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Alternative fuels from pyrolysis of rice crop waste

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Abstract

Rice is one of the most popular crops in the world generating significant amount of crop residues, majorly rice husk and rice straw. In Asia, where more rice and consequently more straw and husk are grown each year to meet the region's growing demand for food grains, managing these crop residues is still a challenge. Burning crop waste is also a fairly common practice in northern India which causes air pollution. The use of clean renewable energies has received a lot of attention due to the scarcity of fossil fuels and serious environmental issues. In such times, biomass like rice straw and rice husk becomes an important source of energy that can be directly transformed into solid and liquid fuels through pyrolysis. In this study, a batch type pyrolysis reactor was used to pyrolyse rice crop residues to produce better alternative fuels. Raw materials were characterized through bulk density, proximate analysis and calorific values to investigate their suitability as feedstock for this consideration. The pyrolysis unit, comprising of reactor, condenser, water pump, grate etc. was developed and operated in limited air supply to produce a hydrocarbon rich bio-oil, a carbon-rich bio-char and a gaseous mixture i.e. pyro-gas. Rice husk and straw were fed into the reactor and bio-oil, char and gases were produced, the liquid and char were collected separately while the gas was released into the atmosphere. The characterization and composition of the product yields were determined separately for the char and oil. The findings demonstrate that to produce 1 liter of crude bio-oil, 8.949 kg of rice husk or 20.548 kg of rice straw was required, to produce 1 kg of bio-char, 1.82 kgs of rice husk or 2.14 kgs of rice straw was required and to produce 1 kg of pyro-gas, 2.95 kgs of rice husk or 2.07 kgs of rice straw was required. The results show that if the pyrolysis conditions are chosen appropriately, it is possible to produce high quality alternative fuels from rice straw and rice husk that are comparable to fossil fuels.

Keywords: Pyrolysis, bio-oil, bio-char, alternative fuels, rice husk, rice straw

1. Introduction

The fundamental element of economic progress and the engine powering the world economy is energy. Nowadays, the most dangerous global problem is the alarming rate of depletion of fossil fuels. Non-renewable energy sources are limited and are unfriendly to the environment, it also drains India's foreign exchange reserves as India imports these kinds of energy sources. Renewable resources have not been used frequently in India due to lack of awareness and acceptability of renewable energy sources by power consumers. Therefore, it is essential to explore other sustainable energy sources. One of these non-conventional sources is biomass energy.

Worldwide, rice is the agricultural commodity with the third-highest worldwide production, with about 761.5 million tonnes production in 2018. India has huge amount of agriculture land area, so massive residues are produced here. Ministry of New and Renewable Energy estimated that 500 Mt of crop residues are generated annually (Anonyms, 2014) ^[1]. Labor shortages in the agricultural sector in India associated with rapid economic, social, and political changes have led to accelerated mechanization, particularly involving combine harvesters in rice-based cropping systems. Compared to traditional harvesting methods, combine harvesters leave rice straw on the field. The intensification of cropping systems is also resulting in a larger volume of rice straw being produced that, in turn, must be managed over shorter turnaround times between crops. Both of these trends have led to an increase in open-field burning of the straw because it is the easiest option for farmers. Open-field burning leads to loss of nutrients and creates air pollution that causes human respiratory ailments. In 2018, burning of rice straw and other agricultural residues which contributes to poor air quality prompted the Indian government to ban open fires in New Delhi. Burning the straw also removes opportunities for adding value to it. More sustainable rice straw management methods are urgently needed to minimize rice production's carbon footprint and its negative effects on human health and to maximize adding value to the straw by-product.

Past research on rice crop residues has focused on isolated topics or component technologies, e.g., to improve straw combustion properties or to analyze nutritional value as an animal feedstock. However, to date, there has been no holistic approach toward the research on potential of rice straw and rice husk as alternative energy source.

Rice straw, as a lignocellulosic biomass, is comprised of three components: lignin, cellulose, and hemicelluloses. Cellulose and hemicelluloses are fiber organics, whereas lignin is the cell wall (Klass, 1998). Each kg of milled rice produced results in roughly 0.7–1.4 kg of rice straw depending on varieties, cutting-height of the stubbles, and moisture content during harvest. Rice straw is separated from the grains after the plants are threshed either manually, using stationary threshers or, more recently, by using combine harvesters.

After the rice is harvested, it undergoes milling to prepare it safe for human consumption. Rice husks are the hard protective coverings of rice grains and it is an abundantly available waste material in all rice producing countries. In the course of a typical milling process, husks are removed from the raw grain to reveal whole brown rice which upon further milling remove the bran layer to yield white rice that can be found in local grocery store or supermarket.

As rice husk and straw contains complex mix of carbon, nitrogen, hydrogen and oxygen, and is largely available, they can be used as an alternative energy source, but the high ash content, relative to other biomass materials, makes its use problematic during co-firing. Despite its potential qualities of using it in the production of amorphous silica, silicon, furfural and as a fuel, it is being wasted in India and no major plan has been made till date to make it useful. Many alternative uses of rice straw and rice husk wastes have been reported recently. Rice straw has excellent potential for soil erosion control, sediment traps, and soil improvement. The straw prevents water erosion and soil loss by retaining moisture, reducing evaporation, controlling soil temperature, preserving fertilizer, paper, composite materials and providing useful soil organic matter as it decomposes. The applications mentioned above still produce other types of wastes. Pyrolysis technologies not only help control mass production of rice straw wastes, but they also assist in converting the wastes into valuable soil amendments and catalysts with a deoxidized bio-char. Also, the bio-oil and the gas generated during pyrolysis can be used in power generation.

Pyrolysis of agricultural waste is a promising route for waste to energy generation. Biomass is heated in the absence of oxygen, or partially combusted in limited oxygen supply to produce a hydrocarbon rich gas mixture, oil like liquid and carbon rich solid residue (Rathore *et al.* 2008) [13]. The process typically occurs at temperatures above 430 °C (800°F) and under pressure. Pyrolysis uses heat to break down combustible materials in the absence of oxygen or in limited supply of oxygen, producing a mixture of combustible gases (methane, complex hydrocarbons, hydrogen, and carbon monoxide etc.), liquids and solid residues.

The thermochemical process produces solid chars, liquid bio-oil and syngas as the final products. Syngas usually contain CO, CO₂, H₂ and CH₄, have relatively high HHV values, and can be co-combusted with natural gases in a combustor. The general composition of pyrolysis oil is cyclopentanone, methoxyphenol, acetic acid, methanol, acetone, furfural, phenol, formic acid, levoglucosan, guaiacol, and their alkylated phenol derivatives. The upgraded bio-oil can be used as the fuels for diesel engines. The solid chars can be co-

gasified with coals in power plants. The mass fraction of the gaseous product, liquid product and the solid product produced from the biomass pyrolysis is influenced by the biomass composition and the operating conditions of the pyrolysis process.

2. Material and Methods

2.1 Raw materials

For the present study rice straw and rice husk were chosen as a raw material. The raw material was collected from university farm and from Shree-Ram Rice Mill, Asud, Dapoli, District Ratnagiri, Maharashtra respectively. Undesirable materials such as small pieces of stones, wood etc. were separated from raw materials by hand picking and sieving. The materials were spread under the sun and stirred regularly to reduce the amount of moisture in product traditionally. The size reduction of raw material was carried out manually by using paddy straw cutter. After the size reduction, the raw materials were stored in an air tight plastic container to ensure continuous supply of materials during the experiment.

2.2 Characterizations of raw product

After cleaning and sun drying the available rice husk and rice straw in the open sun, three different samples each of rice husk and rice straw were randomly selected and analysed for the determination of its characteristics such as bulk density (ASTM D-6683), proximate analysis (i.e., moisture content (ASTM 14 D-3173), volatile matter (ASTM D-3175), ash content (ASTM D-3174) and fixed carbon (method of differences)) and calorific value (HHV) (ASTM E-711).

2.3 Development of the experimental pyrolysis set-up

A 5 kg capacity biomass pyrolysis system was developed in workshop of CAET, DBSKKV, Dapoli and operated in Energy Park, Department of Electrical and Other Energy Sources, CAET, DBSKKV, Dapoli, District Ratnagiri, Maharashtra for pyrolysis of rice straw and rice husk. The experimental pyrolysis set-up mainly consists of a reactor, air blower and insulation, condenser, electric water pump, water storage tank etc.

Table 1: Technical specifications of the pyrolysis reactor developed for study

Sr. No.	Parameter of reactor	Values
1	Diameter (mm)	610
2	Height (mm)	864.4
3	Capacity (kg)	5
4	No. of feeding port	1
5	Diameter of feeding port (mm)	200
6	No. of exhaust port (mm)	1
7	Diameter of exhaust port (mm)	76.2
8	No. of air inlet port	3
9	Diameter of air inlet port (mm)	29.21
10	Height of grate (mm)	250

Table 2: Technical specifications of the condenser developed for study

Sr. No.	Parameters of condenser	Values
1	Diameter of inner cylinder (mm)	150
2	Diameter of outer cylinder (mm)	200
3	Annular space between inner and outer cylinder (mm)	25
4	Length (mm)	970

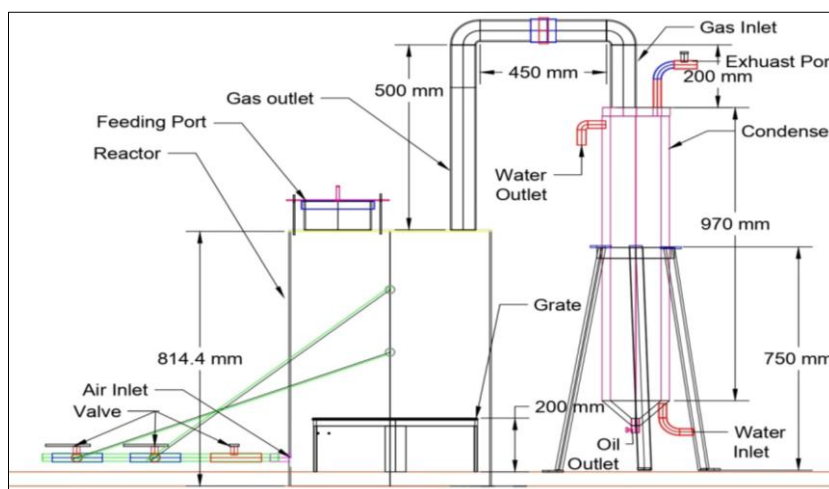


Fig 1: Developed pyrolysis set-up

2.4 Pyrolysis test

The pyrolysis was started in the reactor by the direct limited combustion of rice crop waste, and for that, the minimum amount of air was provided to start the combustion. First, five kg of one of the raw materials was weighed and fed into the reactor chamber through the feeding port. After that, the raw material was set to fire and was left to burn. The feeding port was then closed to make the reactor chamber air tight. Within 1 to 1:30 hours, the temperature of reactor reached 300 to 500°C. When uncondensed gas from the exhaust port of the condenser became less visible, then the pyrolysis process of rice crop waste was considered to be completed. After completion, the reactor was allowed to be cooled down for 2-3 hours for collection of bio-char and bio-oil. The bio-char was collected from the bottom of the reactor and the bio-oil was collected from the bottom of the condenser. Quantity of the final product was measured.



Fig 3: Bio-char from rice husk and rice straw



Fig 4: Bio-oil

Fig 5: Pyro-gas



Fig 2: Pictorial view of complete pyrolysis set-up

2.5 Product yield

Three main products of pyrolysis of rice crop waste were determined in terms of bio-char, bio-oil and pyro-gas yield as per the following formula.

$$\text{Bio-char yield (\%)} = \frac{\text{weight of bio char}}{\text{weight of initial biomass}} \times 100$$

$$\text{Bio oil yield (\%)} = \frac{\text{weight of bio oil}}{\text{weight of initial biomass}} \times 100$$

$$\text{Pyro gas yield (\%)} = 100 - (\text{Bio-char yield} + \text{Bio-oil yield})$$

2.6 Characterization of final products

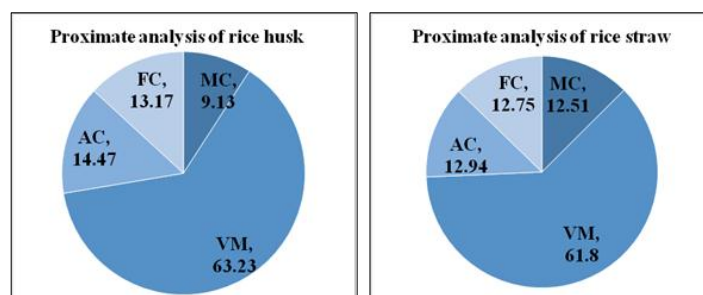
The evaluation of characteristic of bio-char produced from both rice straw and rice husk was carried out separately in terms of bulk density, Calorific value and proximate analysis which included moisture content, volatile matter, ash content and fixed carbon to investigate the quality of product yields, whereas the bio-oil was characterized in terms of liquid fuel properties such as density (ASTMD-6683), calorific value (ASTME-711), viscosity (ASTM D-446), flash point and fire point (ASTMD-92) using the standard method as depicted in the respective brackets.

3 Results and Discussion

The results obtained from the study undertaken on assessment of characterization of rice husk and rice straw, extraction of alternative fuels from rice husk and rice straw by pyrolysis and assessment on properties of pyrolysis products are segmented below. The variety of rice used for the experiment was Ratnagiri Payabhut 13 (BRCKKV-13).

Table 3: Characteristics of raw materials

Sr. No.	Property	Values	
		Rice husk	Rice straw
1.	Moisture content (%)	9.13	12.51
2.	Volatile matter (%)	63.23	61.80
3.	Ash content (%)	14.47	12.94
4.	Fixed carbon (%)	13.17	12.75
5.	Higher heating value (MJ/kg)	15.58	14.67
6.	Bulk density (kg/m ³)	116	69.92

**Chart 1:** Proximate analysis of rice husk and rice straw

There is a direct and strong relationship between moisture content of a biomass fuel and its energy content, or calorific value as it can potentially reduce combustion temperature affecting overall system efficiency. In this study, the average moisture content in rice husk and rice straw was within the acceptable limit (i.e., below 15%) for raw materials to be used for pyrolysis combustion. Moisture content was found to be higher in rice straw largely due to bigger particle size and porous structure of rice straw which would not have let it dry properly as compared to rice husk sample during the sun drying. This might be due to hygroscopic characteristics of material due to its internal structure.

The most significant properties of biomass particles during fuel combustion are volatile matter content and fixed carbon content. Volatile matters are those components of fuel which are readily burnt in the presence of oxygen. It is usually a mixture of aromatic hydrocarbons, short and long-chain hydrocarbons, and sulphur. From the analysis, it was observed that high volatile matter made rice straw and rice husk a good raw material for extracting an alternative fuel through pyrolysis process as it is known that higher amount of volatile matter increases bio-oil, tar and syngas yield.

Ash content in feedstocks also significantly impacts the distribution and quality of pyrolysis products as higher ash content impacts the bio-oil yield negatively. Higher ash content in rice husk implies the higher presence of alkali metals in rice husk as compared to the rice straw.

The fixed carbon content of the biomass accounts for the coke formation in char. It is the solid combustible residue that remains after the volatile matter drive off. The higher fixed carbon content of rice husk sample makes it more efficient biomass fuel as compared to the rice straw sample as fixed carbon content also positively affects the calorific value of a fuel.

The results of proximate analysis obtained in this experiment are in agreement with the findings of previous works done in the subject including Hossain *et al.*, (2017) [5] for rice husk, Biswas *et al.*, (2017) [2] and Makavana *et al.*, (2018) [10] for rice straw.

3.2 Higher heating value of rice husk and rice straw

Higher heating value (HHV) is one of the properties of

3.1 Proximate analysis of rice husk and rice straw

Proximate analysis of a fuel provides the percentage of the material that burns in a gaseous state (volatile matter), in the solid state (fixed carbon), and the percentage of inorganic waste material (i.e. ash), and is therefore of fundamental importance for biomass energy use.

biomass fuels which is essential in investigating their special characteristics and potentialities. It explains the higher energy content and determines the efficient use of biomass. HHV comes to design calculations or numerical simulations of thermal conversion systems for fuels. During reviews, it was found that similar results have also been reported by previous researchers including Jenkins *et al.*, (1998) [7], Yao *et al.*, (2016) [17] and Migo (2019) [11]. These results show that the selected raw materials (rice husk and rice straw) were suitable for pyrolysis. Further, the HHV of rice husk was found to be higher than that of the rice straw mostly because HHV depends largely on the moisture and fixed carbon content of the biomass. Rice husk's moisture content of is less and fixed carbon is higher than that of rice straw resulting in higher HHV for the husk.

3.3 Bulk density of rice husk and rice straw

Bulk density is an important characteristic of biomass that influences directly the cost of feedstock delivered to a bio-refinery and storage cost, it is a major physical property in designing the logistic system for biomass handling. The valuation of bulk density of rice husk and rice straw was found under the standard range in accordance with the results presented by Putun *et al.* (2004) [12] and Kirubakaran *et al.* (2012) [8]. The small variation may be due variety, climate condition and practices followed. The bulk density of rice straw sample was observed to be significantly low as compared to that of rice husk, perhaps due to organic matter of rice straw is lighter than an equal volume of rice husk and is more porous. Moreover, slightly bigger particle size of shredded rice straw, shape and lower particle density could might have lowered the bulk density of rice straw as compared to rice husk.

The knowledge of bulk density of both materials was useful in determination of input quantity of materials into the pyrolysis chamber and in designing of the pyrolysis chamber.

3.4 Product yield from pyrolysis of rice husk and rice straw

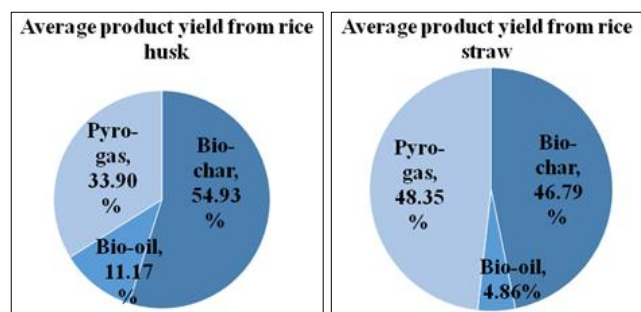


Chart 2: Average product yield from rice husk and rice straw

3.4.1 Bio-char yield from rice husk and rice straw

The solid product obtained from the pyrolysis is bio-char. The average bio-char yield from pyrolysis of rice husk ranged from 54.5% to 55.3% at temperature range from 300°C to 500°C. Same procedure was repeated with the rice straw and bio-char yield from the pyrolysis of rice straw varied from the minimum of 46.56% to the maximum of 47% at same temperature variation. The yield of bio-char is more when compared to bio-oil yield primarily due to the high temperature and bulk quantity of feedstocks used for the pyrolysis in this experiment. Further, higher fixed carbon content of the feedstocks would have also resulted in more bio-char production.

3.4.2 Bio-oil yield from rice husk and rice straw

The main product yield obtained from the pyrolysis process is bio-oil, usually referred as pyro-oil or bio-crude. Pyrolysis bio-oil is the dark brown liquid produced from the condensation of vapour of a pyrolysis reaction. It has potential to be used as a fuel oil substitute. These results obtained from the experiment were in accordance with the results showed by earlier literature on the subject including Doke *et al.*, (2015) [3] and Gopan *et al.*, (2018) [4]. Bio-oil yield is less in comparison to the other researches perhaps because the earlier worker's result in the subject were from small scale laboratory study under controlled conditions whereas this study was primarily focused on more practical use of the pyrolysis for optimum utility of the research.

3.4.3 Pyro-gas yield from rice husk and rice straw

The other by-product, that was obtained from the pyrolysis of rice husk and rice straw was pyro-gas also termed as syngas. The pyro-gas yield was observed to be slightly higher than the yields shown in similar researches and experiments conducted on slow pyrolysis of biomass such as Jahirul *et al.*, (2012) [6]. After analysis, it was observed that all the earlier experiments have been done in laboratory on small scale with very small amount of feedstock sample and in absence of air, whereas this current experiment was done in larger scale (5 kg feedstock in each run) with limited amount of air supply adjusted manually in order to make the experiment more useful and practical for the real users which are farmers in natural limited controlled conditions. The higher amount of feedstock and supply of air (oxygen) would have caused the feedstock to get combusted resulting in error.

3.5 Analysis of product yields

The main product yields obtained in each run from both the

rice husk and rice straw were collected and characterized in terms of proximate analysis and bulk density for the solid product (bio-char) and in terms of fuel properties for the liquid yield i.e. pyro oil.

3.5.1 Characteristics of Bio-char yields

The average moisture content in rice husk char was significantly lower than that of the respective feedstocks due to high temperature in the pyrolysis chamber. From the analysis, it was observed that the value of volatile matter has decreased on percentage basis after the pyrolysis process making it a better solid fuel. The fixed carbon content of rice husk char and rice straw char was significantly increased as compared to their respective feedstocks. Similar result was found by Salunkhe *et al.*, (2021) [14]. The HHV of rice husk and rice straw feedstocks was 15.58 MJ/kg and 14.67 MJ/kg which has increased to 18.65 MJ/kg and 17.70 MJ/kg respectively for bio-chars of both raw materials. The increase in HHV could be attributed to removal of moisture content and volatile matter during pyrolysis as the presence of moisture content and volatile matter had lowered HHV. It indicates that these bio-chars are having better fuel properties in terms of HHV. It can also be observed that small amount of volatile matter was still left in the bio-char and could not be removed fully during experiment. More advanced techniques could help to remove them.

Table 4: Properties of rice husk bio-char

Sr. No.	Property	Value	
		Husk char	Straw char
1.	Moisture content (%)	1.07	1.37
2.	Volatile matter (%)	10.87	12.44
3.	Ash content (%)	14.85	13.32
4.	Fixed carbon (%)	73.21	72.87
5.	Higher heating value (MJ/kg)	18.65	17.70
6.	Bulk density (kg/m ³)	214.6	186.3

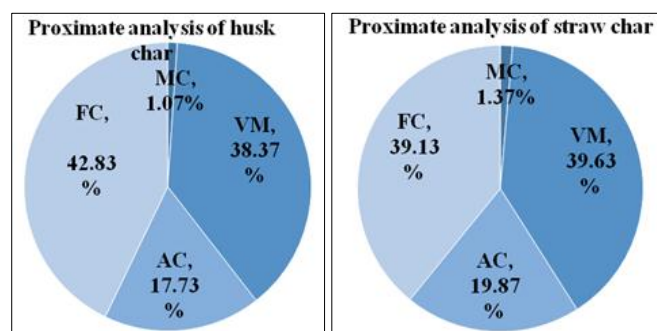


Chart 3: Proximate analysis of rice husk and rice straw bio-char

3.5.1.1 Bulk density of rice husk bio-char and rice straw bio-char

Higher bulk density is beneficial for transportation of fuel as it occupies less space. The bulk density of rice husk char is higher than that of rice straw char as the same of feedstock rice husk was also higher which has reflected in the char yield. The higher density might be due to higher percentage of lignin, cellulose, and hemicelluloses in feedstocks.

3.5.2 Characteristics of Bio-oil yields

The average values of results obtained in respect of characteristics of bio-oil obtained from rice husk and rice straw are depicted in Table 3.5.

Table 5: Properties of rice husk and straw crude bio-oil

Sr. No.	Property	Values	
		Husk bio-oil	Straw bio-oil
1.	Density @ room temperature (g/ml)	1.0842	1.0788
2.	Viscosity @ room temperature (cSt)	63.29	62.67
3.	Calorific value (MJ/Kg)	17.27	16.58
4.	Flash point (°C)	71	73
5.	Fire point (°C)	82	85

Pyro-oil from both the raw materials was found to be similar perhaps because there was not much difference in the characteristics of both the raw materials. The density of the pyro-oil is also observed to be higher than that of water and petroleum fuels such as diesel (i.e., 0.8271 g/ml), presence of significant amount of water, solids and heavy carbonic molecule contamination could be responsible for this.

Viscosity of a liquid fuel also plays an important role on flowability of the fuel. Average values of the viscosity of both the bio-oils at room temperature (28°C) were similar to the other bio-oils but higher than that of diesel (i.e. 2-4 cSt) for the similar reason as for the higher density.

Similarly, the HHV (higher calorific value) of bio-oils was observed to be 17.27 MJ/Kg and 16.58 MJ/Kg for rice husk and rice straw bio-oils respectively. Flash points and fire points help in determining the nature of a fuel's flammability. Determination of flash point and fire point of a liquid fuel is important as knowing the value of temperature in which a fuel catches fire helps in storing and transporting it accordingly. The flash points of both the bio-oils were found out to be 71°C and 73°C whereas the fire point of both bio-oils was obtained 82°C and 85°C respectively. These results are in accordance with Ullah *et al.*, (2014) and Shoaib *et al.*, (2018)^[15]. The fuel quality of rice husk and rice straw bio-oil is better in comparison to the other bio-oils, but is still very low when compared to the fossil fuels like diesel. The raw bio-oil produced in the study has many undesired (impurities) and its characteristics were investigated without any purification. Hence, upgrading of the bio-oil is necessary for better combustion and utilization before it is used as fuel. Some of upgrading techniques which can be used and had previously reviewed in researches included solvent addition, emulsification, esterification, supercritical fluids, hydrotreating, steam reforming, extraction of chemicals from bio-oil etc. Each of these techniques has its own advantages and disadvantages, which can create more research pathways for future improvement.

4. Conclusions

- The proximate analysis results of chars from the pyrolysis rice husk and rice straw show a decrease in volatile matter and an increase in ash content of product yields as compared to the respective raw materials.
- Carbon content in bio-char found from pyrolysis of rice husk (60.04 % increase) and rice straw (60.12 % increase) was more than raw materials.
- Higher calorific value in bio-char of respective raw materials was higher than the raw materials (19.70% increase in rice husk and 20.65% increase in rice straw).
- Higher calorific value in bio-oil produced from pyrolysis of rice husk (10.85% increase) and rice straw (13.02% increase) was more than raw materials.
- Bulk densities of alternative fuels were found higher than the raw materials (98.6 kg/m³ / 85% increase in rice husk and 116.38 kg/m³ / 166.45% increase in rice straw).

- To produce 1 liter of crude bio-oil, 20.548 kg of rice straw was required or 8.949 kg of rice husk was required.

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