SAIP2017 submission ID 174 revised corrections

Layout

The layout revisited for Figure 3.1 and 3.2 and corrected accordingly. Thereafter, the layout was accepted.

Referee 1

- 1. Introduction, line 2: sentence is now starting with "Semiconducting ..." instead of "The semiconducting ..."
- 2. Section 3, line 5: The sentence as whole has been re-written to "The undoped and the double doped (Co-In)-ZnO nanoparticles respond much greater to the NH₂ gas with high current as compared to the In and Co ZnO nanoparticles"
- 3. Section 3, line 8: "The physics being the ..." has been revised to "This suggest that the Co and In combined doping induce more inward strains in the ZnO nanoparticles matrix resulting in decreased volume."
- 4. Section 4, The author feels there is no need to change the sub-title "Conclusion" to "Summary"
- 5. Acknowledgements: "... grateful with ..." has been changed to "... grateful for ..."
- 6. I further re-confirm that no part of this manuscript has been published elsewhere before!

Referee 2

abstract

- For example: 2nd line, the author indicated that XRD study was used to probe the material BUT no rest of xrd is presented in the manuscript. If XRD study is conducted, it should be shown. Also stated that: 4th line: "diffraction from In-ZnO show additional diffraction peak". BUT not shown.
 XRD and SEM publication related to this work have been included as ref [17] on methodology. Statements about XRD and SEM in the abstract have been removed.
- 2. The sentence: 'In all the diffraction patterns observed ...', has been rewritten as: 'In all the current against time plots observed ...'
- 3. All 'Thes' have been removed in the abstract!
- 4. In fact, the abstract has been rewritten to cater for sensing application which is the main topic of the discussion.

Procedure

1. Sufficient information, for example how the gas sensing device is produced: if any interdigitated electrodes and a micro heater device were used, so that it can be repeated elsewhere. In general device design should be included. Example? How is

the ZnO deposited? Does the micro-hotplate sensor form thin film? What is the active area of the device?

Sufficient information about how the samples were tested for sensing has now been included. In **Procedure**: line 7 up to line 16 tackles this challenge. "The KENOSISTEC station equipment was used to characterise the prepared samples for NH₃ gas sensing at various temperatures (200-350 °C) and concentrations (5 -100 ppm). For gas sensing, the ZnO nanoparticles were mixed with ethanol before being coated on the surface of the aluminium substrate. The latter was placed between two Pt electrodes and a heater. The substrates coated with undoped, Co an In doped and Co-In double doped ZnO nanoparticles were then inserted in a chamber within the KENOSISTEC station machine. All the samples were subjected to concentrations of 5, 10, 20, 40, 60, 80, 100 ppm of NH₃ gas. The station was maintained at constant voltage of 5 V. The gas flow in and out was maintained at 5 minutes. Detailed procedures on how a KENOSISTEC sensing station operates can be found on the station website: <u>www.kenosistec.com</u> [18]."

The author stated that using "Williamson-Hall equation... particle size was determined". Again without proper xrd study!
Reference [17] also the procedure tackles the "Williamson-Hall equation ... particle size was determined" matter.

Gas sensing ability

1. The author studied the sensitivity of the sensor for temperature from 200 °C onwards. What about at lower temperatures? At least indicate that metal oxide does not react to ammonia gas at lower temperature.

Information indicating that metal oxide do not react with NH₃ gas at low temperatures has been included. In **Gas sensing applications**, starts from line 3 "It must be mentioned that there is no need to test sensing properties below 200 °C as the metal oxides like ZnO do not react with NH₃ gas at low temperatures [19]."

Figure 3.1 is confusing. The caption indicates that the current-time graph is for different concentration (from 5-100 ppm). What concentration is used to produce these curves? Even stated that, 1 paragraph, 10th line "in Figure 3.1 (a) it has been noted that.....as gas concentration is increased." But Figure 3.1 (a) is current vs time. Please comment.

Figure 3.1 caption has been corrected. It now reads "Figure 3.1: The graphs of current against time for the doped and undoped ZnO nanoparticles at various temperatures for the concentrations of 10 and 40 ppm." 1 paragraph, 10th line has been corrected to "In figure 3.1 (a) it has been noted that at 200 °C the current increases continuously without returning to the reference baseline as the exposure time is increased." 3. 4th line, the author associates the gas sensing ability of the undoped and Co-In doped ZnO to "gran size". But without showing the SEM, again this is meaningless.

Reference [17] in the **Procedure** section tackles this challenge.

1st paragraph, 9th line "All the samples seems to show uneven pattern from 300 °C. And reference [17] is given. Is the author talking his/her own data or that of ref? Need clarity.

That is from my data which was complemented by reference [17], which is now ref [20] ...

5. Why is at lower temperatures (Figure 3.2 (a) and (b)), the sensitivity if more or less constant for all the concentrations? It is to do with saturation? Or related to non-reactivity of ZnO at those lower temperatures? It is good if the author associate this "observations" with the facts/science.

Figure 3.2 (a) and (b) lower temperature behaviour of ZnO complexes has been heightened with scientific facts. Line 16 to 19 of paragraph 2, **Gas sensing applications** section: "Non-sensitivity of In-ZnO and (Co-In)-ZnO nanoparticles could still be attributed to less reactivity of metal oxide complexes with NH₃ gas at low temperatures. Possibly, In³⁺ ions in theses complexes contribute more in retarding the reaction of ZnO and the NH₃ gas [19]."

6. Why is the sensitivity of the co-In double doped show decrease in sensitivity as the concentration increased for a temperature from 200-300 °C? Why it increase after wards? (Page 2 last paragraph (Fig. 3.2 (c and d)). Is it to do with the effect of heat on ZnO? It is well known in ZnO community that 300 °C is an optimum temperature to enhance optical and electrical property of ZnO. What about the reactivity of these nanostructures with increasing temperature?

The last two lines of paragraph 2, **Gas sensing applications** section are addressing this case: "In the case of Co-In double doped ZnO sample, the grain size effect could come into play, as when the size of the nanoparticles are decreased greatly, the melting point also get reduced [22] "

Conclusion

A critical conclusion has now been written; all the figure references have been removed!

In summary all the suggested comments and reasoning has been included with references to all the XRD and SEM results mentioned in the text.