

Effect of additives: γ -Ray sensors based on ionizing organic nanocomposites

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ABSTRACT

We have developed ionizing organic based composites which have demonstrated great promise for radiation sensing. Nickel oxide has been proven as an active material for detecting high energy radiation. The oxidation state of unusual oxides such as nickel oxide in nanocomposites of ionizing organics changes much faster than thin film or bulk, and hence increases the sensitivity for radiation sensing. The resistivity of the oxide composite increases following sequential irradiation processes because of the decrease in holes' concentration. In this paper, we will present the effect of additional oxidizing agent on the morphology, processing and sensing of γ -ray by oxides - urea based nanocomposites. It was observed that addition of MnO_2 decreases the sensitivity. However, it recovers to original properties after irradiation much faster than undoped composites.

Keywords: γ -ray, nanocomposite, urea, oxides, detectors, sensors, radiation, exposure

1. BACKGROUND

There is a strong need for an innovative approach for development of the fast and low cost γ -ray and neutron sensors due to time consuming and very high cost involved in single crystal and helium based radiation detectors. Crystals of semiconductor materials such as cadmium zinc telluride (CZT), thallium arsenic selenide (TAS), thallium gallium selenide and (TGSe) and another binary and ternary halides and chalcogenides of mercury and lead, have been studied [1-3] in details. These crystals have shown great promise. However, growth and fabrication development is very time consuming. In addition to some halides such as hygroscopic materials LaBr_3 and some other materials, nickel oxide film has been used by Arshak and his coworkers [4-6]. These films of nickel oxide sputtered on silicon wafers have shown great promise. We have developed ionizing organic based composites [7] which have demonstrated great promise for increasing the sensitivity for γ -radiation sensing. Since the interaction of γ -ray with composites involves all three interaction processes; photoelectric effect, Compton scattering, and pair production, composites have better chance for enhancing sensitivity. The probability of the photoelectric effect where a γ -ray interacts with an atom resulting in the ejection of an electron from the atom is proportional to the atomic number of the absorbing element and inversely related to the energy of the γ -ray. It is expected that the resistivity of the oxide composite will increase as the result of irradiation processes because of the decrease in holes' concentration. In this presentation, we will describe the effect of manganese dioxide impurity on nickel based composite. These composites were synthesized using urea as the matrix material. Effect of addition on the morphology and performance of composite for γ -ray is described in this study.

2. EXPERIMENTAL METHODS

The preparation of composite was identical to that described in reference 7 for the ethylene carbonate and urea based composite.

2.1 Preparation of composite: We used as supplied urea by Sigma Aldrich. This was listed for 99.99% purity. The active gradients nickel oxide (nickel carbonate) was grinded a wig-L-bug and grounded to make uniform size particles. The particle size ranged between 100 nm. Urea was heated in a beaker on a hot plate well above its melting temperature. The melt was maintained for several minutes at that temperature. The active gradient of nickel and titanium oxide was added in the molten urea and stirred continuously for 15 minutes. Molten material was mixed with

nickel oxide and manganese oxide and then poured in a shaped container placed in ice cold water to freeze. As casted sample is shown in **Figure 1**. Morphology of the composite is shown in **Figure 2**.

2.2 Evaluation of γ -ray on properties: Cs-137 commercially supplied source was used to evaluate γ -ray detection. The resistivity of the composite was measured for the as prepared and radiated composites. Cs-137 was listed for 0.25 μ Ci, 30.2year half-life, β and γ -radiation. Both composites were radiated for 48 hours. Resistance at different voltage and different frequencies were measure. Data for 50 mV to 1 Volt and frequency range of 100Hz to 1000,000Hz are reported to evaluate the effect of frequency and applied voltage. **Figure 3** and **4** show the I-V characteristics of as prepared virgin and radiated composites.

3. RESULTS AND DISCUSSION

Figure 1 shows urea based composites containing NiO_2 as the active gradient and MnO_2 as an additive. When we compare the stability with un doped Urea- NiO_2 composite, MnO_2 doped sample has better mechanical properties for fabrication and silver electrode stability. Unlike the case of ethylene carbonate based composites, this composite did not show sign of rod or needle morphology. This helps in good mixing and ease in casting shaped sample. For electrical measurements, stability of several electrode materials was tested. Silver paste was stable and we used for the electrical measurements.

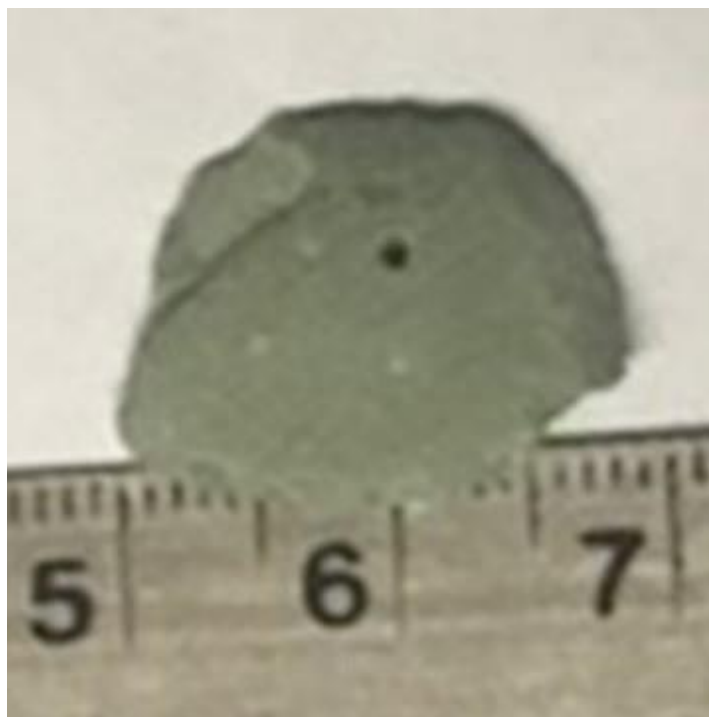
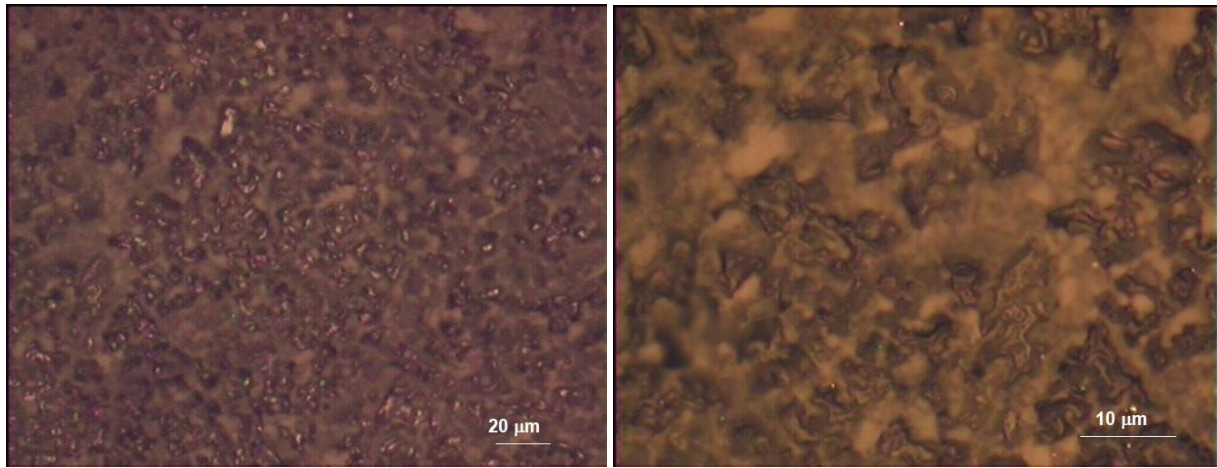


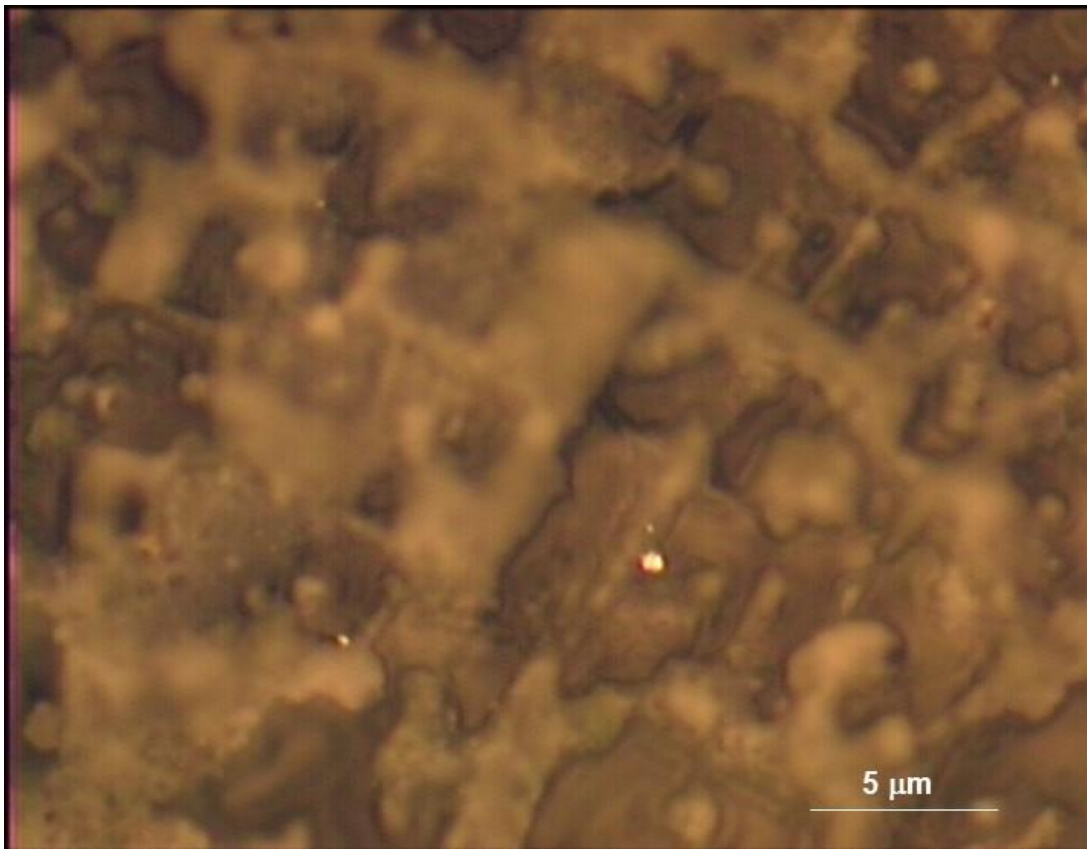
Figure 1. As prepared sample containing NiO_2 , MnO_2 and urea

The micro morphologies showed that shiny particles of oxides were distributed between small crystallites of urea. In some portion of the matrix, there were micrometer size particles and in some portion there were extremely small particles which consisted the composite. **Figure 2** shows the morphology of the sample at different magnification. The high magnification sample (c) shows some glassy behavior of the composite. As shown in Figure 2(b), the morphology appeared as colony structures in the early stage of curing. However, after few hours, we observed that grains disappeared for a large portion of the material and appeared glassy.



(a)

(b)



(c)

Figure 2. Morphology of the sample at different magnification. The high magnification sample (c) shows some glassy behavior of the composite

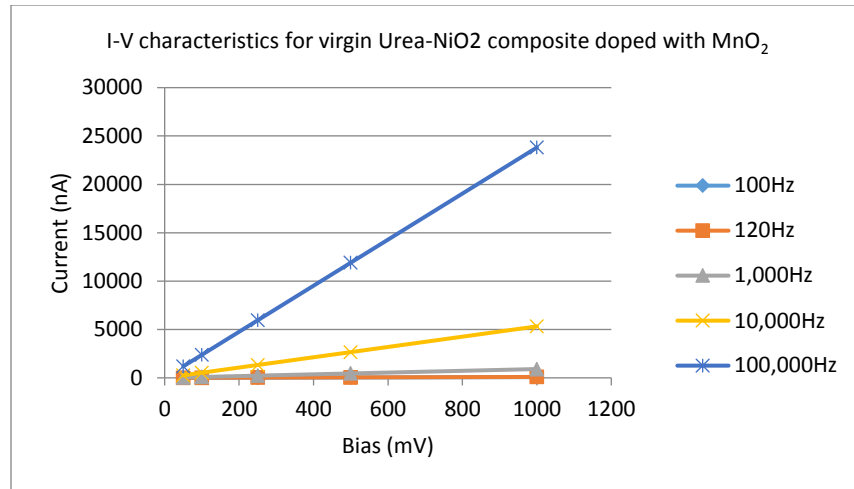


Figure 3. I-V characteristics of as prepared MnO₂ doped composite at different frequencies

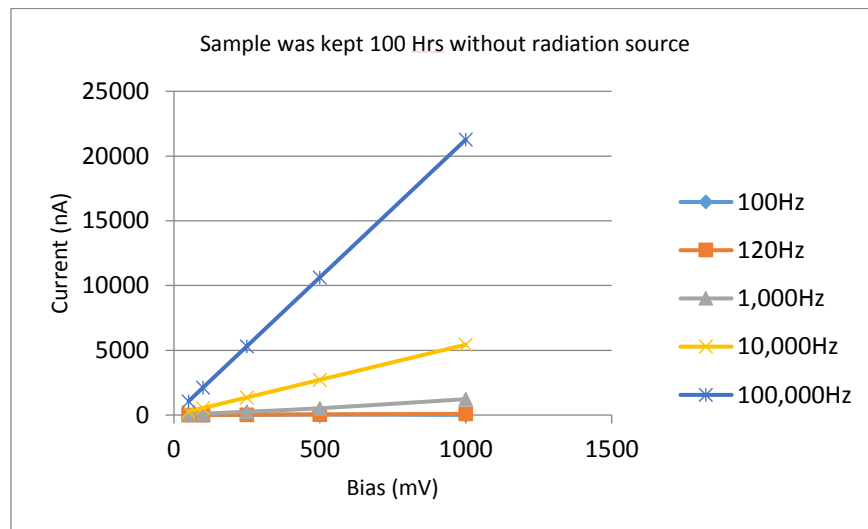


Figure 4. I-V characteristics of 100 hours aged MnO₂ doped composite at different frequencies

Figure 3 shows the current -voltage relationship for the freshly prepared composite at different frequencies. Same sample was kept at room temperature for more than 100 hours to evaluate its stability. The data observed for identical bias and frequencies are plotted in **Figure 4**. There was no significant difference in electrical properties which indicates that composite was stable and properties did not change. **Figure 5** shows results of the composites which was radiated for 48 hours. The resistance of the composite at different voltage and different frequencies was measure. Data for 50 mV to 1000mV, and the frequency range of 100Hz to 100,000Hz were determined to evaluate the effect of frequency and applied voltage. Nature of the curve was identical. However, resistivity was slightly lower than nonradiated composite. **Figure 6** show the I-V characteristics of as prepared virgin and radiated composites at 100, 000Hz. The difference in current is significantly lower compared to the sample which does not have MnO₂. When this sample was kept again in atmosphere, it achieved the current -voltage characteristic (**Figure 7**) of the original composite. This indicates that **Figure 7** which is result of the irradiated sample which was kept to recover in atmosphere after irradiation

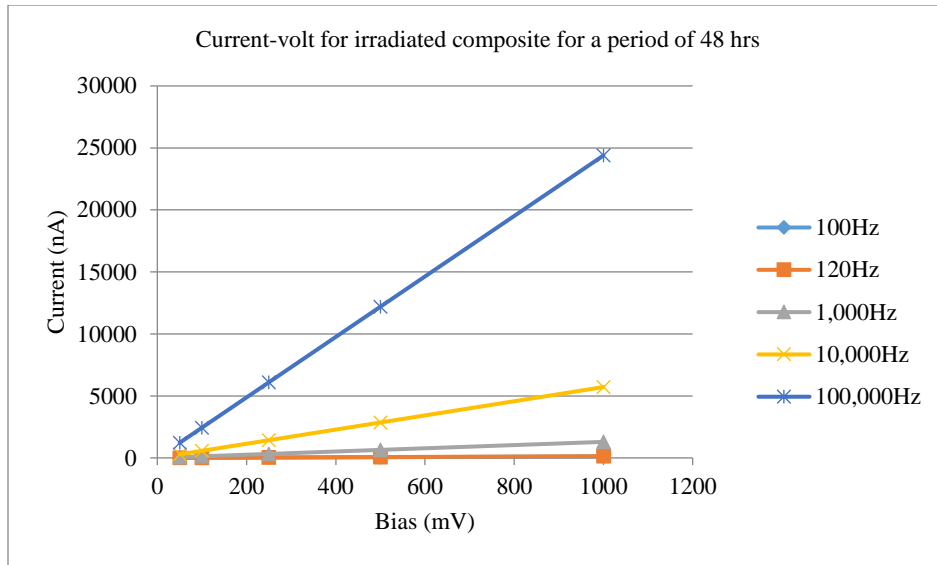


Figure 5. I-V characteristics of radiated MnO₂ doped composite at different frequencies

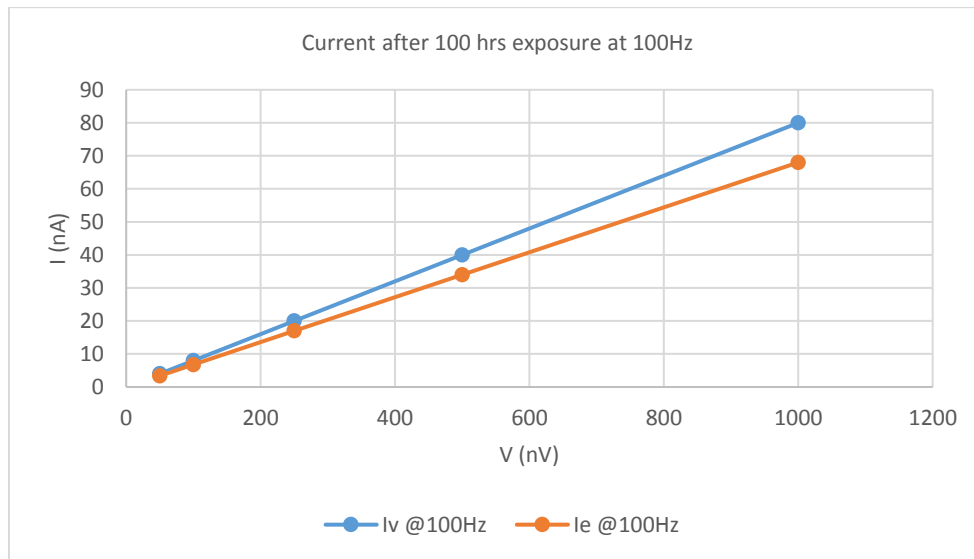


Figure 6. A comparison of I-V characteristics of as prepared and irradiated MnO₂ doped composite at 100Hz

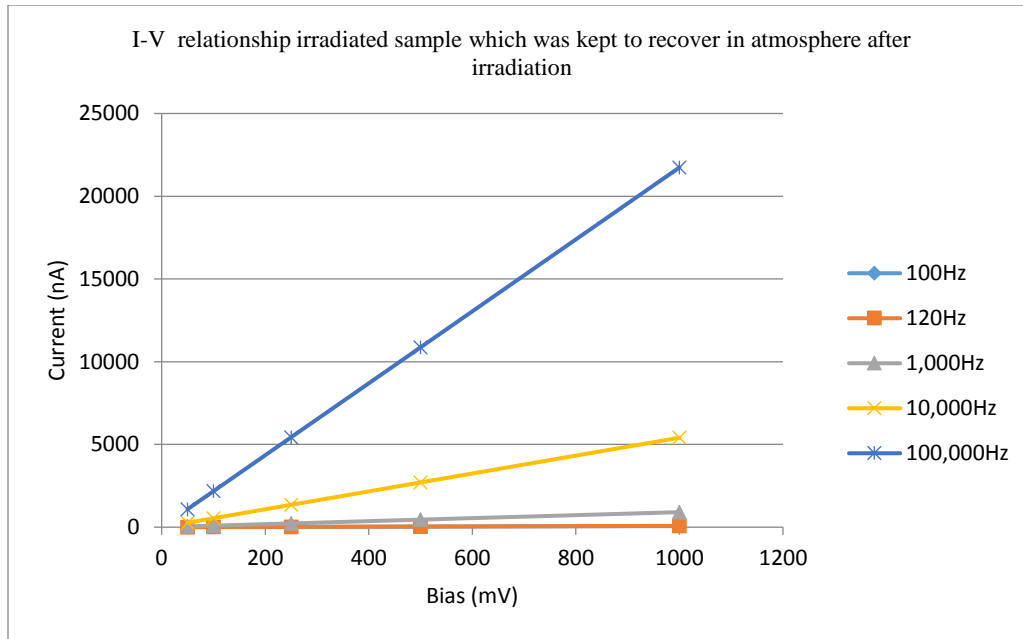


Figure 7. I-V characteristics of MnO₂ doped radiated composite which was placed in atmosphere for 48 hours

4. SUMMARY

Organic matrix doped with NiO₂ has demonstrated significant difference in current – voltage characteristics due to γ -ray radiation even at very low energy of the Cs-137 radiation. Addition of MnO₂ in the composite reduces the sensitivity. However, it was observed that composite recovers the original characteristics when kept in the atmosphere. Frequency had very pronounced effect on the resistivity and hence the current for a particular voltage used as bias.

ACKNOWLEDGEMENT

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5. REFERENCES

- [1]. Knutson, D.J., Singh, N.B., Berghmans, A., Wagner, B., Kahler, D., King, M., Bolotnikov, A., James, R., “Growth of CZT using additional zone-refined raw materials”, *Hard X-ray, Gamma-ray and Neutron Detector Physics*, edited by Ralph B. James, Arnold Burger and Larry Franks and Michael Fiederie, Proceedings of SPIE vol. 8507, 85070X-1 (2012).
- [2]. Kahler, D., Singh, N.B., Knuteson, D. J., Wagner, B., Berghmans, A., McLaughlin, S., King, M., Schwartz K., Suhre, D. and Gottlieb, M., “Performance of Novel Materials for Radiation Detection: Tl₃AsSe₃, TlGaSe₂, Tl₄HgI₆, *J. Nuclear Instrumentation and Methods in Physics Research Section A* 652, 183-185 (2011)
- [3]. Singh, N. B., Duval, W.M.B., Hopkins, R.H., Mazelsky, M., Fox, D. K., Gottlieb, M., Henningsen, T., Coriell, R., Glicksman, M., “Control of polytypes and line defects in radiation detector materials”, *Hard X-ray, Gamma-ray and Neutron Detector Physics*, edited by Ralph B. James, Arnold Burger and Larry Franks and Michael Fiederie, Proceedings of SPIE vol 8507, 8507N-1, 2012.
- [4]. Arshak, Khalil, Korostynska, Olga and Farah Fahim, “Various structures based nickel oxide thick film based gamma radiation sensors”, *Sensors*, 3 (2003) 176-186.
- [5]. Arshak, Khalil, Korostynska, Olga and Harris, J., “Gamma radiation dosimetry using screen printed nickel oxide thick film” Proceedings of MEIL 2002, 23rd International conference on microelectronics (2002) 357-360.
- [6]. Arshak, Khalil, Korostynska, Olga and Hickey, G. “Gamma radiation and Ozon sensing properties of In₂O₃:ZnO:SnO₂ thin film, Proceedings of SPIE 6589 (2007) 658916.
- [7]. Singh, N. B., et al., “Ionizing organic compound based nanocomposites for efficient γ -ray sensor”, *J. Optical Engineering* (Communicated)