TAMBAHAN JURNAL – 1

Judul: New Composites based on low-density polyethylene and rice husk: Elemental and thermal characteristics

PROSES REVIEW

JOOE-D-17-00392 - Submission Notification to co-author

Dari: Journal of Polymers and the Environment (JOOE) (em@editorialmanager.com)

Kepada: m.anshar50@yahoo.com

Tanggal: Senin, 3 Juli 2017 22.51 GMT+8

Re: "New composites Based on Low-Density Polyethylene and Rice Husk: Elemental and Thermal characteristics"

Full author list: Muhammad Anshar; Dahlang Tahir; Makhrami Makhram; Fand Nasir Anis; Ab Saman Kader

Dear Dr. Anshar,

We have received the submission entitled: "New composites Based on Low-Density Polyethylene and Rice Husk: Elemental and Thermal characteristics" for possible publication in Journal of Polymers and the Environment, and you are listed as one of the co-authors.

The manuscript has been submitted to the journal by Dr. Prof. Dahlang Tahir who will be able to track the status of the paper through his/her login.

If you have any objections, please contact the editorial office as soon as possible. If we do not hear back from you, we will assume you agree with your co-authorship.

Thank you very much.

With kind regards,

Springer Journals Editorial Office
Journal of Polymers and the Environment
10-Jul-2017

Dear Dr. Anshar:

A manuscript titled New composites Based on Low-Density Polyethylene and Rice Husk- Elemental and Thermal characteristics (JRP.17.0465) has been submitted by Dr. Muhammad Anshar to Journal of Reinforced Plastics and Composites.

You are listed as a co-author for this manuscript. The online peer-review system, ScholarOne Manuscripts, automatically creates a user account for you and you may receive e-mails and communications from the journal editorial office staff regarding your manuscript based on the authorization you provided to the co-author who created the account.

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Thank you for your participation.

Sincerely,
Journal of Reinforced Plastics and Composites Editorial Office

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Dear Prof. Tahir,

Environmental Engineering Research has received your revised submission. You may check the status of your manuscript by logging onto Editorial Manager at [http://eer.edmrg.com/](http://eer.edmrg.com/).

Kind regards,
Fwd: Your Submission

Dari: dtahir@fmipa.unhas.ac.id (dtahir@fmipa.unhas.ac.id)
Kepada: m.anshar60@yahoo.com
Tanggal: Jumat, 15 Desember 2017 08.14 GMT+8

-------- Pesan Asli --------
Judul: Author Reminder Inv. Manuscript - Before Due Date
Tanggal: 2017-09-30 22:30
Pengirim: "EER" <em@editorialmanager.com>
Penerima: "Dahlang Tahir" <dtahir@fmipa.unhas.ac.id>
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Environmental Engineering Research
Ref.: Proposal No. EER-D-17-00096
New composites Based on Low-Density Polyethylene and Rice Husk-
Elemental and Thermal characteristics

Dear Prof. Tahir,

Just as a reminder, your submission of proposal number EER-D-17-00096 is
due by %INVITED_SUBMISSION_DUE_DATE%.
You can visit our website at http://EER.edmgr.com/.
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To maintain publication deadlines we would be grateful if you would
submit your manuscript as soon as possible.

With kind regards,

Environmental Engineering Research
Dear Prof. Tahir,

Reviewers have now commented on your paper. You will see that they are advising that you revise your manuscript, particularly regarding the English language. I also recommend it for final polishing of your work as a well-written scientific article. If you cannot get such an editorial service from your side, the journal office can help it, but on your own expense. Please let us know. If you are prepared to undertake the work required, I would be pleased to reconsider my decision.

For your guidance, reviewers' comments are appended below.

If you decide to revise the work, please submit a list of changes or a rebuttal against each point which is being raised when you submit the revised manuscript.

In addition, please make sure that your manuscript is well prepared in rendering the English language writing clear and consistently intelligible.

**Your revision is due by Jan 10, 2018.**

To submit a revision, go to [http://eer.edmgr.com/](http://eer.edmgr.com/) and log in as an Author. You will see a menu item call Submission Needing Revision. You will find your submission record there.
Reviewers' comments:

The technical quality of the manuscript appears to be acceptable for publication in the journal. However, its English language should be improved by a native speaker in order to avoid typo and grammatical errors and thereby, to make it constantly intelligible for readers.
Dear Prof. Tahir,

I am pleased to tell you that your work has now been accepted for publication in Environmental Engineering Research.

It was accepted on Feb 06, 2018.

Comments from the Editor and Reviewers can be found below if any.

Please sign and return the Copyright Transfer Agreement to the editorial office within 48 hours. The form is available at the Journal website: [http://eeer.org/authors/authors.php](http://eeer.org/authors/authors.php)

Your accepted manuscript will be published online after the copyright agreement is received by the editorial office. A galley proof will be sent to you as soon as it is prepared. Please contact the editorial office if you have any question on publication of your work.

Thank you for submitting your work to this journal.

With kind regards

Editorial Office
Environmental Engineering Research

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HASIL SIMILARITY CHECK:
Introduction: Clean and efficient utilization of biomass as a source of alternative energy is of major concern and is essential for generating electricity, especially in developing countries. Indonesia, as a developing country, is still lacking in electrical energy supply, with electrification of around 76.56% [1]. About 23.5% of rural communities have not yet used electrical energy, due to the inability of power plants to meet the increasing power needs of the society and industry every year. More than half of the electricity suppliers in Indonesia generate their electricity from coal [2].
# New composites Based on Low-Density Polyethylene and Rice Husk- Elemental and Thermal characteristics

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<tr>
<td>Keywords:</td>
<td>low density polyethylene; plastic bags; rice husk; elemental composition; thermal characteristic.</td>
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<td>Corresponding Author:</td>
<td>Dahlang Tahir</td>
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## Abstract:
We developed new composites by combining the solid waste from Low-Density Polyethylene in the form of plastic bag (PB) and biomass from rice husk (RH) in form of \((\text{RH})x(\text{PB})^{1-x}\ (x=1, 0.9, 0.7, 0.5)\) as alternative fuels for electrical energy sources and best solution to reduced pollutions in the environments. Elemental compositions were obtained by using proximate analysis, ultimate analysis, and X-ray fluorescence (XRF) spectroscopy, and for the thermal characteristics were obtained from thermogravimetric analysis. The composition of carbon and hydrogen from the ultimate analysis shows increases significantly 20-30% with increasing PB in composite. The activation energy for RH is 101.22 kJ/mol, increase by 4 and 6 magnitudes for \(x\) is 0.9 and 0.7, respectively, and remarkable increase to 165.30 kJ/mol for \(x = 0.5\). The range of temperature about 480 to 660°C for combustion of composites \((\text{RH})x(\text{PB})^{1-x}\ (x=1, 0.9, 0.7, 0.5)\) to perform the complete combustion process and produce high energy. In addition, the calorific value (CV) was determined by using bomb calorimetric shows value for RH is 13.44 MJ/kg and increases about 30-40% with increasing PB content, indicated that PB has strong effect to increasing the energy realized to generate electricity.

## Response to Reviewers:
Kwang-Ho Choo, Ph.D. (Editor-in-Chief EER)

Thank you for providing us constructive referee comments on our manuscript Ref No. EER-D-17-00096 and title “New composites Based on Low-Density Polyethylene and Rice Husk- Elemental and Thermal characteristics”

Here we respond the Editor Request and the referee comments:
Reviewer #1: The authors developed an alternative fuels low-density polyethylene plastic bag and rice husk. Limited analyses of proximate, ultimate, X-ray fluorescence and thermogravimetric analyses were determined to characterize the fuels. This research is crucial for development of alternative fuels. The topic is suitable for this journal; however some modifications should be made prior to acceptance.

1. Please recheck the manuscript and modify for efficient and effective sentences.

Thank you for good suggestion of the referee. We have change and improved our revised manuscript.

2. Why RH have been selected, not other biomass products, what is the important chemical properties of RH for your selection?

Thank you for good comment of the referee. In introduction part, we explain as follows: The potential of rice husk (RH) in 2012 was around 1.381×10⁷ tons, equivalent to the electrical energy potential of around 53.702 GWh, the third-largest in the world after China and India [4, 5]. Nonetheless, this potential has not been exploited as a source of energy; only wasted and causing pollutions to the environments. RH has a high carbon and oxygen composition which are fundamental for searching the sources of energy.

3. In the present work waste LDPE and RH were degraded without any catalysts, please also compare your results with other references (using catalysts).

Thank you for good suggestion of the referee. We have compared calorific value with previous reported in Result and Discussion part as follows: For RH shows CV is about 13.44 MJ/kg which is very good agreement with Ref. [34] about 13.30 MJ/kg and Ref. [35] about 13.50 MJ/kg and for PB is about 41.21 MJ/kg in this study also comparable with the previous studies [40-42].

4. The authors should compare their new composite fuels with standard acceptable fuels

We have compared with characteristic calorific value of the fuel standard (ethanol and methanol) in our revised manuscript.

5. What is the optimum conditions for preparation of this fuels?

For the composites (RH)₁(PB)₁₋ₓ (x= 1, 0.9, 0.7, 0.5) combustion as fuel requires range temperature about 480 to 660°C to perform the complete combustion process to produce high energy.

6. L-40 state that by mixture RH and PB to form composites will have a good characteristic element for release energy, could you please to mention the reasons?

Thank you for good question of the referee. We can see from the activation energy for composite in Table 4. Clearly the activation energy of composite was increase with increasing the amount of PB.

Reviewer #2: The manuscript needs major revision.

1. In the abstract, line22, delete "have" after We.

We have deleted in our revised manuscript.

2. In the abstract, line28, how did authors prove their method is the best solution? It is necessary to indicate economically, technically and environmentally.

In this study, we have used direct combustions; time to production is short, easy, and low air pollution as reported in some reference in this study.
3-When Indonesia has huge amount of rice husk, why do not produce pellets which are environmentally low air pollution.

Thank you for good questions of the referee. In this study we have used direct combustions which is easy and effective due to direct conversion (short time to production) compared with producing pellets.

4-It is vital to add literature review in introduction section which supports your study.

We added some reference [5,7,8] in introduction part.

5-Page 3, line 42, delete "have" after "we".

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6-The amount of ash is one of important factor in bioenergy. The amount of the ashes in your study mostly above 10%(Table 1). What do you manage this amounts of ash in industrial factories? Is it possible to use for residential? If yes, what are the amount of air pollution? Air pollution which come from your new fuel does it comply your local standard?

Thank you for good question of the referee. We focus our attention in this study to determining characteristic energy based on data from calorific value, proximate analysis, ultimate analysis and activation energy. In this time we do not consider yet for the possibility in the industrial factory.

7-Page 4 line 35, define TGA and DTG.

We have defined in our revised manuscript

8-Page 6, line 44, change the phrase "can be seen" to other phrase.

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10-Page 7, How did you prove your experimental results have a good agreement with work of Majunder et al. [39]. what was your method to indicate your study has a good agreement with work of Mjunder et al.[39].

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11-Page 8, line 57, you mentioned element of composition which you developed. However, in Table 2, you stated element in ash. Please explain about it.

Thank you for good correction of the referee. The correct is element of ash. We changed in our revised manuscript.

12-Did you consider air pollution occur from your composition which you developed?

Thank you for good questions of the referee. This study is preliminary study; we will consider air pollution for next research.

### Additional Information:

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<th>Question</th>
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Choo, Kwang-Ho,
Editor in Chief of Environmental Engineering Research

Dear Prof. Kwang-Ho Choo,

I am now sending our manuscripts entitled “New composites Based on Low-Density Polyethylene and Rice Husk- Elemental and Thermal characteristics” to be published to Environmental Engineering Research.

We hope this manuscript suitable for publication processes.

Please address any correspondence concerning the paper to me at the below address.

Sincerely Yours,

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Professor at Department of Physics,
Hasanuddin University,
Makassar, 90245, Indonesia
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New composites Based on Low–Density Polyethylene and Rice Husk-
Elemental and Thermal characteristics

Muhammad Anshar¹, Dahlang Tahir²†, Makhrani², Farid Nasir Ani³, Ab
Saman Kader⁴

¹Department of Mechanical Engineering, State Polytechnic of Ujung Pandang, Makassar,
90245 Indonesia
²Department of Physics, Hasanuddin University, Makassar 90245 Indonesia
³Department of Thermodynamics and Fluid Mechanics, Universiti Teknologi Malaysia,
Skudai-Johor, 81310 Malaysia
⁴Marine Technology Centre, Universiti Teknologi Malaysia, Skudai-Johor 81310, Malaysia

†Corresponding author: Email address: dtahir@fmipa.unhas.ac.id
Tel: +62-853-9418-8647, Fax: +62-411-588551

ABSTRACT: We developed new composites by combining the solid waste from Low-Density
Polyethylene in the form of plastic bag (PB) and biomass from rice husk (RH) in form of
(RH)ₓ(PB)₁₋ₓ (x=1, 0.9, 0.7, 0.5) as alternative fuels for electrical energy sources and best
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using proximate analysis, ultimate analysis, and X-ray fluorescence (XRF) spectroscopy, and for
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of carbon and hydrogen from the ultimate analysis shows increases significantly 20-30% with
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RH is 13.44 MJ/kg and increases about 30-40% with increasing PB content, indicated that PB
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Keywords: low density polyethylene; plastic bags; rice husk; elemental composition; thermal
characteristic.
1. Introduction

Clean and efficient utilization of biomass as a source of alternative energy is a major concern and essential for generating electricity, especially in developing countries. Indonesia as developing countries is still lacking for the electrical energy supplies with electrification around 76.56% [1]. It’s about 23.5% of the rural communities not used yet electrical energy due to the inability of power plants to meet the increasing power needs of the society and industries every year. More than half of the electricity suppliers in Indonesia are generated from coal [2]. Coal production in Indonesia is quite large, around $2.751 \times 10^8$ tons in 2010 [3]. However, continuous mining and combusting coal as fuels can damage the natural environment. On the other hand, Indonesia has abundant potential biomass resources that are environmentally friendly. The potential of rice husk (RH) in 2012 was around $1.381 \times 10^7$ tons, equivalent to the electrical energy potential of around 53.702 GWh, the third-largest in the world after China and India [4, 5]. Nonetheless, this potential has not been exploited and few reported as a source of energy [6]; only used as catalyst [7, 8], or wasted and causing pollutions to the environments. RH has been used as fuels for power plants to generate electricity in Thailand [9] and India [10]. RH has a high carbon and oxygen composition which are fundamental for searching sources of energy [9,10]. Generating electricity from biomass provides significant benefits to the environment since it produces clean energy [11].

In addition to the electrical energy shortage, Indonesia also has problems in handling the municipal solid waste (MSW), particularly the low-density polyethylene (LDPE) type plastic bags (PB). The averages household solid waste generated in Jakarta and other big city in Indonesia are 2416.2 kg per household per year with PB (14%) is the second largest and increase every year about 2-5% [12,13]. PB at landfills may take tens to hundreds years to be completely decomposed and subsequently, those particles could contaminate the surrounding
area of soil and water [14]. In fact, PB has calorific value (CV) about 41.5 MJ/kg [15]. PB combustion in the grate bed combustor can be controlled by using exhaust emission control device to minimize harmful exhaust gasses [16-18] and also used as fuel for power plants to generate electric power about 8.9 MW in Malaysia [18-22].

Based on the fuel needed for generating electricity, potentials of solid waste and biomass, in this study, we developed new composites by combination of solid waste and biomass in form of (RH)x(PB)1-x (x=1, 0.9, 0.7, 0.5) as alternative fuels for electrical energy sources and on the other hand giving best alternative solution to reduced pollutions in the environments. We focus in this study to discuss the effect of PB in elemental composition by using proximate analysis, ultimate analysis, and XRF and the thermal characteristics including activation energy by using thermogravimetric analysis.

2. Materials and Methods

2.1. Materials Preparation

The samples in this study are composites (RH)x(PB)1-x (x=1, 0.9, 0.7, 0.5) from rice husk (RH) and plastic bags (PB) as the alternative energy sources. We chose PB type low-density polyethylene (LDPE) by identified from the logo triangle figure with in the middle sign LDPE which is usually used for shower curtains, packaging, films, and clamshell. RH and PB were cutting and grinding to reduce size using a mill after sun dried. Sieving raw materials were done to obtain uniformity of size between 0.125 and 0.3 mm (for detail see Ref. [4, 19]). The physical mixing of samples was performed by weighting each raw samples to form composites (RH)x(PB)1-x (x=1, 0.9, 0.7, 0.5) by using a digital balance. The mass composite samples were fixed about 10 grams. The samples were mixed by stirred for 15 minutes to obtain uniform and homogeneous composites. The samples were heated at the
temperature 105°C for one hour and seven hours for PB and RH, respectively (for detail see Ref. [20]), and for composites were the weighted average time of heating of RH and PB.

2.2. Proximate analysis, ultimate analysis, and calorific value

The chemical composition of composites \((\text{RH})_{x}(\text{PB})_{1-x}\) \((x=1, 0.9, 0.7, 0.5)\) determined from the proximate analysis, ultimate analysis, X-ray fluorescence (XRF) spectroscopy. The proximate and the ultimate analysis were conducted based on the Standard Methods ASTM D 3172 - D 3174, ASTM D 4239, ASTM D 5373, and ISO 565. Meanwhile, the calorific value (CV) was carried out by using bomb calorimeter according to the Standard Methods ASTM D.5865 (for detail see Ref. [23, 24]).

2.3. Oxidative and kinetic study

The oxidative study of the \((\text{RH})_{x}(\text{PB})_{1-x}\) \((x=1, 0.9, 0.7, 0.5)\) was conducted to determine the characteristics of the kinetic parameter based on the some previous studies [25-29] by using Thermal gravimetric analysis (TGA) and differential thermal gravimetric (DTG). The oxidative reaction was carried out from temperature 30°C to 800°C for about 82 min, a heating rate about 10°C/min, and flow rate air about 50 mL/min. The \((\text{RH})_{x}(\text{PB})_{1-x}\) \((x=1, 0.9, 0.7, 0.5)\) samples were performed on the Mettler test with a mass test about 5.5 - 5.8 mg. The characteristics of \((\text{RH})_{x}(\text{PB})_{1-x}\) \((x=1, 0.9, 0.7, 0.5)\) from TGA and DTG were analyzed by using the Sma4Win software. A two-stage reaction kinetic scheme has been proposed for the thermal degradation (TD) of composites under an oxidative atmosphere [28], representing the DTG curves:

The first stage: \[ \text{A (solid) } \rightarrow \text{B (char) } + \text{C1 (gas)} \] (5)

The second stage: \[ \text{B (char) } \rightarrow \text{C2 (gas) } + \text{D (ash)} \] (6)
The kinetic parameters were calculated from the linear regression curve with the correlation factors (above 0.90) based on the Arrhenius law. The activation energy (Ea) that occurs at every TD process [30-32] by use the following equations:

\[ k = Ae^{(-Ea/RT)} \]  

(7)

where \( k \) is the reaction rate, \( Ea \) is the activation energy for the reaction (J/mol), \( A \) is the pre-exponential factor (1/min), \( R \) is the universal gas constant (8.314 J/K mol), and \( T \) is the absolute temperature (K). The equation can be written in a logarithmic form:

\[ \ln k = \ln A - \left( \frac{Ea}{RT} \right) \ln e \]  

(8)

Where \( \ln e = 1 \)

\[ \ln k = - \frac{Ea}{R} \times \frac{1}{T} + \ln A \]  

(9)

Based on previous studies [30, 33, 34] equations (9) were obtained:

\[ \ln k = - \ln \left[ \frac{\ln(1 - \alpha)}{T^2} \right] \quad \text{and} \quad \ln A = \ln \frac{AR}{\beta E} \]

Thus, Eq. (9) can be written as:

\[ - \ln \left[ \frac{\ln(1 - \alpha)}{T^2} \right] = \frac{Ea}{R} \times \frac{1}{T} + \ln \frac{AR}{\beta E} \]  

(10)

Where \( \beta \) is the heating rate (K/min), the mass loss fraction \( \alpha \) (mg) for every stage of the thermal degradation is defined as:

\[ \alpha = \frac{Mi - Ma}{Mi - Mf} \]  

(11)

Mi is the initial mass of the sample at the beginning reaction (mg), Ma is the actual mass of the sample (mg), and Mf is the final mass or the mass after oxidative of the sample (mg). The \( Ea \) may then be extracted from a plot of \( \ln k \) vs. \( 1/T \), which should be linear. Eq. (9) can be analogous to the equation straight line, which is often symbolized by the linear regression
equation \( y = mx + b \). Thus, \( -ln\left(\frac{\ln(1 - \alpha)}{T^2}\right) \) as the y-axis, \( \frac{1}{T} \) as the x-axis, and \( b = \ln \left( \frac{AR}{\beta E} \right) \) as the intercept of the line with the y-axis, \( m = \frac{Ea}{R} \) as slope. The Ea is obtained for every step of degradation (mass loss) [30, 34, 35].

3. Results and Discussion

The results of proximate and ultimate analyses of \((RH)_x(PB)_{1-x}\) \((x=1, 0.9, 0.7, 0.5)\) are presented in Table 1. For RH, it shows good agreement with previous results [35] for elemental composition (FC, VM, A, C, H, and O) with differences < 10%. We compared also with several reported in the previous studies [37-39] for the value of moisture content (MC), volatile matter (VM), fixed carbon (FC), nitrogen (N), sulphur (S), oxygen (O), hydrogen (H), and ash content (A) were the values are similar. This shows that the parameters of the test results and methodology used in this study good accuracy with an acceptable value. The most important element for energy application, which is must be high content are carbon (C), fixed carbon (FC), hydrogen (H), and moisture content (MC) but another element should be low. PB shows high C, H, low ash (A), moisture content (MC), nitrogen (N), oxygen (O) and sulfur (S) and RH shows high FC and MC. By mixture RH and PB to form composites will have a good characteristic element for release energy. Composites \((RH)_x(PB)_{1-x}\) \((x= 0.9, 0.7, 0.5)\) shows homogeneous samples as shown in Table 1, all elemental composition is increased gradually with increasing PB content for an element with higher content in PB and vice versa.

For C, H, and O, we have compared with result from predictive equation by using data from proximate analysis as input parameter, proposed by [40] in the form: 

\[
C = -35.9972 + 0.7698VM + 1.3269FC + 0.3250ASH, \quad H = 55.3678 - 0.4830VM - 0.5319FC - 0.5600ASH, \quad O = 223.6805 - 1.7226VM - 2.2296FC - 2.2463ASH
\]

and proposed by [39] in the
form: $C = 0.47VM + 0.963FC + 0.067ASH$, $H = 0.074VM + 0.012FC - 0.052(VM/FC)$, $O = 0.569VM + 0.012FC - 0.069ASH$. Predictive proposed by Nhuchhen [40] shows reasonable agreement for RH with differences about 10%, for predictive proposed by Thomas Klasson [41] shows good agreement for 10% PB in composites. The C content from the ultimate analysis in this study shows an increase with increasing PB content in composites consistent with the result from predictive equation from Ref. [40] with differences about 15% lower than the ultimate analysis data. The predictive equation from Ref. [41] shows C content is higher for RH and increase about 0.5% with increasing PB content in composites. For H good agreement between predictive equation from Ref. [41] only for RH and from Ref. [40] only for 30%PB and 50%PB compared with ultimate analysis data. For O contents are shows big different about 30-40% between ultimate analysis data with predictive from Ref. [40] and good agreement only for 30% PB with predictive from Ref. [41]. These results indicated the existing predictive equation from Ref. [40] and [41] shows good accuracy result compared with ultimate analysis data only for several compositions in composites RH and PB.

As shown in Table 1, we compared the calorific value (CV) determined by bomb calorimeter in this study with the predictive model developed by Majumder et. al.[42] in the form $CV \text{ (MJ/kg)} = -0.03ASH - 0.11M + 0.33VM + 0.35FC$. For RH shows CV is about 13.44 MJ/kg which is very good agreement with Ref. [37] about 13.30 MJ/kg and Ref. [38] about 13.50 MJ/kg and for PB is about 41.21 MJ/kg in this study also comparable with the previous studies [43-45]. For composites, the CV was increased about 30% with increasing PB content in composites. The differences between predictive equations with the result in this study for 30% and 50% PB in composites are about 10% but become high for pure RH and PB. This indicated that, the predictive CV from Ref. [42] valid for high PB content in composites and good agreement with ethanol (30 MJ/kg) and methanol (23 MJ/kg) as acceptable fuels [46].
For composites \((\text{RH})_x(\text{PB})_{1-x}\) (\(x=0.9, 0.7, 0.5\)), the CV was increase with increasing the amount of PB into RH from 17.85 MJ/kg for \(x=0.9\), 23.97 MJ/kg for \(x=0.7\), to 28.93 MJ/kg for \(x=0.5\). Moreover, increasing the CV showed a positive correlation with C, H, and VM of the fuel. In contrast, the addition of PB lowered the percentage of O, S, N, FC, MC, and A. The physical properties of the fuel depend on the content of CV, MC, and AC that influence to the energy production. The amount of the MC in the fuel affected to the CV and ash (A) of the combustion products [32]. One would expect the fuel mixture to have a maximum CV and minimum A. The energy and the power that can be generated depend on the CV content in the composites for fuel.

The addition of PB into RH can improve the quality of fuel such as increasing CV, C, H, and VM, and decrease FC, MC, S, N, O, and A. The increasing content of C and H means a greater contribution to the energy released [32]. However, the percentage of O decrease in the fuel means increasing the content of C and H as well as increasing CV of the fuel but O required in the process of fuel combustion. The high CV can increase energy production thus; composites in this study become attractive as an energy source. Small amounts of N will reduce NO\(_x\) emissions in the air while small amounts of S can reduce pollution and corrosion. The amount of VM and A is high and low MC are important characteristics required in the process of energy production. The high amount of ash (A) is effected by the reduction of energy production and an increase of handling cost for disposal process [47]. MC is high will aggravate the chemical properties of the fuel; hence, complicating the process of energy production. The amount of PB in composites increase resulting the amount of N and S decrease indicated the formation of NO\(_x\) and SO\(_x\) is decrease, which are both hazardous gasses and very harmful to the environment [48-50]. Thus, the addition of PB into RH to form composites will increase the energy production and also decrease the amount of MC, A, NO\(_x\), and SO\(_x\).
Table 2 shows ash composition of composites from XRF spectroscopy. The main elements for RH are Si and K about 73.61% and 14.31%, respectively, which is similar reported in Ref [23, 24], and that for PB are Ca and Ti about 75.61% and 12.93%, respectively, similar reported in Ref. [51]. For composites shows Si and K as main element from RH decrease with increasing the amount of PB and similar for element from PB is increase with increasing the amount of RH.

(TGA) and (DTG) curves show that there is a difference between the oxidation process of (RH)\textsubscript{1}(PB)\textsubscript{1-x} (x=1, 0.9, 0.7, 0.5) as shown in Fig. 1. The TGA and DTG curves of (RH)\textsubscript{x}(PB)\textsubscript{1-x} (x=1, 0.9, 0.7, 0.5) show the dehydration process (DP) with degradation temperature (DT) from 40 to 100°C and mass loss (ML) from 2.3 to 5.2% as moisture evaporation indicated the dehydration processes. The thermal degradation (TD) of composites is two stages for x ≥ 0.7 and three stages for x=0.5. TD in the first zone (TDZ 1) are at in the range of temperature in between 240 and 360°C with ML in between 40.4 and 43.7%, while TDZ 2 are at in the range of temperature in between 370 and 490°C with ML in between 25.4 and 32.6%. Furthermore, TDZ 3 is in the range of temperature in between 600 and 670°C with ML of about 2.8%. The total ML that during the degradation of (RH)\textsubscript{1}(PB)\textsubscript{1-x} (x= x=1, 0.9, 0.7, 0.5) are in between 71.6 and 79% and generated ash (A) approximately in between 21 and 28.4%. This shows that the addition of PB into RH influences the thermal characteristics of composites by enlarges the energy production and reduces the ash (A) residues. The parameters of kinetic study for the oxidative environment of (RH)\textsubscript{1}(PB)\textsubscript{1-x} (x= x=1, 0.9, 0.7, 0.5), are shown in Table 3.

The activation energy (E\textsubscript{a}) that occurred in the TDZ1 of the (RH)\textsubscript{1}(PB)\textsubscript{1-x} (x= x=1, 0.9, 0.7, 0.5) are shown in Fig. 2. The activation energy for the TDZ1 of RH is 56.49 kJ/mol were obtained on the basis of the linear regression: y\textsubscript{1} = 6794x + 1.902, with R\textsuperscript{2} = 0.915 for DT from 240 - 360°C (Fig. 2a). The E\textsubscript{a1} of 10%PB is 55.55kJ/mol was based on the
the equation: $y_1 = 6681x + 2.340$ with $R^2 = 0.978$ for DT from 240 - 380°C (Fig. 2b). For 30%PB the $E_a_1$ is 64.63kJ/mol based on the equation: $y_1 = 7774x + 0.721$ with $R^2 = 0.959$ for DT from 250 – 380°C (Fig. 2c). Furthermore, the $E_a_1$ of 50%PB is 54.42kJ/mol based on the equation: $y_1 = 6546x + 3.005$ with $R^2 = 0.979$ for DT from 250 – 380°C (Fig. 2d). By the same method, the equation and $E_a$ for the TD process in the second (TDZ2) and the third zones (TDZ3) obtained by linear regression are shown in Table 4.

From the Table 4 clearly shows that the amounts of PB in composites are important in increasing the $E_a$ value. The higher contents of PB in composites are the higher value of $E_a$. The oxidative process of TD processes for RH was perfect after reaching a temperature about 480°C and increase to 490°C for 10%PB and 30%PB. For 50%PB, the TD process was perfect after reaching a temperature about 660°C and from temperature from 660 to 800°C there is no TD and ML. This shows that the composites $($RH$)_x($PB$)_{1-x}$ ($x=$ $x=$ 1, 0.9, 0.7, 0.5) combustion as fuel requires range temperature about 480 to 660°C to perform the complete combustion process to produce high energy.

4. Conclusion

New composites $($RH$)_x($PB$)_{1-x}$ ($x=$ 1, 0.9, 0.7, 0.5) have been obtained and shows elemental composition is increased gradually with increasing PB content for an element with higher content in PB and decrease with increasing PB for lower content in PB indicated homogeneous composite sample. The calorific value (CV) determined by bomb calorimeter shows about 13.44 MJ/kg for RH and about 41.21 MJ/kg for PB, for composites was increased about 30% with increasing PB content in composites, indicated PB has a strong effect in composites.

The temperature range to produced highest energy and perform completed combustion process of composites $($RH$)_x($PB$)_{1-x}$ ($x=$ 1, 0.9, 0.7, 0.5) are at 480 to 660°C. The
activation energy for RH is 101.22 kJ/mol, increase by 4 and 6 magnitudes for x is 0.9 and 0.7, respectively, and increase remarkable to 165.30 kJ/mol for x =0.5. From thermal characteristics, calorific value, and the elemental analysis shows activation energy, calorific content, carbon, and hydrogen content, respectively, strongly affected by PB content which is contributed to increasing the energy realized.

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**References**


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FIGURE CAPTION

Fig. 1. TGA and DTG spectra for the temperature 30 - 800°C with a heating rate of 10°C/min
and flow rate of air about 50 mL/min from composites (RH)ₙ(PB)₁₋ₙ (x=1 for (a),
x=0.9 for (b), x=0.7 for (c), and x=0.5 for (d)).
Fig. 2. The first degradation (TDZ1) stage of the oxidative process to determine activation energy (E_a) and R^2 values from the linear regression of -ln [ln (1- α)/T^2] vs. 1/T for composites (RH)_x(PB)_{1-x} ((x=1 for (a), x=0.9 for (b), x=0.7 for (c), and x=0.5 for (d)).
Table 1. Elemental composition by using proximate and ultimate analysis, and calorific value from bomb calorimeter in oxygen environment for composites \((RH)_x(PB)_{1-x}\) (x=1, 0.9, 0.7, 0.5). We have included elemental composition for RH from Ref. [31], calculation based on predictive equation for carbon, hydrogen, and oxygen from Ref. [35,36], and for calorific value from Ref. [37] for comparison.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Present Study</th>
<th>RH [31]</th>
<th>10% PB</th>
<th>30% PB</th>
<th>50% PB</th>
<th>PB</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Proximate Analysis</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fixed carbon (FC)</td>
<td>(%)</td>
<td>14.81</td>
<td>16.95</td>
<td>13.62</td>
<td>10.89</td>
<td>8.00</td>
<td>0.72</td>
</tr>
<tr>
<td>Volatile matter (VM)</td>
<td>(%)</td>
<td>55.62</td>
<td>61.81</td>
<td>58.65</td>
<td>65.21</td>
<td>72.39</td>
<td>89.04</td>
</tr>
<tr>
<td>Moisture content (M)</td>
<td>(%)</td>
<td>10.46</td>
<td>9.68</td>
<td>7.53</td>
<td>5.26</td>
<td>2.22</td>
<td></td>
</tr>
<tr>
<td>Ash (A)</td>
<td>(%)</td>
<td>19.11</td>
<td>21.24</td>
<td>18.05</td>
<td>16.37</td>
<td>14.35</td>
<td>10.02</td>
</tr>
<tr>
<td><strong>Ultimate Analysis</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon</td>
<td>(%)</td>
<td>37.48</td>
<td>38.50</td>
<td>40.21</td>
<td>47.00</td>
<td>53.60</td>
<td>67.49</td>
</tr>
<tr>
<td>Predictive [35]</td>
<td>(%)</td>
<td>32.68</td>
<td>33.09</td>
<td>33.97</td>
<td>35.01</td>
<td>36.76</td>
<td></td>
</tr>
<tr>
<td>Predictive [36]</td>
<td>(%)</td>
<td>41.91</td>
<td>42.13</td>
<td>42.49</td>
<td>42.98</td>
<td>43.57</td>
<td></td>
</tr>
<tr>
<td>Hydrogen</td>
<td>(%)</td>
<td>5.08</td>
<td>5.20</td>
<td>5.82</td>
<td>7.10</td>
<td>8.61</td>
<td>10.25</td>
</tr>
<tr>
<td>Predictive [36]</td>
<td>(%)</td>
<td>4.09</td>
<td>4.28</td>
<td>4.64</td>
<td>4.98</td>
<td>0.17</td>
<td></td>
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<tr>
<td>Oxygen</td>
<td>(%)</td>
<td>37.81</td>
<td>34.61</td>
<td>35.45</td>
<td>29.13</td>
<td>23.08</td>
<td>11.92</td>
</tr>
<tr>
<td>Predictive [35]</td>
<td>(%)</td>
<td>51.92</td>
<td>51.73</td>
<td>50.29</td>
<td>48.91</td>
<td>46.19</td>
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<tr>
<td>Predictive [36]</td>
<td>(%)</td>
<td>24.92</td>
<td>26.39</td>
<td>29.56</td>
<td>33.04</td>
<td>41.08</td>
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<tr>
<td>Nitrogen</td>
<td>(%)</td>
<td>0.43</td>
<td>0.38</td>
<td>0.31</td>
<td>0.28</td>
<td>0.26</td>
<td></td>
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<tr>
<td>Sulfur</td>
<td>(%)</td>
<td>0.09</td>
<td>0.09</td>
<td>0.09</td>
<td>0.08</td>
<td>0.06</td>
<td></td>
</tr>
<tr>
<td>Ash</td>
<td>(%)</td>
<td>19.11</td>
<td>18.05</td>
<td>16.37</td>
<td>14.35</td>
<td>10.02</td>
<td></td>
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<tr>
<td><strong>Calorific and Bulk</strong></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Calorific value</td>
<td>(MJ/kg)</td>
<td>13.44</td>
<td>17.85</td>
<td>23.97</td>
<td>28.93</td>
<td>41.21</td>
<td></td>
</tr>
<tr>
<td>Predictive [37]</td>
<td>(MJ/kg)</td>
<td>21.81</td>
<td>22.52</td>
<td>24.01</td>
<td>25.68</td>
<td>29.31</td>
<td></td>
</tr>
<tr>
<td>Bulk density</td>
<td>(kg/m³)</td>
<td>265</td>
<td>228.5</td>
<td>198.8</td>
<td>167.6</td>
<td>112</td>
<td></td>
</tr>
</tbody>
</table>
Table 2. Elemental composition by using X-ray fluorescence (XRF) for ash of composites

\[(RHa)(PB)_{1-x} \text{ (x=1, 0.9, 0.7, 0.5)}\]

<table>
<thead>
<tr>
<th>Element</th>
<th>RH</th>
<th>10%PB</th>
<th>30%PB</th>
<th>50%PB</th>
<th>PB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>73.61</td>
<td>63.35</td>
<td>34.61</td>
<td>31.99</td>
<td>2.98</td>
</tr>
<tr>
<td>K</td>
<td>14.31</td>
<td>6.81</td>
<td>4.00</td>
<td>2.40</td>
<td>-</td>
</tr>
<tr>
<td>Ca</td>
<td>3.51</td>
<td>18.25</td>
<td>52.89</td>
<td>57.75</td>
<td>75.61</td>
</tr>
<tr>
<td>Cl</td>
<td>2.78</td>
<td>2.55</td>
<td>1.64</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Px</td>
<td>2.33</td>
<td>3.84</td>
<td>0.73</td>
<td>0.57</td>
<td>-</td>
</tr>
<tr>
<td>Fe</td>
<td>2.06</td>
<td>0.97</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Ti</td>
<td>-</td>
<td>1.26</td>
<td>5.57</td>
<td>6.16</td>
<td>12.93</td>
</tr>
<tr>
<td>Mg</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>7.05</td>
</tr>
<tr>
<td>Pb, Zn, Cr, Mn, Nb, Mo, In</td>
<td>-</td>
<td>2.97</td>
<td>0.56</td>
<td>1.13</td>
<td>1.43</td>
</tr>
</tbody>
</table>
Table 3. Parameters of the kinetic study for the oxidative environment of composites (RH)_{x}(PB)_{1-x} (x=1, 0.9, 0.7, 0.5) for thermal degradation zone (TDZ1, TDZ2, TDZ3) based on FIG. 1.

<table>
<thead>
<tr>
<th>Materials (mg)</th>
<th>BP</th>
<th>DP</th>
<th>TDZ1 ML (%)</th>
<th>TDZ1 T (°C)</th>
<th>TDZ2 ML (%)</th>
<th>TDZ2 T (°C)</th>
<th>TDZ3 ML (%)</th>
<th>TDZ3 T (°C)</th>
<th>Ea (kJ/mol)</th>
<th>Ash (%)</th>
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<tbody>
<tr>
<td>RH 5.8</td>
<td>5.2</td>
<td>50 - 100</td>
<td>41</td>
<td>240 - 360</td>
<td>25.4</td>
<td>370 - 480</td>
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<td>101.22</td>
<td>28.4</td>
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<tr>
<td>10%PB 5.5</td>
<td>4.4</td>
<td>50 - 100</td>
<td>43.7</td>
<td>240 - 360</td>
<td>25.8</td>
<td>370 - 490</td>
<td>-</td>
<td>-</td>
<td>104.03</td>
<td>26.1</td>
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<tr>
<td>30%PB 5.6</td>
<td>2.3</td>
<td>50 - 100</td>
<td>42.4</td>
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<td>390 - 490</td>
<td>-</td>
<td>-</td>
<td>110.95</td>
<td>24.4</td>
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<tr>
<td>50%PB 5.5</td>
<td>3.2</td>
<td>50 - 100</td>
<td>40.4</td>
<td>250 - 380</td>
<td>32.6</td>
<td>390 - 460</td>
<td>2.8</td>
<td>600 - 670</td>
<td>165.30</td>
<td>21</td>
</tr>
</tbody>
</table>
Table 4 Thermal degradation process for zone 1, 2, 3 to determine activation energy (Ea) by using linear regression (as shown in Fig. 2 for TDZ1) of Arrhenius law from analysis data of TGA and DTG in Fig 1.

<table>
<thead>
<tr>
<th>Samples</th>
<th>DT (°C)</th>
<th>Linear regression of Arrhenius law</th>
<th>(R^2)</th>
<th>Ea (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Ea1</td>
</tr>
<tr>
<td>RH</td>
<td>240 – 360</td>
<td>y_1 = 6794x + 1.902</td>
<td>0.915</td>
<td>56.49</td>
</tr>
<tr>
<td></td>
<td>370 - 480</td>
<td>y_2 = 5380x + 4.792</td>
<td>0.916</td>
<td>-</td>
</tr>
<tr>
<td>10%PB</td>
<td>240 – 380</td>
<td>y_1 = 6.681x+ 2.340</td>
<td>0.978</td>
<td>55.55</td>
</tr>
<tr>
<td></td>
<td>390 - 490</td>
<td>y_2 = 5831x + 4.216</td>
<td>0.961</td>
<td>-</td>
</tr>
<tr>
<td>30%PB</td>
<td>250 – 380</td>
<td>y_1 = 7774x+ 0.721</td>
<td>0.959</td>
<td>64.63</td>
</tr>
<tr>
<td></td>
<td>390 - 490</td>
<td>y_2 = 5571x + 4.627</td>
<td>0.971</td>
<td>-</td>
</tr>
<tr>
<td>50%PB</td>
<td>250 – 380</td>
<td>y_1 = 6546x + 3.005</td>
<td>0.979</td>
<td>54.42</td>
</tr>
<tr>
<td></td>
<td>390 – 460</td>
<td>y_2 = 6444x + 3.637</td>
<td>0.915</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>600 - 660</td>
<td>y_3 = 6892x + 4.695</td>
<td>0.924</td>
<td>-</td>
</tr>
</tbody>
</table>