Dear Dr Radak,

We are sending to you the revised article: “Application of non-thermal plasma reactor for degradation and detoxification of high concentrations of dye Reactive Black 5 in water”. We have revised this manuscript along the lines that reviewers have suggested. All corrected or added text is marked with red color in the manuscript.

**REVIEWER A**

1. **p.5 lines 141-143 in sentence "This type of reactor was successful in phenol removal from water38 and also in dye removal, pesticide removal, medicaments removal and surfactants removal." statements about reactor efficiency in" dye removal, pesticide removal, medicaments removal and surfactants removal" should be supported by citing appropriate references.**

Answer: In line with the Reviewer's request reference list is now extended for two references under numbers 39. and 40.

# 24. B. P. Dojčinović, G. M. Roglić, B. M. Obradović, M. M. Kuraica M. M. Kostić, J. Nešić, D. D. Manojlović, *J. Hazard. Mater.* 192 (2011) 763

25.M. Jović, D. Manojlović, D. Stanković, B. Dojčinović, B. Obradović, U. Gašić, G. Roglić, *Chem. Eng. J.* **260** (2013) 1092

39. M. Marković, M. Jović, D. Stanković, V. Kovačević, G. Roglić, G. Gojgić-Cvijović, D. Manojlović, *Sci. Total Environ.* **505** (2015) 1148

40. M. M. Aonyas, J. Nešić, M. Jović, M. Marković, B. Dojcinović, B. Obradović, G. M. Roglić, *CLEAN-Soil, Air, Water* **43**(2015) 1

**2. p.5 in "Methods of analysis" subsections "Decolorization efficiency" and "Determination of decolorization efficiency at high concentrations of Reactive Black 5" should be merged in order to omit some repetition.**

Answer: Subsections “Decolorization efficiency” and “Determination of decolorization efficiency at high concentrations of Reactive Black” are now merged into subsection “Determination of decolorization efficiency at high concentrations of Reactive Black 5” (now stated on page 5, line 157-166 and page 6 line 167-168).

**3. The same sentences in Introduction part (p.4 lines 110-113 sentences " The degradation efficiency and specific degradation products were monitored by UV-VIS spectrophotometry, COD and ion chromatography (IC). The toxicity of the dye solution was determined before and after the plasma treatment by means of Artemia salina test organisms.") and RESULTS AND DISCUSSION (p.8 lines 239-241 sentences " The degradation efficiency and specific degradation products were monitored by UV-VIS spectrophotometry, COD and ion chromatography (IC). The toxicity of the dye solution was determined before and after the plasma treatment by means of Artemia salina test organisms.").**

**This repetition should be omitted.**

Answer: On page 8, line 239-241, two sentences: “The degradation efficiency and specific degradation products were monitored by UV-VIS spectrophotometry, COD and ion chromatography (IC). The toxicity of the dye solution was determined before and after the plasma treatment by means of *Artemia salina* test organisms.” are deleted from the section Results and Discussion. On page 7, line 234, part of the sentence is now added: “and the toxicity of the solutions before and after plasma treatment was obtained using the test organisms *Artemia salina*.”

**4. p.8 lines 247-48 sentence "Decolorization efficiency was measured 5 minutes and 24 hours after the plasma treatment." should be deleted as this is described in experimental part. Instead of that sentence obtained results 5 minutes and 24 hours after the plasma treatment should be commented.**

Answer: On page 8, line 247-248, the sentence: “Decolorization efficiency was measured 5 minutes and 24 hours after the plasma treatment.” is now deleted. Decolorization of Reactive Black 5 24 h after the plasma treatment is greater than after 5 minutes. Decolorization of dye is mainly attributed to OH radicals as highly reactive and species with short life-time in the experiment, and in smaller part to other molecular and longer living reactive species present (ozone, H2O2) which show their effect after 24 h from plasma treatment.

The following text is added on page 8, line 249-250: “Higher decolorization after 24 h, in comparison with decolorization 5 minutes after the plasma treatment, has been observed because of longer living oxidative species (such as O3 and H2O2) remaining after treatment in the dye solution which promote further degradation and decolorization after degradation mainly caused by OH radicals.”

**5. p.8 lines 261 and 263 in "... than OH mean free path ..." and "... were closer to OH and increased ..." should be written OH radical or OH˙.**

Answer: Sentences in lines 259 – 263 are corrected and now state: “OH radical non-selectively reacted with surrounding molecules and, as its life-time was very short (order of micro-seconds in a gas phase or order of nano-seconds in a liquid phase), it reacted with the dye molecule only if it was closer than OH radical mean free path. Otherwise, OH radical will react with water molecule. The increase of dye concentration provided that the dye molecules were closer to OH radical and increased the possibility for their interaction and, consequently, the increase of the decolorization efficiency.”

**6. Fig. 4 caption "Dependence of decolorization of the concentration reactive dye Reactive Black 5 (c0=  40-1000 mg L-1) for applied energy densities of 66 and 242 kJ L-1 (24 h after the treatment). " should be corrected, for example " Dependence of decolorization ON (or FROM) the reactive dye CONCENTRATION... ".**

Answer: Fig. 4 caption is corrected and now states: “Dependence of decolorization from the reactive dye Reactive Black 5 concentration (c0=40-1000 mg L-1) for applied energy densities of 66 and 242 kJ L-1 (24 h after the treatment).”

**7. p.9 lines 280-283 sentence is not clear and should be corrected.**

Answer: The sentence on page 9, lines 280 – 283 is corrected and now states: “It was determined that reactor capacity had linear dependence of applied energy density in the tested concentration range for which effectiveness of 50 % decolorization is achieved after 5 minutes and 24 h after the plasma treatment (Figure 5b).”

**8. p. 9 in subsection " Determination of COD " data concerning to experimental procedures and earlier explained in Experimental section should not be repeated. For example lines 297-99 should be deleted, they just confuse reader.**

Answer: On page 9, line 297-299, sentences: " The use of microwave digestion at high temperature and pressure (up to 100 bar) shortened the time needed for oxidation of organic substance during the sample preparation for COD analysis. For all three experiments results were 900 ± 10 mgO2 L-1." are deleted.

**9. p.10, line 314, some → same**

Answer: In sentence on page 10 line 314 “some” is now changed to “same”.

**10. on page 11, lines 357-59 sentence is not clear and should be corrected.**

Answer: Sentence on page 11 lines 357 – 359 is corrected and now states: “For applied energy density of 66 kJ L-1, the concentration of oxalates was increased for dye concentrations up to 200 mg L-1, while it dropped with further increase of dye concentration.”

**11. In title and abstract in Serbian, the name of the color Reactive Black 5 should not be translated, it should be just written in italic.**

Answer: In title and abstract in Serbian, the name of the color Reactive Black 5 is written in original and formatted as italic text.

**REVIEWER C**

1. **The reasons for using such high concentrations of the dye (up to 1 g/L) should be mentioned. Is there any correlation between these values and the concentrations is real wastewater?**

Answer: Reason for using such high concentration of the dye is for determining the DBD reactor capacity, and for evaluating of DBD reactor usage in potential industrial accident situation. The sentence has been added on page 8, line 239-240.

1. **In the experimental section:**

**-       The gaseous atmosphere should be specified; it is not clear from the figure if the plasma reactor is open or closed, or if a gas is introduced in the system.**

Answer: Reactor used is an open type of reactor, which does not use any gas except air. Sentence has been modified on page 4, line 133-134 in order to clarify: "Coaxial DBD reactor used in this research was designed as a non-thermal plasma reactor which is an open type of reactor operating in the air at an atmospheric pressure and it is described in Kuraica *et al.*37 and Dojčinović *et al.*24  "

1. **The initial pH and conductivity of the solution should be mentioned. It should also be stated if the solutions were prepared with distilled water or tap water.**

Answer: Initial values for pH and conductivity are measured and their values in following ranges: pH from 8.44 (for 40 mg L-1 solution) to 10.45 (for 1000 mg L-1 solution) and conductivity from 32 (for 40 mg L-1 solution) to 722 μS cm-1 (for 1000 mg L-1 solution). Appropriate sentence regarding pH and conductivity of initial solutions has been added on page 5, line 160. Initial pH value of the solutions was not adjusted because in our previous papers,24,35 adjusting of initial pH value had no effect on degradation of dyes and 4-chlorophenols (now stated on on page 5, line 160).

Solutions were prepared by dissolving of the dye in deionized water, which was already written on page 5, lines 159 – 160.

1. **In the results section:**

**-       In the end of page 8, it appears that the degradation of the dye is attributed solely to OH radicals. Is there any evidence that the other reactive species formed in plasma (e.g. ozone) do not contribute to degradation?**

Answer: Degradation of the dye is mainly attributed to OH radicals as most reactive species in the experiment because it has the highest oxidation potential. There are, of course, other reactive longer living oxidative species present (ozone and H2O2) which may still react after treatment and promote further degradation and decolorization after the degradation mainly caused by OH radicals (now stated on page 8, line 249-250).”

1. **It is written (lines 267-269) that “oxidation species at lower concentrations of the dye were used just for decolorization and at higher concentrations evenly for decolorization reactions and for oxidation of intermediates”. It is true that Fig. 3a shows the faster decolorization at lower concentrations, however, the competition between the parent compound and its degradation products exists for all dye concentrations. I believe the assumption that the oxidative species selectively react with the parent compound when its concentration is low (while at high concentration they equally react with the dye and its by-products) is not supported by the experimental data.**

Answer: The authors agree with Reviewer's comment. Oxidative species equally react with dye and its byproducts both in low and high concentration of the dye. In text in the manuscript on page 8, lines 267 – 269 now stands: “Based on results presented in Figure 3a, it can be concluded that oxidation species at both lower and higher concentrations of the dye are used evenly for decolorization reactions and for oxidation of intermediates.”

1. **Line 272: the expression “DBD reactor capacity” should be defined.**

Answer: DBD reactor capacity can be defined as compound concentration at which specific energy stops decreasing even the compound concentration is still being increased. Sentence on page 9, line 272 has been modified: “Specific energy can be useful for DBD reactor capacity estimation which represents concentration of dye at which specific energy stops decreasing even the compound concentration is still being increased.”

1. **The evolution of COD removal as a function of concentration (Fig 7) shows an oscillatory behavior for the energy density of 242 kJ/L. Is this behavior an effect of some physico-chemical phenomena, or is it within experimental errors? Perhaps showing error bars in the figures would be helpful.**

Answer: This type of behavior is attributed to experimental error. Measurement uncertainty for this method was estimated following EURACHEM / CITAC Guide CG 4 “Quantifying Uncertainty in Analytical Measurement”. Measurement uncertainty can be expressed as VALUE ± COMBINED UNCETRAINTY (%) \* VALUE. Combined measurement uncertainty is 15 % for this method. From previously mentioned formula it can be seen that error bars based on measurement uncertainty overlap for Δ COD values for dye concentration of 200 and 500 mg/L. This confirms that this type of behavior is in limits of experimental error.

In order to clarify Figure 7 was changed to show error bars based on estimated measurement uncertainty, and following sentence was added in text on page 10, line 310: “Variation of Δ COD values for the energy density of 242 kJ L-1 shown in Figure 7 lies within measurement uncertainty (± 15 %) for COD determination method thus having no effect on overall data interpretation.”

1. **Some comments on the higher decolorization observed at 24 h after the end of plasma treatment as compared to the decolorization determined immediately after plasma treatment would be useful.**

Answer: The following text concerning decolorization difference after 5 minutes and 24 h has been added on page 8, line 249-250: “Higher decolorization after 24 h, in comparison with decolorization 5 minutes after the plasma treatment, has been observed because of longer living oxidative species (such as O3 and H2O2) remaining after treatment in the dye solution which promote further degradation and decolorization after degradation mainly caused by OH radicals.”

Hoping that we have readjusted our manuscript to the demands of Journal of the Serbian Chemical Society,

Kind regards,

Dr Biljana Dojčinović