

Response to Reviewer 3

The author reviewed several electrochemical detection methods for nanoparticles using printed electrodes. The manuscript highlights advances of electrochemical strategies and the examples of different techniques incorporated with the printed electrodes for nanoparticle detection. In general, the topic of the manuscript is interesting and appropriate to the review for Biosensors. However, the contents in the manuscript should be more systematic and informative. There are several specific major/minor changes I would recommend and the following items need to be explained and corrected in a revision.

I would like to thank the reviewer for the good reception of our manuscript and for the valuable comments raised. I have tried to improve the manuscript as suggested, and more specifically, my answers are the following:

First of all, it is likely that the authors just wrote down each paragraph based on just one or two papers and put them together without good interconnection. This is unacceptable for a review paper. The author should work on revising the manuscript to have a good flow.

The review has been organized in different sections following different electrochemical detection strategies. Then, in every section there is an initial introduction paragraph and then every paragraph describes different types of nanoparticles/reactions/detection. For instance, section 2.1 is organized in one introduction paragraph and one paragraph for different types of nanoparticles (gold, silver, quantum dots...) since the electrochemical strategies are usually different and independent between them. Having said that, I honestly think that there are fair interconnections between the different sections and paragraphs in many cases. For instance, and not limited to only these ones:

* reasoning why the direct electrochemical detection of silver nanoparticles is easier than gold nanoparticles (lines 181-185). This interconnects paragraph from gold nanoparticles and silver nanoparticles (section 2.1).

* comparison between detection of QDs using direct or digestion methods, which are described in different sections.

* Initial lines of section 2.2 describe why the digestion method could be different from the direct method described in section 2.1.

Anyway, I tried to reorganize some short paragraphs to reduce the feeling that they only come from one or two references. I hope that the reviewer can see this as an acceptable issue since this is more like a style concern and not a scientific one.

In page 1, lines 9 and 11, the author would use different sequence works, instead of repeating the similar words. Revise the sentence with removal of either “On the one hand” or “On the other hand.”

I have corrected the manuscript as suggested by the reviewer.

In page 1, line 14, suggest revising the sentence, “In this review, I will introduce...” and avoiding the first person voice (I, me, mine), but use the passive voice in scientific writing.

I have followed the recommendation by the reviewer and changed all the manuscript to passive voice. Nevertheless, I would like to mention that this is a choice of style and that the first-person voice is totally adequate in modern scientific writing.

In page 1, line 31, some critical literatures related to nanomaterials (e.g. peptide nanostructures, graphitic carbon nanomaterials) used for electrochemical biosensors are missed. They should be introduced to develop the solid rationale and contribution of the present study. The suggested articles are as follow: Biosens Bioelectron, 2012, 38, 295-301; Chem. Mater., 2013, 25, 2803-2811

I thank the reviewer for the suggestion to add these references. However, these references are not significantly related to the scope of the review or the sentences in lines 26-32 since, in that part, nanomaterials with potential uses in commercial products are described. The suggested references: 1) use nanomaterials in a biosensing device and 2) describe synthesis of a very specific graphitic nanomaterial. These are not relevant to the manuscript at all, and therefore, I have decided not to add them.

In page 2, lines 52 and 59, suggest removing “very” It sounds subjective.

I have removed that word from the manuscript.

In page 2, line 86, revise the sentence to be a passive voice.

Passive voice is now used in the manuscript.

In page 2, line 87, the author should introduce types of printed devices or electrodes, as they are major contents in this review.

I have added some lines to introduce the type of printed electrodes which are reported in the reviewed works. I have also added more information about the type of device in every electrochemical detection strategy described throughout the manuscript.

In page 3, line 103, the reasons to choose the strategies over other electrochemical techniques should be clearly stated. For instance, what are the advantages of the selected methods for nanoparticle detection, compared to other electrochemical techniques (e.g. EIS, electrochemical impedance spectroscopy, CV, etc.)?

This review describes different general strategies for electrochemical detection of nanoparticles reported in the literature using printed devices. These strategies can be mainly organized in direct detection, detection after digestion step, electrocatalytic detection, surface-enhanced methods and nanoparticle impacts. However, these strategies are not directly related to electrochemical techniques, and it is not possible to compare a direct detection strategy with, for example, EIS. In some cases, authors have compared different strategies for detection of the same nanoparticles, and I have mentioned that in the manuscript in the relevant sections. I had already mentioned the actual electrochemical technique applied (voltammetry, amperometry, etc.) in some cases throughout the manuscript and I tried to mention more details about the specific electrochemical techniques in the revised version.

In page 3, line 111, the author should provide an overview table in terms of different electrochemical techniques, target detection nanomaterials, quantitative values regarding detection performance, types of printed device and electrodes.

I thank the reviewer for this suggestion. However, I honestly think that a Table would not be very valuable in this case due to the heterogeneous strategies used for electrochemical detection of nanoparticles reviewed in the manuscript. In some cases, these strategies are used for nanoparticles in solution, other for nanoparticles adsorbed on electrode and in most cases in biosensing applications. There is no usually quantitative analytical information about the nanoparticle detection in the latter since authors usually only provide information about biosensing sensitivity. Furthermore, in cases where the nanoparticles are detected in solution, there are very different ways to report the analytical performance (for example: particle concentration, metal concentration within the nanoparticle, weight concentration). Therefore, the comparison between them would not be very informative. The scope of the review has been to highlight different strategies that lead to the possibility to detect a wide range of different nanoparticles using printed devices, but I did not focus

on quantitative information because of the heterogeneous way to report analytical information by different authors. I have tried to mention in the text the type of printed devices/electrodes used in the different strategies described throughout the manuscript.

In page 4, line 145, how close the AuNPs contact to the electrode surface? Any references or quantitative data should be presented in the manuscript.

I have added some references where the influence of the distance between nanoparticles and electrode surface was studied in detail as suggested also by other reviewer.

In page 11, lines 405-507, suggest revising the sentences to avoid using “in this case” repeatedly.

In page 15, line 591, “have” should be replaced with “has.”

In page 16, line 611, corrected as “because of”

In page 16, line 617, “I am sure that” sounds subjective. Revise the sentence to be a passive voice.

In page 16, line 621, suggest replacing “many” with “various”

I have modified the manuscript as suggested by the reviewer.