BUDAPEST UNIVERSITY OF TECHNOLOGY AND ECONOMICS

Laboratory of Plastics and Rubber Technology

Budapest, 20. December, 2020

Dear Editor,

Please find enclosed the manuscript **"Effect of particle characteristics and interfacial adhesion on the properties of PP/sugarcane bagasse fiber composites"** by András Bartos, Judit Kócs, Juliana Anggono, János Móczó and Béla Pukánszky for your kind consideration to be reviewed and published in the Polymer Testing. The corresponding author of the manuscript is András Bartos. The paper deals with PP/sugarcane bagasse fiber composites prepared from two fractions of fibers with different particle characteristics.

Because of the continuous search for new materials and the increasing environmental awareness of the industry as well as the public, the interest in materials from renewable resources increases continuously also in the plastics industry. Many products are prepared with natural reinforcement, however, besides their advantages, natural reinforcements have several drawbacks like their small transverse strength and their adhesion to the polyolefin matrices is often quite weak. The most important characteristics of the fibers, besides their transversal strength, are their dimensions, i.e. length, diameter and aspect ratio. The dimensions of the fibers change during the processing of composite materials by traditional thermoplastic technologies. However, the attrition of the fibers has a considerable impact on the properties of the composites, fiber dimensions and attrition are analyzed thoroughly only in very few papers dealing with natural fiber-reinforced polymers. Often even the original dimensions of the fibers are not reported, but their change during processing is frequently neglected. Considering these facts, the goal of this work was to study the effect of particle dimensions and interfacial adhesion on composite properties. Furthermore, local deformation processes were followed by acoustic emission testing.

Overall, we believe that the evaluation, interpretation and conclusions give novelty for our study. We hope that this paper will provide a comprehensive and detailed knowledge for the scientific community of PP/natural composites including the effect of sugarcane bagasse fiber characteristics, local processes occurring during deformation and interfacial adhesion.

Hereby we also confirm that the manuscript or its contents in some other form, has not been published previously by any of the authors, it is not under consideration for publication in another journal at the time of submission, and all the authors have seen and approved the submission of the manuscript.

I thank you for your care and patience and send you my best regards.

Yours sincerely
András Bartos

Bartos András

Feladó: em.pote.0.70c529.1f8ddd9c@editorialmanager.com Polymer Testing <em@editorialmanager.com> nevében
Küldve: kedd 2021. január 19 22:35
Címzett: András Bartos
Tárgy: Decision on submission to Polymer Testing

Manuscript Number: POTE-D-20-00717

EFFECT OF PARTICLE CHARACTERISTICS AND INTERFACIAL ADHESION ON THE PROPERTIES OF PP/SUGARCANE BAGASSE FIBER COMPOSITES

Dear Mr Bartos,

Thank you for submitting your manuscript to Polymer Testing.

I have completed my evaluation of your manuscript. The reviewers recommend reconsideration of your manuscript following major revision. I invite you to resubmit your manuscript after addressing the comments below. Please resubmit your revised manuscript by Mar 05, 2021.

When revising your manuscript, please consider all issues mentioned in the reviewers' comments carefully: please outline every change made in response to their comments and provide suitable rebuttals for any comments not addressed. Please note that your revised submission may need to be re-reviewed.

To submit your revised manuscript, please log in as an author at https://www.editorialmanager.com/pote/, and navigate to the "Submissions Needing Revision" folder.

Polymer Testing values your contribution and I look forward to receiving your revised manuscript.

Kind regards,
Ulf W. Gedde
Editor

Polymer Testing

Editor and Reviewer comments:

Reviewer #1: PP/sugarcane bagasse fiber composites by injection molding from two fractions of fibers with different particle characteristics have been proposed, with provided the obvious improvements of good adhesion, the stiffness and strength of PP and its impact resistance. The paper is well structured, and the subject of the study is within the scope of the journal. Experimental approached regarding materials and analysis are novel enough for its publication after revision being taken into accounts.
1. How to prepare composite materials with the similar ratio of MAPP/fiber each time. And explain how the ratio of 0.1was chosen?
2. The writing needs some improvements.
3. Clearly state how many independent samples were tested for each type of measurements?
4. As we know, good interfacial adhesion may not always result in the satisfactory overall properties of composites. The enhancement of the interfacial adhesion is usually accompanied by a corresponding decrease in the impact toughness. It ascribes to the tightly bound but stiff interface generating high stress

concentrations and limiting energy absorption. It is need one degree to keep more toughness. So, the author should give one reference value for the interfacial adhesion strength.
5. To study valuable information about the mechanism of the local processes, other micrographs should be examined in the paper or in the section of supplementary materials.
6. Some related literatures about fiber surface modification for enhancing interfacial adhesion are needed to be cited (Composites: Part A 2017, 99, 58; Composites: Part A 2017, 101, 511; Composites Science and Technology 2017, 138,144)

Reviewer #2: The authors investigate the reinforcing effects of sugarcane bagasse in PP in regard to the particle size of the fibres as well as the presence of a coupling agent. Interfacial adhesion, morphology and mechanical properties are investigated. The paper is well written and presented, and the findings are based on the data, supported with references. I only have minor remarks for considerations by the authors which are given below.

p5, "…In order to determine fiber dimensions after processing, composite samples were put into boiling xylene (Molar Chemicals Kft., Halásztelek, Hungary) for 8 hours to dissolve the polymer…." Here it would be nice to have some details, e.g. the approx. sample mass and the approx. volume of xylene being used. Also, was the filtration carried out at raised temperature (as PP tends to precipitate rather quickly) and with vacuum? Residual PP can "glue" particles together and therefore influence the particle dimension measurements.
p6, "…Interestingly the final dimensions of both fractions are very similar, although the aspect ratio of the long fibers is slightly larger than that of the short fraction…."
Only a comment from my side (as we have experienced similar in processing natural fibres) - this could correlate with the internal dimensions of the smallest slit/narrow passage in your extruder between the screw/kneading elements. You can check these, and maybe there is a possible correlation between these dimensions.
Figure captions 2, 3, 4, 7, 8 - I think the poor and good adhesion should be skipped here, as this is a conclusion from the data and not a label of the data itself Figure caption 6 - In my opinion, it should read "Resulting morphologies from local processes are indicated by arrows", as the processes itself cannot be seen.
Reference 4 seems to miss the date of publication.

Reviewer #3: POLYMER TESTING, ANDRAS BARTOS ET AL.

The manuscript is well written and generally is suitable to be published in first class scientific journal. I appreciate especially thorough characterization of fibres including changes occuring during processing. This should be definitely the standard procedure for any fibre - containing composites.
I have few comments which might be considered by the authors for accepting or explained to the reviewer the reason in case they are not accepted. The comments are as follows:
1. Part 3,3 p. 5 The dimensions of 5-600 fibers - what is this? Number of fibres or some code? If number of fibres, I recommend ...of five to 600 fibers...
2. P. 6 bottom - Interestingly the final dimensions of both fractions are very similar, although the aspect ratio of the long fibers is slightly larger than that of the short fraction - This is well-known basic theoretical conclusion coming from equilibrium between interfacial adhesion on the boundaries of fibre surface and the matrix (if prevailing, pull out of the fibres occurs) and strength of the fibres (breaking the fibres to shorter length, if prevailing), thank you for very nice example of this, I believe that you paper will

be accepted and I will be able to use your Table in my textbook of polymer physics under preparation for undergraduate students. Re your paper. perhaps it would not be difficult to find a quotation of this phenomenon, which is known for years, in published literature.

3. Previous experience shows that debonding usually occurs at small deformations and stresses, while the second process might be fiber fracture and/or fiber pullout - since you have nice data on breaking the fibres during processing it should be easy to extend this experiment to materials after mechanical testing, both tensile and impact, you could tell exactly whether in your case the reason is fiber fracture or pullout.

4. I would appreciate to insert 1 - 2 short sentences how the characteristic stress is defined. By the way is „characteristic stress" a terminus technicus? If not, perhaps better name could be found, in my view it looks like stress onset or something similar.

5. As shown by Fig. 7, the first occurs at the same and quite small stresses for the two fractions UNCLEAR SENTENCE, please reformulate the statement to be understandable without reading twice.

6. The largest particles debond first and thus not aspect ratio or average size, but the size and number of large particles determine the initiation stress of this process - Perhaps a bit surprising since in my view, debonding depends on overall strength of the interactions on the particle - matrix interface which should be proportionte to the overall surface area of the fibre, obviously this is higher for longer particles. Moeover, I am somewhat confused, since after processing the size of long and short particles is more or less the same.

7. A comment to the same part: In the part Local processes the discussion is aimed to large and small particles, anyway, at the beginning you are discussing fibres. In that case, large particles means longer fibre (the fibre are more or less of the same length after processing - Table 1) or something else. I suggest to unify the terms so that it will be clear what is larger or longer and , especially, you are discussing the same fillers (fibers) through whole paper.

RECOMMENDATION: ACCEPT AFTER MINOR CHANGES

Reviewer #4: Major comments

1. What exactly is the novelty of the study? PP/sugarcane bagasse composites have been commonly described in the literature. The necessity of coupling in composites of polyolefines and natural fibres is well known, and so are its effects on micromechanical deformation processes. Maleated polyolefins are commonly used for this purpose. The effect of particle size and aspect ratio on natural fibre/polymer composites' properties has been widely studied. The identification of micromechanical deformations based on acoustic emission tests and SEM micrographs as well as estimating load-bearing capacity from the composition-dependence of mechanical properties have been discussed in several papers, some of them published by the same research group. Attrition is indeed a neglected topic, and its study presents some novelty, which should be better highlighted, perhaps, and even included in the title.

2. What explains the relatively large differences in the estimated degree of attrition in the few studies that exist? Is it due to different characterization methods, processing conditions or perhaps the inherent properties of the fibres? Since the article is being considered for publication in Polymer Testing, some more focus on methodology would be welcome, in my opinion.

3. Sugarcane bagasse is not a homogeneous material: it consists of fibres derived from different plant tissues, mainly so-called 'pith' and 'stem' fibres. Since the former tend to be shorter, the short fraction in the study is likely richer in pith fibres. This might explain some of the differences in the properties of composites prepared with long and short fractions, respectively. Have the authors attempted to determine the composition of the two fractions to ensure that the only differences between them are indeed their particle size and aspect ratio?

4. Figure 3: What explains the lower strength values determined in the composites made with short fibres at higher filler contents?

5. Page 6: "Increased adhesion does not have any effect on attrition; practically the same dimensions are obtained after processing in both the presence and the absence of the MAPP coupling agent." One would expect adhesion to have some effect on attrition during processing. How do the authors explain the lack of it?
6. "…the composites containing the long fraction have somewhat larger modulus because of their larger aspect ratio." Why does a larger aspect ratio result in higher modulus?
7. Page 7: In the section about properties, the effects of aspect ratio, orientation and adhesion are mixed. It is hard to determine how exactly the authors aim to interpret the complex relationship between these three factors and composite properties. Some clarification would be desirable.
8. Figure 7: The composition-dependence of characteristic stress should be discussed in some detail.
9. Page 11: The observation according to which initial particle dimensions have a very limited effect on properties due to attrition during processing is an important one, and should be emphasized. On the other hand, the fact that coupling changes interactions and local deformation processes has been widely reported and should not be presented as new information.
10. My last major comment is admittedly highly subjective. Nevertheless, I believe the strict adherence to a certain structure, in which macroscopic properties, local processes, adhesion, etc., are discussed separately, does not necessarily serve clarity or readability. Although the form chosen by the authors is certainly logical, it is often difficult to follow their reasoning, since cause and effect are presented in different parts of the text. A more integrated interpretation of the results might be desirable.

Minor comments
1. Language of the manuscript is adequate, although it contains some grammatical and stylistic errors. A few typical examples are listed below.
a) Incorrect word order in some sentences
b) The relative pronoun "which" is frequently, and incorrectly, used instead of "that".The two are not interchangeable.
c) Missing hyphenation in some compound words, e.g. fiber-reinforced polymers (page 2)
2. Page 3: "In fiber-reinforced polymer composites frequently local deformation processes take place which are related to or occur around the fibers." The phrasing seems a little awkward. The local deformation processes in question are always "related to" the fibres: they are either initiated by them or they happen to them.
3. Page 3: "Fiber fracture occurs more frequently in the case of strong interfacial interactions, when they are improved…" It is unclear what "they" refers to.
4. Page 4: "…the goal of this study was to prepare composites from a locally derived natural reinforcement, sugarcane bagasse fiber…" Locally derived where? Hungary of Indonesia?
5. Page 4: "The bagasse fibers were obtained directly from the sugar mill." Which sugar mill? Can the authors be more specific?
6. Page 4: "The ratio of MAPP/fiber was 0.1 in all composites." Is that weight ratio?

*****

Data in Brief (optional):

We invite you to convert your supplementary data (or a part of it) into an additional journal publication in Data in Brief, a multi-disciplinary open access journal. Data in Brief articles are a fantastic way to describe supplementary data and associated metadata, or full raw datasets deposited in an external repository, which are otherwise unnoticed. A Data in Brief article (which will be reviewed, formatted, indexed, and given a DOI) will make your data easier to find, reproduce, and cite.

You can submit to Data in Brief when you upload your revised manuscript. To do so, complete the template and follow the co-submission instructions found here: www.elsevier.com/dib-template. If your manuscript is accepted, your Data in Brief submission will automatically be transferred to Data in Brief for editorial review and publication.

Please note: an open access Article Publication Charge (APC) is payable by the author or research funder to cover the costs associated with publication in Data in Brief and ensure your data article is immediately and permanently free to access by all. For the current APC see: www.elsevier.com/journals/data-in-brief/2352-3409/open-access-journal

Please contact the Data in Brief editorial office at dib-me@elsevier.com or visit the Data in Brief homepage (www.journals.elsevier.com/data-in-brief/) if you have questions or need further information.

*****

We invite you to submit a method article alongside your research article. This is an opportunity to get full credit for the time and money spent on developing research methods, and to increase the visibility and impact of your work. If your research article is accepted, we will contact you with instructions on the submission process for your method article to MethodsX. On receipt at MethodsX it will be editorially reviewed and, upon acceptance, published as a separate method article. Your articles will be linked on ScienceDirect.

Please prepare your paper using the MethodsX Guide for Authors: https://www.elsevier.com/journals/methodsx/2215-0161/guide-for-authors (and template available here: https://www.elsevier.com/MethodsX-template) Open access fees apply.

More information and support

FAQ: How do I revise my submission in Editorial Manager?
https://service.elsevier.com/app/answers/detail/a_id/28463/supporthub/publishing/

You will find information relevant for you as an author on Elsevier's Author Hub:
https://www.elsevier.com/authors

FAQ: How can I reset a forgotten password?
https://service.elsevier.com/app/answers/detail/a_id/28452/supporthub/publishing/kw/editorial+manager/

For further assistance, please visit our customer service site: https://service.elsevier.com/app/home/supporthub/publishing/. Here you can search for solutions on a range of topics, find answers to frequently asked questions, and learn more about Editorial Manager via interactive tutorials. You can also talk 24/7 to our customer support team by phone and 24/7 by live chat and email.

In compliance with data protection regulations, you may request that we remove your personal registration details at any time. (Use the following URL: https://www.editorialmanager.com/pote/login.asp?a=r). Please contact the publication office if you have any questions.

Answer

to the comments of the Referees on the manuscript “Effect of particle characteristics and interfacial adhesion on the properties of PP/sugarcane bagasse fiber composites” by Bartos, A., Kócs, J., Anggono, J., Móczó, J. and Pukánszky, B. submitted to Polymer Testing (POTE-D-20-00717)

We appreciate the comments of the Referees as well as their suggestions to improve the quality of the paper. The comments helped considerably in revising the paper thus we took as many as possible into account during revision. The modifications carried out and the answers to the questions are listed below. Questions are printed by normal letters, while answers are written in italic. Modifications are made in the revised manuscript and a clean version is submitted now.

Reviewer #1

We are very glad that the Referee found that "the paper is well structured, and the subject of the study is within the scope of the journal. Experimental approached regarding materials and analysis are novel enough for its publication after revision being taken into accounts". We considered the questions and suggestions of the Reviewer very thoroughly and modified the paper accordingly. Our answers and the modifications are listed below.

1. How to prepare composite materials with the similar ratio of MAPP/fiber each time. And explain how the ratio of 0.1was chosen?

   The coupling agent was always added to the composite in the same 0.1 ratio calculated for the weight of the fiber and it was added on the expense of the matrix polymer. The text was modified to make the composition clearer. Earlier we studied the effect of the amount of the coupling agent on composite properties and found this ratio as the optimum [A1]. Accordingly, we use the same ratio in all of our fiber-reinforced composites.
2. The writing needs some improvements.

   We thank the Referee for the advice. We checked the paper several times and did our best to correct the language according to our best ability.
3. Clearly state how many independent samples were tested for each type of measurements?

   We followed the standards indicated in the experimental part in the selection of the number of specimens tested. We measured five specimens in tensile and ten in impact testing. The number of specimens is indicated explicitly in the revised paper, now.
4. As we know, good interfacial adhesion may not always result in the satisfactory overall properties of composites. The enhancement of the interfacial adhesion is usually accompanied by a corresponding decrease in the impact toughness. It ascribes to the tightly bound but stiff interface generating high stress concentrations and limiting energy absorption. It is need one degree to keep more toughness. So, the author should give one reference value for the interfacial adhesion strength.

   The Reviewer is completely right that good adhesion is not always beneficial and strong bonding might lead to the decrease of impact strength. This statement is strongly supported by the results of this study, showing somewhat smaller impact strength in the *presence of the coupling agent. However, the effect depends very much on the components and the mechanism of interaction; strong adhesion is not so deleterious in our composites than in thermoset matrices in which chemical coupling may lead to very stiff materials with small impact resistance. We estimated the strength of adhesion in the paper and the values are compiled in Table 2 showing that adhesion strength is about 140 mJ/m^2 in the absence of MAPP and about ten times as large, around 1600 mJ/m^2 when the coupling agent is added.*

5. To study valuable information about the mechanism of the local processes, other micrographs should be examined in the paper or in the section of supplementary materials.

   We thank the Referee for the comment. We prepared a large number of micrographs during the study of the structure and the mechanism of failure by electron microscopy. We included only two micrographs into the paper in order to save space and also because additional micrographs would not have offered new information compared to those two. Based on the advice of the Reviewer, we added several micrographs to the Supplementary Information for viewing.

6. Some related literatures about fiber surface modification for enhancing interfacial adhesion are needed to be cited (Composites: Part A 2017, 99, 58; Composites: Part A 2017, 101, 511; Composites Science and Technology 2017, 138,144).

   We thank the Referee for calling our attention to these valuable publications. We went through them and added them as references to the revised paper.

Reviewer #2

We are delighted that this Referee thinks that "the paper is well written and presented, and the findings are based on the data, supported with references". He or she had only minor remarks for consideration. The action taken and the answers to the questions/remarks are listed below

1. p5, "…In order to determine fiber dimensions after processing, composite samples were put into boiling xylene (Molar Chemicals Kft., Halásztelek, Hungary) for 8 hours to dissolve the polymer…." Here it would be nice to have some details, e.g. the approx. sample mass and the approx. volume of xylene being used. Also, was the filtration carried out at raised temperature (as PP tends to precipitate rather quickly) and with vacuum? Residual PP can "glue" particles together and therefore influence the particle dimension measurements.

   *The Reviewer is completely right, the dissolution of the matrix and the determination of fiber dimensions is not straightforward in PP composites. We compressed about 2 g of the composites into thin films, cut them up and then put the pieces into 200 ml xylene. The solvent containing the pieces was boiled for 8 hours and then the suspension was filtered while hot. The fibers remaining on the filter were washed with hot xylene. The residue on the filter was dried and then the dimensions of the fibers were measured with the help of a digital optical microscope. The Referee is right again that PP tends to glue the particles together, but the procedure used made possible their separation and the determination of fiber dimensions. We enclose a micrograph (**Fig. A1**) to prove that individual fibers could be distinguished and their dimensions determined quite well.*

Fig. A1 *DOM micrograph used for the determination of fiber dimensions after processing.*

2. p6, "…Interestingly the final dimensions of both fractions are very similar, although the aspect ratio of the long fibers is slightly larger than that of the short fraction….". Only a comment from my side (as we have experienced similar in processing natural fibres) - this could correlate with the internal dimensions of the smallest slit/narrow passage in your extruder between the screw/kneading elements. You can check these, and maybe there is a possible correlation between these dimensions.

   We agree with the Reviewer completely that narrow slits may determine fiber dimensions after processing. However, as indicated in the experimental part, we used a twin-screw compounder which has short screws with large diameter, deep channels and the flights are cut through to increase mixing volume. The diameter of the capillaries used as dies was also much larger than the fibers (4 mm). Accordingly, not machine dimensions, but the forces acting in the melt determined the dimensions of the fibers.
3. Figure captions 2, 3, 4, 7, 8 - I think the poor and good adhesion should be skipped here, as this is a conclusion from the data and not a label of the data itself.

   We thank the Referee for the comment. He is right, and the captions were corrected, the reference to good and poor adhesion was deleted from those indicated by him or her.
4. Figure caption 6 - In my opinion, it should read "Resulting morphologies from local processes are indicated by arrows", as the processes itself cannot be seen.

   The Reviewer is right again, the caption of Fig. 6 was modified accordingly.
5. Reference 4 seems to miss the date of publication.

   We thank the Referee for calling our attention to the missing information. The revised text contains now the details of publication.

Reviewer #3

It is nice to read that according to this Referee "the manuscript is well written and generally is suitable to be published in first class scientific journal. I appreciate especially thorough characterization of fibres including changes occuring during processing. This should be definitely the standard procedure for any fibre - containing composites". The answers to the questions and remarks of the Reviewer are listed below.

1. Part 3,3 p. 5 The dimensions of 5-600 fibers - what is this? Number of fibres or some code? If number of fibres, I recommend ...of five to 600 fibers...

   We thank the Referee for the remark, the text was modified accordingly.
2. P. 6 bottom - Interestingly the final dimensions of both fractions are very similar, although the aspect ratio of the long fibers is slightly larger than that of the short fraction - This is well-known basic theoretical conclusion coming from equilibrium between interfacial adhesion on the boundaries of fibre surface and the matrix (if prevailing, pull out of the fibres occurs) and strength of the fibres (breaking the fibres to shorter length, if prevailing), thank you for very nice example of this, I believe that you paper will be accepted and I will be able to use your Table in my textbook of polymer physics under preparation for undergraduate students. Re your paper. perhaps it would not be difficult to find a quotation of this phenomenon, which is known for years, in published literature.

   We thank the Reviewer for his or her nice words and we are glad that he or she thinks that our paper is textbook material.
3. Previous experience shows that debonding usually occurs at small deformations and stresses, while the second process might be fiber fracture and/or fiber pullout - since you have nice data on breaking the fibres during processing it should be easy to extend this experiment to materials after mechanical testing, both tensile and impact, you could tell exactly whether in your case the reason is fiber fracture or pullout.

   *We thank the Referee for the suggestion, it is a great idea. However, we must consider the fact that the distribution of both fiber length and diameter is rather wide (see **Fig. A2**), fibers of all sizes are dispersed in the matrix. We had to measure the dimensions of a large number of fibers in order to obtain the distributions. Only a few fibers break on the surface of the specimens during fracture which would not change the distributions significantly. Moreover, depending on the orientation of the fibers and interfacial adhesion, debonding, fiber pullout and fracture occur simultaneously during failure which further complicate the determination of the dominating mechanism. In a number of previous studies, we found that the best way to identify the local processes taking place during deformation is the analysis of the acoustic emission results and the SEM micrographs recorded on the fracture surfaces simultaneously.*
4. I would appreciate to insert 1 - 2 short sentences how the characteristic stress is defined. By the way is „characteristic stress" a terminus technicus? If not, perhaps better name could be found, in my view it looks like stress onset or something similar.

   The procedure for the determination of the characteristic stresses is shown in Fig. 5a and b. The text was extended with a few sentences to facilitate understanding and make it clearer. The Reviewer is completely right, the determined quantities are stress values indicating the initiation of the given process. We could call them initiation stress as well, but characteristic stress became a terminus technicus for us indeed in recent years.
5. As shown by Fig. 7, the first occurs at the same and quite small stresses for the two fractions UNCLEAR SENTENCE, please reformulate the statement to be understandable without reading twice.

   We apologize for the unclear formulation of the sentence. The text was changed in the revised paper to make it clearer.

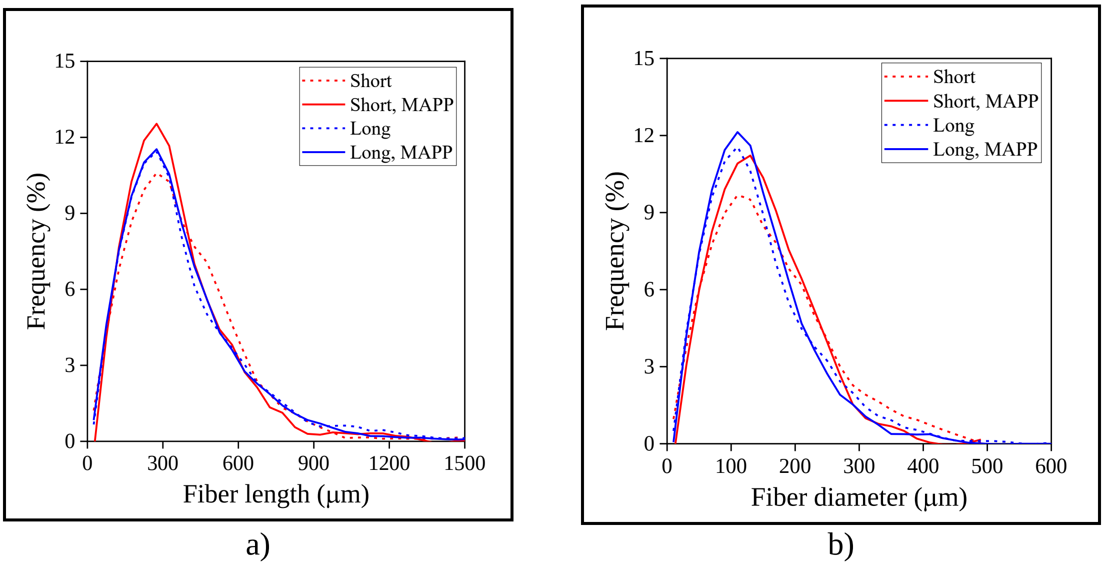

Fig. A2 *Distribution of fiber length and diameter after processing.*

6. The largest particles debond first and thus not aspect ratio or average size, but the size and number of large particles determine the initiation stress of this process - Perhaps a bit surprising since in my view, debonding depends on overall strength of the interactions on the particle - matrix interface which should be proportionte to the overall surface area of the fibre, obviously this is higher for longer particles. Moeover, I am somewhat confused, since after processing the size of long and short particles is more or less the same.

   Debonding stress depends on thermal stresses (σ^D), the stiffness of the matrix (E) interfacial adhesion (F_a) and the size of the particles (R), the diameter of the fibers in this case, as shown by Eq. 2 in the paper (Eq. A1 in the answers)

$$\sigma^D = -C_1\,\sigma^T + C_2\left(\frac{E\,F_a}{R}\right)^{1/2} \tag{A1}$$

   Accordingly, debonding is initiated first at the fibers with the largest diameter. The size distribution and average dimensions of short and long fibers are very similar leading to the very similar initiation (characteristic) stresses of debonding (first step) in Fig. 7 strongly supporting the last sentence of the Reviewer.

7. A comment to the same part: In the part Local processes the discussion is aimed to large and small particles, anyway, at the beginning you are discussing fibres. In that case, large particles means longer fibre (the fibre are more or less of the same length after processing - Table 1) or something else. I suggest to unify the terms so that it will be clear what is larger or longer and , especially, you are discussing the same fillers (fibers) through whole paper.

   The Referee is completely right, terms are confused in this section. The main reason is that the model (see ***Eq. A1****) was developed for particulate-filled composites. However, it can be applied equally well for composites reinforced with wood flour and natural fibers, like the sugarcane bagasse fibers used in this study. The only difference that the diameter of the fiber must be introduced into the model, since debonding occurs with the largest probability at the surface of the fibers vertically to the axis of the fiber. We* *checked the text and unified the terminology used to facilitate understanding.*

Reviewer #4

This Referee was the most critical with our paper and questioned especially the novelty of the work. He or she had a number of questions and remarks, most of them very knowledgeable and justified. We tried to take them into account during revision, but in some cases it was not really possible without the complete reconstruction of the structure of the paper (see Q10). The questions and remarks are below as before together with our answers.

1. **Major comments.** What exactly is the novelty of the study? PP/sugarcane bagasse composites have been commonly described in the literature. The necessity of coupling in composites of polyolefines and natural fibres is well known, and so are its effects on micromechanical deformation processes. Maleated polyolefins are commonly used for this purpose. The effect of particle size and aspect ratio on natural fibre/polymer composites' properties has been widely studied. The identification of micromechanical deformations based on acoustic emission tests and SEM micrographs as well as estimating load-bearing capacity from the composition-dependence of mechanical properties have been discussed in several papers, some of them published by the same research group. Attrition is indeed a neglected topic, and its study presents some novelty, which should be better highlighted, perhaps, and even included in the title.

   The Reviewer might be right that particular questions discussed in the paper, like sugarcane bagasse as reinforcement, the use of maleated polyolefins, local deformations have been studied before. However, we think that the novelty of our paper is not in the individual measurement, but in the complex analysis of the results. We might even question the statements that PP/sugarcane bagasse composites have been "commonly" described in the literature; there are a few studies, but not many. Similarly, the effect of particle size and aspect ratio might be mentioned in some papers, but again not in too many, and the measurements and/or results are not always adequate. However, besides the analysis of the results we think that the strength of our paper is the quantitative estimation of the strength of interfacial adhesion, the establishment of correlation between local processes and macroscopic properties and, as the Referee also state, the thorough analysis of fiber attrition during processing and its effect on properties. And we did not measure only the length, but also the diameter of the fibers that has never been done before, at least according to our knowledge. We also should like to call the attention of the Referee to the fact that Reviewer #3 found our paper textbook material. Nevertheless, we agree completely with the Referee that attrition is one of the most interesting aspect of our work and accepted his or her suggestion and included this into the title of the revised paper.
2. What explains the relatively large differences in the estimated degree of attrition in the few studies that exist? Is it due to different characterization methods, processing conditions or perhaps the inherent properties of the fibres? Since the article is being considered for publication in Polymer Testing, some more focus on methodology would be welcome, in my opinion.

   As the Referee states, only a few papers deal with the effect of particle characteristics and especially the attrition of the fibers on the properties of natural fiber-reinforced composites. None of them studies the changes in the diameter of the reinforcement. We are convinced that the differences observed are caused by the different conditions *prevailing during processing. Vazquez et al. (Ref. 21), for example, produced their composites in an internal mixer. On the other hand, Jimenez et al. (Ref. 22) prepared their specimens by injection molding, but they did not give any information about the actual processing conditions. Since the source of the fibers, their initial length, the molecular weight of the polymer, the type and conditions of processing all differ in the reported cases, it is impossible to draw general conclusions about attrition from these or other published information.*

3. Sugarcane bagasse is not a homogeneous material: it consists of fibres derived from different plant tissues, mainly so-called 'pith' and 'stem' fibres. Since the former tend to be shorter, the short fraction in the study is likely richer in pith fibres. This might explain some of the differences in the properties of composites prepared with long and short fractions, respectively. Have the authors attempted to determine the composition of the two fractions to ensure that the only differences between them are indeed their particle size and aspect ratio?

   The Reviewer is very knowledgeable about the structure and properties of sugarcane bagasse fibers and he or she is completely right about the possible differences caused by the different plant tissues. Unfortunately, we obtained the fibers as a large batch from the sugar factory and it was impossible to separate different tissues. We could have analyzed the short and long fraction chemically, but we have not done so, mostly because our previous experience with wood fibers indicated that the chemical composition of the fibers is less important for composite properties than their particle characteristics. We also have results that indicate that the chemical composition, structure and characteristics of the two fractions do not differ significantly. We determined the inherent strength of the two fractions with a technique developed earlier [A2] and found that it is practically the same as shown by ***Fig. A3****. The results indicate that fiber characteristics are more important, indeed, than the source of the fibers.*

4. Figure 3: What explains the lower strength values determined in the composites made with short fibres at higher filler contents?

   We are convinced that the slightly larger length and the larger aspect ratio of the long fibers result in the difference in their reinforcing effect at strong adhesion. We considered the question and could not find any other explanation. The text was extended in the revised manuscript to offer this explanation.

5. Page 6: "Increased adhesion does not have any effect on attrition; practically the same dimensions are obtained after processing in both the presence and the absence of the MAPP coupling agent." One would expect adhesion to have some effect on attrition during processing. How do the authors explain the lack of it?

   This is a very interesting question again. Mainly large particles break during processing as shown by the difference in attrition for the short and the long fibers. Interfacial adhesion improves, but shear forces are basically the same in the presence or absence of the coupling agent. Improved adhesion may result in the fracture of some smaller fibers as well, but the difference is small. In fact, the Reviewer is right questioning the use of the word "any" and we modified the text and changed it to "not much". According to the values listed in Table 1 coupling has some effect on attrition, indeed, but it is very small.

6. "…the composites containing the long fraction have somewhat larger modulus because of their larger aspect ratio." Why does a larger aspect ratio result in higher modulus?

   Interfacial shear stress has a distribution along the fiber. Shear stress and thus stress transfer is zero at the ends of the fiber, increases linearly towards the center and achieves a constant value in the middle if the length of the fiber exceeds the critical *length depending on interfacial adhesion. Accordingly, if the aspect ratio of the fiber increases the average stress transferred to it also increases and thus modulus and usually also strength increases. A general goal in the preparation of short fiber composites is to increase aspect ratio (long fiber-reinforced composites) and the somewhat smaller reinforcing effect of natural fibers can be explained partly by their smaller aspect ratio compared to traditional glass and carbon fibers.*

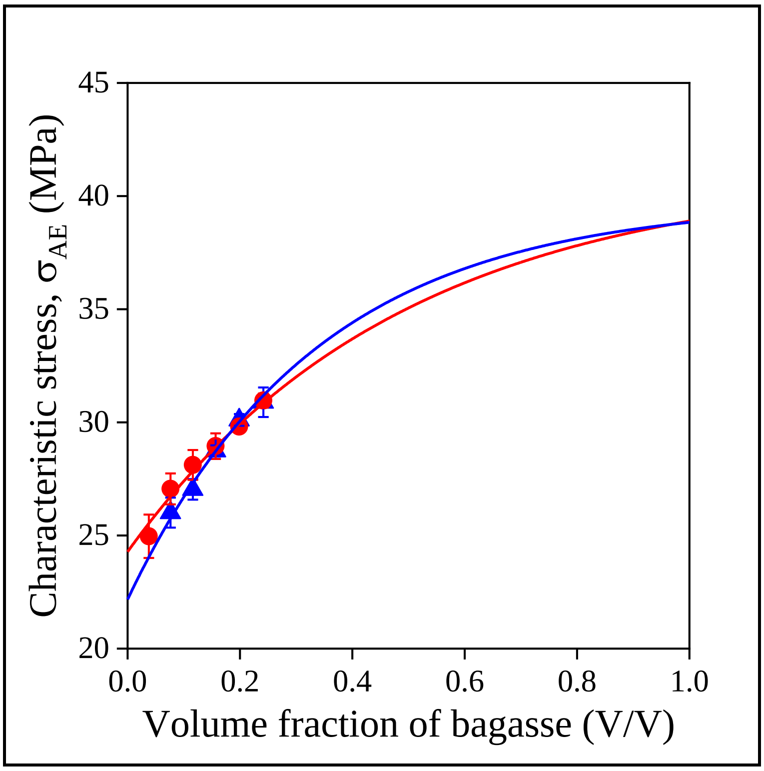

Fig. A3 *Determination of the inherent strength of the sugarcane bagasse fibers used in this study. Symbols:* (●) *short,* (▲) *long fraction.*

7. Page 7: In the section about properties, the effects of aspect ratio, orientation and adhesion are mixed. It is hard to determine how exactly the authors aim to interpret the complex relationship between these three factors and composite properties. Some clarification would be desirable.

   The Reviewer might be right that the explanation could be clearer in the property section. However, as he or she describes, the relations are complex among the three factors. Moreover, we have limited differences in aspect ratio, very little information about orientation and have quantitative values only for adhesion. Originally, we hoped for more information on particle characteristics and aspect ratio, that is why we separated the fibers into two fractions, but the different extent of attrition in the case of the two batches hindered the drawing of further conclusions about the effect of these factors. Nevertheless, the conclusions of the paper on fiber attrition, interfacial adhesion and the correlation of local processes and macroscopic properties are clear and unambiguous and we hope that also useful for the public.
8. Figure 7: The composition-dependence of characteristic stress should be discussed in

some detail.

We thank the Referee for the remark, checked the section discussing characteristic stresses and extended the text to explain the differences among the correlations presented including both the specific processes and the values belonging to them.

9. Page 11: The observation according to which initial particle dimensions have a very limited effect on properties due to attrition during processing is an important one, and should be emphasized. On the other hand, the fact that coupling changes interactions and local deformation processes has been widely reported and should not be presented as new information.

   We agree with the Referee completely that less information is available about the attrition of natural fibers than on the effect of coupling on interfacial adhesion and local processes. We acknowledge this latter fact by referring to some papers published earlier. We do not dwell extensively on this question either, but discuss more the quantitative relationship of local processes and macroscopic properties. The discussion section (Section 3.5) consists of three paragraphs, the first discussing attrition, the second the correlation mentioned above and the third the role of fiber fracture in the increase of impact strength. We do not think that the discussion of the effect of coupling on adhesion and local processes is out of proportion.

10. My last major comment is admittedly highly subjective. Nevertheless, I believe the strict adherence to a certain structure, in which macroscopic properties, local processes, adhesion, etc., are discussed separately, does not necessarily serve clarity or readability. Although the form chosen by the authors is certainly logical, it is often difficult to follow their reasoning, since cause and effect are presented in different parts of the text. A more integrated interpretation of the results might be desirable.

    The Referee might be completely right that the results could have been presented differently. As he or she says himself or herself, the structure of the paper is logical and we thought that discussion must be divided into parts in order to facilitate reading and understanding. The discussion section was hoped to amalgamate the results presented in the preceding sections and to show the connection among the factors studied in the project. We might not have succeeded in this effort. However, we should like to stick to our own logic and thus did not modify the structure of the paper during revision.

11. **Minor comments.** Language of the manuscript is adequate, although it contains some grammatical and stylistic errors. A few typical examples are listed below. a) Incorrect word order in some sentences. b) The relative pronoun "which" is frequently, and incorrectly, used instead of "that". The two are not interchangeable. c) Missing hyphenation in some compound words, e.g. fiber-reinforced polymers (page 2)

    We thank the Referee for calling our attention to grammatical and stylistic errors. We checked the paper, took all minor comments into account during revision and corrected all errors according to our best ability. Word order was changed where necessary, the use of "that" and "which" was revised and missing hyphenation was corrected.

12. Page 3: "In fiber-reinforced polymer composites frequently local deformation processes take place which are related to or occur around the fibers." The phrasing seems a little awkward. The local deformation processes in question are always "related to" the fibres: they are either initiated by them or they happen to them.

    The sentence was modified to avoid ambiguity and make its meaning clearer.

13. Page 3: "Fiber fracture occurs more frequently in the case of strong interfacial interactions, when they are improved…" It is unclear what "they" refers to.

    This sentence was also rewritten to make the meaning of the pronoun clear. In fact, *the pronoun was deleted from the sentence completely.*

14. Page 4: "…the goal of this study was to prepare composites from a locally derived natural reinforcement, sugarcane bagasse fiber…" Locally derived where? Hungary of Indonesia?

    We apologize for the missing information. Naturally, the fibers came from Indonesia. The sentence was modified accordingly and the information is included now.
15. Page 4: "The bagasse fibers were obtained directly from the sugar mill." Which sugar mill? Can the authors be more specific?

    Similarly, the missing information on the sugar mill is included into the revised paper.
16. Page 4: "The ratio of MAPP/fiber was 0.1 in all composites." Is that weight ratio?

    The Referee is completely right, it was weight ratio. The experimental part was modified to include the information.

References

1. Dányádi, L., Janecska, T., Szabó, Z., Nagy, G., Móczó, J., Pukánszky, B.: Wood flour filled PP composites: Compatibilization and adhesion, *Compos. Sci. Technol.* **67**, 2838-2846 (2007).
2. Faludi, G., Link, Z., Renner, K., Móczó, J., Pukánszky, B.: Factors determining the performance of thermoplastic polymer/wood composites; the limiting role of fiber fracture, *Mater. Design* 61, 203-210 (2014)

Bartos András

Feladó: em.pote.0.725ced.c6736814@editorialmanager.com Polymer Testing <em@editorialmanager.com> nevében
Küldve: péntek 2021. április 2 10:23
Címzett: András Bartos
Tárgy: Decision on submission to Polymer Testing

Manuscript Number: POTE-D-20-00717R1

EFFECT OF PARTICLE CHARACTERISTICS AND INTERFACIAL ADHESION ON THE PROPERTIES OF PP/SUGARCANE BAGASSE FIBER COMPOSITES

Dear Mr Bartos,

Thank you for submitting your manuscript to Polymer Testing.

I am pleased to inform you that your manuscript has been accepted for publication.

My comments, and any reviewer comments, are below. Your accepted manuscript will now be transferred to our production department. We will create a proof which you will be asked to check, and you will also be asked to complete a number of online forms required for publication. If we need additional information from you during the production process, we will contact you directly.

We appreciate you submitting your manuscript to Polymer Testing and hope you will consider us again for future submissions.

Kind regards,
Ulf W. Gedde
Editor

Polymer Testing

Editor and Reviewer comments:

Editor: Thank you for your very careful revision and your response letter. We are delighted. Ulf W. Gedde
Reviewer #2: Thank you, my comments were addressed, I like the paper and it is suitable for publication.
Reviewer #3: The authors accepted most of my remarks and the reason for those which were not fully accepted, was acceptably explained. Recommendatkion ACCEPT

More information and support

FAQ: When and how will I receive the proofs of my article?
https://service.elsevier.com/app/answers/detail/a_id/6007/p/10592/supporthub/publishing/related/

You will find information relevant for you as an author on Elsevier's Author Hub:
https://www.elsevier.com/authors

FAQ: How can I reset a forgotten password?
https://service.elsevier.com/app/answers/detail/a_id/28452/supporthub/publishing/kw/editorial+manager/

For further assistance, please visit our customer service site: https://service.elsevier.com/app/home/supporthub/publishing/. Here you can search for solutions on a range of topics, find answers to frequently asked questions, and learn more about Editorial Manager via interactive tutorials. You can also talk 24/7 to our customer support team by phone and 24/7 by live chat and email.
