Manuscript: Effect of chemical treatments on chemical composition and properties of flax fibers

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Dear Prof. Petrović,

We appreciate the reviewer critical reading and useful comments to our manuscript. We carefully considered the comments and revised the manuscript as described below. We submitted the revised version with changes highlighted in red in the text and described below the point-by-point response to each comment.

Comments to the Author and authors' answers:

GENERAL

In general, the manuscript » Effect of chemical treatments on chemical composition and properties of flax fibers « presents an interested topic, but what was the main goal of this work respectively the scientific contribution?

The manuscript presents research on the chemical processing of flax fibers in order to clarify the effect of lignin and hemicelluloses removal on the morphology, chemical and physical properties of flax fibers. The effect of lignin and hemicelluloses removal on structure and properties of fibers such as jute,31 hemp26,32 and agave33 fibers, have been studied in detail, while such studies of flax fibers are still limited. Gained comprehensive knowledge should allow an increase in refinement quality and alteration of flax fiber properties in defined manner.

The manuscript has fundamental flaws in the science and writing. Significant additional work is needed to bring up to publisher quality. The comments are provided within the document, marked in yellow and summarize the major deficiencies, but does not address the all grammatical errors in the manuscript. Double click on the text marked in yellow will allow reading and/or answering of the provided comments. Figure quality is very poor. Figures have been corrected. If all recommendations are taking into account, the article is worth of publishing.

In my opinion, this manuscript should:

be published after major revision and additional review

The comments included within the manuscript:

Abstract:

lines 13-15: to long sentance - the word "properties" is used twice

According to this comment the sentence in question is corrected.

lines 17-19: this is alredy clear from the first sentance

We cannot agree with this reviewer comment, from the first sentence is only clear that these treatments lead to hemicelluloses and lignin removal, while in the sentence in question we specified that alkali treatment leads to hemicelluloses removal and chlorite treatment leads to lignin removal.

the efect of both treatments on the chemical composition and properties should be emphasis since this is the title of this manuscript

According to this comment we added data about effect of both treatment on the chemical composition and emphasis their effect on fiber properties.

**INTRODUCTION**

The introduction is rather too long - it should contain only the lliterature review on the topic dealing within the manuscript - (e.g. the 1st and 5th paragraph could be deleted9

According to this comment we deleted the 1st paragraph, as well as parts of 5th and 6th paragraphs.

p. 2, line 35: in what sence "significant" - needs to be clarified

According to previous reviewer suggestion 1st paragraph containing this statement was deleted.

p. 2, line 40: some literature cited is needed

Actually, literature concerning to "industrial applications" has been cited at the end of the statement, for the sake of clarity we cited literature after term industrial application: "...industrial applications,6-10 such as pulp and composite materials."

p. 2, line 45: in what sence - needs to be clarified

According to this suggestion we clarified our statement about fiber modification in order to impart special or modify existing fiber properties, such as hydrophilicity, hydrophobicity, sorption, antimicrobial, and other properties, and supported it with additional literature cited.

p. 2, line 57: unclear for what applications - needs to be clarified

According to this comment we clarified this statement.

p. 3, line 72: why emphasis both?

We left only the term sorption properties since this term include also water-fiber interaction.

p. 3, line 82: what changes? - above the variety of treatments were mentioned - some of them should be pointed out

Part of the 5th paragraph containing this statement is deleted according to previous reviewer's suggestion.

p.2, lines 93-94: treatments could not be used as methods to study structure...this is not correct - the author is strongly adviced to re-write this

We rewrite this sentence.

p.4, line 103: needs to be more specific - what is the scientific contribution of the manuscript since the author stated that both treatment methods have been alredy used for years (see page 3 lines 93-95) - this needs to be re-written

According to this suggestion we changed the last sentence of the 5th and the first sentence of the 6th paragraph of Introduction section.

p. 4, line 110: in which year and what were the climate conditions of growth since the quality of fibres depend on that

We agree with reviewer's comment that the quality of fibers depend on the climate conditions of growth. This is especially important in the case when you compare properties of fibers grown under different climate conditions caused by different year of growth or different climate area. Considering general comments that the length of the manuscript is not appropriate and the manuscript needs condensation, but also the fact that we used only long flax fibers obtained from the same harvest we deleted superfluous data concerning to starting material. Just to answer reviewer's question flax stems were pulled at the end of June 2005, and the climate conditions data were recorded and stored by Republic Hydrometeorological Service of Republika Srpska.

p. 4, line 112: how was this determined - needs to be added

As we explained above, we deleted superfluous data concerning to starting material.

p. 4, lines 113-114: which fibres were excatly used - the grown or technical ones - needs to be explained

As we stated, we used technical fibers, to be more precise we added word multi-cellular.

p. 4, line 114: how was this determined? needs to be written

According to this suggestion we made correction.

p. 4, line 123: was this performed until the constant conductivity was obtained?

We added asked data.

p. 5, Table I: why the duration of the treatment timeis differnt for NaOH and NaClO2 - as such it is difficult to compare the efect of time vs concentration - this needs to be explained

We are not sure that we understand this comment, since it is difficult to compare these two chemical treatments directly considering used concentrations (in the case of NaOH: 5,7, 10 and 18%, and for chlorite: 5, 10, 15 and 20 g/L, that corresponds to 0.5, 1, 1.5 and 2%) and duration of treatment (chlorite treatment as oxidative one is more effective - up to 60 min almost all lignin is removed, while in the case of alkali treatment even after 120 min only 70 % of hemicelluloses are removed), we compared them considering achieved changes in chemical composition and structure, and consequently their effect on properties.

p. 5, line 132: the metods used could be divided as methods for determining chemical composition changes, structural and physical and sorption properties - in this way in could be more clear

According to this suggestion we reorganized experimental section.

p. 6, line 160: was it tested only in dry or also in wet conditions - needs to be explained

The tensile strength of single flax fibers was determined in dry state only. We did not make any correction in the manuscript since the common rule if it is not otherwise stated that means fibers are characterized in dry state only. Furthermore, we did not characterized flax fibers in wet state considering well known fact that natural cellulose fibers, including flax fibers, have higher tensile strength in wet conditions (up to 10%). The tensile strength in wet state is very important parameter for man-made cellulose fibers having lower tensile strength in wet state (up to 50%).

p. 8, line 189: what dis it meant by this?

We made correction in order to make sentence in question clear.

p. 9, line 210: what samples?

We corrected it in order to precise about which samples we discussed.

p. 9, lines 210-214: this needs to be more detailed explained

According to this suggestion we made additional explanation.

p. 9, line 233: by some modified samples the alpha cellulose content is higher compared to non-treated one - why - this needs to be explained

According to this suggestion we made additional explanation.

p. 9, line 235: in latter sentance it was steted "without decrease" - this is rather confusing and the author is strongly advised to re-write this

We are not sure that we understand this comment, since in "latter sentence" we stated "without increase" not "without decrease". We pointed out "without increase" since increase in copper number means higher aldehyde group content and cellulose degradation. Furthermore, term "without increase" cover both cases: no changes of copper number and decrease of Cu-number. Last one we discussed further. To avoid any further misunderstandings we corrected this sentence.

p. 10, line 241: why in general there is higher decrease of cooper number by naOH tretament compared to naOCl2 treatment? What is the influenece of concentration and temperature?

According to this suggestion we made additional explanation.

p. 10, line 242: below anre not explained the results of fineness - rather the morphology - this needs to be clarified

As the first we changed subtitle to correspond more to discussed results. We discussed with the intention changes in morphology and fineness of modified fibers together since they are interrelated. Changes in fiber surface morphology have effect on divisibility of flax fibers and consequently fiber fineness, as we discussed.

p. 12, lines 273-277: taher too long sentance - needs to be shortned to make it more understandable

We divided sentence in question to be more understandable.

p. 12, line 288: this is for treament tiome 120 min - needs to be clarified

Actually, increase in fineness observed for fibers modified with 18% NaOH at room temperature, treatment time longer that 30 min (i.e. 60 and 120 min), was explained by some disorientation of the fibrils. Fig. 2c was cited as illustration for this phenomenon. From the discussion it is clear that this is true for 60 and 120 min treatment time.

p.12, line 290: any explanation why is strongly adviced to the autor to provide it

According to this suggestion we made additional explanation.

p. 13, line 301: based on mentioned there is no need to provide both results i.e. ISV and crystallinity

We believe that both ISV and CrI data should be provided in paper, and we will try to explain why.

Considering sorption properties it is important to know ISV as a measure of the accessibility of fibers to aqueous solutions. On the other hand for structure consideration it is important to know CrI. Indeed these two values are correlated and knowing ISV, crystallinity index can be calculated using equations:

 / %

But from given equation it is clear that knowing only ISV value you cannot get, without calculation, information about changes in CrI; for example ISV changes from 74.3 to 90.0 (increase for 15.7 units), but CrI changes from 82.0% to 78.2% (decrease for 3.8%) (Table III).

Furthermore, considering general comments that the length of the manuscript is not appropriate and the manuscript needs condensation we decided to present Table III as Supplementary material (Table S-II).

p.13, line 307: this is dificult to undestand - if the noncellulosic material is removed the sorption properties areusuly indicating the improved sorption properties i.e. ISV increase - as it was discussed before along with ISV results

Removal of easily accessible noncellulosic material such as hemicelluloses and lignin is followed by decrease in sorption properties, as we discussed in the case of chlorite treated samples. In the case of alkali treated samples, removal of noncellulosic material is followed by the polymorphic transformation of cellulose I to cellulose II, later one responsible for increase in sorption properties. As we explained exception was observed in the case of alkali treated samples only for low concentration (5%), treatment not followed by the polymorphic transformation. Obtained results have been compared and discussion supported by literature data.

According to this suggestion we made additional explanation.

p. 13, lines 317-319: the latter should be more detailed described - the influence of the T, t and concentration since this is the main topic of the manuscript as I understood

According to this suggestion we made additional explanation.

p. 14, lines 325-327: not clear enought between whom?

According to this suggestion we made additional explanation.

p. 15, line 343: hydrophobic layer does NOT enable better hydrophilicity - it makes it rather worse - this needs to be re-written

According to this suggestion we corrected this sentence.

p. 15, line 344: why the modifyed fibres show worse water retention value - they should show improved sorption properties since the results of ISV value were improved indicating the increase amorphous phase and thus proving a capable area for retention of water - this is rather confusing - it is strongly advised to the author to re-writte this

Reviewer is right, fibers having higher ISV should show improved water/moisture sorption properties. But here we are discussing water retention, properties determined not only by accessible hydroxyl groups but also by all water absorbing and holding surfaces, cracks, and cavities. Our results show that alkali modified fibers are incapable of keeping water molecules in their structure during centrifugation due to the simultaneous removal of hemicelluloses which were placed in interlamellar layer and amorphous areas. Furthermore, similar decrease in water retention capacity of alkali treated fibers was also observed during the alkali scouring of flax fibers, as well as alkali treated hemp fibers, and reported in literature24,25,27.

p. 16, line 387: why the tensile strenght were not pšerformed also in wet conditions - needs to be explaind why?

We explained it above answering reviewer's comment about method for determination of tensile strength.

p. 17, line 398: This needs to be explained why!

According to this suggestion we made additional explanation.

p. 17, line 410: any proof of that?

According to this suggestion we made additional explanation.

p. 18: sopme general conclusion is missing e.g. i) what was the treatment effect on the structural properties, chemical composition, sorption,..ii) was there any influence of T, t and concentration - and the explanation why, iii) why the obtained knowledge accuired within presented study is usefull

According to this suggestion we added some general conclusions.

Does the manuscript contain enough significant original material?:

no

We believe that we clearly explain what was the main goal of this work respectively the scientific contribution and improve manuscript to be bring up to publishing quality.

Is the manuscript clearly and concisely written?:

no

According to the reviewer's suggestions we made additional explanations and corrections.

Are the conclusions adequately supported by the data?:

yes

Does the manuscript give appropriate credit to related recent publications?:

yes

Are the references appropriate and free of important omissions?:

yes

Is the length of the manuscript appropriate?:

no

Considering general comments that the length of the manuscript is not appropriate and the manuscript needs condensation we decided to present details about chemical treatment scheme and list of samples (Table I), iodine sorption value and crystallinity index (Table III), and whiteness index (Fig 6) of untreated and treated flax fibers as Supplementary material (Tables S-I and S-II, Fig.S-1).

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Does the manuscript need condensation or extension?:

yes

Please see above.

Is the quality of the figures (including legends and axes labelling)

satisfactory?:

no

Figures have been corrected.

Are the nomenclature and units in accordance with SI?:

yes

Are the English grammar and syntax satisfactory?:

no

The manuscript is carefully edited and grammar mistakes corrected by a fluent English reader/writer.

Yours sincerely,

Mirjana Kostic