July 8, 2019

Polymers, MDPI
Dear Professor,

We are resubmitting our revised paper entitled *Cellulose fibres modification via a hybrid chemical modification* by, Stefan Cichosz, Anna Masek with a request to reconsider it for publication in *Polymers*.

We have carefully considered the Editor and Reviewers' comments. The manuscript was revised exactly according to these comments. The list of responses to the reviewer’s comments and corrections made in the manuscript is attached.

The manuscript has not been previously published, is not currently submitted for review to any other journal, and will not be submitted elsewhere before a decision is made by this journal.

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Yours sincerely,

Ph. D., D.Sc. Anna Masek
Answers to reviewer #1 comments

Reviewer #1: Manuscript entitled “Cellulose Fibres Hydrophobization via a Hybrid Chemical Modification” presented by authors is good work and can be accepted for publication after minor corrections. Few comments are as follows.

The comments are listed below.

1. **Author need to improve quality of figure: 1.**
   
   **Answer:** Fig. 1 has been deleted according to the comment of Reviewer #3.

2. **Author need to correct typos throughout the manuscript (like °C).**
   
   **Answer:** We have taken Reviewer’s comment into consideration. All typos were corrected.

   
   **Answer:** We are very grateful for gathering such a broad range of references that may enrich our article. All of mentioned research studies has been cited: Nowadays, cellulose fibres are the subject of various research studies [1-12]. In the following article the stress is put on the moisture content influence on the mentioned biopolymer properties.

4. **Why authors have chosen ethanol and either hexane for solvent exchange.**
   
   **Answer:** We are grateful for this comment. Nevertheless, data is available in Supplementary Materials. It is mentioned in 2.2. section - *Moreover, solvents*
of different polarity employment influence has been examined (ethanol and hexane chosen on the basis of sorption experiments – data available in a Supplementary Materials).

5. **How and why different solvents contribute to the higher yield of the modification process; author need to provide short expalination in the abstract part of the manuscript.**
   
   **Answer:** This is a valuable advice. We have added this information in Abstract section: *With the use of Fourier-transform infrared spectroscopy (FT-IR), it has been proved that the employment of different solvents may contribute to the higher yield of the modification process as they causes rearrangements in hydrogen bonds structure, swell the biopolymer and, therefore, affect its molecular packing.*

6. **Author need to include some interesting data in the abstract part to make it more interesting for the readers.**
   
   **Answer:** We are grateful for this comment. The abstract part has been improved and some interesting results were presented, e.g., *Furthermore, according to the thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC), the improvement in fibres thermal resistance has been noticed, e.g., shift in the value of 5% temperature mass loss from 240 °C (regular modification) to 306 °C (while solvent employed).*

7. **What is the % of lignin present in the cellulose sample used by authors in their experiments because properties of cellulose is highly effected by lignin and hemicellulose contents.**
   
   **Answer:** As it is mentioned in experimental section, the Arbocel® UFC100 Ultrafine Cellulose for Paper and Board Coating from J. Rettenmaier & Soehne was the type of cellulose used in this research. It is in a powder form (white, odourless) and its density is about 1.3 g/cm³. It is insoluble in water and fats. Nevertheless, this material exhibits a high water binding capacity (even at high temperatures and shearing forces). Its average fibre length is about 8 μm. pH value varies between 5-7.5.

   It is a commercially bought cellulose. Nevertheless, we are not able to present the data considering the lignin and hemicellulose content.
8. **Author need to label FT-IR peaks with functional groups in the figure 3.**
   
   **Answer:** We are thankful for Reviewer’s comment. However, data considering the assignment of the peaks presented in Fig. 3 is available in Table 3. There are the exact values given. The table has been improved.

9. **TGA Fig 9, Mass change should be replaced by Wt%.**
   
   **Answer:** The mistake has been corrected.

10. **What is the residual weight in each TGA graph author need to provide explanation.**
    
   **Answer:** We thank Reviewer for paying attention to this problem. Some more information has been given, e.g., *Moreover, the residual weight in each TGA graph are pyrolysis reaction products, which were not oxidized, as the experiment has been performed in an argon atmosphere. A complexity of cellulose thermal degradation was widely described in literature [58,59].*

11. **Why ethanol employment highly contributes to the lowering of water absorption ability of cellulose author need to provide proper explanation with suitable reference.**
    
   **Answer:** We are thankful for Reviewer’s comment. The explanation has been given: *In each of mentioned modification process ethanol was used. It is believed that due to the strong interactions between cellulose and ethanol, in comparison with hexane-fibre interaction forces, the biopolymer is swelled well and the fibre structure becomes more available to the solvent. As a consequence, higher amount of water is desorbed and the fibre reactivity is improved – the effect of cellulose structure opening and reorganization of hydrogen bonds [26,28,71–73]. Therefore, it may be claimed that the employment of this solvent contributes to the UFC100 of a low moisture content obtaining.*

12. **Author need to compare their results with previously reported similar research.**
    
   **Answer:** As presented chemical modification is a new approach, unfortunately, it is hard to compare the results with different research studies, especially considering the water content. Nevertheless, when possible, the comparison is given, e.g., *Furthermore, in order to assess the differences between the modified samples*
in a more precise way, the activation energies of decomposition steps have been calculated (Table 7). Presented data may be a useful tool in describing the water content in analysed samples and goes in a well correspondence with the literature [60]. Literature says that various chemical treatments [68–70] and solvent exchange [25] could alter cellulose fibres water uptake ability.