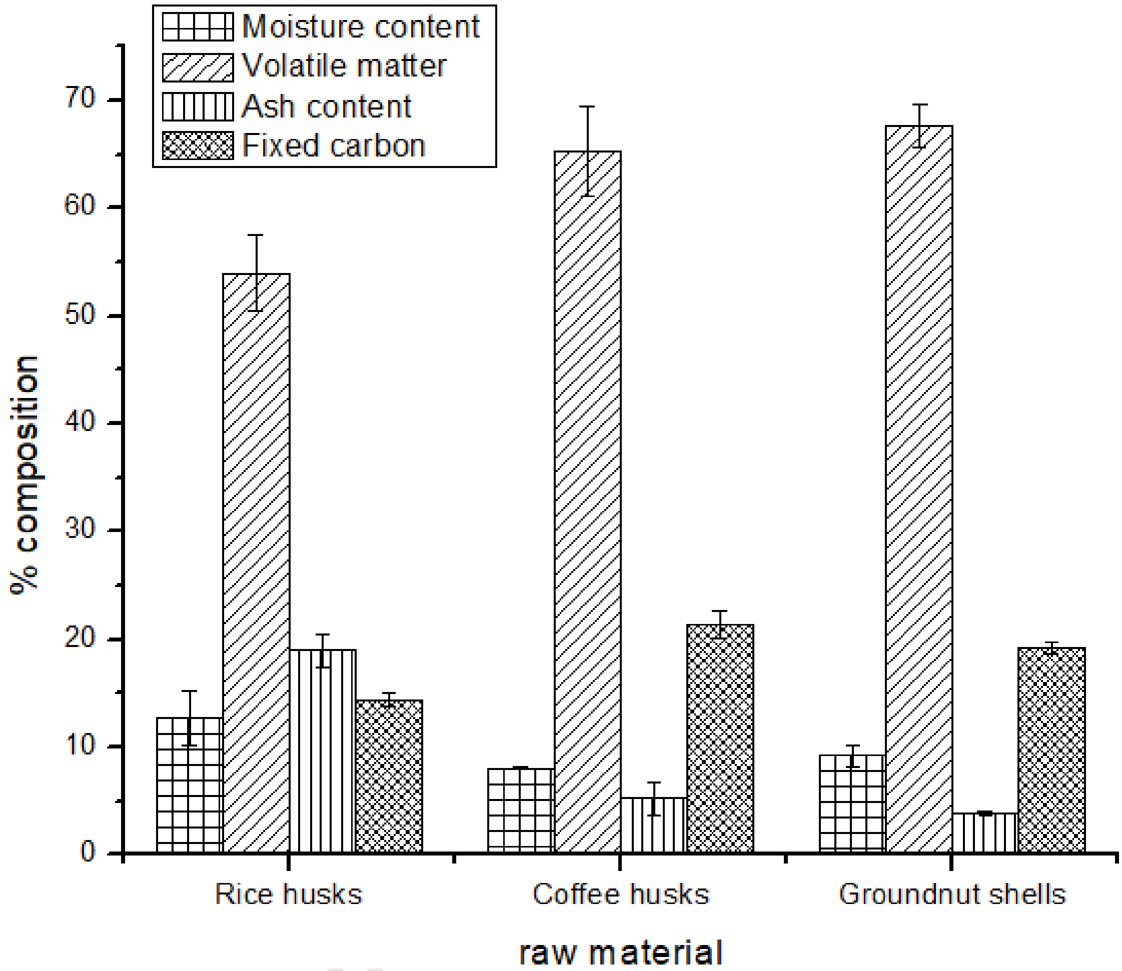

Figure 3: Physical properties for rice husks, coffee husks and groundnut shells agricultural wastes

Physical properties for the developed bio-composite briquettes are shown in Table 1. Moisture contents ranged from 10.98 % - 25.62 %, ash content from 12.21 % - 34.61 %, fixed carbon from 25.7 % - 38 % and volatile matter from 24.61 % - 39.45 %. Volatile matter content for all of the developed bio-composite briquettes were below 40 % which is acceptable for briquette development, especially for enhancing ignition properties [57,58]. During carbonization volatiles are expelled as temperature increases. This reduction in volatiles is partly responsible for reduced smoke during utilization of carbonized briquettes. Fixed carbon contents for the developed bio-composite briquettes were higher than for the agricultural wastes (See Figure 3). This result is expected and can be directly attributed to the carbonization process [4,5]. The carbonization process tends to reduce the amount of water and volatiles in the biomass resulting in an increase in the fixed carbon content [59]. Higher values of fixed carbon are synonymous with higher values of calorific value for the developed bio-composite briquettes [10,53,60]. Ash content in the bio-composite briquettes was higher compared to ash content levels in the agricultural waste raw material. This is particularly noted for bio-composite briquettes that had rice husks constituting 50 % or more in the briquette composition. This is disadvantageous because ash is incombustible and does not provide useful energy for briquette utilization in domestic cooking applications [10,54]. However, previous studies on briquette development from a single agricultural waste reported even higher ash content values [4,5,61]. This implies that development of bio-composite briquettes had a net positive impact on ash content levels.

Table 1: Physical properties for the developed bio-composite briquettes

Composite briquette (%)	Briquette composition (Bio-char)	Moisture content %	Volatile matter %	Ash content %	Fixed carbon %
30C70R	Coffee husks + Rice husks	10.98	29.81	28.97	30.24
50C50R	Coffee husks + Rice husks	11.43	30.69	24.10	33.77
70C30R	Coffee husks + Rice husks	13.72	34.82	15.87	35.59
90C10R	Coffee husks + Rice husks	12.27	39.45	12.21	36.07
30C70G	Coffee husks + Groundnut shells	12.62	38.84	12.53	36.01
50C50G	Coffee husks + Groundnut shells	15.10	33.00	15.09	36.80
70C30G	Coffee husks + Groundnut shells	14.05	36.61	12.02	37.32
90C10G	Coffee husks + Groundnut shells	13.23	35.09	14.25	37.42
30R70G	Rice husks + Groundnut shells	18.71	33.01	16.82	31.45
50R50G	Rice husks + Groundnut shells	22.39	27.98	20.08	29.54
70R30G	Rice husks + Groundnut shells	11.48	30.34	28.26	29.93
90R10G	Rice husks + Groundnut shells	11.39	24.61	34.61	29.39
30C20R50G	Coffee husks + Rice husks + Groundnut shells	23.67	26.40	19.42	30.51
50C30R20G	Coffee husks + Rice husks + Groundnut shells	14.77	28.91	21.17	35.15
20C50R30G	Coffee husks + Rice husks + Groundnut shells	25.62	25.67	23.00	25.70
20C30R50G	Coffee husks + Rice husks + Groundnut shells	16.38	30.75	14.88	37.99

3.2 Calorific value

Results for calorific value for the developed bio-composite briquettes are shown in Figure 4. As expected bio-composite briquettes with higher fixed carbon percentages also had higher calorific values. The experimental analytical results match very well with the model developed by García et al., (2014) [51]. Bio-composite briquettes with 70 % rice husks biochar and 30 % groundnut shells biochar had the least calorific value of 16.6 MJ/kg and the composition of 30 % coffee husks biochar and 70 % groundnut shells biochar had the highest calorific value of 22 MJ/kg. The developed bio-composite briquettes have calorific values that are sufficient enough for use as fuel for cooking, compared to firewood [62]. Generally, bio-composite briquettes made from coffee and groundnut shell bio-chars attained the highest calorific values. When compared to briquettes developed with only a single agricultural residue, the calorific values of the bio-composite briquettes are similar [4,5,63]. This means that the briquettes developed in this study have equal or even higher calorific values than the briquettes earlier developed with individual agricultural waste material, hence ease adaption as domestic energy sources. The calorific values obtained in this study are higher than those reported in other studies on bio-composite briquette development. Jittabut, (2015) reported values between 16.33 MJ/kg - 17.83 MJ/kg [15]. Islam et al., (2003) and Olugbade & Mohammed, (2015) reported values between 14.29 MJ/kg - 16.32 MJ/kg and 13.54 MJ/kg - 14.25 MJ/kg respectively, which were much lower than calorific results obtained in this study [14,27]. Suryaningsih & Nurhilal, (2018) reported calorific values between 17.18 MJ/kg and 20.46 MJ/kg [10].

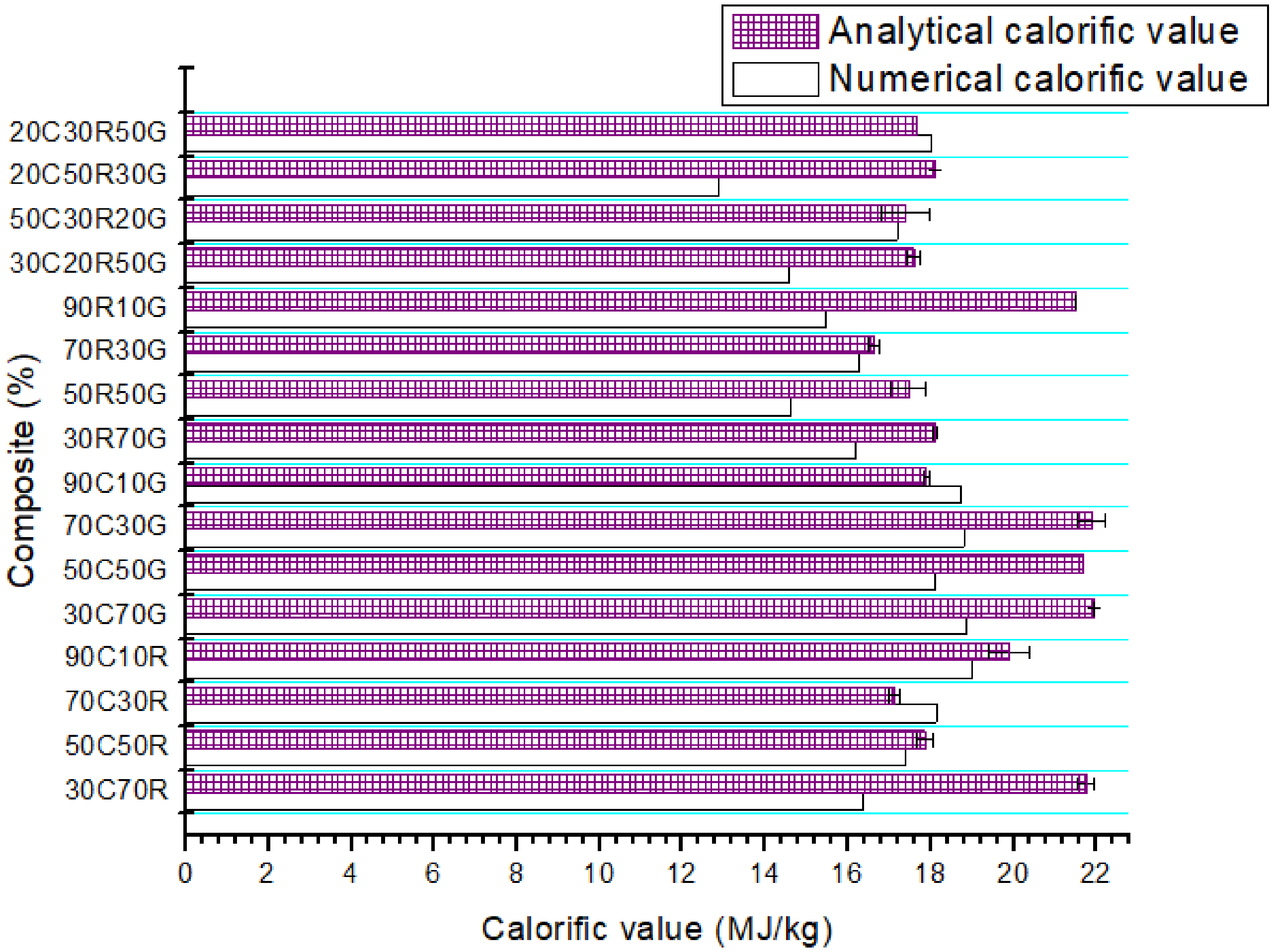

Figure 4: Comparison of analytical values for calorific values for developed bio-composite briquettes with numerical model by Garcia et al., (2014).

3.3 Water boiling test

Water boiling test results indicated that 200 g of each of the bio-composite briquettes was sufficient to boil 1000 ml of water in less than 25 minutes (See Figure 5). It is important to recognize that 25 minutes reported here also includes the ignition time for each of the developed bio-composite briquettes. The time taken to boil was less than the time recorded for similar studies that focused on single agricultural residue briquettes [4,5]. This specific characteristic is important for domestic users of the briquettes in sub-Saharan Africa because it is easily relatable to what users do when cooking domestically. Higher calorific values due to higher fixed carbon levels and considerable amounts of volatiles allowed ignition of the bio-composite briquettes to take place much faster and higher calorific values enables greater heat to be released within and between the fuel [11,53]. Mopoung and Udeye, (2017) obtained 36 minutes and 48 minutes for banana brunch charcoal briquettes and banana peel charcoal briquettes, respectively [64]. Huangfu et al., (2014) obtained 19.4 minutes - 30.5 minutes for water boiling [65].

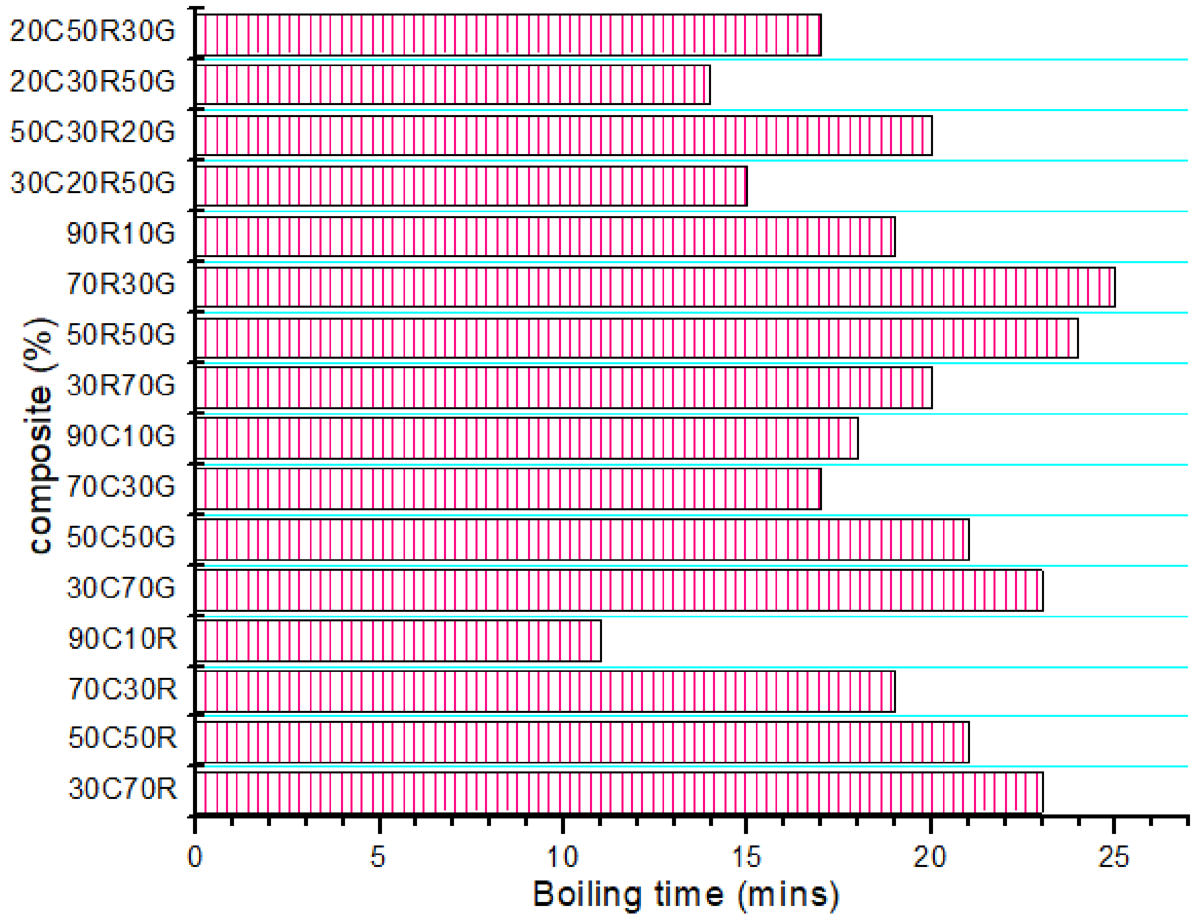

Figure 5: Total boiling time including ignition time for developed bio-composite briquettes

A relationship between calorific values and water boiling times for the developed composite briquettes has been provided (see Figure 6). This relationship helps to identify how calorific values of briquettes affect the water boiling experiment. It should be noted that high calorific values provide for less water boiling times using the developed briquettes. This is attributed to the fact that these briquettes produce more energy for combustion and therefore low times are required to boil water. Similar results were obtained in previous research [4,5]. At low amounts of coffee husks in the composite briquettes however, calorific value tends to have a negative correlation with boiling times (see Figures 6a and 6b). For these cases, high calorific values contribute to higher water boiling times.

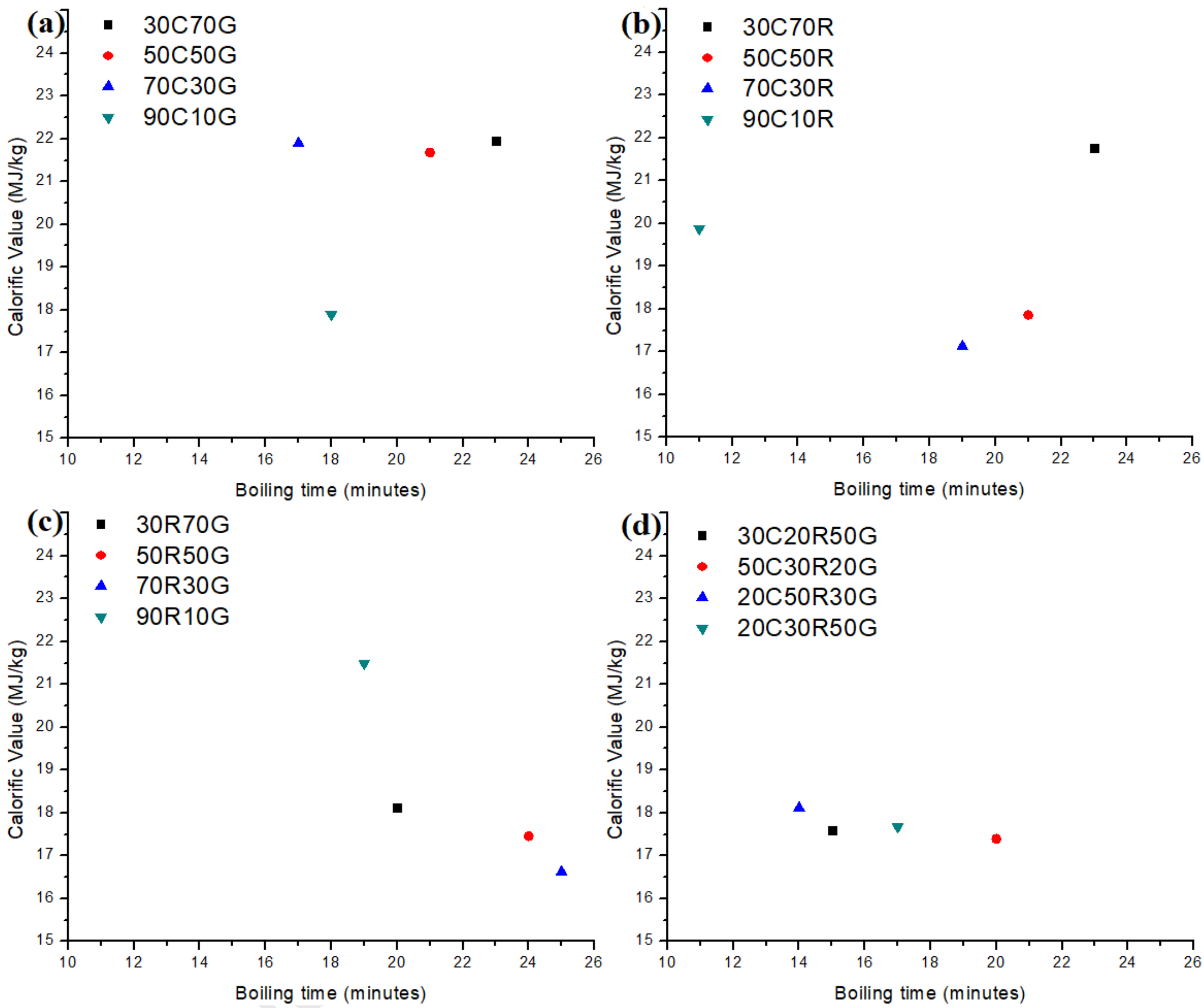

Figure 6: Calorific Values vs. boiling times for coffee husks and groundnut shells (a); coffee husks and rice husks (b); rice husks and groundnut shells (c); and, coffee husks, rice husks and groundnut shells (d); bio-composite briquettes

3.4 Thermogravimetric Analysis

Thermogravimetric analysis (TGA) was used to evaluate the effect of composites on combustion characteristics of the developed bio-composite briquettes. TGA results (see Figure 7) show the weight loss thermograms as a function of temperature. Clearly, the incorporation of different agricultural waste materials as constituents in the developed bio-composite briquettes has resulted in thermogravimetric behavior that is different from typical biomass materials, with clear peaks for volatilization due to presence of hemicellulose, cellulose and lignin difficult to predict [5]. This is attributed to the fact that each individual bio-char material has its own degradation pattern. From the TGA thermograms, the developed briquettes remain at plateau from combustion at room temperature to 104 ^{o}C, followed by undergoing a major weight loss at about 105 ^{o}C. This weight loss occurred due to the evaporation of moisture from the composite briquettes. The low peaks in the de-watering process are related to a lower amount of internal moisture in bio-composite briquettes as a result of the carbonization treatment process. Carbonized briquettes adsorb less moisture due to the destruction of hydroxyl groups, which are hydrophilic in nature. Volatile matter contents are equally low after carbonization [4]. Onset of devolatilization varies in the developed bio-composite briquettes from about 230 ^{o}C to 330 ^{o}C. From the onset of devolatization to about 600 ^{o}C, the aliphatic side chains start splitting off from aromatic rings [66]. These variations in initial degradation temperatures of the bio-composite briquettes are due to the differences in the elemental and chemical compositions of the individual constituents of the developed composite briquettes [67]. Between 600 ^{o}C and 900 ^{o}C, a stage governed by the thermal decomposition of inorganic minerals like carbonates and clay, the last decomposition phase was observed, owed to the degradation of lignin in the developed briquettes [68]. At approximately 900 ^{o}C, lignin in the developed bio-composite briquettes had decomposed off, implying that remaining weight percentage was mainly composed of residues, including, ash, tars and fixed carbon. Lignins contain both aliphatic and aromatic constituents and thus signify ability of the developed briquettes to resist hydrolysis [69]. An analysis of the percentage weight remaining corresponding to residual matter tallies very well with summation of ash and fixed carbon contents already shown in the results for physical properties of the developed bio-composite briquettes (see Table 1).

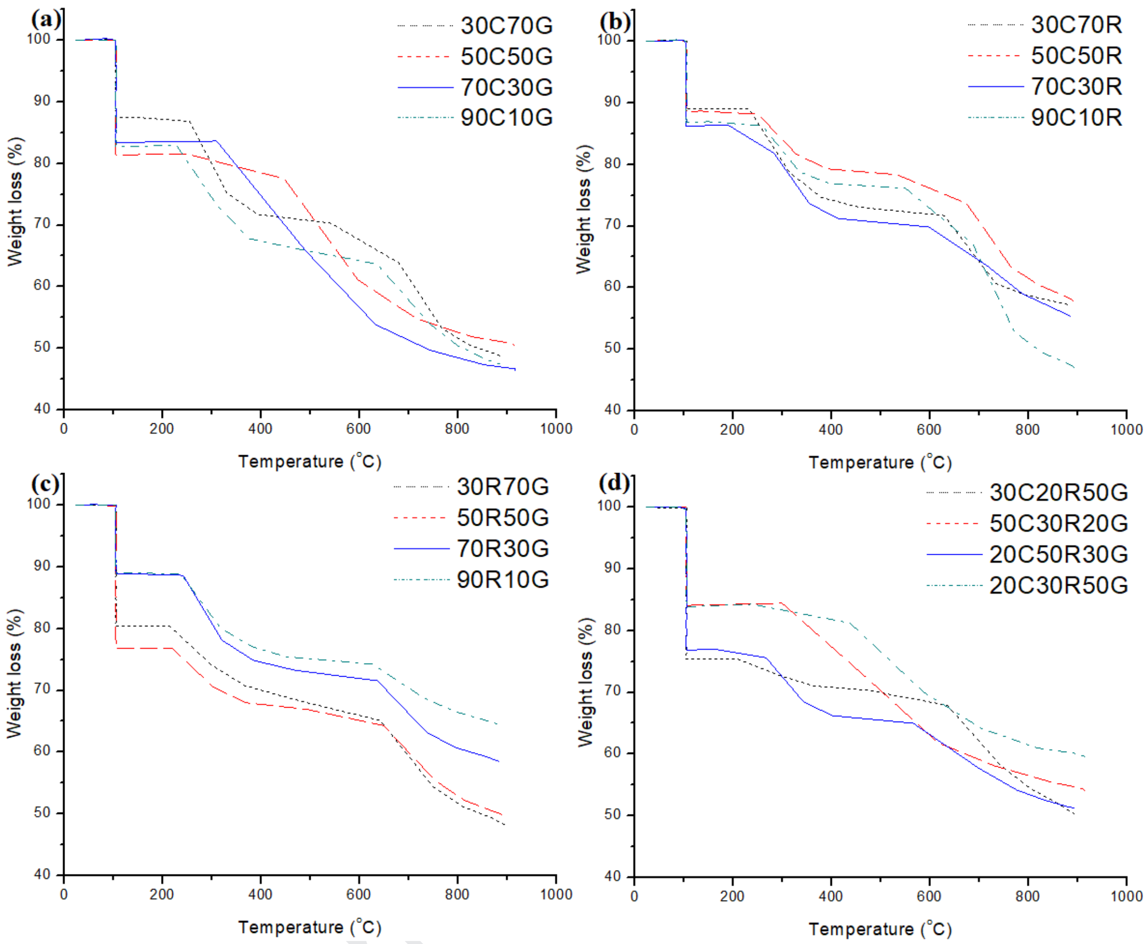

Figure 7: Weight loss thermograms for coffee husks and groundnut shell (a); coffee husks and rice husks (b); rice husks and groundnut shell (c); and, coffee husks, rice husks and groundnut shell (d); bio-composite briquettes

The lowest total percentage weight losses at the highest combustion temperature was 36.0 %, obtained for bio-composite briquettes consisting of 90 % rice husks and 10 % groundnut shells (90R10G), while the highest percentage weight loss was 53.69 % obtained for bio-composite briquettes consisting of 70 % coffee husks and 30 % groundnut shells (70C30G). This result is similar to results reported by Munir et al., (2009) [67]. This particular bio-composite briquette had high volatile matter and the lowest ash content. This shows that bio-composite briquettes 70C30G had the lowest thermal stabilities. Additionally, from Figures 7b and 7c, it was observed that an increase in rice husks content in the developed bio-composite briquettes resulted in a delay in thermal degradation of the developed composite briquettes. This is because rice husks inherently have high silica content, which reacts with oxygen to form various forms of silica dioxide, which enhances flame retardancy [70,71]. The bio-composite briquette with the highest thermal stability consisted of 90 % rice husks and 10 % groundnut shells as constituents.

3.5 Burning Rates Analysis

Results for the burning rates of the developed bio-composite briquettes are shown in Figure 8. Burning acceleration was observed to proceed significantly until 50 minutes of combustion after which a deceleration in burning rates is observed after 160 minutes. The initial peaks immediately combustion commences were due to addition of weight from moisture initial decomposition in the bio-composite briquette. The highest burning rate of 0.0175 %/min was obtained in the 177th minute by the 30R70G briquettes set. The highest rates of change in burning rates for the developed bio-composite briquettes were obtained between the 160th and 195th minute. An increase in rice husks contents in bio-composite briquettes reduced the burning rates with increase in time (see Figures 8b and 8c). This was attributed to the fact that rice husks have high ash and lower volatile matter contents, compared to coffee husks and groundnut shells (see Figure 3 and Table 1). Low volatile matter and high ash content is synonymous with suppressed ignitability with increase in time [4,71]. Additionally, the formation of SiO_2 during oxidation of silica in the rice husks has a natural thermal resistance property that it imparts on the developed bio-composite briquette [71]. The main effect of the natural thermal resistance is that the combustion of such a rice husks-containing briquette is gradually retarded with time, thus reducing the burning rates of these particular briquettes during combustion. This means that such briquettes will be able to burn at lower rates over time.

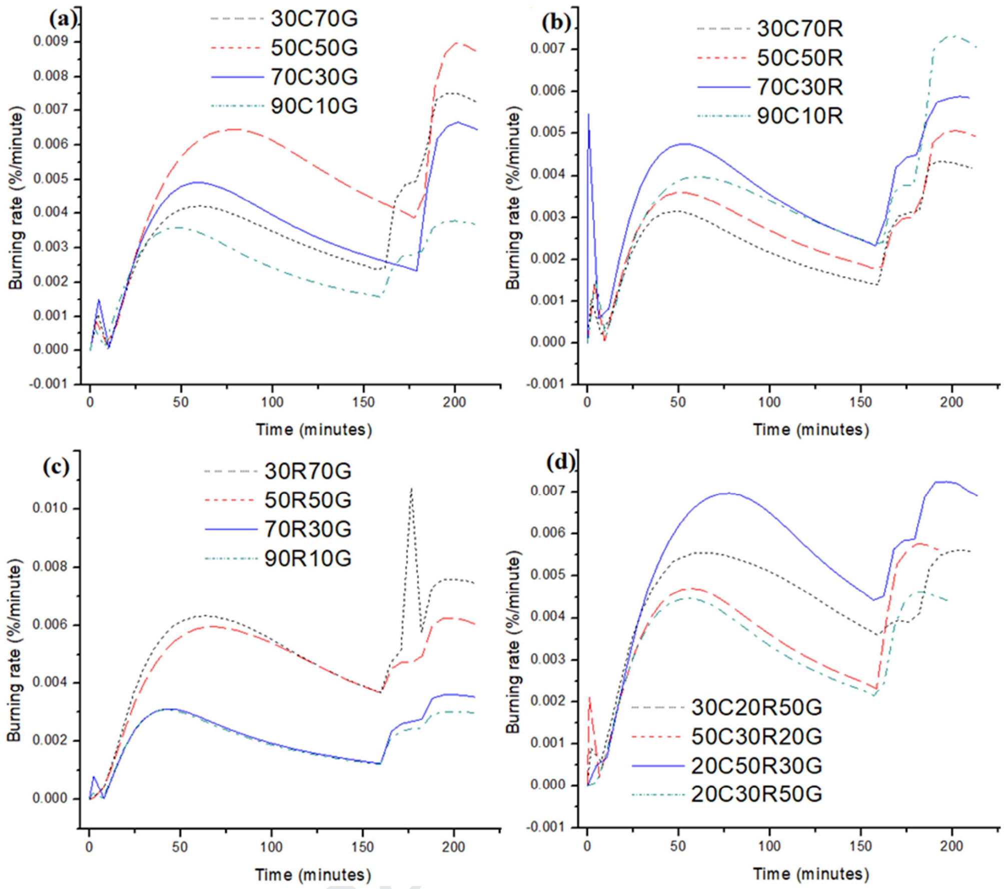

Figure 8: Burning rates for coffee husks and groundnut shell (a); coffee husks and rice husks (b); rice husks and groundnut shell (c); and, coffee husks, rice husks and groundnut shell (d); bio-composite briquettes

3.6 Heat transfer analysis along composite briquettes

Heat flow through the developed composite briquettes was studied. As the constituents of the composite briquettes have the same length and area, the heat flux through the composite briquette depends on the individual heat transfer coefficients as well as thermal conductivities of the constituent briquettes. Considering a series composite wall as shown in Figure 2, Heat flux flows as a result of the lengths and thermal conductivities of the individual husks, i.e, rice husks (L_r,k_r), coffee husks (L_c,k_c)and groundnut shells (L_g, k_g) (see Figure 2a). Additionally, with inclusion of binder, the lengths and thermal conductivities (L_b, k_b) also contribute to the heat flux across the briquettes (see Figure 2b). Table 2 shows the calculated heat flux ranges per briquette, considering that the heat transfer coefficient of air, h_a is 2 W/m^2/K [52]. Rice husks have a 0.039 W/mK thermal conductivity [72], coffee husks have a 0.11 W/mK thermal conductivity [73] and that for cassava binder is 0.24 W/mK [74]. Wang et al., (2018) showed that thermal conductivity of groundnut shells is 0.0006 W/mK [75]. An assumption made in this study is that the constituent lengths of the raw materials (rice husks, coffee husks and groundnut shells) is the same and is 1 m. Another assumption is that the briquette burns in room temperature from the highest attainable temperature of about 900 ^{o}C. The heat flux therefore is given by Equation 6.

$$q'' = \frac{\Delta T}{\left(\frac{2}{h_a} + \sum_{i=1}^{n} \frac{L_i}{k_i}\right)} \qquad \text{Equation 6}$$

From Table 2, heat flux ranges from 0.052 W/m^2 to 12.887 W/m^2 obtained during combustion of 90R10G and 30C70R composite briquettes respectively. Briquettes developed with rice and coffee husks bio-chars have higher heat fluxes due to the fact that these two have the highest thermal conductivities compared to that of groundnut shells. An observation from Table 2 is that incorporation of groundnut shells strongly reduces heat flux through the briquette. This is because the thermal conductivity of groundnut shells is 98.5 % and 99.5 % less than that of rice husks and coffee husks respectively. In fact, similar to the trend in Figure 7b, heat flux increased with increasing rice husks content. When the effect of binder is brought into consideration, the heat fluxes slightly reduce due to a 4.167 m^{2}K/W resistance term induced in the Equation 6.

Table 2: Heat flux through the developed bio-composite briquettes

Composite briquette (%)	**Briquette composition (Bio-char)**	**ΔT K**	**$\frac{2}{h_a}$ (m^{2}K/W)**	**$\frac{L_c}{k_c}$ (m^{2}K/W)**	**$\frac{L_r}{k_r}$ (m^{2}K/W)**	**$\frac{L_g}{k_g}$ (m^{2}K/W)**	**q''(Without starch binder (W/m^2)**	**q'' (With starch binder (W/m^2)**
30C70R	Coffee husks + Rice husks	875	1	30.3	36.6	0	12.8866	12.14154
50C50R	Coffee husks + Rice husks	875	1	18.2	51.3	0	12.41135	11.71875
70C30R	Coffee husks + Rice husks	875	1	13.0	85.5	0	8.79397	8.440514
90C10R	Coffee husks + Rice husks	875	1	10.1	256.4	0	3.271028	3.220859
30C70G	Coffee husks + Groundnut shells	875	1	30.3	0	2381.0	0.362724	0.362099
50C50G	Coffee husks + Groundnut shells	875	1	18.2	0	3333.3	0.260999	0.260675
70C30G	Coffee husks + Groundnut shells	875	1	13.0	0	5555.6	0.157103	0.156985
90C10G	Coffee husks +	875	1	10.1	0	16666.7	0.052465	0.052452

	Groundnut shells							
30R70G	Rice husks + Groundnut shells	875	1	0	85.5	2381.0	0.35461	0.354012
50R50G	Rice husks + Groundnut shells	875	1	0	51.3	3333.3	0.258448	0.25813
70R30G	Rice husks + Groundnut shells	875	1	0	36.6	5555.6	0.15644	0.156324
90R10G	Rice husks + Groundnut shells	875	1	0	256.4	16666.7	0.051701	0.051689
30C20R50G	Coffee husks + Rice husks + Groundnut shells	875	1	30.3	128.2	3333.3	0.250515	0.250217
50C30R20G	Coffee husks + Rice husks + Groundnut shells	875	1	18.2	85.5	8333.3	0.103698	0.103646
20C50R30G	Coffee husks + Rice husks + Groundnut shells	875	1	45.5	51.3	5555.6	0.154774	0.15466
20C30R50G	Coffee husks + Rice husks + Groundnut shells	875	1	45.5	85.5	3333.3	0.252503	0.2522

3.7 Drop strengths

All the developed briquettes had drop strengths of over 85 % (see Figure 9). Least drop strength of 85.4 % was obtained from briquettes with 70 % coffee husks and 30 % groundnut shells (70C30G) while the highest drop strength of 100 % was obtained from briquettes with compositions of 30 % coffee husks band 70 % rice husks (30C70R). Drop strength results are particularly important because they provide an indication of handle-ability, transportation and storage integrity of the developed bio-composite briquettes [7,58]. Drop strengths is also an important parameter in briquetting because it shows the extent to which briquettes can be stacked on top of each other during domestic or kiln cooking. Having higher drop strength enhances this purpose and thus saves time during cooking, thereby increasing coking efficiency. Generally, higher drop strength values were obtained for bio-composite briquettes developed from rice and coffee husks agricultural wastes. The high drop strengths are possibly due to the fact that the binder used in the development process was cassava starch. High drop strength is an indication of possible strong C-C bonds formed after carbonization [76]. Durability has been noted to improve in other studies containing composite briquettes [32,77,78].

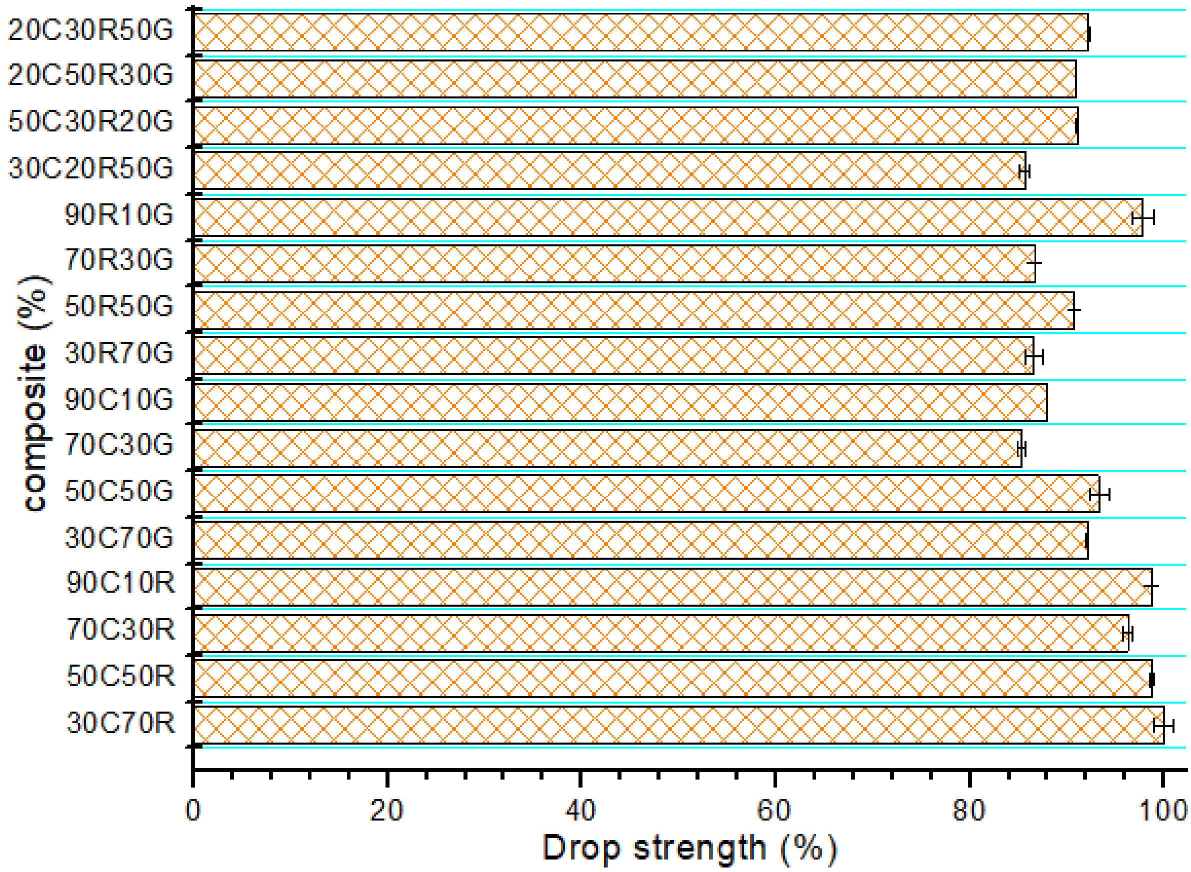

Figure 9: Drop strengths for developed bio-composite briquettes

A relationship between calorific values and drop strengths of the developed composite briquettes has been provided (see Figure 10). Generally, high calorific values corresponded to high drop strengths. High drop strength indicates the possibility of strong C-C bonds in the developed composite briquettes [70]. It is these bonds that improve the calorific values of the developed briquettes. Much as all the developed briquettes had high drop strengths of over 85 %, for each category, briquettes with the highest drop strengths also yielded the highest calorific values. This is so because of the use of cassava starch as binder which provides double advantages in terms of heating value addition due to their protein nature, as well as drop strength increment due to formation of intermolecular hydrogen bonds between the amylose and amylopectin components of starch. [4,5].

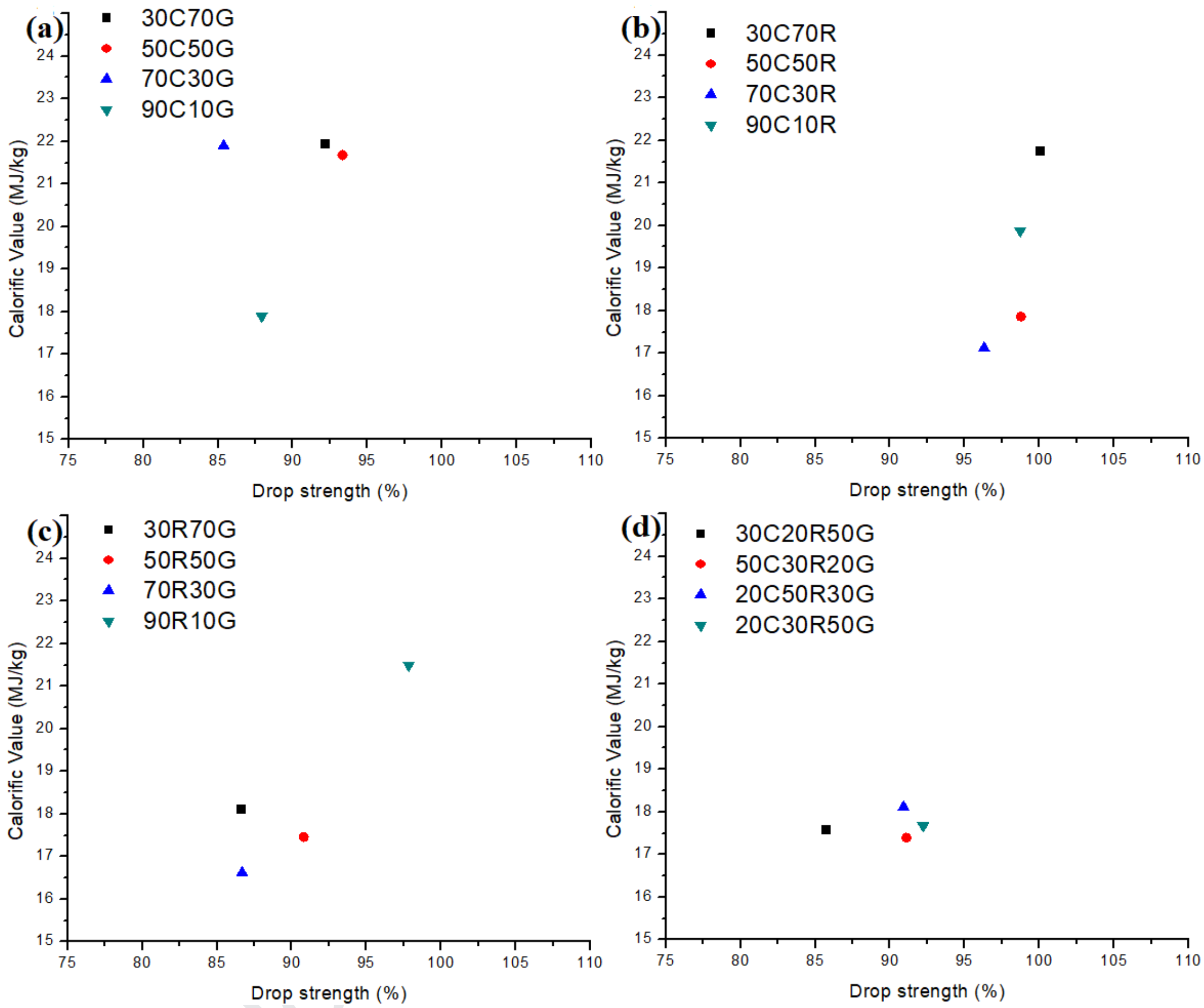

Figure 10: Calorific Values vs. drop strength for coffee husks and groundnut shells (a); coffee husks and rice husks (b); rice husks and groundnut shells (c); and, coffee husks, rice husks and groundnut shells (d); bio-composite briquettes.

3.8 Particle density

Particle densities for developed bio-composite briquettes were between 476 kg/m^3 and 578 kg/m^3 (see Figure 11). According to Atan et al., (2018), particle density shows the compactness of a briquette and the strength of the briquette directly proportional to its density [11]. Particle densities of the developed bio-composite briquettes exhibit higher values when compared to the previously developed briquettes based on single agricultural wastes [5]. Therefore, the composite briquetting process enhances the mechanical properties as well as dimensional stability of the biomass, as opposed to its loose bulk condition. This is due to the combined aggregated weighted effect of the individual agricultural waste bio-chars. Additionally, high particle densities have also been reported to enhance the calorific values of the developed fuel [53,64]. Particle densities of bio-composite briquettes developed in this study were above values that ranged between 366 kg/m^3 and 570 kg/m^3 reported by Muazu and Stegemann, (2015) [9]. However, the particle densities obtained in this study were lower than particle density values between 599 kg/m^3 and 741.5 kg/m^3 reported by Atan et al., (2018) [11]. Carbonization enhances compaction as bio-chars of representative agricultural wastes have radicals exposed that are free to bond with each other. Depolymerisation and de-volatilization occur where the lignin of the raw material softens and makes the raw materials more suitable for densification [79].

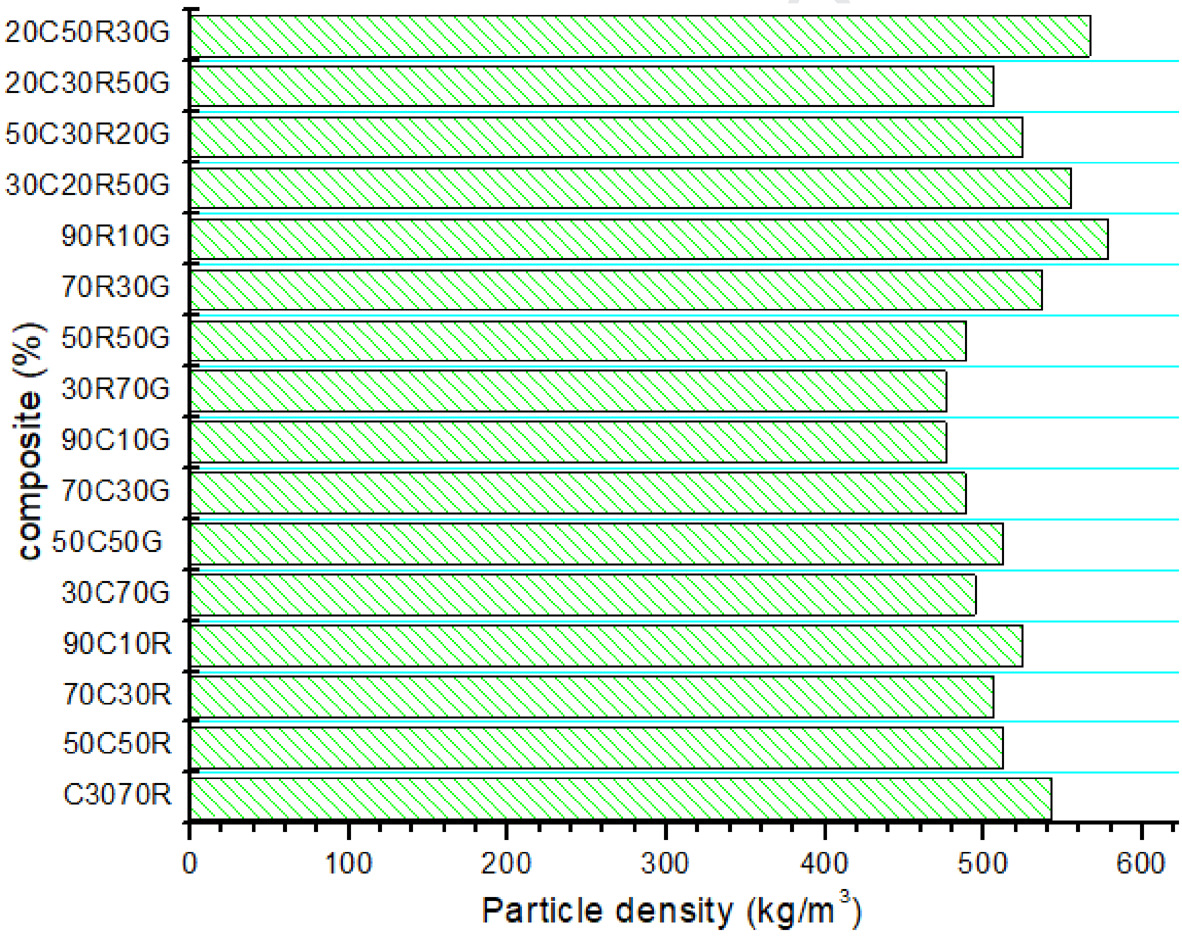

Figure 11: Particle densities for developed bio-composite briquettes.

A relationship between calorific values and particle density of the developed composite briquettes has been provided (see Figure 12). Similar to the trends obtained in Figure 10, high calorific values are obtained in briquettes with highest particle densities. This is due to the fact that higher particle densities show compactness of the developed briquettes and therefore increased fixed carbon components and thus increased calorific values. Fixed carbon greatly affects the calorific value of a fuel because it decreases the residence stage until combustion is completed [51]. In fact, fixed carbon has a positive coefficient compared to other physical properties in the analytical representation of calorific value (see Equation 2). Similar results between calorific values and particle densities have been obtained in previous work [5]. Other researchers also found positive relationships between calorific values and densities in fuel sources [80,81].

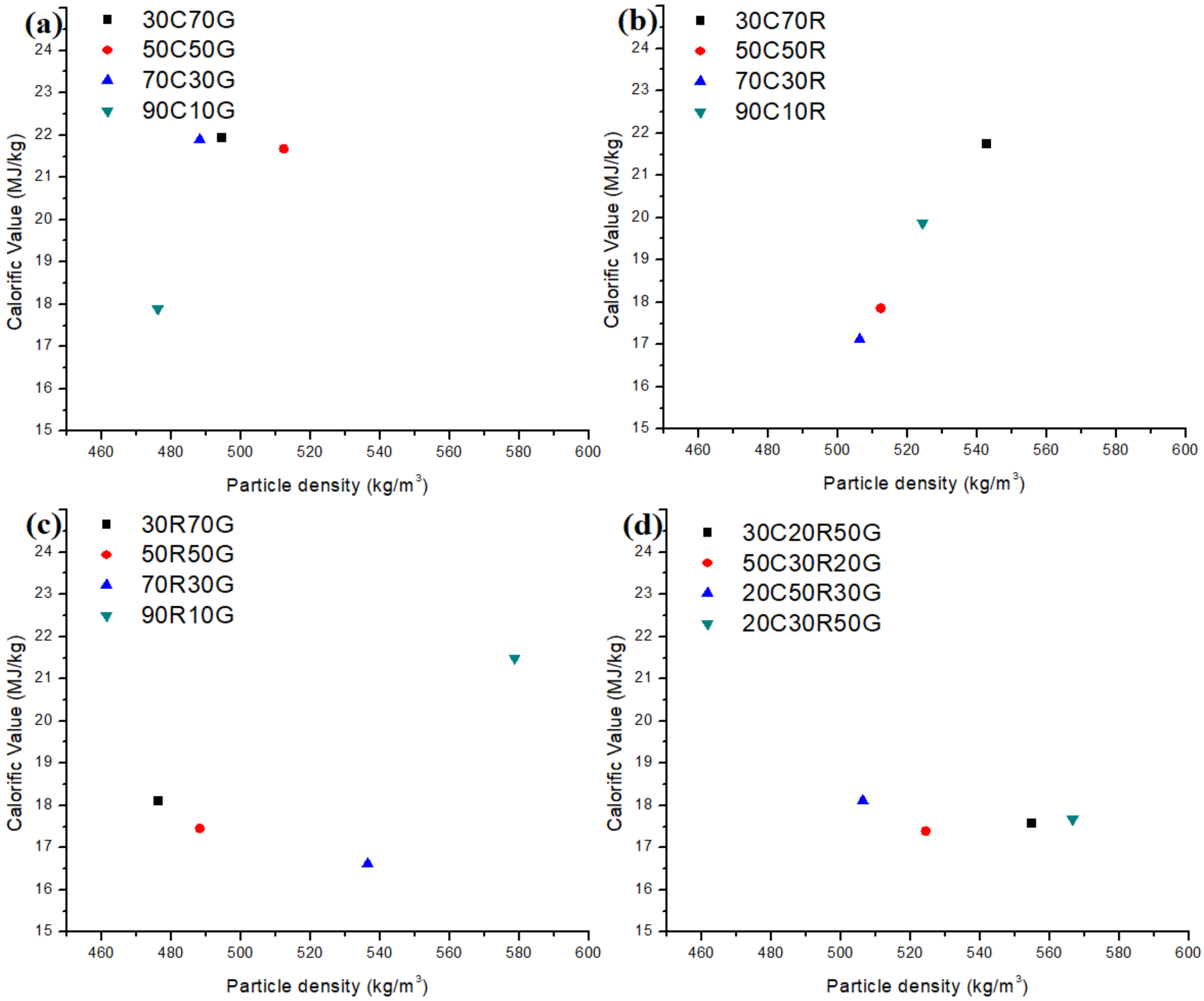

Figure 12: Calorific Values vs. particle density for coffee husks and groundnut shells (a); coffee husks and rice husks (b); rice husks and groundnut shells (c); and, coffee husks, rice husks and groundnut shells (d); bio-composite briquettes

Conclusions

This work aimed at producing composite briquettes from three common agricultural residues in Uganda, namely rice husks, coffee husks and groundnut shells. Low pressure technique and cassava starch binder was employed in the briquette development process. The physical properties of the developed composite briquettes were determined by using thermogravimetric analysis (TGA). Bomb calorimetry was used to determine the calorific values of the briquettes. TGA and bomb calorimetry provided the combustion properties of the developed briquettes. Additionally, the flow of heat within the component materials constituting the developed briquettes was studied with and without binder material. The mechanical integrity of the briquettes was determined using the drop tests and particle density. A water boiling test was carried out with each set of the developed briquettes. Developed composite briquettes from this study had improved properties compared to those developed previously from individual bio-chars. Moreover, the carbonization process significantly improved the fixed carbon contents and this later was verified by the increased calorific values of the developed briquettes after carbonization. Rice husks had higher moisture and ash contents compared to groundnut shells and coffee husks. The developed briquettes yielded higher volatile matter and ash contents compared to their constituent raw materials. The calorific values of the developed briquettes ranged between 16.6 MJ/kg and 22 MJ/kg. Drop strengths ranged between 85.4 % and 100.0 % while particle densities ranged between 476.14 kg/m^3 and 578.1 kg/m^3. This means that the developed briquettes will withstand forces during transportation and storage more than briquettes developed from individual bio-chars. TGA confirmed that the developed briquettes have high residuals and thus high fixed carbon contents. The highest rates of change in burning rates for the developed bio-composite briquettes were obtained between the 160th and 195th minute. The lowest ignition and boiling time was obtained when composite coffee husks and rice husks bio-chars were used in ratios of 90:10 respectively. The lowest total percentage weight losses at the highest combustion temperature was 36.04 %, obtained by the 90R10G briquette while the highest total percentage weight loss was 53.69 % obtained by the 70C30G briquette, showing that the former yielded the best thermal stabilities, owing to its lowest volatile matter content (24.61 %). Heat flux ranged from 0.052 W/m^2 to 12.887 W/m^2. Briquettes developed with rice and coffee husks bio-chars had higher heat fluxes due to the fact that these two have the highest thermal conductivities compared to that of groundnut shells. This work showed that calorific values have no linear relationship with water boiling time, drop strength and particle density, even though it varies with them. The results provide vital information that shows composite briquettes have higher potentials for use as fuel for cooking purposes compared to briquettes developed from individual residues. There is however need to explore the use of different binders, most preferably those not associated to food material.

Data Availability

Datasets related to this article can be found at http://dx.doi.org/10.17632/fsnn5w4fyb.1 an open-source online data repository hosted at Mendeley Data [82].

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Highlights

- Bio-char bio-composite briquettes were developed from agricultural residues
- Calorific values for the developed briquettes ranged between 16.6 MJ/kg and 22MJ/kg.
- Drop strength for the bio-composite briquettes were all above 86%
- Particle densities ranged between 430 kg/m^3 and 580 kg/m^3.
- Heat transfer was enhanced when no binder was present

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: