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Fuel characteristics of agropellets fabricated with rice straw and husk

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Abstract−Our aim was to identify the potential of rice straw (RS) and rice husk (RH) as raw materials for pellet production. Compared to woody biomass, RS and RH can be easily dried, but contain significant levels of ash. Higher heating values of oven-dried RS and RH are slightly lower than those of commercial wood pellets. RS and RH contain substantially more silicon, potassium and calcium than larch sawdust. However, ash and moisture contents of RS was significantly reduced following a 15-week exposure period on rice paddy. These results suggest that RS and RH present suitable alternatives to wood as raw materials for pellet production due to their availability, relatively high calorific value and low moisture content. The durability of RS and RH pellets improved steadily with increasing pelletizing temperature and time. Pelletization under appropriate conditions also enabled the durability and bulk density of RS and RH to be improved, enhancing their potential as alternative combustion fuels.

Keywords: Rice Straw, Rice Husk, Pellet, Ash Content, Outdoor Exposure

INTRODUCTION

For the last two decades, there has been tremendous interest in using biomass materials as a carbon dioxide gas neutral and sustainable alternative to fossil fuels for heat and power generation. Among the biomass-based fuels, wood pellets are the most popular and are widely used in many European countries and in North America, because of their lower manufacturing, transportation and handling costs, compared to other biofuels [1]. To date, wood pellets have mainly been produced from sawdust, planer shavings or dried chips obtained from sawmills and wood industries [2]. Global consumption of wood pellets in 2008 was 11 million tons (MT) on an oven-dry basis, and that was projected to increase to more than 22 Mt by 2014 [3]. More interestingly, demands for wood as a construction material and as a source of energy are expected to increase greatly in the future. It is therefore reasonable to assume that woody biomass for pellet production might become increasingly scarce, or that its price might increase. As a result, it has become necessary to search for new biomass material to secure a safe and cost-effective supply of raw materials for pellet production. The raw material has not yet been used for this purpose, and there will not be competition with food production or other uses.

As biomass materials satisfying these criteria, agricultural residues have become an object of attention for pellet production in several European countries and in North America. Relative to other biomass materials, these constitute a socially and environmentally acceptable option for the creation of new, abundant, clean-burning, greenhouse gas-friendly fuel for heat-related energy applications. For example, switchgrass was chosen as a model herbaceous energy crop by the U.S. Department of Energy in the early 1990s [4]. Canada was also considering the use of switchgrass and alfalfa for fuel. At present, various grass straws, including miscanthus, straw, reed canary grass and peat are the most commonly used raw materials for solid bio-fuels [5]. Straw, in particular, is the most important alternative biomass material, since nearly 23 MT of dry straw biomass are harvested annually. This amount exceeds European usage of wood pellets (10 million ton) and production of alternative pellets (0.4 million ton). Reed canary grass in Northern Europe and miscanthus grass in Central Europe are also popular for use as fuels, because their combustion characteristics are better than those of straw.

However, there is a serious problem with the thermo-chemical behavior of fuel pellets produced from agricultural residues (agropellets). The relatively high nitrogen, sulfur, chlorine and ash content of agricultural residues, and their low ash melting temperature can lead to clinker formation and corrosion of boilers, as well as to high emission levels [6]. Furthermore, the physio-mechanical and chemical properties of agropellets differ from those of wood pellets. Thus, the pelletizing process to produce agropellets is more challenging compared to that involved in producing wood pellets, owing to low bulk density of agricultural residues. The durability of agropellets is also quite limited, since most agricultural residues have lower lignin content than wood sources, and a high concentration of hydrophobic waxes on their outer surface [7]. Such problems with agricultural residues have resulted in slow commercial acceptance of agropellets. The technical problems might, however, be efficiently resolved by mixing agricultural residues with additives, with other agricultural residues or even with woody biomass, managing delayed harvest and using advanced combustion systems [8].

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In South Korea, rice is the most commonly grown crop, and annual production in 2011 was about 40 million tons [9]. Rice straw and husk biomass therefore have significant potential for heat and power production, and could act as possible alternatives to woody biomass in South Korea, which has few forest resources. This study was conducted to identify the potential of rice straw and husk as raw materials for pellet production.

EXPERIMENTAL

1. Raw Materials

Rice straw (RS) was collected from rice paddies, located in Cheongju, South Korea. The RS was air-dried in the shade for one week, and then chopped using a chopping mill (YM-450BM, Yulim Co. Ltd., Kyeongsan, South Korea). Rice husk (RH) was obtained from Jukyung rice mill (Ohchang, South Korea) as ground particles. The RS and RH were screened through two sieves with size opening 1.41 mm (18 mesh) and 3.17 mm (8 mesh). Particles (1.41 mm< size of particles<3.17 mm) obtained by screening were used as raw materials for the fabrication of agropellets (Fig. 1). Wood pellets provided by the Forest Products Distribution Information System (Yeoju, South Korea) of the National Forestry Cooperative Federation (NFCF) were used as control pellets. The wood pellets were mainly made of larch (*Larix kaempferi C.*) sawdust.

The RS, RH and wood pellets were ground to powder using a Polytron medium-capacity homogenizer (PT-MR 2500 E; Kinematica AG; Lucerne, Switzerland), and then screened through a sieve with size opening of 0.42 mm (60 mesh). The fine powder passed through the sieve was used to analyze elemental and chemical compositions as well as to identify fuel characteristics (ash content and higher heating value), of agropellets and wood pellets.

2. Analysis of Chemical Composition

Moisture content of RS and RH was determined using an oven-drying method [10]. For the measurement of ash contents, 1 g of the oven-dried RS and RH was placed into a dried and pre-weighed porcelain crucible, and then burned away the RS and RH in the muffle furnace at 575 °C for 6 h [11]. The crucible was weighed after cooling to room temperature in a desiccator. Ash content was calculated as the ratio of weights of ash and original sample. For the determination of holocellulose and lignin content, standard procedures of the Association of Official Analytical Chemists and National Renewable Energy Laboratory were used, respectively [12,13].

Fig. 1. Images of rice straw and husk used for the fabrication of pellets in our study.

Agricultural residues, especially straw, have high ash content, affecting poor operation in conventional small-scale boilers by corrosion. Reducing the amount of ash contained in agricultural residues is required in order to be used as a raw material of solid biofuels. Several studies showed that elemental concentration of alkali metals on fresh biomass was reduced by leaching behavior under a variety of leaching media [14-16]. In this study, RS was subjected to outdoor exposure, from November 4, 2011 to February 17, 2012, to examine the reduction of ash content by washing with water or sunlight. The exposed RS was examined biweekly, to identify the effect of outdoor exposure on moisture and ash contents.

3. Elementary Analysis and Higher Heating Value

For the elementary analysis of RS and RH, a specimen of 2 mg was put into the reactor chamber and combusted at 990 °C, with a sufficient supply of oxygen. The specimens were converted into a gas mixture with complete oxidation. The gas mixture was passed to a gas chromatographic system with helium gas as a carrier gas, and separated into CO_2, N_2, H_2O and SO_2 gases. Each gas was transformed to an electrical signal using a thermal conductivity detector. Each gas was calibrated through elementary analysis of standard samples provided by the instrument manufacturer, and then the content of carbon, hydrogen, nitrogen and sulfur was measured. The amount of chlorine content contained in RS and RH was determined by the inductively coupled plasma - atomic emission spectroscopy (ICP - AES, Optima 4300 DV, PerkinElmer, Waltham, USA). The results of elementary analysis were calculated based on the average of three measurements.

The adiabatic oxygen bomb calorimeter (Parr 6400 Automatic Isoperibol Calorimeter, Parr Instrument Inc., Moline, Illinois, USA) was used for the determination of higher heating value (HHV) of each specimen based on the standard for the quality of wood pellet designated by the Korea Forest Research Institute (KFRI) [17]. Initially, 1 g of each specimen was placed in a nickel crucible and burned inside the bomb calorimeter. The specimen was ignited by a pure cotton thread in the presence of oxygen. Upon ignition, the released heat was transferred to the temperature sensor [18]. The increase in temperature was used to calculate the HHV of the specimen. The HHV was expressed in MJ/kg. Each specimen was tested three times to obtain the most accurate results.

4. Qualitative Analysis of Ash

Qualitative analysis of ash contained in RS and RH was measured according to the standard for the quality of wood pellets designated by the KFRI [17]. All constituents except for ash were dissolved by 65% nitric acid solution, and then the ash was separated on the glass filter (1G4). Subsequently, silicon (Si), calcium (Ca), potassium (K), sodium (Na), magnesium (Mg), zinc (Zn) and chromium (Cr) were analyzed by ICP - AES, and arsenic (As), cadmium (Cd), mercury (Hg), copper (Cu) and lead (Pb) by ICP - mass spectrometry (ICP - MS).

5. Pelletizing and Pellet Characterization

To minimize the experimental deviation resulting from differences in the moisture content of raw materials and pellet size, moisture content and amounts of RS or RH particles used for the fabrication of an agropellet were adjusted to 11±0.1% and 1 g, respec-

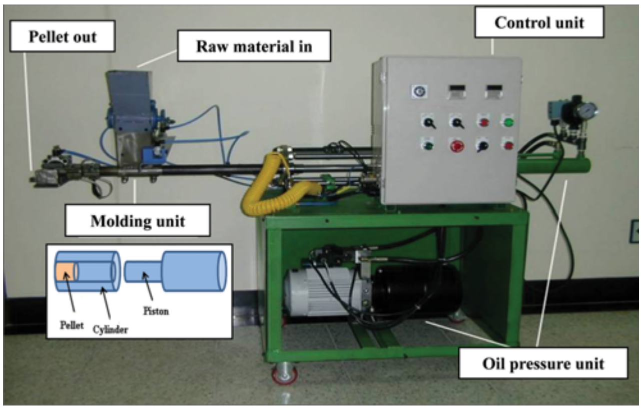

Fig. 2. Image of piston-type pelletizer used for the fabrication of pellets in our study.

tively. Agropellets were fabricated by using a single pellet press installed at the laboratory of Chungbuk National University (Cheongju, South Korea), as shown in Fig. 2 [19]. The press consisted of a 7 mm-cylindrical die in diameter, made of hardened steel, and lagged with heating elements. The end of the die was plugged by using a removable backstop. Pressure (150 MPa) was applied to the biomass using a piston made out of hardened steel and connected to a hydraulic press. The die temperature was adjusted to 90, 120, 150 and 180 °C, and pellets were pressed for 1, 2 and 3 min, to examine the effects of pelletizing temperature and time on the durability of the agropellets. The fabricated agropellets were placed in an incubation room (25 °C and 50% RH) for at least 24 h before the durability test.

Durability of agropellets was tested by the tumbling can method [20]. The tumbling can consists of a rectangular box with the inside dimension of 30.5×30.5×12.7 cm^3. The box was sealed so that no dust could escape during tumbling. Agropellets (50 g) were placed in the tumbling can and tumbled for 500 rounds at 0.83 Hz. Following tumbling, the tumbled pellets were sieved using a 3.15 mm sieve. The durability of the agropellets was calculated as the weight ratio after tumbling to that before tumbling.

RESULTS AND DISCUSSION

1. Chemical Composition

The chemical composition of RS and RH is given in Table 1. Moisture content (MC) of RS and RH was lower than that of larch sawdust, which is the most commonly used for the production of wood pellets in South Korea. These results indicate that RS and RH can be easily dried, when compared to woody biomass. Both contained large amounts of holocellulose to the same degree as woody biomass, but showed lower lignin content than larch. Most importantly,

Table 1. Chemical composition of rice straw and husk

Biomass	Moisture (%)	Total solids (%)	Chemical composition (% w/w)[a]			
			Holocellulose	Lignin	Ash	Others[b]
Rice straw	6.72	93.28	65.47	12.83	9.44	12.26
Rice husk	5.59	94.41	65.21	22.00	10.87	1.92
Larch pellet[c]	10.55	89.45	68.80	28.62	0.34	2.24

[a]It means the percentage based on dry weight
[b]It includes silicon, extractives and so on
[c]Wood pellets produced in the Forest Products Distribution System of National Forestry Cooperative Federation. The wood pellets were fabricated with larch (*Larix kaempferi C*) sawdust

Table 2. Elemental contents and higher heating values of rice straw and husk

Biomass	Elemental content (%)					Higher heating value (MJ/kg)
	C	H	N	S	Cl	
Rice straw	43.02	5.64	0.70	0.06	0.14	17.3
Rice husk	46.01	5.87	0.66	0.06	0.12	17.8
Larch pellet[a]	46.87	5.99	0.08	0.01	<0.01	20.1
3rd-Grade pellet[b]			<0.70	<0.05	<0.05	≥16.9
ENplus-B[c]			≤1.00	≤0.05	≤0.03	16.0<Q<19.0
Agro+[d]			≤1.50	≤0.20	≤0.20	
Cereal straw pellet[e]			≤0.70	≤0.10	≤0.10	Minimum value to be stated

[a]Wood pellets produced in the Forest Products Distribution System of National Forestry Cooperative Federation. The wood pellets were fabricated with larch (*Larix kaempferi C*) sawdust
[b]Standard for the quality of wood pellets designated by Korea Forest Research Institute
[c]European wood pellet standards (EN 14961-2)
[d]French agropellet standards
[e]European non-woody pellet standards (EN 14961-6)

Table 3. Content of major metals contained in rice straw and husk (unit: mg/kg)

Biomass	Si	K	Na	Ca	Mg
Rice straw	70,488	12,745	108	10,644	1,317
Rice husk	99,374	4,023	326	3,266	424
Larch pellet[a]	146	115	66	471	84
Typical average value of wheat, rye, barley straw[b]	110,000	10,000	500	4,000	700

[a]Wood pellets produced in the Forest Products Distribution System of National Forestry Cooperative Federation. The wood pellets were fabricated with larch (*Larix kaempferi C*) sawdust
[b]Refer to Lu & Ralph (2010)

Table 4. Content of other metals contained in rice straw and husk (unit: mg/kg)

Biomass	Pb	As	Cd	Cu	Zn	Cr	Hg
Rice straw	ND	0.3	ND	18	48	ND	ND
Rice husk	ND	ND	ND	16	26	ND	ND
Larch pellet[a]	ND	0.2	ND	2	2	0.2	ND
ENplus-B[b]	≤ 10	≤ 1	≤ 0.5	≤ 10	≤ 100	≤ 10	≤ 0.1
Agro, Agro+[c]	≤ 10	≤ 1	≤ 0.5	≤ 40	≤ 60	≤ 10	≤ 0.1
Cereal straw pellet[d]	≤ 10	≤ 1	≤ 0.5	≤ 20	≤ 100	≤ 50	≤ 0.1

[a]Wood pellets produced in the Forest Products Distribution System of National Forestry Cooperative Federation. The wood pellets were fabricated with larch (*Larix kaempferi C*) sawdust
[b]European wood pellet standards (EN 14961-2)
[c]French agropellet standards
[d]European non-woody pellet standards (EN 14961-6)
ND means that each element was not detected

as expected, RS and RH contained significant levels of ash.

2. Elementary Analysis and Higher Heating Value

Table 2 presents the elementary analysis and higher heating value (HHV) results for RS and RH. Carbon and hydrogen content of RH were higher than those of RS, but lower than those of larch sawdust. Additionally, RS and RH exhibited significant quantities of nitrogen, sulfur and chlorine when compared to larch. The content of nitrogen, sulfur and chlorine contained in RS and RH did not meet the KFRI standard for 3rd-grade wood pellets [17], but satisfied the European standards for B-grade wood pellet (ENplus-B), the French standard for Agro^{+}-grade agropellet (Agro^{+}) and the European standard for nonwoody pellets (EN cereal straw pellet), respectively [21-23].

Table 2 also lists the HHV of RS and RH. HHV of oven-dried RS and RH was lower than that of commercial wood pellets. The low HHV of RS and RH was probably due to their low lignin and carbon content, as shown in Tables 1 and 2. According to White, Dhamodaran, Cordero et al. [24-26], HHV of a lignocellulosic or carbonaceous material is proportional to its carbon and lignin content. The content of lignin and carbon contained in RS and RH was lower than that of larch, and consequently HHV of RS and RH might be lower than that of larch sawdust. Additionally, HHVs of RS and RH were slightly higher than that of the KFRI standard for the 3rd-grade wood pellets, and satisfies the ENplus-B standard. From the results of the elementary analysis and HHV, RS and RH might possibly have a potential as raw materials for agropellet production.

3. Qualitative Analysis of Ash

To identify metals and heavy metals contained in RS and RH, qualitative analysis of ash was conducted. The results are summarized in Tables 3 and 4. Si, K and Ca content of RS was notably higher than those of larch sawdust. In particular, the high K content of RS is the main reason for the production of clinker or slag formation in a boiler during combustion [27,28]. Further work on reducing the potassium content of RS is therefore necessary to be used as a raw material for agropellet production. RH, meanwhile, contained less K, Ca and Mg than RS, but the relative values were still high when compared to those of wood pellets. The results indicate that RH might be a more suitable raw material for the production of agropellets than RS.

When other metals included in RS were analyzed qualitatively, Cu, Zn and minor quantities of As were found (Table 4). RH contained only large amounts of Cu and Zn. The amount of Cu and Zn contained in RS and RH largely exceeded that found in wood pellets, but below the standards of ENplus-B, Agro^{+} and cereal straw pellets. On the other hand, no Pb, Cd, Cr and Hg were detected in RS or RH. The results indicate that RS and RH might be usable raw materials for the production of agropellets.

4. Effect of Outdoor Exposure on Chemical Behavior of RS

When RS is used as a raw material for agropellet production, its high ash content has a negative effect on the thermochemical behavior of the agropellets. One way to resolve the problem is to leach the elements contained in RS with water and sunlight from its outdoor exposure [14-16,27,29]. RS was therefore exposed to the outdoors for 15 weeks after harvesting, and then the ash and moisture content of the exposed RS was measured biweekly (Fig. 3). The initial ash content of RS was around 12.3%, and the ash content

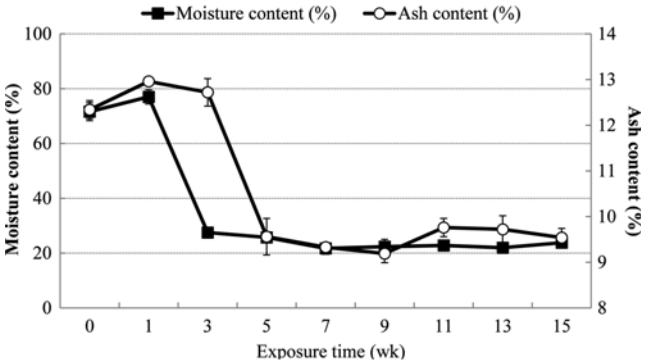

Fig. 3. Effect of exposure time on the moisture and ash contents of rice straw.

had slightly increased until the third week. However, the ash content decreased considerably to 9.6% in the fifth week, and then remained at approximately 9.0% until the fifteenth week. The results indicate that the outdoor exposure of RS for a certain period can effectively reduce its ash content.

In the case of MC, outdoor exposure of RS showed a quite similar result to the ash content of the exposed RS. Based on the results of ash and moisture content, the outdoor exposure of agriculture residues might be an effective way to improve several thermochemical properties of agropellets.

5. Effect of Pelletizing Temperature and Time on Durability

Durability of RS pellets was higher than that of RH pellets (Fig. 4). RS is more flexible than RH, and RS particles might thus come into closer contact than RH particles during the pelletizing process, resulting in high durability of RS pellets. In addition, differences in the particle shape of RS and RH might have an effect on the durability of the respective pellets. In other words, as shown in Fig. 1, the slender shape of RS particles might contribute to increasing the durability of pellets [20]. This inference could be ascertained from the result that the bulk density of RS pellets (692 kg/m^3) is higher than that of RH pellets (654 kg/m^3).

Effects of pelletizing temperature and time on the durability of RS and RH pellets are shown in Fig. 4. The durability of RS (p=0.01) and RH pellets (p=0.01) increased with increasing pelletizing temperature (Figs. 5 and 6). In particular, the durability of RS

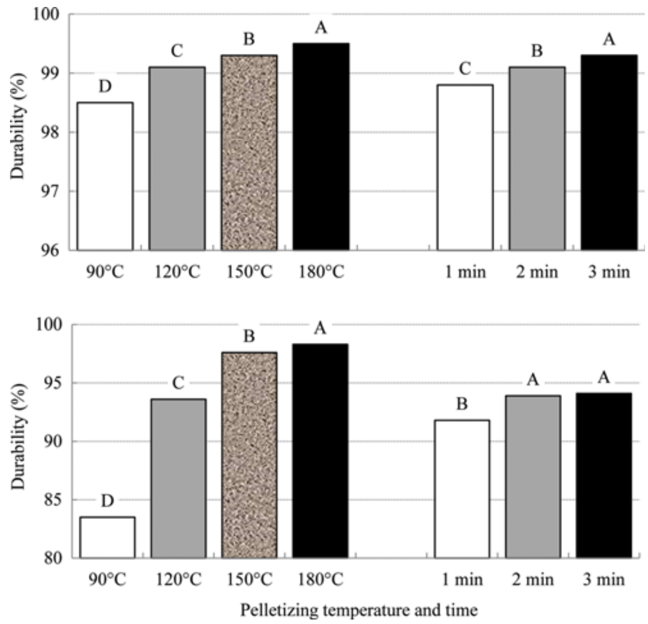

Fig. 4. Effects of pelletizing temperature and time on durability of agropellets fabricated with rice straw (top) and rice husk (bottom). The same capital letters over each column indicate that results are not significantly different from each other at α=0.05 (student's t-test).

and RH pellets greatly improved when the pelletizing temperature increased from 90 °C to 120 °C. In addition, increasing pelletizing time had a positive effect of the durability of RS (p=0.01) and RH (p=0.01) pellets. The increased durability of RS and RH pellets fabricated at higher temperatures and for longer times is probably due to the greater auto-adhesive action of lignin contained in RS and RH [19-20,30]. For instance, lignin likely acted as a natural binder between RS or RH particles, and thus may be thermally softened at elevated temperatures and extended pelletizing times. Consequently, the softened lignin helps enhance the binding strength between RS or RH particles. However, the durability of RH pellets fabricated for 3 min was not significantly different from that for 2 min (p=0.22). The result suggests that the durability of RH pellets might be less affected by increase in pelletizing time than by

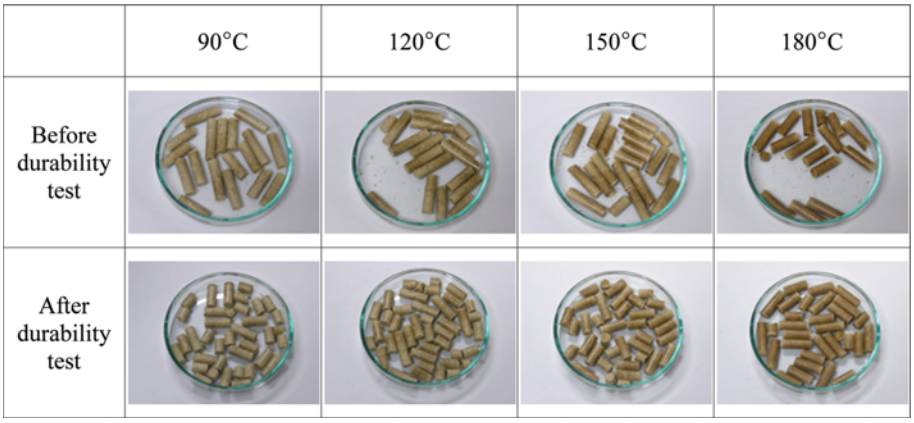

Fig. 5. Images of rice straw-based agropellets pelletized for 3 min before and after the durability test.

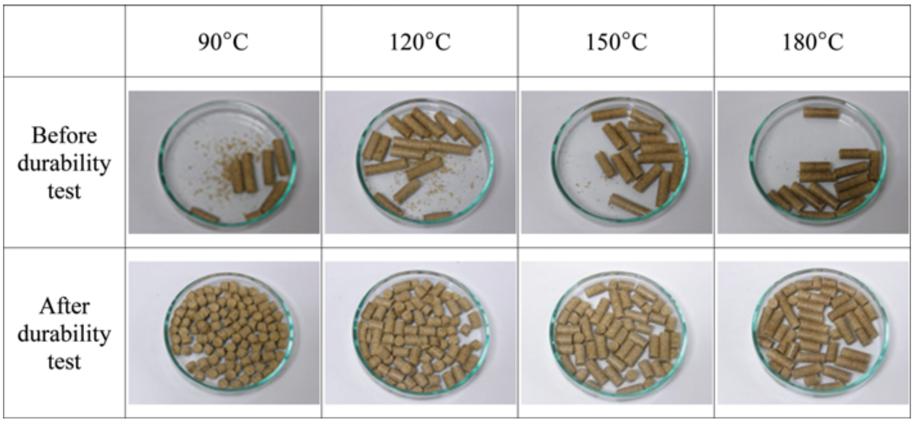

Fig. 6. Images of rice husk-based agropellets pelletized for 3 min before and after the durability test.

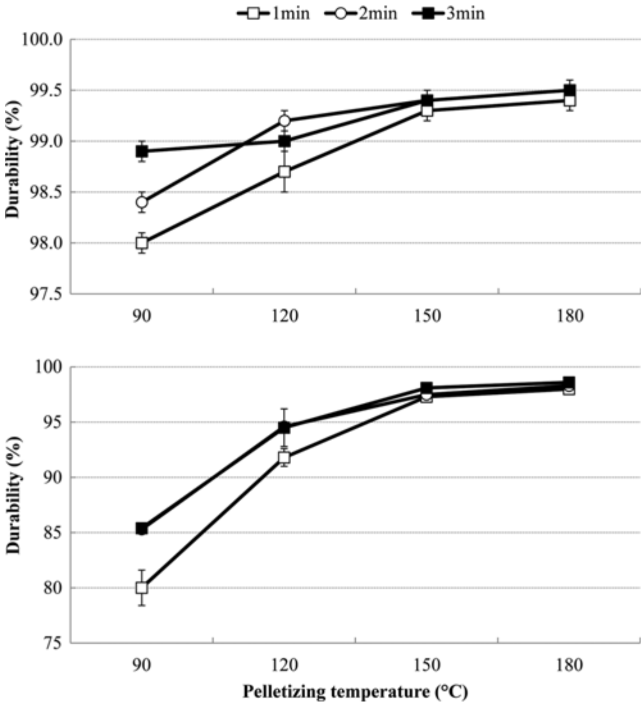

Fig. 7. Interaction effects of pelletizing time and temperature on the durability of agropellets pelletized with rice straw (top) and rice husk (bottom).

pelletizing temperature.

When the effect of pelletizing temperature within a certain pelletizing time was compared, the durability of RS (p=0.01) and RH pellets (p=0.01) showed a significant difference (Fig. 7). For example, at pelletizing temperatures of 90 °C and 120 °C, there were significant differences between the durability of RS and RH pellets fabricated for 1 min and 2 min, but these differences were not evident or were not noticeably lower at a pelletizing temperature of 150 °C and above. This result indicates that pelletizing temperature is a more significant factor for improving the durability of RS and RH pellets than pelletizing time.

When the durability of RS and RH pellets was compared with that of commercial wood pellets, the durability of RS pellets was slightly lower than that of larch wood pellets (99.5%). Furthermore, all RH pellets showed substantially lower durability than larch wood pellets. The low durability of RS and RH pellets might be associated with their low lignin content, compared to larch sawdust [19-20,30]. Meanwhile, all durability values of RS pellets exceeded the standards of the KFRI for the 1st-grade wood pellets (≥97.5%), ENplus-B (≥96.5%), Agro+ (≥95.0%) and EN cereal straw pellets (≥97.5%) regardless of pelletizing conditions. On the other hand, RH was required to be pelletized at a temperature of 150 °C or higher to satisfy the standards as stated above.

Overall, this study demonstrated that RS and RH present a suitable alternative to wood as a raw material for pellet production, due to their availability, relatively high calorific value and low moisture content. Pelletization under appropriate conditions also enabled the durability and bulk density of RS and RH to be modified to improve their potential as alternative combustion fuels. In addition, when RS was exposed to the outdoors for 15 weeks after harvesting, significant reduction of its ash and moisture content was found. However, further work is required to determine whether the observed reduction is of practical significance or not.

CONCLUSIONS

Energy obtained from the combustion of biomass is a renewable alternative to fossil energy. Biomass alternatives are needed to reduce reliance on wood, which is currently the predominant biomass for combusting fuels. We investigated the potential of RS and RH as a raw material of agropellets from the results of their chemical composition, elemental analysis, qualitative/quantitative analysis of ash and fuel characteristics. RS and RH can be easily dried compared to woody biomass, but contain significant levels of ash. The carbon and lignin contents of RS and RH are lower than those of larch sawdust, but RS and RH exhibit significant quantities of nitrogen, sulfur and chlorine compared to larch. HHV values of oven-dried RS and RH are slightly lower than that of commercial wood pellets. The ash analysis of RS and RH showed that these contained more Si, K, Ca, Mg and Na than larch sawdust. The amounts of Cu and Zn contained in RS and RH largely exceeded those of larch wood pellets, but Pb, Cd, Cr and Hg were not detected in either RS or RH. The high ash content of RS can be a main causative factor for the production of clinker or slag formation in a boiler during combustion. For this reason, RS was exposed to the outdoors for 15 weeks after harvesting, and the ash and moisture content of the exposed RS was measured biweekly. Outdoor exposure of RS for this period reduced its ash and moisture content effectively. In the RS and RH pellets, the increases of pelletizing temperature and time had a positive effect on the durability. All durability values of RS pellets exceeded the standards of the KFRI for 1st-grade wood pellets, ENplus-B, Agro+ and EN cereal straw pellet regardless of pelletizing conditions. On the other hand, RH was required to be pelletized under appropriate conditions for satisfying the standards as stated above.

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