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Assessment of the factors that affect the properties of Transparent Conducting Oxide [TCO]/ Perovskite Oxide based Nanocrystal.

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ABSTRACT

In this work, we assessed different growth mechanisms of transparent conducting oxide and perovskite oxide based nanocrystals [TCO/perovskite] for both the one in powdered form and the ones in thin film form along with such parameters being anticipated to influence their properties. In view of this the material, (TCO/perovskite) grown with different growth techniques were assessed in conjunction with other parameters such as annealing, doping and ligand in order to ascertained how they contribute in the modification of the properties material during the growth processes. However, based on this assessment and survey whereby different growth techniques were highlighted in conjunction with the use of the aforementioned parameters, it was seen after the analysis that the TCO/perovskite nanocrystal properties were in all cases influenced more by annealing dopant rather than growth mechanism. Also the assessment on the impact of the modification on their properties as it affects them as a major building block in many applications in electronic industries for production of electronic devices for both now, in this present dispensation and in the next generation of electronic devices production.

1. INTRODUCTION

Transparent Conducting Oxide which a times exhibits perovdskite oxide based nanocrystal characteristics has a unique structural and optical properties that makes it useful to so many applications. This one's of the reasons why such materials have become point focus in material science research over the years, coupled with the fact that most such material invariably belong to group III-IV type of semiconductors that are well known for their high technological applications in the field of optoelectronics and other solid state devices. In addition they have been tailored for use as transparent electrodes in solar cells and other electronic devices and this is because TCO thin films based has unusual unique properties of high visible wavelength transparency and metal like conductive characteristics. However apart from the aforementioned unique properties possessed by them, they can be as well tailored into n-type semiconductor materials by doping them with other elements such as transition element as the may contribute in the enhancement of the already possessed property of their visible transparency and high conductivity to higher percentage. These materials however have been discovered to have their several significant problems because of the difficulties encountered especially during their growth that hampers process that lead to realize a large area deposition of its film on a surface in order to achieve proper utilization of its properties enhance their applications in various areas one of which is particularly due to the scarcity and high cost of the materials and the technology involved in growing them apart from other problems of the characteristics optical absorption edge within blue-green region, chemical and temperature instability their likelihood of fracturing on flexible substrates. These are the problems and challenges posed by the growth of such class of thin films material to scientists and researchers.[1-5] These encountered problems that are encountered are being highlighted here to provide an insight to others beforehand so as to be cautious beforehand when embarking on research in the area that involves such materials so as to widen their horizon in their quest on the ways to precaution in overcoming these challenges and focus appropriate mechanisms that would enhance development the thin film appropriately with an improved properties that would optimize their applicability in the needed areas either by varying the parameters used for their deposition such as lingands or by annealing them at various temperature or doping them with other element or still by exploring a better result yielding deposition methods or conditions in way of preparation of the precursors. However we have to comment here that out of these well-known deposition techniques such as Chemical Vapor deposition Chemical Bath Deposition .sol-gel, Ionic Layer Adsorption and Reaction (SILAR), Atomic Layer Deposition, spray Pyrolysis, etc., that have been used to prepare some of these oxide based thin films especially CdO, CuO_2, SnO_2 ,ZnO, TiOx,[6-7] none of them has been found or reported in any case to be a better method amongst them yet, Rather each of them has been found to be peculiar on its own irrespective of the type of crystal it produces. However there must have been an anticipation that out of one of these techniques one may have advantage over others in deposition of any one of the particular TCO or Perovskite oxide based nanocystal [8-9] ,but that is not on general expectation because as generally observed, the advantage might only base on cost effectiveness, simplicity, reproducibility and optimization influence on the properties as it is often generally inferred due to the fact that the processes that are involves it enhances creating a precursor such as anion and cation that offer a better control over the deposition parameters such as PH, temperature may enhance the growth of some of these oxide based thin films with a glaring expectation to impact on the influencing the optimization of the structural and optical properties of these thin film'. In view of this we deemed it necessary to comment on the various growth techniques of TCO / Perovskite oxide nanocrystals and assess the applicability and the techniques that can be explored to enhance its properties that may likely lead to enhancement those properties that can optimize the applications the material nanocrystal in question to various uses. [10-11]

In general the deposition technique varies from various physical and chemical technique that may invariably lead to formation of perovskite oxide/TCO nanocrystal that is in form of powder or thin film formed on substrate for which the reaction mechanism may involve solid- state reaction that has three stages vis diffusion process, sintering and calcination coupled with milling process[12-18] and there is melt-solid reaction type that uses salt as a medium that involves heating of reactants and the salt above the melting point of salt in order to ensure that the particles are formed leads to the production of the salt solvent for use in the formation of the material[19-29]. However, the method which is our main focus is wet- chemical methods which has been found more popular and useful in deposition of TCO/peroviskite oxide based nanocrystal that involve some processes are highlighted here. They are hydrothermal, sol-gel of which the former involves heating of an aqueous suspension of insoluble salts a system where it crystallize to a desired phase and then subsequently form fine and homogeneous nanocrystal with a controllable size distribution and morphology, while the latter case alkoxide-hydroxide sol- precipitation methods and chemical bath methods [30-41]. A brief processes of the duo are for the growth nanocrystals briefly outlined below.

2. HIGHLIGHT ON GROWTH MECHANISMS

The brief highlight of some of the popular deposition techniques that have been utilized in depositing TCO/Perovskite nanocrystal either in powdered form or as thin film are to be outlined here after which some of the results obtained will analyzed in order to assess the properties of the crystal.

2.1 Hydrothermal Method

The hydrothermal method is one of the useful technique for synthesizing perovskites and the method is found to depend on solubility of minerals in hot water under high pressure, and the method has been used for many syntheses of perovskites for catalytic using various advancements approach The method enhances control of the particle size which carried out by controlling the reaction temperature, pH, time, and concentration of reactants. For instance, hydrothermally synthesized bismuth ferrite (BFO) nanoparticles at a low temperature of 180°C within 1 h is different from the one when compared with solid state reaction process, because the former yielded submicron crystallites of BFO with enhanced homogeneity unlike the latter method synthesized $LaFeO_3$ via a hydrothermal microwave-assisted synthesis at a relatively low temperature of 240°C and pressure of 60 bar, where the precursors were mixed in deionized water with the addition of KOH gradually while the system is being continuously stirred. The presence of microwave as the heating source assisted in an enhanced crystallization rate of nanoparticles. Also in another example, synthesized $BiFeO_3$ using nitrates of bismuth and iron via a hydrothermal technique where KOH was added as a mineralizer to assist in the co-precipitation of Bi_3+ and Fe_3+.After which the XRD result of the crystal shows that a single-phase cube-like $BiFeO_3$ was successfully synthesized and the investigation carried out on the effects of reaction time, KOH concentration, and organic dispersant on the $BiFeO_3$ particle morphology size indicated that the method of synthesizing perovskite oxide photo catalyst yields equally a good result. [42-44]

2.2: Sol–Gel Method

The citrate sol–gel method which is one of the sol-gel methods for instance that usually used to prepare nanosized materials is found to be defective due to fact that the application is limited its stability and its precursor system that invariably leads to difficulty in controlling the chemical composition of complex oxides and as such it is not always advocated for use when it comes to high precision growth as far as sol-gel technique is concerned .The method popularly used in sol–gel procedure is that which involves an aqueous medium that uses inorganic salts and a chelating agent of carboxylic acid such as citric acid as a precursor. This technique has widely been used in making thin films with low temperature .However reported has been for site–deficient perovskite prepared via the classic sol–gel calcination method, a method in which the nitrates of the metal ions were dissolved in deionized water, citric acid, and ethylene glycol to form a homogeneous solution at a certain pH, calcination temperature, and time.[46- 49]

2.3: Chemical bath method

ZnO thin films prepared by CBD method is based on the heating of alkaline bath of zinc salt containing the substrates immersed in it. 0.1M of zinc sulphate was used as a source of zinc, to make the solution alkaline, aqueous ammonia solution was added with constant stirring. Firstly, the solution became milky-turbid due to the formation of zinc hydroxide Zn (OH) $^{2-}$. Further addition of excess ammonia dissolved the turbidity and made the solution clear and transparent. The pH value of the resultant solution was ~11.0. The substrates were immersed in the bath at room temperature and the bath was heated at a temperature of 343K for 2hours, heterogeneous reaction occurred and the deposition of ZnO, AlZnO and CuZnO took place on the substrates respectively. After the deposition, ZnO coated substrates were removed from the bath washed with distilled water, dried in air and preserved in an air-tight container. These procedure leads to deposition on undoped ZnO on the glass substrate. However a better yield of the material can be achieved by annealing, doping the thin film or varying other parameters such as ligand temperature and deposition time using the same method as in the literature.

3. RESULTS AND DISCUSSION

The results shown below were as recorded from the instrument used in the analysis the TCO/perovskite oxide based nanocrystal for each of the deposition techniques in which consideration was given to annealing/doping as captioned in this commentary.

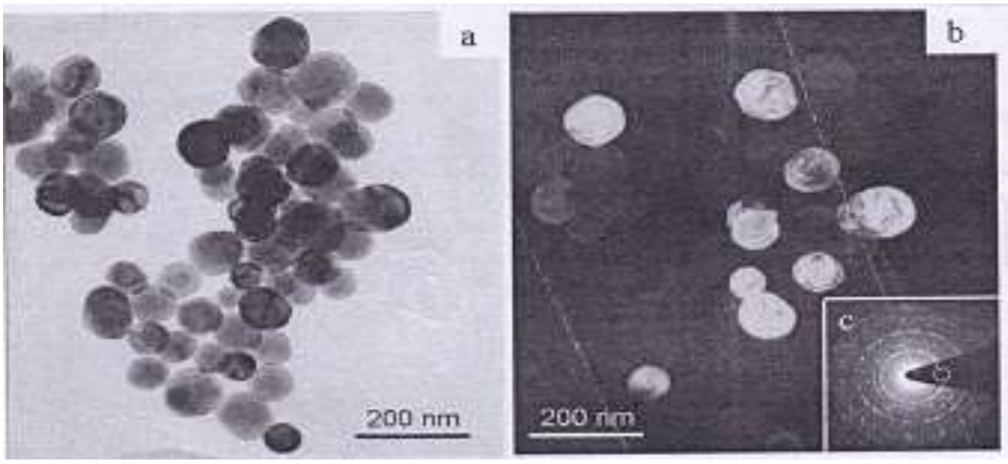


Fig.1; TEM image of hydrothermal deposited $BaTiO_3$ Powdered nanoparticle

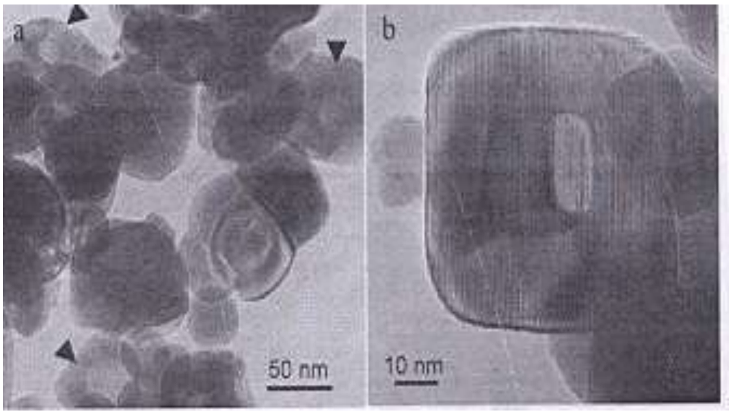


Fig. 2; TEM image of BaT_iO_3 Powder Oxide nanoparticle; (a) Un-annealed, (b) Annealed Temperature of 673K.

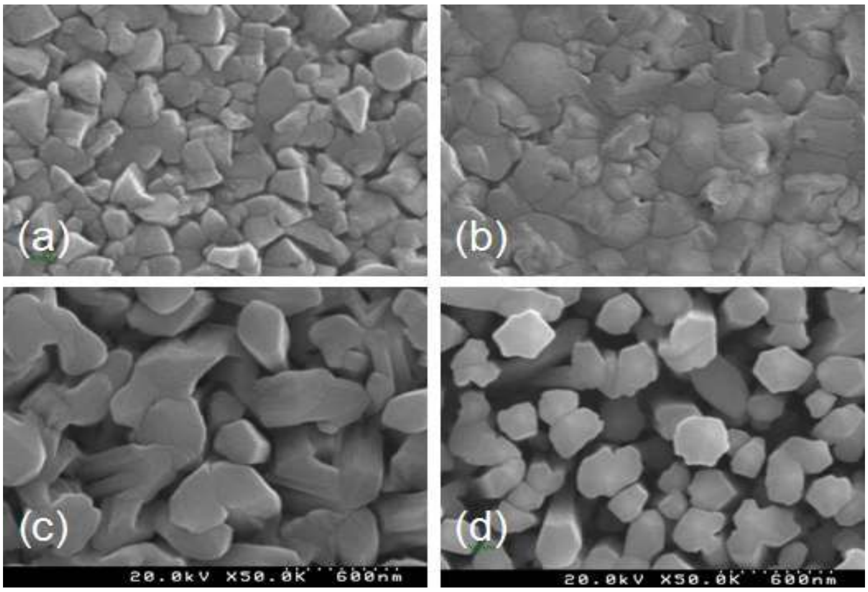


Fig.3; SEM Surface mophology[a, b,c and d] of pure ZnO thin film annealed at temperatures of100°C, 200°C, 300°C and 400°C respectively

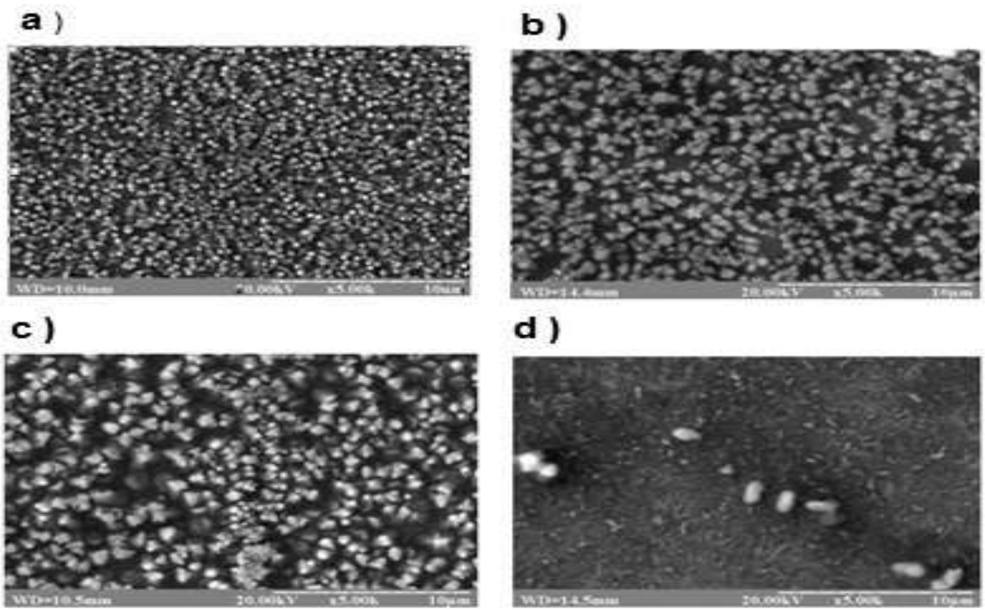


Fig. 4; SEM morphological Structure of cu-doped ZnO thin film [a, b, c and d] as deposited and annealed at temperature;500°C, 700°C and 850°C

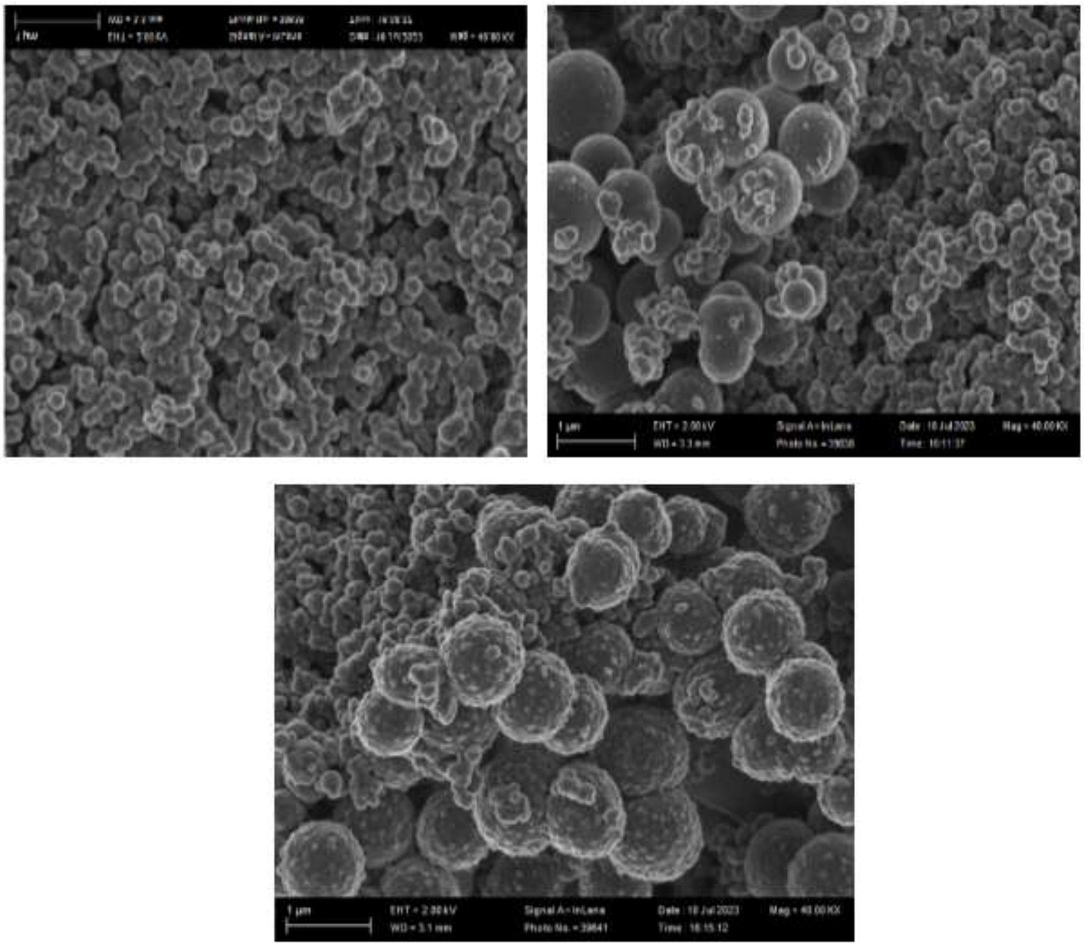


Fig.5; SEM image of the sample of pure SnO_2 (T) and aluminum doped SnO_2 (T_1) and (T_2) nanocystal

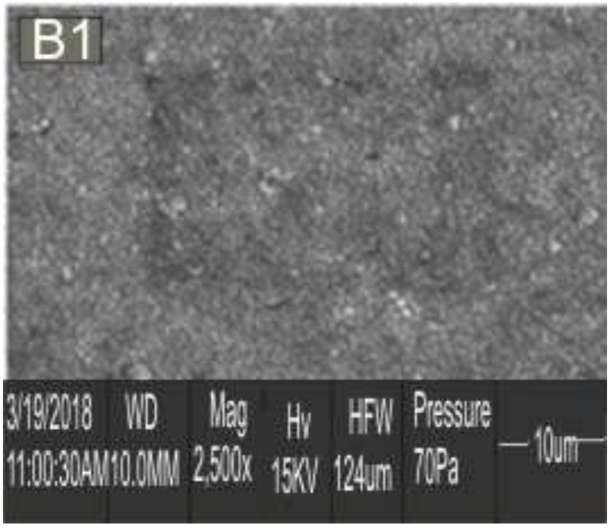


Fig.6; SEM Image Un-annealed AlZnO

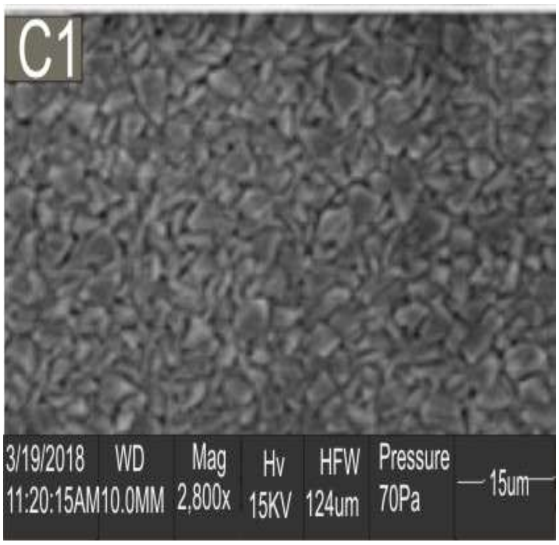


Fig 7: SEM Image of AlZnO annealed @ 100°C.

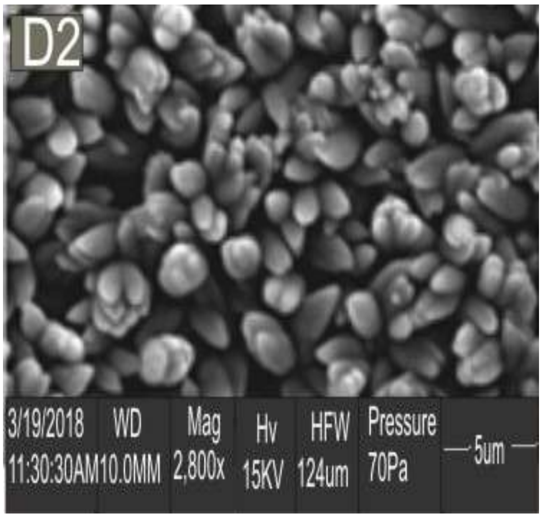


Fig 8: SEM Image of AlZnO annealed at 150°C.

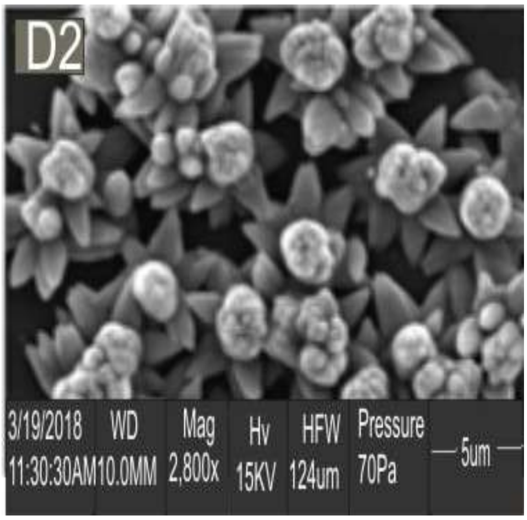


Fig 9: SEM Image of AlZnO annealed @ 100°C.

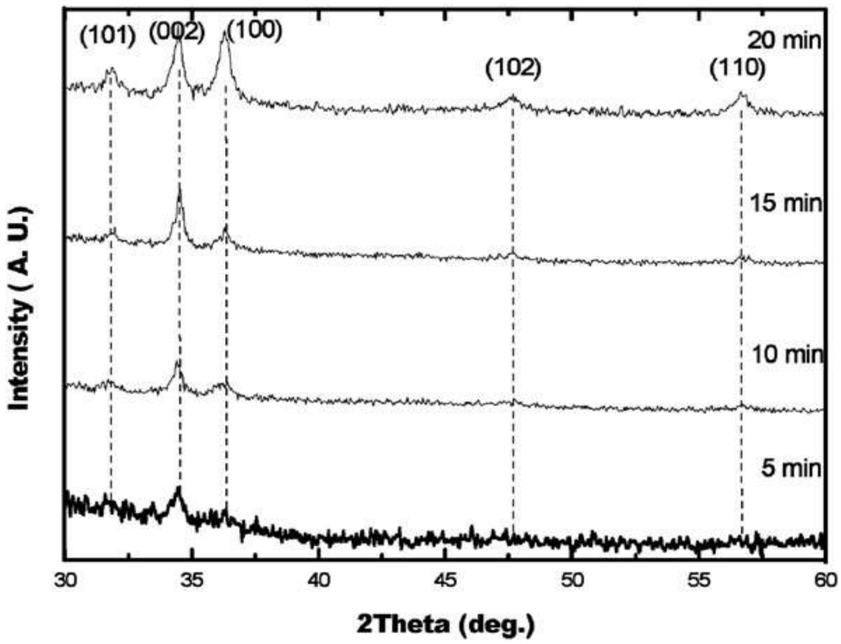


Fig.10; XRD Spectra for various percentage Boron doped ZnO thin film

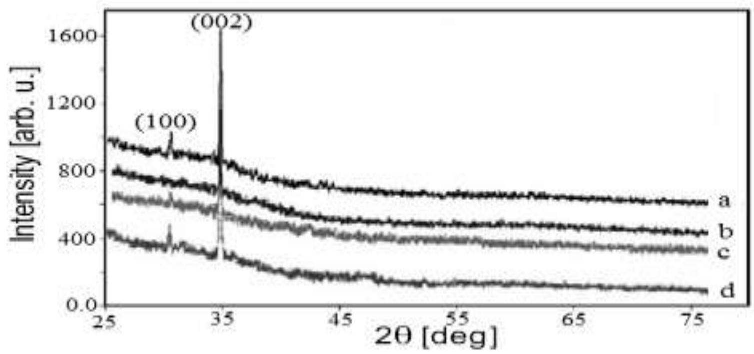


Fig.11; XRD Spectra of pure and CuZnO thin film annealed at Temperatures; 500°C,700°C and 850°C

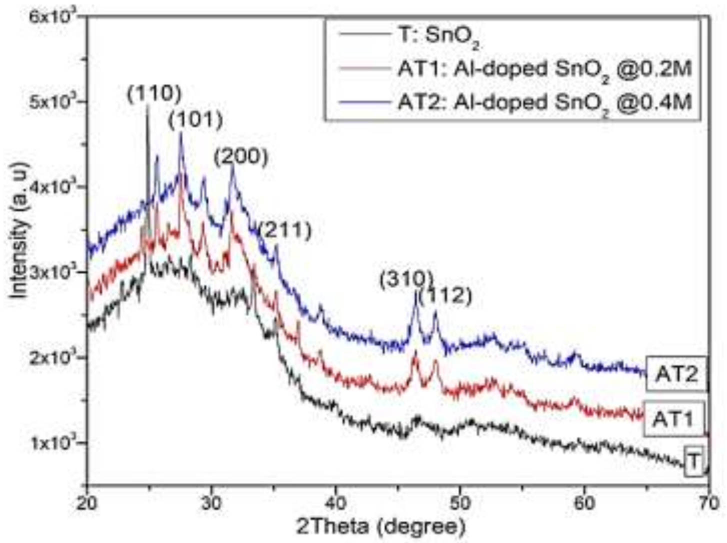


Fig.12; X-Ray Diffraction pattern of as-deposited SnO_2 and Al-doped SnO_2 Thin Films

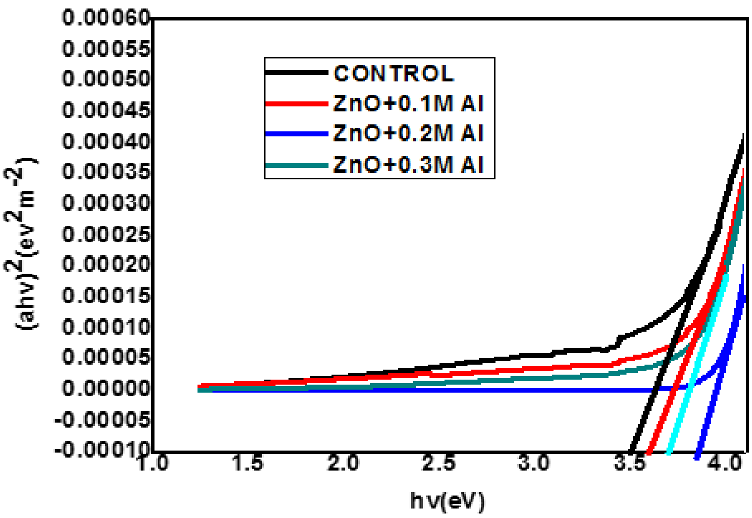
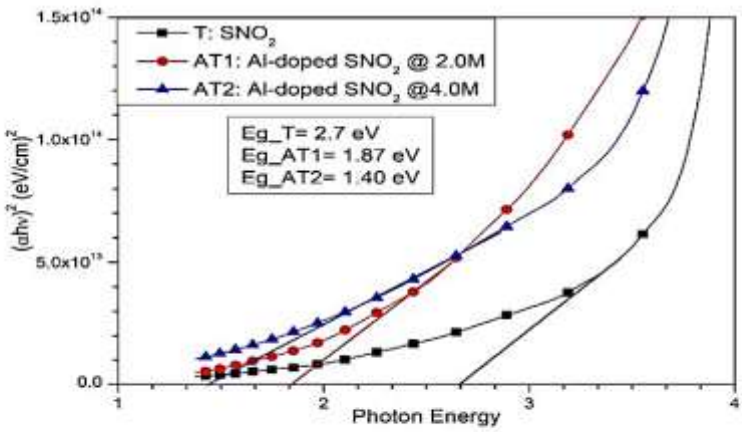


Fig.13 $(\alpha h\nu)^2$ **against photon energy of as-deposited ZnO and AZnO with different Al concentrations**



Fiug.14; $(\alpha h\nu)^2$ **as a function of Photon Energy for the three samples**

3. RESULT AND ANALYSIS

The XRD characteristics deposited of TCO/ perovskite oxide nanocrystals deposited using different deposition techniques were determined in order to assess the crystal orientation of their structure. The diffraction patterns of each crystal were depicted in figures 5,6,7,8 and 9 respectively and from the analysis, it was seen that each of them had an intense peaks at different diffracting angle irrespective of the parameter at the focus when producing the crystal. Orientations that were most prominent were (100), (002) and (101) as in figures 5 and 6. In the case of Cu-doped and boron doped ZnO thin film, intense peaks were observed at (100), (002),(101 and (102) respectively all occurring within $30^o and 40^o$ with increase in the intensity of the peak as annealing temperature increases as in figure7.The XRD for boron doped ZnO as in figure7 exhibited defined intense peaks along (100),(002), (110) (103), (200) and (112) within $32^o to 37^o and 48^o and 67^o$ respectively while for Al-doped, intense peaks were identified along (100), (002) and (101) at $2\theta = 36.24^o, 32.37^o and 36.24^o$ respectively. Generally it was seen that irrespective of annealing growth technique, doping element used notwithstanding some many have high diffraction peaks elaborated at (100) and (002) in all cases This observation indicated that many TCO/perovskite oxide nanocrystals are strongly c-axis oriented with characteristic of wurzite structure although increase in the per cent of doping element affects and often shifts the diffraction peak slightly to a lower angle side with report that crystal structure of the film deteriorate at a higher doping concentration of doping element as it decreases the c-lattice as in the literature.[1;6-7]

3.1 ANALYSIS OF THE ENERGY BAND GAP

The band gap of ZnO thin film as recorded in the all the experiment for both as deposited, annealed and doped was based on Tuac model which involved a plot of a curve of $(\alpha h\nu)^2$ as function of photon energy, $h\upsilon$ (eV). In the plot, the band gap is obtained by extrapolating the straight line portion of the curve / tangential line to the photon energy axis .from the extrapolation as in figure 10,it was observed that the band gap for pure an annealed ZnO shifts/narrows from3.13eV at $100^o C$ to 3.09eV at $200^o C$ and finally to 2.69eV at $400^o C$ respectively. This is in accordance with the report of the works as in the literature, in particular as seen in the Aluminum, Cu-doped etc. doped ZnO thin film, using the same Tauc model, it was noted that the undoped film has its band gap as shown in figures 13 & 14 where the band gap varied with the percentage increase in the concentration of the dopant element. In some cases the band gap was increased up to maximum value of the band gap ranging to the tune of 3.42eV. Therefore that fact remains as shown here that the Alminium doping concentration as observed clearly affected the band gap of the nanocrystal of TCO/Perovskite oxide materials which invariably may be associated with their well noted uniform nanostructured texture and grain size with tetragonal structure having a unique characteristics that make them to exhibit relatively good and excellent dielectric constants possessing low dielectric loss which is a property that make them a promising good candidate for nanomaterial capacitance coupled with their observable ferroelectric polarization behavior in conjunction with associated unique and peculiar magnetic properties due to their particle size and large surface-area to volume ratio with uniform size and curie temperature well above room temperature which is one of the key properties highly required for their applications for use in biomedical, magnetofluidic and other important applications because of their saturated magnetization and magnetic transition temperature which increases linearly with increasing average particle size. Report have been given based on an observation especially from the sample of $BiFeO_3$ particle by [50-52] revealed that TCO/ perovskite oxide based crystals exhibit size-dependent characteristics that invariably correlate with increased suppression of spiral spin structure when the particle size is decreased, uncompensated spins and strain anisotropic at the surface. In general, many TCO/perovskite nanoparticles exhibit combined ferromagnetic and ferromagnetic behavior in the same phase with coupling between the two orders indicates that they actually have spontaneous polarization characteristics which can be re-oriented by an applied magnetic field and the other one that can be re-oriented by applied magnetic field also These processes often generate magneto- electric memory effects and magnetic switching of ferroelectric domain and again coupled with their high performance in solar energy harnessing .From this it is clearly observed from the foregoing comments that based on high dielectric, ferroelectric, piezoelectric, pyro-electric properties, TCO/perovskite nanocrystal have numerous devices applications that are made from them, such as multilayered ceramic capacitors, ferroelectric memories, [53-57] voltage tunable capacitors, surface acoustic wave devices Infra-red detectors and again their unique performance in high efficiency in solar energy harnessing etc.. These numerous applications associated with them and their wide range of characteristics in the electronic gadgets and instruments have placed any material made from them to be in high demand in this present dispensation, and that is also the more reason why there has been much quest on miniaturization of ferroelectric, magnetoelectric and other associated materials with them [58-64] in order to achieve the reality of their amenability to these many applications. And off course these are the more reasons why there have been a evolution for quest for research in TCO/perovskite oxide based nanocrystal especially in electronic industry especially in the recent time and also for the next generation of electronics. However, we wish to emphatically stress that there are some challenges in the synthesis of such type of materials with high– purity and homogeneity that would yield desired structure of TCO/perovskite oxide based material that will be capable of enhancing the desired properties that would optimize the materials ‘properties that would lead to full realization of that anticipated applications as outlined here.Thus, there is need for material scientists to dig more on technique or procedure that would be aimed in overcoming this obstacle.

4. CONCLUSION

It has been generally known that TCO / pervoskite oxide based nanocrystal are very flexible so that it can adapted for so many useful applications since the characteristics and properties can easily be modified by doping and annealing. From this assessment, it becomes imperative state that it is worthy to noted that with temperature variation coupled with dopant concentration, the morphology of these categories of nanocrystal appeared to have coarse grain size as a result of randomly oriented fine-grained polycrystals structure that is formed during the growth, but at higher temperature or when doped with other element the smoothness of the morphology become more pronounced with preferred c-axis orientation which is of the solid state features that likely affects the properties because in all cases when they are doped with different elements, the grain size as seen in SEM images increased with increase in the percentage concentration of the dopants or with high annealing temperature which is an indication that the dopants and annealing temperature does influence the physical properties of TCO/ perovskite nanocrystal as the properties are invariably enhanced. Their surface morphologies were found generally to be good with the stoichiometric formation the nanocrystals shape which demonstrates good aggregation of the particles and this was suggested to have been originated from the large specific surface area and high surface energy as observed from the structural analysis. From the XRD analysis in all the cases the TCO/ pervoskite nanocrystal materials and their doped counterpart annealed at various temperatures depicted high and pronounced intensity at some major planes as shown in the graphs respectively where they it was expressed that there is an increase in peak intensity as annealing temperature or dopant concentration is increased in all the cases respectively irrespective of the growth technique and the dopant used during the growth of nanocrystal.

It was also found that the materials are good candidate for UV filter and good infra-red transmitter coupled with their direct band gap characteristics band gap [58],it could also inferred that the material having a visibly transparent and heating characteristics could be useful in a cold climate area for selective windows to transmit only visible and infra-red radiation into buildings while shutting off UV radiation which will help to warm indoor temperature in any buildings if have their windows coated with such nanocrystal materials, apart from the fact that they can be miniatured to form large surface area solar cell for solar energy harnessing. These observed wonderful features of TCO/ perovskites nanocrystals materials and especially their functionalities in electronic devices have placed them to tremendous enviable opportunities for research motivation on them. [48; 51-52]

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