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RESEARCH ARTICLE

RESEARCH ON PHYSICAL AND CHEMICAL PROPERTIES OF PELLETS PRODUCED USING PADDY STALKS FOR GASIFICATION PROCESS

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ABSTRACT

Pellets which are produced using paddy stalks are the main type of biomass solid fuel and have great potential as a Bioenergy resource in the future of Turkey. But it also showed important problems because of its high content of ashes and its low gross calorific value, reducing the possibility to be used in domestic heating. In this research, the paddy stalks were chopped and then mixed with different materials such as coal powder, PVA (poly (vinyl alcohol)) and molasses before producing of pellets. In addition to these, only paddy stalk pellets without additive were produced to compare the other pellets. After preparation of the pellets, some physical and chemical properties of them were determined to investigate the effects of additives on the changing of these properties and gasification process. Pellet samples used in the tests were labeled according to additive percents are pellet produced using paddy stalk (PRF), paddy stalk + 3% PVA (PVA3), paddy stalk + 5% molasses (PML5), paddy stalk + 5% coal powder (PC5) and paddy stalk + 15% coal powder (PC15). Pellets were tested using downdraft fixed bed Gasifier. The volatile matters of PRF, PVA3, PML5, PC5 and PC15 were found to be in the range of 61.73 - 66.25%. Highest volatile mater found in pellets of PRF. Bulk densities were determined in the range of 494-606 kg/m^3. Maximum bulk density was found for the PVA3 pellets. Highest ash fusion temperature was found to be 1436 °C for PRF pellets. The ash fusion temperatures indicate slag formation. The calorific values were changed in the range of 12, 18-12, 92 MJ.kg^{-1}. The PC15 pellets has highest calorific value compared to the other pellets. According to research results, it can be recommended that the pellets produced from pellets produced using only paddy stalk for gasification by using appropriate equipment.

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INTRODUCTION

Rice is one of the most important grains consumed after wheat in the world. In Turkey, production of rice has started to increase also in recent years. According to data of 2013, 900.000 tones rice in 110.600 hectares cultivated area is produced in Turkey. Approximately, 70% of the production area of rice is located in Thrace region. Edirne City retains a portion of 39% of the rice production areas in our Turkey with 43.000 hectares production area and 362.000 tones rice production. The highest amount of production is in Ipsala district in Edirne City.

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Rice production is rather important for regional agriculture. However, one of the most significant problems of rice production is the paddy stalks that remains on the field surface after harvesting. Rice has a high stalk/grain ratio of 1/2. Therefore, 350-400 kg/da of rice stalk remains on the field surface after harvesting. Also, since there is a high ratio of silicon in the stalk of paddy, it is very much difficult to be decomposed. Moreover, the stalk does not decay easily until next cultivation consequently. For this reason, rice producers' burn the paddy stalks on the field every year even though it is prohibited. In Edirne province, approximately 180.000 ton/year of paddy stalk are burned every year. In this case, it creates some negative environment conditions as well as it leads to a substantial amount of loss of energy. The energy content of paddy stalk is almost 15MJ/kg (Jain, 2006). Every year 2,7× 10^9 MJ of energy is wasted due to burning of paddy stalk on the field surface in this region. Whereas paddy stalk can be recycled with convenient techniques and different methods to contribute to the economy as well as it will help adverse environmental conditions to be lower. Gasification is one of these methods. When it is possible to gasify paddy stalk with roughly 65% yield, it will lead to 1,75×109 MJ energy gain in the region every year. This corresponds to a 55,5 MW power of a plant that works for 24 hours a day. Obtained gas can be used by being burnt directly in the rice grain drying process that needs a lot of energy requirement or in energy power plants to produce power generation by being converted to mechanical energy in gas engines. Thus, acquired biomass energy from paddy stalk that is a renewable energy source will contribute to national economy and will be an alternative option for fossil fuels.

Researches related to paddy stalk to be used as an energy source has showed an increase in recent years. Besides, energy content and density of paddy stalk is lower than the other biomass such as wood, corn stalk, wheat stalk and its alkali and alkaline components is higher compared to others. This situation complicates the process of picking, transportation and storage of the paddy stalk. However, these inconveniences can be eliminated by applying some pre-treatments to paddy stalk. These operations, which alter the physico-chemical characteristics of stalk, increase its energy content and reduce scum formation, can be ranged as sizing, bailing and pelletizing. In this way, to produced energy from paddy stalk can be more economic (Kargbo *et al*., 2009). Kirubakaran *et al*. (2009), determined that volatile substances ratio is 80,2%, ash rate is %19,8 and fixed carbon rate is 19,8% for paddy stalk. The same rates were determined respectively as 81,6% , %23,5 and %18,4 for rice husk. Calvo *et al*. (2004), investigated heating of paddy stalk and calcining process and point out inconveniences of burning it in an open field.

Ultimate analysis results of paddy stalk were given as 37.87% C, 4.61% H, 0.63 % N, 0.14% S, 1.01% CI and 34.87% O_2, respectively. They also indicated that its moisture content, ash content, fixed carbon rate and upper calorific value were determined as 7.43 %, 67.95 % 12.98 and 14.71 MJ/kg, respectively. The use of biomass for thermal energy is ancient but the biomass as a renewable energy source implying clean combustion process is more recent. Therefore the objective of this research is to test a clean and more efficient renewable source to obtain energy from crop residues. For this reason, chemical and physical characteristics of biomass pellets produced using paddy stalk should be known in order to choose the appropriate energetic conversion process.

MATERIALS AND METHODS

Paddy stalk that was used as material was obtained from Ipsala province of Edirne City. Stalks were chopped into suitable size to produce pellets using a stalk chopper ekuipment. Pellets were prepared in 5 different compositions using different additives with different percents as seen in Fig.1 and Table 1.

Figure 1. The prepared pellets sample

Table 1. Components of pellets prepared as Biofuels

Sample codes	Pellet component
PRF	No additives (reference sample)
PVA3	Paddy stalk + 3% PVA
PML5	Paddy stalk + 5% molasses
PC5	Paddy stalk + 5% coal dust
PC15	Paddy stalk + 15% coal dust

The moisture content of the pellets

The moisture content of a solid is defined as the quantity of water per unit mass of the wet solid. The moisture content plays an important role in the formation of pellets and subsequently its combustion process. Moisture content of biomass at the time of harvesting varies drastically. The moisture content of biomass was measured by oven dry method. Initially the sample with the known weight is kept in oven at 105°C for 24 hours. The oven dry sample is then weighed. The moisture content of sample was calculated by following formula.

$$M = \frac{W_1 - W_2}{W_1} \, x \, 100$$

Where,

W_1 = Weight of sample before drying (g),
W_2 = Weight of sample after drying (g).

Pellet dimensions

The pellets were produced as cylindrical shape. In order to determine dimensions and unit mass, a hundred pellets were randomly selected for each experiment. The length and diameter of each pellet were measured using a digital vernier caliper. The mass of each pellet was also weighed using a precision digital balance.

Particle density of pellets

Particle density of each pellet was determined by weighing the individual pellet and calculating its volume based on its length and diameter with the following equations (Liu *et al*., 2013).

$$V_u = \frac{\pi d^2 l}{4}$$

$$\rho_u = \frac{m_u}{V_u}$$

Where,

V_u= the volume of pellet (cm^3),
D = the diameter of pellet (mm),
L = the length of pellet (mm),

ρ_u = the density of pellet (g/cm^3),
m_u = the mass of pellet (g).

Bulk density

Bulk density of pellets were determined based on EN 15103 Standard Specification. Bulk density was calculated as the ratio of the material mass to the container volume as given in following equation. The pellets were leveled to the top surface of the container and were weighed using a digital balance. The container volume was calculated by measuring its length and diameter.

$$\rho_b = \frac{m_b}{V_b}$$

Where,
ρ_b = the bulk density (g/cm^3),
V_b = the volume of container (cm^3),
m_b = the total mass of pellets (g).

Durability

Durability of pellets were determined based on EN 15210-1 Standard Specification. Pellet durability was determined by mass loss of samples. Some pellets were randomly selected and weighed using a precision digital balance. The initial mass was recorded. They were then put into a vibrating sieve with screen size 3.17 mm (1/8 in). After 10 min, they were weighed again and the final mass was recorded. Pellet durability was calculated by using the following equation:

$$D_d = \frac{m_f}{m_i} \times 100$$

Where,
D_d= pellet durability (%),
m_i= initial mass of samples (g)
m_f= final mass of samples (g)

Pellet fracture resistance

In order to determine fracture resistance of pellets, 3 pellets were taken randomly from bulk , were dropped 4 times from a height of 1.85 m. Pellets were sifted by using a 3.15 mm round sifter. Depending on the loss of weight during the test, the fracture resistance was defined as % by proportioning pellet weight after test to pellet weight before test (Yılmaz, 2014).

Pellet compression resistance

Degree of compaction of pellet is defined as the maximum breaking load that pellet can resist before its disintegration and it is determined by compression test (Kaliyan and Morey 2009). In this test, 10 pellets, from each pellet sample that has different composition, were inserted between two plaques separately and a simplex compression force was applied from the top. Applied compression force continued by increasing at a constant rate until pellet was broken and the applied loads were transferred to computer during the test (The compression resistance of pellets (N)). The specific compression resistance were calculated as N/mm by proportion of compression resistance to pellet length. Tensile strength of pellets was calculated as the pressure applied to the half of the surface area of pellet calculated depending on pellet diameter and pellet length (Tabil, 1996, Shaw *et al*., 2009, Kashaninejad and Tabil, 2011).

$$\sigma_x = \frac{2F}{\pi d l}$$

Where,
σ_x = tensile strength (Pa)
F= Maximum compression force (N)
d= Pellet diameter (m)
l= Pellet length (m)

Elemental, proximate, calorific value and fusibility analyses of pellet samples

The elemental analyses namely C, H, N, S and O percent and proximate analyses namely moisture, ash, volatile matter and fixed carbon of prepared pellet samples were performed in TUBITAK MARMARA RESERCH CENTER (MAM). In addition to the elemental and proximate analyses, calorific value, fusibility tests (IDT, ST, HT, and MT) and ash content analyses were made in TUBITAK MARMARA RESERCH CENTER (MAM).These analyses and tests were made according to standard methods given in Table 2 as below.

Table 2. Methods of biomass fuel analysis

Property	Analytical method
Moisture	ASTM D 7582-12
Ash	ASTM E 1755-01
Volatile material	ASTM D 7582-12
C (Fixed)	ASTM D 3172-13
C	ASTM D 5373-14
H	ASTM D 5373-14
N	ASTM D 5373-14
S	ASTM D 4239-14
O_2	ASTM D 3176-09
Lower calorific value	ASTM D 5865-13
Upper calorific value	ASTM D 5865-13
IDT (Initial deformation temperature)	ASTM D 1857/D1857
ST (Softening temperature)	ASTM D 1857/D1857
HT (Hemispheric temperature)	ASTM D 1857/D1857
FT (Melting temperature)	ASTM D 1857/D1857
Ash content analysis	ASTM D 2795

RESULTS AND DISCUSSION

Analyses and test results performed for pellets that were prepared in 5 different content, are given as in tables and figures. Moisture content of pellets altered between the rates of 7.01% and 8.94% as seen in Figure 2. Pellets which are containing coal powder have a higher moisture content compared to the other pellet samples (Figure 2). Pellet size is determinant of pellet strength, as longer pellets can be more easily broken. Pellet dimensions are important factors in the context of fuel feeding properties and thermochemical processes. Thinner pellets allow for a more uniform combustion rate than thicker ones. With the shorter the pellets, the easier the continuous flow can be arranged (Said *et al.*, 2015).In this research, the pellet diameter changed in the range of 6.03 to 6.16mm and a length of 51.47 to 56.67 mm and a weight of 1.94 to 2.17 g (Table 3). Bulk density plays a crucial role on the efficiency of transportation and storage. Higher mass density leads to requirement of higher transportation yield and less storage capacity area (Liu *et al*. 2013). In this research, the pellet bulk density of 494-606 kg/m^3 ranged (Table 3 and Figure 3). Many researchers have reported that the value of 450-700 kg /m^3 (Said *et al*., 2015). Pellet particle density ranged from 1176 to 1326.67 kg/m^3. The highest particle density was reached for PML5 pellets. High fuel density represents the value of high energy. The volume density of five different pellet fuels are available to be used as a flatulent fuel.

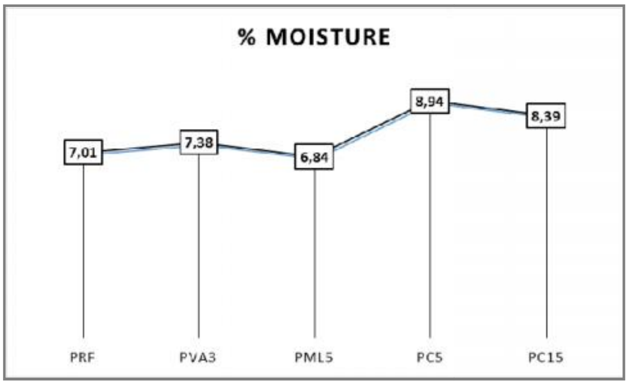

Figure 2. The moisture content of pellet samples

Researchers indicated also that material moisture content has great effects on particle density and bulk density of pellet. Since density parameter in pellets is directly related to pellet quality, when material moisture content increases, quality decreases and when moisture content increases, density of pellet declines (Yılmaz 2014).

Table 3. Physical Properties of different pellet samples

	PRF	PVA3	PML5	PC5	PC15
ρ_b(kg/m^3)	562,3	606,0	559,8	494,6	555,1
ρ_u (kg/m^3)	1226,4	1236,0	1326,7	1176,5	1284,2
d (mm)	6,16	6,08	6,07	6,03	6,12
L(mm)	53,61	55,69	56,67	52,8	51,47
m_b (g)	1,95	2	2,17	1,9	1,94

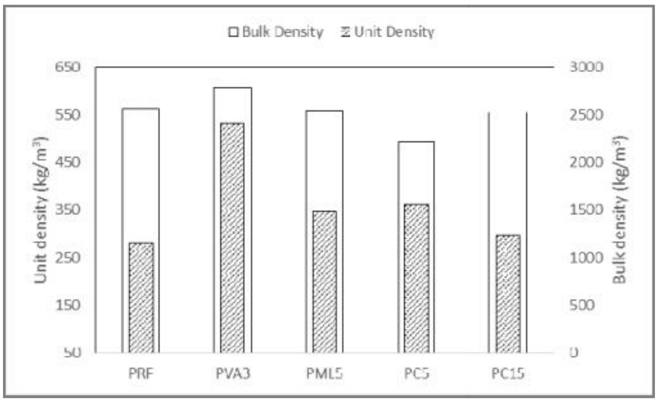

Figure 3. Bulk and particle (unit) density of pellet samples

Durability is one of the most significant quality factor of biomass pellets. High durability means high qualified pellet. Disorders like low durability in the systems of pellet feeding lead to problems like dust emission and increase the risk of fire and explosion during manufacturing and storage. Its recommended durability value is >97.50% (Said *et al*., 2015). According to this research results; the resistance degree (durability) of pellets was found higher this value for all pellet samples and changed between 97,8-99,3%. The highest resistance degree was achieved in PML5 pellets and PML5 pellets have the lowest moisture content. The ultimate strength (impact resistance or dropout resistance) test is conducted especially for pellets which confronts with force during defueling or packaging (Kaliyan and Morey, 2009). Pietsch (2002) stated that these tests can be used for identification of safe height of pellet production. Pellet durability and fracture resistance values are given in Table 4. As the degree of resistance of all tested pellet types is higher than the degree of 97,5%, it can be said that they are qualified from the point of resistance. Compression test is conducted for simulation of the burden pellets apply to each other during storage in packs or silos and transportation (Kaliyan and Morey, 2009).

Table 4. Pellet durability and fracture resistance values

Pelet	Durability(%)	FractureResistance (%)
PRF	97,8	98,67
PVA3	98,9	99,82
PML5	99,3	99,94
PC5	98,7	99,34
PC15	98,1	99,96

Compressive strength of the pellets, the specific compression resistance and tensile strength values are given in Table 5. The fact that the pellet has the highest value of PH is evaluated as high quality. Increasing material moisture content leads to a decreasing pellet density. Therefore, it is defined as decreasing pellet resistance (Yılmaz, 2014). In our study, compaction resistance of pellet was between 1910,2 N and 2652,97 N. The highest value was gained with PC15 pellets.

Table 5. Compressive strength of the pellets, the specific compression resistance and tensile strength values

Pelet	Compressive strength(N)	Specific compression resistance (N/mm)	Tensile strength (Mpa)
PRF	1910,2	55,57	5,69
PVA3	2282,7	68,48	7,14
PML5	2311,7	60,95	6,34
PC5	2584,5	66,66	6,93
PC15	2653,0	67,62	6,96

Ash contents were between 18,27% and 20,91%. Ash contents were a little bit higher in the containing coal dust pellets. Volatile contents are between 61,73% and 66,25%. The volatile content is higher in the additive-free pellet sample (PRF) (Figure 4). Fixed carbon rate of pellet samples was found between 15,02 and 17,36%. These values were determined as 15.24, 15.02, 16.06, 16.29, and 17.36 for the samples of PRF, PVA3, PML5, PC5, PC15, respectively. Results of elemental analyses are summarized as followed: carbon rate: 39,10-40,16%; hydrogen rate: 4,79-5,05%; nitrogen rate: 1,24-1,69%; Sulphur rate: 0,17-0,32%; oxygen rate: 32,32-35,71% (Table 6). Results of elemental analyzes show coherence with the former studies (Jenkins *et al*., 1998).The results of calorific value and fusibility analyses of the pellet samples are given in Table 7. Although sub-calorific value of pellet samples were found higher for the pellets that contain 15% coal powder pellets, they have almost same values (min: 12,18 MJkg $^{-1}$for PRF sample- max: 12,92 MJkg^{-1} for PC5 sample). Sub-calorific values of pellets are lower than the former studies. Jankins *et al*. (1998) found that calorific value of paddy stalk was 15 MJkg^{-1}.

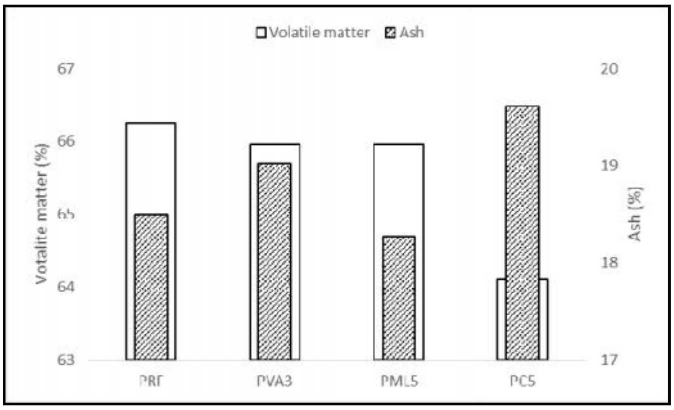

Figure 4. Ash and volatile matter content of the pellet sample (%)

Table 6. Results of elemental analysis of pellet samples (%)

Pellet Sample	PRF	PVA3	PML5	PC5	PC15
C	39.90	39.42	39.10	39.37	40.16
H	4.89	4.95	5.05	4.80	4.79
N	1.24	1.32	1.69	1.60	1.50
S	0.20	0.17	0.18	0.24	0.32
O_2	35.27	35.12	35.71	34.38	32.32

The most important parameter that affects the process of glaze is initial deformation temperature because of the percentage of silicon in paddy stalk, it is between 948°C and 1045°C. Tuğ (2016) found that glass transition started when the temperature in core zone exceeds 800°C during gasification of paddy stalk straw pellets. The best result was obtained in gasification process with upper air intake and excess air coefficient of 0,20. It was observed that the mixing process have not positive result during the gasification of paddy stalk straw. Initial deformation temperature (IDT) of pellet samples in our research are appropriate for gasification according to the results of Tuğ (2016) (Table 7). Ash contents of pellets are in Table 8. The ash content of paddy stalk used only compared with studies conducted before. Jenkins *et al*. (1998) determined that the rate of SiO_2 is 74, 67% in ash of paddy stalk. In this research, the SiO_2 rate of ash content is lower than 65,9% stated (Table 8). The other values are similar to the former studies.

Table 7. The results of physical analysis of the pellet sample (%)

Pellet Sample	Lower calorific value (MJ.kg^{-1})	Initial Deformation Temperature (IDT) (°C)	Softening Temperature (ST)(°C)	Hemispheric temperature (HT) (°C)	Melting Temperature (FT)(°C)
PRF	12,18	1045	1239	1436	1436<FT<1500
PVA3	12,71	951	1066	1305	1305<FT<1500
PML5	12,78	944	1078	1275	1275<FT<1500
PC5	12,48	971	1109	1284	1284<FT<1500
PC15	12,92	1018	1127	1262	1262<FT<1500

Table 8. Ash content analysis results (%)

PelletSample	Al_2O_3	K_2O	Na_2O	SiO_2
PRF	0,9	12,7	4,2	65,9
PVA3	1,4	11,6	3,9	63,8
PML5	1,5	12,1	4,1	63,6
PC5	3,2	10,8	3,3	59,6
PC15	5,3	9,5	3,2	65,8

Conclusions

Paddy stalk straw could be used with the current heat and power technologies in many rice producer countries to replace fossil fuels to reduce sulphur dioxide and greenhouse emission as well as prevent pollution from paddy straw open burning. Determination of its physical and chemical properties is required before the using as biomass energy resource to reduce the associated costs of transport, handling and storage which is a major hurdle with respect to its chemical and energy production. Paddy stalk straw has low calorific value and low bulk density compared with other biomass. There is no big difference between the chemical and physical analyzes of pellet samples gained from different mixtures using additives. However, calorific value of pellets is lower than the reported values in literatures. Pellet preparation process in this research showed that that there is no problem in pelletisation which is made without any additive. So It can be said that it is not necessary to use additives in pelleting of paddy stalk straw when using suitable pelleting techniques. As a result of this research, it is occurred that paddy stalk can be easily pelletized and gasified. In the Thrace Region in Turkey, 300.000 ton paddy stalk were burned every year. If this amount can be put to good use by pelleting and gasification process, it will contribute to the national economy. The other advantages of this biomass are its high amount as raw material and its very cheap cost, even cost-free. So it can be used directly for thermochemical processes as pellet form in many countries that are important rice producer.

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