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Prof. Dr. Jasna Djonlagic

Sub Editor

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Title: **“Effect of nano- and micro-alumina fillers on some properties of poly(methyl methacrylate) denture base composites”**

Thank you for giving us opportunity to improve our manuscript. We have tried our best to make the necessary corrections according to the Reviewers’ Comments. The itemized corrections are appended below. With all these corrections and improvements, we would like to resubmit this manuscript to your esteem journal for your kind consideration. We are really hoping that the manuscript will be acceptable for publication in the Journal of the Serbian Chemical Society.

Thank you.

Yours sincerely,

Fathie A.M Kundie

On behalf of my co-authors.

Universiti Kebangsaan Malaysia

**Answers to Reviewers' Comments**

**Reviewer #1**

**Comments:**

* **In this paper authors investigated influence of micro and nano sized fillers on mechanical properties of denture base materials. The results are presented and discusses well.**

**Response to the comments**

Thank you very much for this very encouraging comment. It is a great honor to our hard work. Once again, thank you very much.

* **The main objection is related to the used nano particles. Namely authors stated that they used nanoparticles with average particle size of 7 nm but SEM images of neat particles showed agglomerated particles with diameter between 50-150 micrometer.**

**Response to the comments**

We have added TEM micrographs (Figure 1b) that show the actual particle size of nanoparticles.

* **If these agglomerates were kept during mixing and ultrasonic bath treatment and are presented in final product then, in my opinion, it is not correct to use term nano. Thus, authors should investigate morphology of composite materials and if agglomerates of nanoparticles are present in composite material then word nano should be excluded from this paper.**

**Response to the comments**

The dispersion of nanoparticles in the matrix can be clearly seen in FESEM micrograph (Figure 2b) of the composites

* **If authors investigate morphology of prepared composites using micro and nano fillers and correlates mechanical properties and water absorption with filler particle size, this paper would be suitable for publishing in JSCS.**

**Response to the comments**

Yes, our research is about the effects of silanized Al2O3 micro- and nanoparticles fillers on the mechanical properties, water absorption, and solubility of PMMA denture base composites.

1. **In the Introduction authors described how micro and nano fillers affect various properties but authors excluded specific values. For example, authors stated ‘’Ellakwa et al. (2008) investigated the effect of Al2O3 powder addition on the flexural strength of PMMA denture base; the flexural strength of the composite significantly increased after incorporation of 10% Al2O3’’, here I would suggest to add exact values, like …the flexural strength of the composite increased more than xx% after incorporation of 10% Al2O3… I would suggest authors to make this change in the majority of cited work.**

**Response to the comments**

The suggested information (percentages) have been added to those references that include control material values.

* **Also, I would suggest authors to add following two references in the part where they discuss poor properties of neat PMMA denture base materials: D.C. Smith, The acrylic denture base — mechanical evaluation of dental poly(methylmethacrylate), Brit. Dent. J. 111 (1961) 9–17; The effect of the accelerated aging on the mechanical properties of the PMMA denture base materials modified with itaconates, Hemijska industrija 2011 Volume 65, Issue 6, Pages: 707-715.**

**Response to the comments**

The suggested references have been added in the manuscript.

14. D. C. Smith, *Br. Dent. J.*, **111** (1961) 9

15. P. M. Spasojević, M. Zrilić, D. S. Stamenković and S. J. Veličković, *Hemijska industrija* **65** (2011) 707

1. **Line 111. Authors wrote ...typical molecular weight of 996 000… please add unit for molecular weight and state is it number average molecular weight or weight average molecular weight.**

**Response to the comments**

The sentence has corrected as “…PMMA with weight average molecular weight of Mw = 996,000 g/mol (product no. 182265, Sigma Aldrich, USA)…”.

1. **Line 118. Authors wrote …26N-0801G has an average particle size of 7 nm with a specific surface area of 2.1 m2/g… as I mentioned earlier in my opinion it is not correct to state that this particles have average particle size of 7 nm if they are agglomerated. The fact that particles specific surface area is 2.1 m2/g means that they are agglomerated, otherwise specific area would be much bigger >200m2/g.**

**Response to the comments**

TEM micrographs (Figure 1b) that show the actual particle size of nanoparticles has been added.

1. **Table 1. I would suggest authors to change samples codes in order to make it clear what is the filler content from sample code name. I would change M1 to M0.5; M2 to M1; M3 to M2; M4 to M5 and so on... Same with the N samples.**

**Response to the comments**

The samples codes in Table 1 have changed as mentioned in the comment.

1. **Equation 1. Explain what is y**

**Response to the comments**

The symbol has explained.

where P= load at peak (N), S = span length (mm), a = notch length (mm), y = the geometrical correction factor, t = specimen thickness (mm), and w = specimen width (mm).

1. **Line 311. Remove the following sentence ‘’According to 311 ISO 1567-2000, the water absorption value of denture base materials should be less than or 312 equal to 32 μg/mm3 after 7 days of storage.’’ because there is almost the same sentences two sentences before.**

**Response to the comments**

The repeated sentence has deleted.

**Reviewer #2**

**Comments:**

1. **The authors are dealing with PMMA matrix reinforced using micro and nano alumina fillers. The surface of the inorganic particles was treated in order to improve the contact among phases in the composite. The introduction is well written and has a broad literature presentation even if this field is so waste that there could always be added some more references but this part of the manuscript is well done.**

**Response to the comments**

Thanks so much for the words of encouragement.

1. **In the introduction the statement about the aim of the research should be better presented so that the reader should be prepared what to find in the body of the paper.**

**Response to the comments**

We have added the following sentences in the Introduction part.

“In the present study, we sought to improve the mechanical properties of the PMMA denture base. To this end, Al2O3 micro- and nanoparticles fillers were chosen as the preferred additives to this material. Different ratios of silanized Al2O3 micro- and nanoparticles fillers were employed, and the aim of this study was to evaluate their effects on the mechanical properties, water absorption, and solubility of PMMA denture base composites.”

1. **Line 132 the authors say that they used the sonication for 10 min. The usual duration time for this operation is about 30 min. It seems that this time is short for to arrive to an appropriate result of dispersion.**

**Response to the comments**

Based on previous research works, sonication for long time leads to monomer evaporation. For examples in these two papers; 5 and 10 min were used, respectively.

(1) Žukas, Tomas, et al. "The influence of nanofillers on the mechanical properties of carbon fibre reinforced methyl methacrylate composite." Materials Science 18.3 (2012): 250-255.

(2) Ash, Benjamin J., et al. "Mechanical properties of Al2O3/polymethylmethacrylate nanocomposites." Polymer Composites 23.6 (2002): 1014-1025. Sonication for long time leads to monomer evaporation.

1. **Line 182 the best specimens from both series were used for this water absorption test. The authors don’t define what defines the best specimen is that the mechanical properties, or aspect of the specimen or some other characteristic.**

**Response to the comments**

Best specimen is based on the best mechanical properties.

1. **Line 202 the figure capture but the comment is for the figures. It seems that micro particles are smaller than the nano particles. I understand that those are agglomerates but could the authors give us un idea about the real size of individual particles. The bar in the figure is not clear enough could this be made more clear in the image.**

**Response to the comments**

Figures 1 (a) and 2 (a) were checked and improved. The bar now is clearer.

1. **Line 211 The authors identify several peaks that could be of some interest, but discuss only 2 of them Why are other peaks pointed out and what do they correspond to. I would add to the image the short descriptive of the groups identified and point out what is interesting for the contact achieved with the matrix using the surface prepared in this way. Does this preparation give something on the aspect of the particles? Is it possible to make some more comments about what was achieved and why this is important for the particles characteristics.**

**Response to the comments**

Thank you for your valuable comment on the improvement. The other peaks have identified and the result has connected to the other properties.

1. **Line 231 the presentation of data obtained from mechanical testing in a table is inappropriate. It is really difficult to follow the results and to compare them, some visualization would be appreciated, at least by me.**

**Response to the comments**

A diagram has been added instead of the table.

* **On the other hand, was it possible to obtain some information about the flexural modulus from those measurements? Generally, if the displacement and the** **force are measured some more data could be calculated from the result.**

**Response to the comments**

The flexural modulus result has been added.

1. **Line 277 the data discussion is based mostly on speculation and no data about the morphology of the fracture surface of the specimens are given. Are those microcracks visible, are there any voids in the specimen visible under some optical microscroscopy? SEM could be of some use too. The authors did not give any analysis of the composite some images of the prepared samples, on the macro scale even are not present. The crack surface could give a lot of responses and it would be useful to analyze it using some microscopy. Also the dispersion of the particles in the composite is not even mentioned or presented. No microstructure analysis was done.**

**Response to the comments**

FESEM micrograph of the (Figure 2b) fracture surface has been added and discussed.

1. **The thermal characterization for the composite giving any idea about the interaction particle/matrix is not present.**

**Response to the comments**

The TGA result (Figure 7) has been added and discussed.

* **The authors could also use the FTIR analysis to describe the interaction particles/matrix. The article, especially the results presentation should be carefully revised and presented with more detail so the speculation in results discussion could be omitted and supported by more data on the material itself.**

**Response to the comments**

The results have been improved, discussed and supported.

1. **I recommend that this article should be rewritten and resubmitted to this journal if the authors wish so.**

**Response to the comments**

All the mentioned items (*i.e., text and missed results*) have been corrected, improved and given in the new resubmission manuscript.

1. **The introduction and literature review are well done and give a lot of data about what was done in this field. This is the best part of the article.**

**Response to the comments**

Thank you once again. It's appreciation for the work that we are doing; it's also great to hear words of encouragement.

1. **In the preparation of samples my personal opinion is that the ultrasonication time was not long enough to enable the dispersion of the particles. No argument about what was achieved with this dispersion are given.**

**Response to the comments**

The results showed increased fracture toughness. FESEM for fracture surface shows the dispersion of nanoparticles in the matrix has been added.

1. **Mechanical testing data are given in the form of table but it is difficult to follow what happens like that. It would be easier to follow if the data were presented in the form of a diagram or some visual interpretation of data.**

**Response to the comments**

A diagram has been added instead of the table.

1. **The article misses a lot of analysis from the point of view of microstructure, the character of the bond achieved in the composite and the dispersion of the particles in the matrix.**

**Response to the comments**

FESEM for fracture surface shows the dispersion of nanoparticles in the matrix has been added.

1. **As stated in comments all those parts are missing and the article is unacceptable without this analysis. I suggest that the article should be rewritten and resubmitted to the journal to a novel revision.**

**Response to the comments**

All the missed parts (*i.e., FTIR peaks, TGA, flexural modulus and SEM*) have been added and given in the new resubmission manuscript.