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Research Article Lagerstroemia speciosa (Banaba) seed as a new and prospective bioenergy resource Riturani Tamuly^{1,2} and Prasenjit Saikia ^{1,2*}

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Abstract

Bio-oil, a highly valuable product obtained from biomass pyrolysis after proper up-gradation and refining, can be utilised in a variety of downstream applications which is gaining attention to the researcher from last few decades. Extraction and fractionation are two viable techniques to upgrade bio-oil those divide the complex mixture of bio-oil compounds into distinct fine chemicals and fractions enriched classes of chemical compounds. In the current investigation, a new biomass plant (*Lagerstroemia speciosa*) seed, commonly known as Banaba underwent thermo-chemical conversion at four terminal temperatures viz., 350 $^{\circ}$ C, 450 $\rm{^0C}$, 550 $\rm{^0C}$ and 650 $\rm{^0C}$ with a rate of heating 10 $\rm{^0C/min}$. The highest bio-oil yield (59.36 % out of that 41.68 % is of oil phase) was attained at pyrolysis temperature of 550 °C. The optimum condensable liquid (bio-oil) was collected and analysed. Properties of Banaba were investigated by physico-chemical, biochemical, ultimate analysis, TGA/DTG and FTIR analysis. Characterization of condensable liquid (bio-oil) was investigated by FTIR, NMR and GC-MS. Fuel property of the condensable liquid was further analysed through flash point, pour point, viscosity, Calorific value, and ultimate analysis. The Hydrogen/Carbon molar ratio (1.49) of the bio-oil was determined to be on a level with petroleum-based diesel. The oil derived from Banaba is found to have considerable fuel properties and may be substituted for commercial fuel with chemical modification or through blending with conventional diesel for utilization in IC engine which need to be addressed further. **Keywords:** Banaba, thermo-chemical conversion, char, bio-oil

1. Introduction

With a GDP of US\$ 2.87 trillion, India ranked fifth in the world in terms of economic size [1]. At 1.38 billion, it possessed the second highest population as of 2020. By 2050, with roughly 1.641 billion people, it is predicted to become the second largest economy in the world. A growing population and robust economic growth are both major factors in the rising demand for energy [2]. While coal attributes almost 44% of India's preliminary energy demand in 2020, oil and gas made up 31% of the total, primarily from imports [3]. India has initiated eight national missions to address climate changerelated issues, with the main cause being the accumulation of greenhouse gas emissions [4]. One of the missions focuses on solar energy and the upgrowth of technologies to harness it to meeting a portion of nation's total energy demand [5, 6]. Biomass has emerged as a promising renewable resource for energy production due to significant environmental issues, including pollution [7].

Organic matter created by water and carbon dioxide through photosynthesis, which transforms solar energy into biochemical energy, is referred to as biomass [8]. Data from 2017 show that, on a dry basis, the mass of biomass on Earth is 55 billion tons of carbon, of which 45 billion tons are derived

from plants [9]. Research priorities are thus shifting for the proper development of highly efficient and environment friendly technologies for producing new carbon materials from biomass [10]. Utilizing biomass-based carbon materials has the added benefit of helping to achieve long-term carbon retention and lessen the greenhouse gas problem by lowering $CO₂$ emissions [11]. The two primary process technologies for converting biomass to energy are biochemical/biological and thermochemical [12]. The pyrolysis of lignocellulosic biomass (on moderate temperature range of $350-600$ °C) is currently the most promising and sophisticated thermochemical conversion method for producing biofuel [13]. Biochar, liquid (bio-oil), and biogas are the three valuable products of this process that turns biomass [14]. Phenolic compounds, acids, alcohols, aldehydes, ketones, esters, and heterocyclic derivatives are typically present in bio-oil which can serve as an attractive biofuel for gas turbines, diesel engines, thermal power plants, and petroleum refineries [15]. Since they are unstable, corrosive, and have a high viscosity, they can be converted into lighter, more valuable hydrocarbon fuels that can be transported and stored more easily [16, 17].

To obtain this goal first important work is the selection of raw biomass materials. Thus, availability of raw materials is an important factor for the industrial growth of a country [18]. The naturally available organic wastes can be an excellent choose for this purpose, the utilization of which not only solves the disposal problem and ensures rich surpluses to the farmer, but can also provide access to a natural capital reservoir for overcoming various challenges in the development of new industries [19]. Production of biooil from energy crops and waste biomass is gaining attention to cater the future energy need. Biooil contains several oxygen containing functional groups leading to have the potentiality to polymerization due to which viscosity increases. Although there are so many unique applications of biooil, but it can be used as a low grade fuel in the boilers. It can also be used as high grade fuel after up-gradation [20, 21]. Apart from this, biooil can also be used as a base materials for production of resin, adhesives or as binder [22, 23, 24, 25] and useful

chemicals [26, 27]. Numerous studies have been made with different biomass sources like Sugarcane bagasse [28], Hornbeam shell [29], Switchgrass [30], Cherry seed [31], Rape seed [32], Oil palm residue [33], Grape bagasse [34], Almond shell [35], Corncob [36] and Palm kernel shell [37]. In these studies they performed the pyrolysis operation in a fixed bed reactor with optimum terminal temperature ranges from $400\degree$ C ${}^{0}C$ to 600 ⁰C. The optimum yield of biooil percentage were reported from 24% to 59% with high heating value (HHV) were observed from 19.1 MJ/kg to 38.4 MJ/kg.

In this study, banaba seeds are chosen as the raw material for this study.Banaba trees are found in grown in South East Asia including India. In India it is available in Assam, Kerala, Madhya Pradesh, and Maharashtra. Locally it is found throughout Assam mainly in alluvial sites and along rivers with sandy to clay soils. Banaba seeds are one of the best feedstocks for energy production as it contains high volatile matter content. More over the fixed carbon contain is very high. Also, it has low nitrogen and sulfur content and higher carbon percentage resulting in reduced emissions of sulfur and nitrogen oxides and increased biomass heating value. Also, pyrolysis of Banaba seeds has not been done yet by any researcher although it has excellent physical properties to act as a significant feedstock for bio-oil generation.

This paper represents a study on the characterization of the liquid phase (bio-oil). Moreover, compositional analysis of the raw material and characterization as well as the proper physical and chemical analysis of the aqueous oil phase are also reported.

2. Experimental Section

2.1. Sample Collection

The location for collection of biomass samples used for our study represent the bank of Kakodunga river lies in the Upper region of Assam. The sites located within two adjoining districts viz, Jorhat (latitude of 26°44'47.47"N and a longitude of 94°12'9.31"E) and Golaghat (Latitude: 26° 30' 42.01" N Longitude: 93° 57' 34.24" E).

Figure 1. Digital photograph of a) Banaba tree (*Lagerstroemia speciosa*) b) Banaba Seed, c) Banaba flower.

2.2. Sample Pr epar ation

During collection time, the biomass samples (Banaba seeds) were collected with due care from the above cited location. The preliminary weights of the samples were recorded and then air dried for 2-3 days. The larger samples were broken up and sieved through a 212 mm sieve using a Retsch AS 200. The entire samples were further micronized and homogenized by using a grinder and then dried in oven for 24 hours at 60°C. The prepared samples were kept in a desiccator for further use (Figure 2).

Figure 2. A digital photograph of the sample prepared for the study.

2.3. Pyrolysis Experiment

The pyrolysis of the biomass was carried out in a laboratory scale batch-type reactor. The reactor is associated with a furnace, PID controller, condenser and measuring cylinder [38]. The reactor containing electrical heating facility has a length of 300, thickness of 52–53 mm internal diameter. A highly sensitive PID controller using Al/Rh thermocouple was used to control the furnace temperature (with average reaction temperature errors within ± 5 °C). A glass condenser (kept at ice cooling environment) was fitted with the reactor to collect the condensable phase (oil).

At the beginning of the pyrolysis experiment, sample was poured in a silica retort (10g) and was placed in the reactor that was attached to a glass condenser (kept at ice cooling environment) and non-condensable gas measuring unit to collect the condensable phase (bio-oil) in a glass tube and non condensable phase (bio-gas) in a beaker containing water. An inert atmosphere was created within the retort using nitrogen gas during pyrolysis.The pyrolyzing experiments were performed at 350 °C, 450 °C, 550 °C, and 650 °C temperatures with a rate of heating 10 °C/min. The condensable phase collected was determined to be in heterogeneous phase with two phases (aqueous and heavy oil fractions). The two fractions were then separated using separation funnel and stored in glass bottles for further analysis. The optimum temperature where the highest bio-oil yield obtained was taken in concern for further characterization and analysis. The product yield was calculated by using the following formula: Percentage yield of liquid phase $=$ weight of liquid/weight of dry biomass \times 100%

Percentage yield of char $=$ weight of char/ weight of dry biomass \times 100%

Percentage yield of gas = weight of dry biomass - (weight of char + weight of liquid)/ weight of dry biomass \times 100%

2.4. Char acterization methodology

2.4.1. Feedstock and product char acteristics

Proximate analysis is used to determine the moisture, ash, volatile matter, and fixed carbon contents. It is done in a thermogravimetric analyser TGA-701 which performs the analysis according to ASTM D3173, ASTM D3175 and 3. ASTM D3174 for determination of moisture content, volatile matter and ash content in an inert atmosphere Proximate analyser sample was performed on dry basis and fixed carbon was determined by the difference.

Ultimate analysis is determined using CHN analyser (Perkin Elmer, 2400 Series-II). The oxygen content was determined by the difference. It is basically used to determine the carbon, hydrogen, sulfur, nitrogen, and oxygen content of the samples. Biochemical analysis was done with the help of gravimetric method [39]. Flash point, pour point and Calorific values were determined according to ASTM D92-05, ASTM D9705 and using Bomb Calorimeter (LECO, AC 350 LECO model) 80 respectively. The iodine number of the activated carbon was $\frac{70}{6}$. determined by using Gimba and Musa method. Acid value was Determined as the number of milligrams of KOH required to

neutralize the free acid in 1 gram of the oil.

All the analysis carried out above has been done in triplicate
 $\frac{26}{5}$ 40
 $\frac{36}{5}$ $\frac{36}{5}$ $\frac{36}{5}$ $\$ neutralize the free acid in 1 gram of the oil.

neutralize the free acid in 1 gram of the oil.

All the analysis carried out above has been done in triplicate $\frac{25}{5}$ 40 $\frac{3}{5}$ 40 $\frac{3}{5}$ and the mean value has been reported.

Thermogravimetric analysis (TGA) is performed either in an inert nitrogen environment or in an oxidative environment (air). Thermogravimetric analysis (TGA) is performed either in an and analysis (TGA) is performed either in an analysis (TGA) and the thermal behaviour of the biomass TGA and a set of the biomass TGA analysis has been carried out to see its degradation profile
Biochemical analysis Proximate analysis considering the terminal temperature of 700 °C.

Functional group was determined by Fourier-transform **Characterization of Banaba** infrared spectroscopy (FTIR) using FTIR-system-2000, Perkin Elmer, USA in the range of $400-4000$ cm-1 with a resolution of 4 cm-1.

Nuclear magnetic resonance spectroscopy (NMR) value was carried out by Advanced DPX-300 MHz, FT-NMR. Rheology was performed according to ASTM D445-04 at 40 °C and 100 \degree C in a MCR 500. Gas chromatography–mass spectrometry (GC–MS) was performed in Perkin Elmer Clarus 80 GC/ 600C MS Gas Chromatography (GC), and the compounds were determined using mass selectivity (MS) detector equipped with HP-5MS column (30.0 m \times 0.25 mm \times 0.25 μm) based on GC retention time. The mass spectra were computer matched with those of standards available in mass spectrum libraries.

3. Results and discussion

3.1. Feedstock char acterization

The physico-chemical and bio-chemical analysis of the raw materials was done to optimize the conditions to carry out the thermo-chemical conversion process. Bio-chemical analysis (Figure. 3) of the raw material is very important as it gives an overview of the composition and extent of transformation to form a bio-oil mixture as a pyrolysis product. Bio-chemical analysis has been done gravimetrically in Fibre Tech equipment resulting high cellulose and hemicellulose content showing the potentiality on high yield of bio-oil production.

The inherent moisture content of the biomass has a notable effect on its conversion efficiency and calorific value. The proximate analysis (Figure 3) shows the raw material contains minimum amount of moisture content and thus it will not decompose easily and there will be less energy loss during storage.

Figure 4. TG and DTG analysis of (a) Banaba seeds (b) oil phase.

Figure 5. (a) FT-IR analysis of the pyrolysis liquid derived from Banaba-seeds, (b) Comparison of the FTIR spectra of raw Banaba seed and Bio-oil

Ultimate analysis (Figure 3) of the raw material shows that oxygen content in the oil phase is high as the biomass is composed of cellulose, hemicellulose and lignin, the oil derived during pyrolysis will contain large percentage of oxygenates [40]. Carbon content is much higher than hydrogen. In the IR absorption spectrum of Banaba seeds, the presence of free and intermolecular bonded hydroxyl group peak is confirmed by the peak around 3400 cm−1 [41]. For the stretching vibration of the C–H group the peaks are observed at 2850 cm⁻¹ [42]. The C-C stretching vibration at ~1650 cm⁻¹

are due to the contribution of lignin aromatic groups [43] The strong C-O band at 1100 cm^{-1} is due to the presence of carbonyl groups connected to the lignin structure in Banaba. The C-H bending modes of aromatic compounds are established by the peaks at 600 cm^{-1} (Figure 4).

In thermogravimetric (TG) analysis, significant values were found for the constitution of cellulose, hemicellulose, and lignin fractions in biomass. In the TG curve (Figure 4) we have obtained the first stage of weight loss below 100 °C which is due to release of light volatile matters (like water).

DTG peak at 48 °C shows a slight weight loss of \sim 13%. The second stage of weight loss is called an active pyrolysis zone which is due to the degradation of the intermolecular, other weaker chemical bonds and the side aliphatic chains and some small gaseous molecules may form at this temperature.

Figure 6. ¹H NMR analysis of the pyrolysis liquid derived
modused from B from Banaba-seeds (in ppm).

Figure 7. ¹³C NMR analysis of the pyrolysis liquid derived from Banaba-seeds.

The second stage of weight loss of about 56% has been observed between 200 and 500 °C (DTG peak at 319 °C) represents the maximum degradation peak. In this stage decomposition of hemicellulose followed by the cellulose takes place. From about 500 °C, the degradation rate become almost constant. In the third stage the weight of the biomass becomes less than half of the original weight as about 70-75% of biomass consist of volatile matters and 20-25% of contains only carbon [44]. The highest temperature needed to degrade lignin which takes place slowly over a wide temperature range 160-900°C [45]. Above 600°C temperature due to the decomposition of lignin, bigger molecules break to smaller

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molecules with the breaking of chemical bonds. Considering the TG and DTG analysis, the temperature range between 350 °C and 650 °C were chosen to estimate the optimum bio oil yield.

3.2. Pyrolysis of Banaba seed

From the pyrolysis experiment as described in the section 2.1, the maximum bio-oil yield(59.36%) was found at the terminal temperature of 550 ⁰C. The 59.36% which includes both aqueous and oil phase of the condensable liquid. Out of 59.36%, oil phase contributes 41.68% and the remaining 16.68% contributed by the aqueous phase (Supporting documents). The oil phase thus produced at the terminal temperature 550 ⁰C has been collected and studied has been carried out for further analysis, characterization, and fuel property evaluation. From the TGA analysis of the biooil produced from Banaba seed shows an initial weight loss of 18% at around 100 ⁰C due to moisture removal from the oil. Further weight loss (73%) was obtained from 200 $\rm{^0C}$ to 400 $\rm{^0C}$ ${}^{0}C$ and then from 500 ⁰C onwards the rate of weight loss was observed almost constant.

3.3. Characterization of the oil phase derived through pyrolysis

Figure 5a, and 5b represents the FT-IR analysis of the Banaba seed and pyrolysis liquid that describes the structure and functional groups present in the molecules [46] (Details in the Supplementary file Table S1). In the IR absorption spectrum of banaba seeds, the presence of free and intermolecular bonded hydroxyl groups peak is confirmed by the peak around 3400 cm⁻¹ [38]. For the stretching vibration of the C–H group the peaks are observed at 2850 cm−1 . The C-C stretching vibration at \sim 1650 cm⁻¹ are due to the contribution of lignin aromatic groups. The strong C-O band at 1100 cm−1 is due to the presence of connected to the lignin structure in banaba [47, 48, 49]. The presence of ketone, quinone and aldehyde groups etc. are confirmed by the presence of $C=O$ stretching vibration at \sim 1710 cm⁻¹. FT-IT also confirms the presence of alkenes and aromatics in the oil from the absorbance peak at $~1650$ cm−1 for C=C stretching vibrations. The peaks at ~1225-980 cm−1 describes the presence of primary, secondary and tertiary

alcohols, phenols, esters and ethers in the oil [38]. The absorption peaks between 770 and 950 cm−1 are due to the presence of mono- and polycyclic and substituted aromatic groups. The C-H bending modes of aromatic compounds are confirmed by the peaks at 600 cm^{-1} (Figure 5b). The highly oxygenated functional groups make the oil little acidic as well as low in calorific value.

40 °C and 100 °C.

Figure 9. Change in viscosity with constant shear rate at 40 °C and 100 °C temperatures.

The $\rm{^1H}$ peaks in the up-field region (Figure 6), from 0.5 to As shown in the $\rm{^{13}C}$ s 1.05 ppm, are due to the presence of aliphatic protons attached to beta or gama carbon atoms linked to C=C bond or

heteroatom (especially O or N). The region from 1.5 to 3.0 ppm corresponds to the protons linked to a aliphatic carbon bonded to a C=C double bond of an aromatic or olefinic compound [50]. The peaks between 4.3 and 5.0 ppm are due to the aromatic ether protons (i.e., lignin derived methoxyphenols) and the protons linked to carbohydrates [51]. The region 6.0-8 ppm the NMR spectrum corresponds to the aromatic protons present in the oil. These protons may be connected to benzenoids heteroaromatics containing O and N. Some heteroaromatic also resonates above 8.5 ppm. The presence of aldehydic, carboxylic acid protons are confirmed by the peaks in the downfield region 9.3-10 ppm (Supplementary file, Table S2).

Figure 10. Change of viscosity with temperature.

Figure 11. GC-MS analysis of the oil fraction.

As shown in the 13 C spectrum (Figure 7), carbons for short aliphatic chain $(-CH_3, -CH_2CH_3, -CH(CH_3)_2, -C(CH_3),$ etc.) resonates in the up-field region (0-25 ppm). Chemical shift

value for carbon attached to long, branched chain, methoxyphenols, paraffinic methylene (CH2), allylic or benzylic carbons, cyclic carbons are found below or near ∼55 ppm. The carbon atoms linked to a heteroatom (O) atom in carbohydrates, ethers, alcohols resonate between 55 and 80 ppm. The presence of aromatics due to lignin and olefins are confirmed by the peaks shown in the range of 100-155 ppm. Carbonyl carbon atoms of acid and ester in the 155-180 ppm range and that of ketone and aldehyde are in more downfield region, >180 ppm. Thus, NMR analysis showed that the oil contains both organic and aqueous phase containing the functional groups (Supplementary file Table S3). From the fuel properties of the oil, it has been found that the flash point and the iodine value can be compared to petroleum-based fuels. Due to the presence of large branched components and oxygenated hydrocarbons flash point of the bio-oil is high [52]. Due to the same reason acid value is also high which is a drawback of the oil for storage and handling. The average chemical composition of the oil was found to be $CH_{0.5}O_{0.5}N_{0.03}$. The nitrogen content is also found to be low and thus the oil derived from Banaba can be contemplate as an alternative for commercial utilization although some chemical modification is necessary for the oxygenated hydrocarbon components present [53].

Table 1. GC-MS analysis of the oil derived from banaba seeds.

Retention time	Compounds detected	Chemical formula
4.185	Phenol, 2-methoxy-	$C_7H_8O_2$
5.451	Phenol, 5-methoxy-5-methyl-	$C_8H_{10}O_2$
6.546	Phenol, 4-ethyl-2-methoxy-	$C_9H_{12}O_2$
7.060	Ethanone, 1-(2-hydroxy-5-methylphenyl-)	$C_9H_{10}O_2$
7.574	Phenol, 4-allyl-2-methoxy-	$C_{10}H_{12}O_2$
8.742	Phenol, 2-methoxy-4-propenyl-, (E) -	$C_{10}H_{12}O_2$
9.875	Methyl vanillyl ketone	$C_{10}H_{12}O_3$
11.716	Phenol, 4-allyl-2,6-dimethoxy-	$C_{11}H_{14}O_3$
12.593	2-pentanone-1-(2,4,6-trihydroxyphenyl)	$C_{11}H_{14}O_4$
20.481	Thiocyanic acid, 6,6'-diamino-3,3'-biphenylene ester	$C_{18}H_{18}O_4$

Table 2. Fuel properties of the biooil and its comparison with conventional diesel

Flow behaviour was carried out by exposing the sample to a constant shear stress by taking a cone-and-plate geometry. The flow properties were analysed from the strain produced in the process. The applied shear stress was the controlled parameter throughout the process.The stress measured in different steps were measured and figured against the constant shear rate (Figure 8). Also, with the help of a temperature control unit the viscosity change of the sample with change in temperature was studied. The curves obtained from the plotting of shear stress against constant shear rate are non-linear. Thus, the oil sample shows non-Newtonian behaviour (Figure 8). As a result, the flow behaviour of the oil changes with applied stress. The sample will behave either as a thicker fluid called shear thickening or like a thin runnier fluid called shear thinning. If we apply stress to it and when the stress is removed it will come to its original state (Figure 9). When temperature rises the viscosity of the oil decreases (Figure 10). In this case the sample shows unique flow behaviour under different stress and $\frac{4}{1}$ temperature. From Figure 9 and Figure 10, it has been evident that the biooil thus produced has shear thinning behaviour.

The bio-oil is further analysed with GC-MS (Figure 11). This study of the undivided bio-oil gives us the idea about the most probable compounds present and using the standard library (NIST) GC-MS analysis proves that the oil is composed complex mixture of organic compounds [54]. From the analysis it has been found that the pyrolysis oil fraction mainly consisting of branched/long chains, phenol and its derivatives, mixed acids, esters, ketones, and various aromatics (Table 1). The oxygenated and aromatic compounds that found present in the oil is due to the hemicellulose and cellulose texture of the parent material [38]. Presence of oxygenated compounds makes the oil more susceptible towards polymerization leading to increase its viscosity further. Moreover, it also enhances the corrosive nature of the oil. Therefore, to reduce the oxygenate structure, the oil required to be up-graded into lower oxygenated compounds so that it can be able to use as a potential liquid fuel.

Since, the oil fraction consist of oxygenates and aromatic compounds, the calorific value of the oil (26.1 MJ/kg) will be comparatively less than the conventional diesel (44 $MJ/kg - 46MJ/kg$). Although the flash point (92) and iodine value (138) are comparable with commercial diesel (∼55 to 80 and 80 to135 respectively) but due to the oxygenated compounds pour point (7) and acid value (95) of the oil is significantly in the higher side than diesel (−40 to −30 and 0 respectively). Thus, prior to commercial use, these components need to be removed first and upgraded to modify their physical properties (Table 2). The results are in the same line even more in case of yield and HHV for most of the current studies on biomass such as Sugarcane bagasse, Hornbeam shell, Switchgrass, Cherry seed, Rape seed, Oil palm residue, Grape bagasse, Almond shell, Corncob and Palm kernel shell as described by Gupta et al. [21].

4. Conclusion

In the present study a new biomass seed from banaba plant has been considered for biooil production. The raw material contains high cellulose and hemicellulose content with high volatile matter and high carbon content. The optimum temperature for biooil production during pyrolysis was found to be at 550 \degree C. The pyrolysis oil phase was characterized by FT-IR, NMR and GC-MS analysis which showed the presence of oxygenated hydrocarbons as found by the other researchers [55]. The fuel properties of the biooil has been evaluated and compared with the conventional diesel. The flash point and pour point of the oil phase was found to be 92°C and 7°C respectively, out of which flash point is comparable with the conventional commercial diesel. The oil has the value of unsaturation, i.e., iodine value of 103, which is also comparable with commercial fuel. It has a high acid value of 95 as the structure contains various oxygenates. Hence, modification of the structure is necessary. The flow behaviour of the oil was studied at different temperatures and shows that the oil behaves both as shear thinner and shear thickener liquid

at low and high temperatures respectively. Hence the oil 2. derived from banaba may be assumed to have the qualities to be used conventional commercial fuel through blending to run a IC engine for which further study can be carried out. The study also reveals that the Banaba seed can be a prospective feedstock for production of bio-oil in the biorefinery approach. The finding of this study will certainly assist the researcher to introspect this banaba resources for harnessing new energy products in near future.

Authors Contribution

Prasenjit Saikia (PS) conceived the presented research idea, $\frac{4}{3}$ supervised the research activities, verification of the data, critical analysis and wrote the manuscript. Riturani Tamuly (RT) have carried out the experiments, optimization studies, characterization and manuscript preparation.

Conflicts of Interest

There is no conflict of interest between the authors.

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Data Availability statement

The data presented in this study are available on request from the corresponding author.

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