Dear prof. Dr. Dušan Mijin,

Thank you very much for your efforts in processing our Manuscript submitted to your Journal (Hemijska industrija). The authors decided to revise manuscript and to resubmit the revised manuscript to Hemijska industrija.

We have thoroughly contemplated Reviewers remarks and corrected our manuscript according to their comments and suggestions. Authors would like to thank for useful remarks that have improved the manuscript and hope that each important Reviewers’ point was taken into consideration.

In attached file is a final copy of manuscript with changes to the text highlighted in yellow. Point by point list of responses to the Reviewers’ comments are given in following text.

We hope You will be satisfied with quality of corrected manuscript and responses to Reviewers’ comments.

Sincerely Yours,

Jelena Rusmirović

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**Reviewers’ comments**:

**Reviewer E**:

**General comments**:

In this work organic solvent free laboratory and industrial procedure for preparation of oxidized starch using hydrogen peroxide as oxidant was described. The influence of hydrogen peroxide concentration, type of used catalyst, reaction temperature and type of used plasticizer on the properties of synthesized oxidized starch was examined. The paper provides useful results for environmentally friendly starch oxidation method. Therefore I recommend this paper for publication in journal Hemijska industrija after following corrections:

**Comments**:  
1. Page 3, line 50, the text “The starch also” should be changed to “The starch powder also”.

**Answer**: The authors agree with this comment. The correction is inserted in the revised manuscript (yellow marker).

2. Page 7, line 134, the text “The details of copper ricinoleate” should be changed to “The details of copper ricinoleate synthesis”.

**Answer**: The authors agree with this comment. The correction is inserted in the revised manuscript (yellow marker).

3. Page 7, lines 139-141, the text “After that, in the obtained mixture was added catalyst (FeSO4·7H2O, CuSO4·5H2O, copper citrate or copper ricinoleate) at quantity shown in Tab S1 (Supplementary materials)” should be changed to “After that, in the obtained mixture catalyst (FeSO4·7H2O, copper citrate or copper ricinoleate) was added at quantity shown in Tab S1 (Supplementary materials)”, because the data for CuSO4·5H2O catalyst was not shown in Table S1.

**Answer**: The authors accept this comment. The correction is inserted in the revised manuscript ( yellow marker).

4. Page 8, lines 171-172, the text “final product which can be application in a paper industry” should be changed to “final product which can be used in the paper industry”.

**Answer**: The authors accept this comment. The correction is inserted in the revised manuscript (yellow marker).

5. Page 9, the text in lines 179-181 should be deleted and the text in lines 188-190 “The hydrogen peroxide concentration, reaction temperature and presence of plasticizer RA/DIPT/ESO/ELO/ESFO have significant effect on the carboxyl and carbonyl contents.” should be changed to “According to the results of optimization of starch oxidation determined by response surface methodology (RSM), given in Supplementary material (Tab S3), the hydrogen peroxide concentration, reaction temperature and presence of plasticizer RA/DIPT/ESO/ELO/ESFO have significant effect on the carboxyl and carbonyl contents.”.

**Answer**: The authors accept this comment. The text is deleted while the sentence is corrected and inserted in the revised manuscript (yellow marker).

6. The successful oxidation of starch molecules by this procedure was proved by FTIR spectroscopy, so the results obtained by FTIR spectroscopy should be given at the beginning of Results and discussion, before the results obtained by other characterization methods. Also, the Figure 1a and

the text related to FTIR spectra of synthesized plasticizers (lines 273-280) should be moved to the Supplementary material, because the details of their synthesis, as well as the results obtained by elemental analysis and NMR spectroscopy are given in Supplementary material.

**Answer**: Authors accept this suggestion. The results obtained by FTIR spectroscopy are moved to the beginning of Results and discussion while the Figure 1a and the discussion related for Figure 1a are moved to the Supplementary material (yellow marker).

7. Page 17, lines 336-344 and Table S9, “DSC profiles of both, native and oxidized/modified starch, display one value at about 310 °C (Fig 3c), which represents starch gelatinization endothermic peak. The amount of heat necessary for gelatinization is provided by exothermic creating of starch-lipid complex, which additionally reduces ΔH value (Tab S9). Conversely, the great decrease in ΔH value for Exp 16 in comparison to native starch suggests that the introduction of fatty acid inhibits melting of starch crystallites [24].”, the endothermic peak at 310 °C cannot be assigned to the starch gelatinization because the starch gelatinization occurs at the much lower temperature (see Ref. 44). At this temperature, thermal degradation of starch occurred, as it can be seen from the TG and DTG curves.

**Answer**: Thank you for this comment! The correction is inserted in the revised munuscript (yellow marker).

8. This paper is scientific paper. It is not common that scientific paper contains any economical calculations.

**Answer**: Accepted. The economic analysis is deleted.

9. Supplementary material, page 8, “The viscosity of oxidized starches obtained at laboratory level was determined using capillary viscometer methods” should be changed to “The viscosity of oxidized starches obtained at laboratory level was determined using capillary viscometer”.

**Answer**: Comment accepted and incorporated in the revised munuscript (yellow marker).

10. Supplementary material, page 8, “The viscosity of oxidized starches obtained at industrial level was measured using Brookfield method (Brookfield DV-II + Pro Viscometer) (B Method) according to [6] with some changes:” should be changed to “The viscosity of oxidized starches obtained at industrial level was measured using Brookfield rotational viscometer (Brookfield DV-II + Pro Viscometer) (B Method) according to already described procedure [6] with some changes:”.

**Answer**: Comment accepted and incorporated in the revised munuscript (yellow marker).

11. Supplementary material, page 11, Fig. S5b, one of the axes is unmarked.

**Answer**: Thank you for this comment! Y-axis on Fig. S5b is marked.

**Reviewer F**:

**General comments**:

The manuscript “Inovative Environmentally Friendly Method of Starch Oxidation by Hydrogen peroxide” by N.Karic et al describes process for industrial starch oxidation. Although the manuscript donates a number of valuable data it needs improvements before acceptance.

**Comments**:

• Title of the manuscript is too general and it should be changed in such way to reflect the content of the manuscript.

**Answer**: The authors accepted the suggestion and changed title of the manuscript (yellow marker).

• In abstract authors claim: “To develop environmentally friendly processess, i.e. replacement of aforementioned hazardous technologies, a simple and efficient laboratory and industrial procedure for preparation of oxidized starch using hydrogen peroxide as oxidant is developed.” It turns that these authors are the first which use hydrogen peroxide as oxidant for the preparation of oxidized starch.

**Answer**: The authors accepted the comment and incoporated the additional information in abstract (yellow marker).

• The authors should clearly state what the novelty in the introduction part is.

**Answer**: The authors accept this comment completely. The corrections are inserted in the revised manuscript (yellow marker).

• The authors should check the names of compounds. See line 127 “diisopropil tartarate”.

**Answer**: The authors accepted this comment and check the names of all compounds in the revised munuscript (yellow marker).

• The authors should check text (also SM) for errors like in line 127 “was done added”.

**Answer**: The whole manuscript has been checked and the appropriate corrections have been made.

• There is no need for the techno-economic analysis in the scientific paper.

**Answer**: Accepted. The economic analysis is deleted.

• It is unusual to use materials from “local grocery store” in the scientific paper. Please at least give the producer and the specification of wheat starch from label.

**Answer**: Authors agree and additional the the producer and the specification of wheat starch in the revised munuscript (yellow marker).

• Please check characterization data for the synthesized compound especially NMR data (e.g. ricinoleic acid, methyl group).

**Answer**: Thank you for this comment! The characterization NMR data for the synthesized compound has been checked and the appropriate corrections have been made.

• Please check Fig. S3 and decomposition of H2O2.

**Answer**: The authors accepted this comment and Fig. S3 has been checked. In the hydrogen peroxide oxidation reaction a hydroxide ion and a hydroxyl free radical are formed. The hydroxyl radical formed oxidizes an alcohol group forming a radical. This radical reacts with ferric ion, and hydrogen peroxide, affording a carbonyl group. Hydrogen peroxide decomposes inevitably to oxygen and water (reference: [22] Parovuori P, Hamunen A. Oxidation of Potato Starch by Hydrogen Peroxide. *Starch/Staerke*. 1995; 47: 19–23; and [23] Tolvanen P, Mäki-Arvela P, Sorokin AB, Salmi T, Murzin DY. Kinetics of starch oxidation using hydrogen peroxide as an environmentally friendly oxidant and an iron complex as a catalyst. 2009; 154: 52–59.).

• The authors should check the names of compounds in Serbian abstract like: vodonik-peroksid, bakar(II)-sulfat, bakar(II)-citrat, bakar(II)-ricinoleat, diizopropil-tartarat.

**Answer**: Accepted and corrected.