Annealing Temperature Effect on Structural, Morphological and 1/f Noise Possessions of ITO/TiO_2 Thin Films using Chemical Spray Pyrolysis Method for Applications in solar cell

P. Ananda, S. Victor Vedanayakam, K. Thyagarajan, V. Manjunath, G. Manjunatha, K. Murali Mohan Achari, B. J. Babu, S. Basamma, B. Rajitha

ABSTRACT: ***In present work indium Tin oxide/titanium Dioxide (ITO/TiO_2) based thin films prepared on glass substrate by chemical spray pyrolysis (CSP) method at 300 °C, 350 °C, 400 °C and 450 °C annealing temperatures. From X-ray diffraction (XRD) the amorphous and crystalline nature of ITO/TiO_2 structure was analysed at all the temperatures. As the grain size becomes larger, indirectly it develops the crystalline quality of the TiO_2 films studied from Atomic Force Microscopy (AFM). The surface of TiO_2 films and the crystallite size of the sample are increased gradually with respect to temperature which is observed in Scanning electron microscopy (SEM). The elemental compositions of thin films are determined by the energy dispersive analysis of X-ray (EDAX) showed that TiO_2 thin films were highly stoichiometric. The TiO_2 is promising material for relatively low cost; manufacturing is so simple and high-efficiency in present generation of solar cells. In dye-sensitized solar cells (DSSCs) the presence of a TiO_2 is necessary to avoid short circuits in between electrodes, this layer determines by fact that compared with surface having in flat, the DSSCs absorbent surface area is increase the absorption and hence conduct to the rise of solar cells performance. In this paper we are presented samples by using spray method and ITO/TiO_2 structure was investigated, and deferent annealing measurements are acted and showed most useful differences in between annealing temperatures (300 °C, 350 °C, 400 °C and 450 °C). And the noise is also affected the efficiency of the cells, the 1/f noise analyses are achieve the impartiality at all the samples, the results proved that these work is most useful for recent trend of solar cell applications.***

KEYWORDS: ***ITO/ TiO_2 thin film, structural analysis (XRD), Surface Morphology and crystallite size of the sample (AFM and SEM), elemental composition (EDAX) and 1/f noise, Chemical Spray Pyrolysis (CSP), Annealing Temperature***

I. INTRODUCTION

Indium tin oxide (ITO), n-type degenerate wide band gap semiconductor (SC) has an optical band gap of 3.7 eV. It is transparent and has low electrical resistivity (6×10^{-4} Ω cm.) and due to its large free carrier density ($10^{20}/cm^3$ to $10^{21}/cm^3$) refractive in the infrared (IR) spectral regions [1-4], this type of thin films have a number of applications in area like high electrical conductivity nature, transparent electrodes, light emitting diodes (LED's), flat panel displays sensors, window materials in solar cell [5-10]. These materials accommodate fragment wide gap SC. The development of TiO_2 thin films has attracted the attention of scientific community due to their outstanding electrochemical and photo catalytic properties. Photo catalysis has been widely applied in life, especially after the discovered hydrophobic nature on the surface of TiO_2 thin films. This application is then constantly evolving as anti-fogging, self-cleaning [11], and some of them applications in fields like water treatment, fuel cells and air purification (remove organic pollutants) [12-14]. It is very much interest to do research in ITO/ TiO_2 based thin films, because of scientific curiosity as well as practical reasons.

Some research groups have done research contributing to the formation of ITO/ TiO_2 based thin films [15-20]. For instance, G. Kenanakis et al. [15] have Immobilized TiO_2 and Degussa (Enovik) P-25 catalysts was coated on glass by using sol-gel method at 600 and 800 °C, and studied AFM, XRD, SEM and optical transmittance spectra, the grain size changes from 15.8 ± 2 nm -19 ±3 nm. R Valencia-Alvarado et al. [16] studied the SEM, XRD, EDS, XPS and Raman spectra of the prepared TiO_2 thin films, studied properties, colour changes of the thin films are observed and concluded that the prepared anatase, rutile phases are not require any additional annealing after the oxidation. F. Gherardi et al. [17] synthesized TiO_2 films by using a TTIP precursor via spin coating method. The film at different temperatures varied of 250 °C, 300 °C and 350 °C and was calculated. At 350 °C the transmittance test results of thin films revealed their crystalline nature by XRD films. S. N. Sadikin et al. [18] the properties and their performance of the DSSC device fabricated with TiO_2 as the photo anode was influenced by the things of TiO_2 thin films. TiO_2 shoal was produced by the hydrolysis of tetrabutyl titanate, Yu Liang et al [19] succeeded in doing this by using the sol-gel method. Davinder singh et al. [20] fabricated high dielectric constant pure and iron doped TiO_2 films by optimized sol-gel process.

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P. Ananda, Research Scholar, Department of Physics, JNTUA, Ananthapuramu, A.P. India- 515002

S. Victor Vedanayakam, Department of Physics, Madanapalle Institute of Technology and Science, A.P. India-517325

K. Thyagarajan[d]**,** Department of Physics, JNTUCEP Pulivendula, A.P. India-516390

V. Manjunath[c*]**,** Department of Physics, Sri Padmavati Mahila Visvavidyalayam (Women's University),Tirupati, A.P. India-517502

G. Manjunatha[b]**,** Department of Physics, Madanapalle Institute of Technology and Science, A.P. India-517325

K. Murali Mohan Achari[e]**,** Department of Chemistry, Sri Padmavati Mahila Visvavidyalayam (Women's University),Tirupati, A.P. India-517502

B. J. Babu[b]**,** Department of Physics, Madanapalle Institute of Technology and Science, A.P. India-517325

S. Basamma[c]**,** Department of Physics, Sri Padmavati Mahila Visvavidyalayam (Women's University),Tirupati, A.P. India-517502

B. Rajitha[c]**,** Department of Physics, Sri Padmavati Mahila Visvavidyalayam (Women's University),Tirupati, A.P. India-517502

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The TEM, XPS, dielectric and frequency analysis results showed that the doped TiO_2 films has high dielectric constant, low leakage current, less number of interfacial states, high break down strength and low band offset.

In this Present work, the ITO/TiO_2 layers on glass substrate prepared by Chemical spray pyrolysis method [5, 25] have been studied for structural, morphological and i/f noise properties for solar cell applications. To the best of our knowledge, the 1/f noise spectrum on variation of 1/f noise on structural and morphological properties of ITO/ TiO_2 film at deferent annealing temperatures has not been reported. Hence, the main aim of this work is to investigate the new features of annealing temperature effect of prepared ITO/ TiO_2 thin films using XRD, SEM with EDAX, AFM and I/f noise spectrum studies [17, 20,24]. And the prepared films suggest that it is a hopeful applicant of high k gate dielectrics and solar cells.

I. EXPERIMENTAL METHOD

Titanium dioxide (TiO_2) (Sigma Aldrich; > 99%) films are coated on ITO glass substrate by spray pyrolysis technique; it was clearly shown in Fig. 1. The spraying chemical solution was prepared by mixture of titanium isopropoxide (IV) solvent at containing 40% of ethanol, 5% of acetic acid, and 5% of water were stirred with magnetic stirrer for 1 h. To eradicate the damaged surface layer and unwanted impurities from the surface of the thin films, the ITO microscope glass was cleaned with distilled water followed by rinsed with ethanol using ultrasonic agitation for 5 min during each step [24, 25].

Using a muffler furnace system with thickness of 1000 nm the TiO_2 film was annealed at 300 ºC, 350 ºC, 400 ºC and 450 ºC for 1 hour. The phase purity and the crystalline of the thin films at all the annealing temperatures are observed by XRD. SEM and AFM have the crystallographic orientation/dislocation density, surface morphology and grain structures. The result obtained from EDAX was clearly showed the constituent element percentages. At last the 1/f noise dependence was studied in different annealing temperatures to achieve the impartiality of 1/f noise studies. The role of defects and impurities were finding other synonyms by different structural analysis.

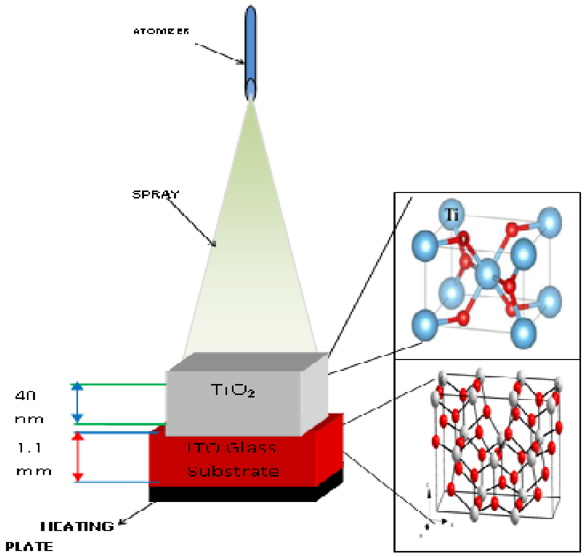

Fig 1. The schematic diagram of cross-sectional view with various layers, and ITO/ TiO_2 structure.

II. RESULTS AND DISCUSSION

In Fig. 2 the results of of XRD measurements, the X-ray diffraction profiles of ITO/TiO2 thin films prepared using chemical spray pyrolysis method on glass substrate are shown. Fig. 2 demonstrates the XRD plot of thin films at 300 oC, 350 oC, 400 oC and 450 oC for 2 hour annealed in nitrogen ambient. Fig. 2(a) demonstrations the 300 oC TiO2 thin films on ITO substrate shows TiO2 characteristic peaks of (101), (103), (200), (211), (215), (224) and ITO (204), (301), the 350 oC annealed film Fig. 2(b), identified extra peaks as ITO (222), (311), for the 400 oC annealed film Fig. 2(c), there is an extra peaks observed that is ITO (110), (400) and 450 oC annealed films Fig. 2(d), there is a repeated peak observed which is identified as ITO (204), (301). The films have three strong (101), (222) and (211) crystal orientations. The (101) diffraction was decreases compared by the large intensity of the (222) intensity up to the increasing annealing temperature 400 oC and then start to decrease and increasing the (211) diffraction to 450 oC, this is correlated with by D. Dzibrou and Ya-Qi el al. [21, 22] These results are indicative of the formation of interfacial phases after increasing annealing temperatures at 300 oC to 450 oC at the ITO/TiO2 interfaces and it's demonstrated that the ITO/TiO2 thin films strongly formed on glass substrate identified amorphous and crystalline natures. The 2-Dimetianal and 3-Dimentianal AFM images of TiO_2 films annealed at 300 ^{0}C, 350 ^{0}C, 400 ^{0}C and 450 ^{0}C with 2×2 μm^2 areas are displayed in Fig. 3. The thin films uniformly present in surface sample this has been confirmed by the 2-Dimetianal image. The grain size of the TiO_2 films increases and the surface morphology of the films is influenced because of the annealing temperature. From 3D images are clearly show that height of the sample was estimated to be about 381 to 406 nm at 300 ^{0}C, 290 to 324 nm at 350 ^{0}C, 214 to 244 at 400 ^{0}C and 120 to 165 nm at 450 ^{0}C annealed samples respectively, similar results are reported by Hosseini et al.

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[23] Finally, the AFM images are obviously shown that the surface roughness and thickness of TiO_2 thin films are strongly dependent on the annealing temperatures. The dramatic topography changes on crystallization of the films were due to the increasing of annealing temperature. As the grain size becomes larger, indirectly it develop the crystalline quality of the TiO_2 films.

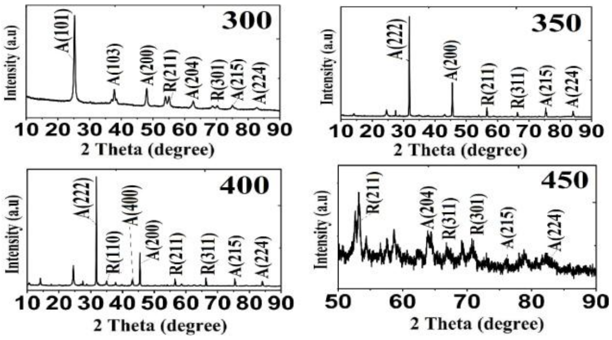
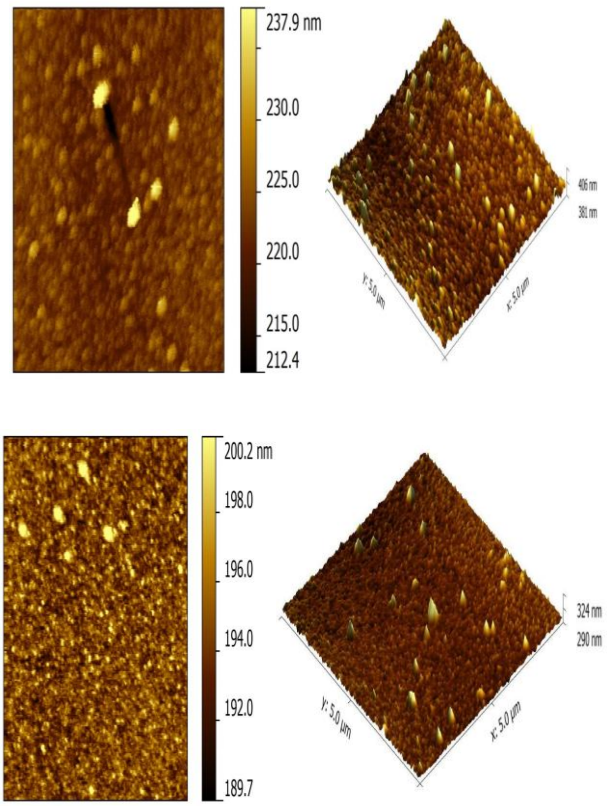


Fig.2 XRD patterns of TiO_2 fimls deposited on ITO coated glass substrates at different annealing temperatures 300 °C, 350 °C, 400 °C, and 450 °C.

Scanning electron microscopy (SEM) images (Fig. 4(a) to (d), respectively) has been exploited for the study of morphology, size, shape, scale and distribution of TiO_2 thin films. Fig. 4(a) to (d) shows the SEM images of TiO_2 deposited onto ITO substrate at different annealing temperatures (300 ^{0}C, 350 ^{0}C, 400 ^{0}C and 450 ^{0}C) during 1 hour under N_2 atmosphere.



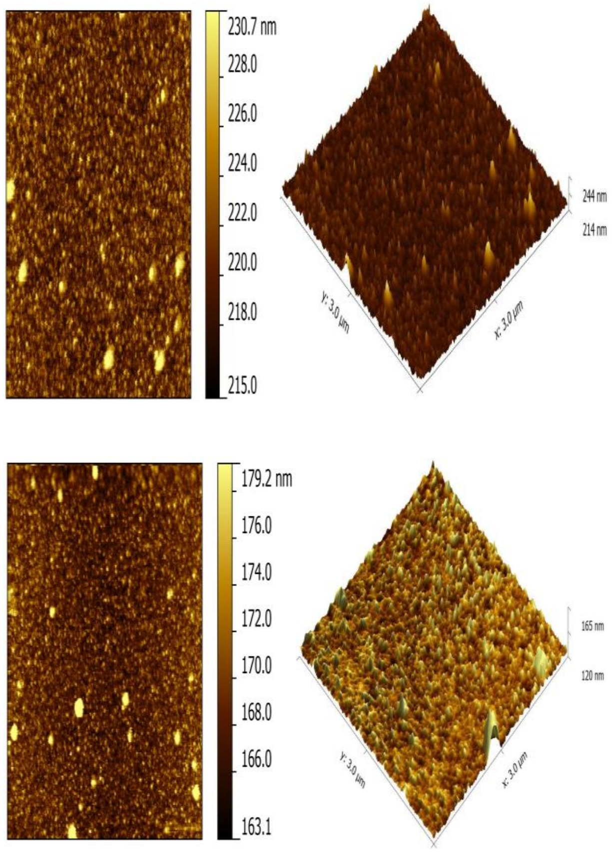


Fig.3 *AFM images (with 2D and 3D) of TiO_2 films deposited on ITO coated glass substrates at 300 °C, 350 °C, 400 °C, and 450 °C annealing temperatures.*

The results of SEM images indicated the increment of annealing temperature can change the dispersal of grains on the surface of TiO_2 films. And observed crystallite size of the sample is increased gradually with respect to temperature. In addition, the typical energy-dispersive X-ray spectroscopy (EDAX) spectrum shows the TiO_2 films on ITO substrate at different annealing temperatures. The EDAX spectrum observes the characteristic peaks corresponding to the binding energy of the elements. The EDAX spectrums confirms the presence (strong peaks) of the Ti and O elements along with the indium tin oxide of the thin films at various annealing structures, which is in agreement with Khuram Ali et al. [22, 24] These analyses showed that TiO_2 thin films were highly stoichiometric. The elemental Ti/O atomic ratios at all the annealing structures are given in tables. 1 to 4, results obtained from EDAX it showed the constituent element percentages. The increase in particle size agrees well with both AFM and SEM with EDAX results.

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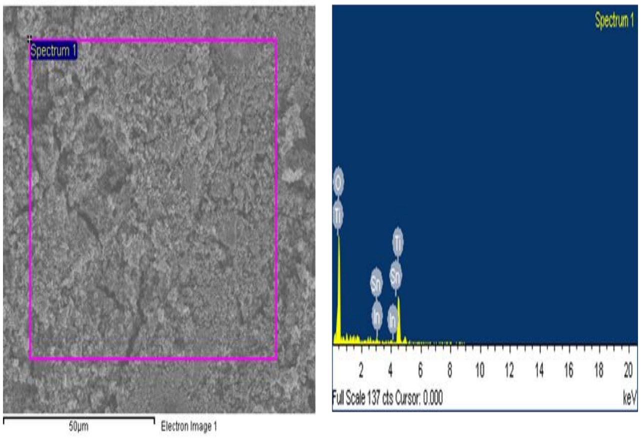
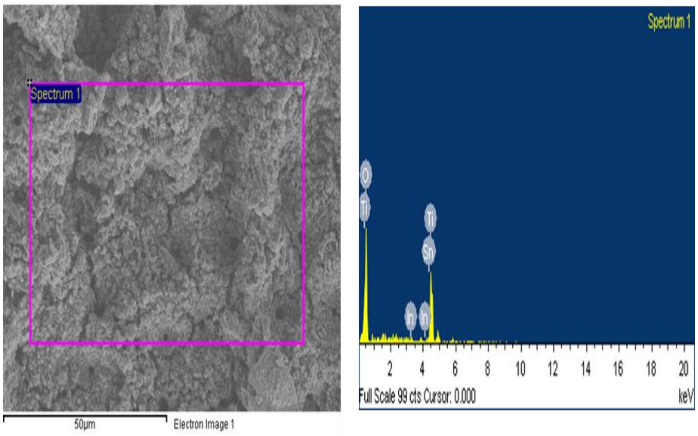
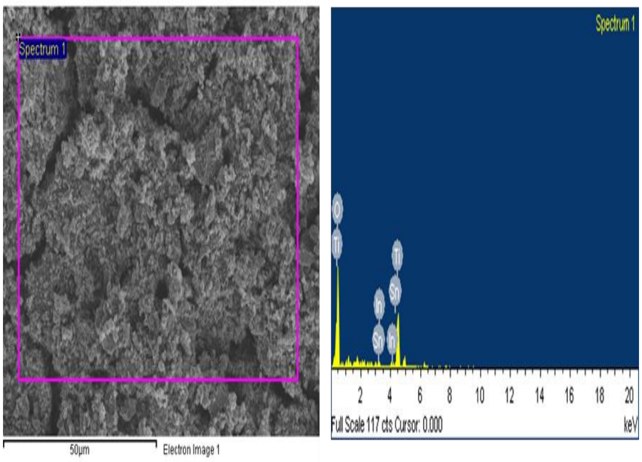


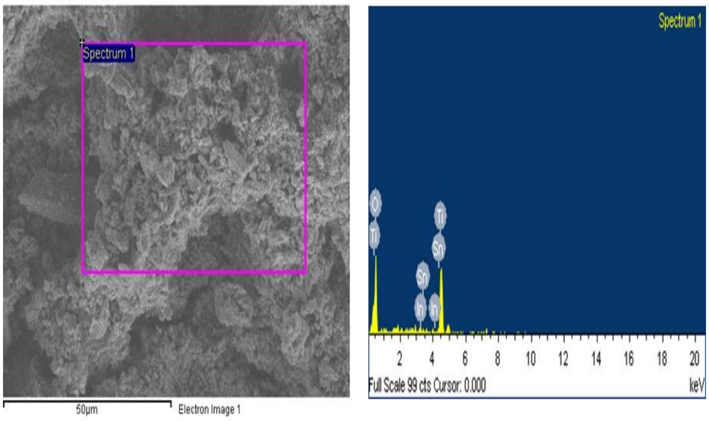
Element	Weight%	Atomic%
O K	50.33	75.21
Ti K	49.67	24.79
In L	0.00	0.00
Sn L	0.00	0.00
Totals	**100.00**	**100.00**



Element	Weight%	Atomic%
O K	46.31	72.09
Ti K	53.69	27.91
In L	0.00	0.00
Sn L	0.00	0.00
Totals	**100.00**	**100.00**



Element	Weight%	Atomic%
O K	43.16	69.45
Ti K	56.84	30.55
In L	0.00	0.00
Sn L	0.00	0.00
Totals	**100.00**	**100.00**

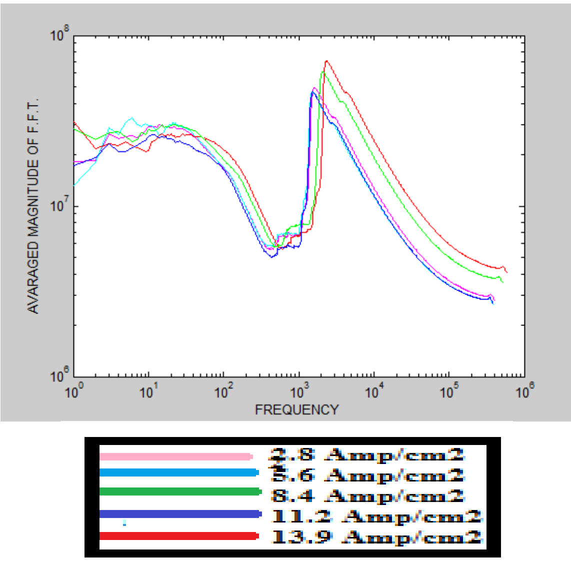


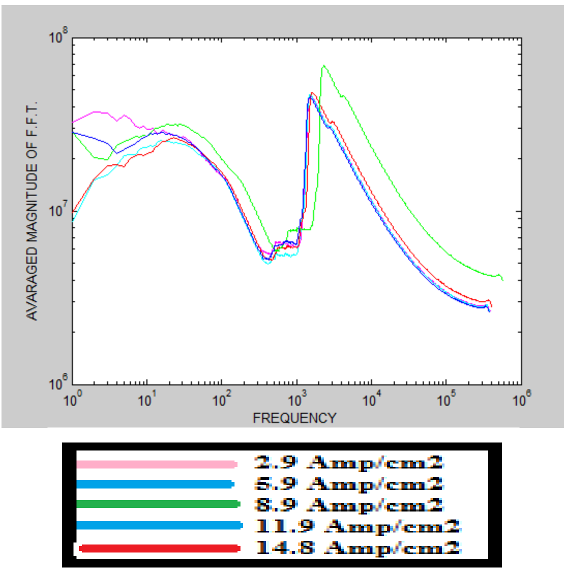
Element	Weight%	Atomic%
O K	41.76	68.22
Ti K	58.24	31.78
In L	0.00	0.00
Sn L	0.00	0.00
Totals	**100.00**	**100.00**

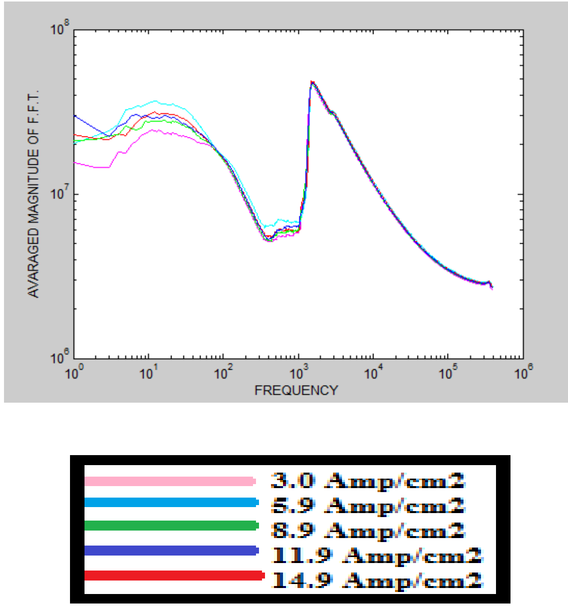
***Fig.4** SEM with EDAX images of TiO_2 fimls deposited on ITO coated glass substrates at different annealing temperatures.*

The frequency versus averaged magnitude of FFT plot for TiO_2 films are shown in Fig 5 with thickness of 400 A^0 (same thickness and environment) for different annealing temperatures are 300 ^oC, 350 ^oC, 400 ^oC, and 450 ^oC [26].

The theoretical calculated γ = -1, while the values of - γ are given by 0.560, 0.561, 0.579, and 0.581. For 300 ^{o}C values of current density (Amp/cm^2) is 2.8, 5.6, 8.4, 11.2 and 13.9 with an average of 8.38, for 350 ^{o}C values of current density (Amp/cm^2) is 2.9, 5.9, 8.9, 11.9 and 14.8 with an average of 8.88, for 400 ^{o}C values of current density (Amp/cm^2) is 3.0, 5.9, 8.9, 11.9 and 14.9 with an average of 8.92 and finally, 450 ^{o}C current density (Amp/cm^2) values are 3.4, 8.7, 13.1, 17.5 and 21.9 with an average of 12.92 value. According the vague error of ± 3%, the γ values are nearly constant for the three current densities. Now the 1/f noise analysis was studied at various annealing temperatures was shown in Table 5, by increasing annealing temperature the corresponding γ values also increases, but increases the magnitude of all the samples it may be due to samples/oxides deposition defects on the ITO/glass substrate. Finally, the careful analyses of the 1/f noise plots are achieve the impartiality at all the annealed samples.







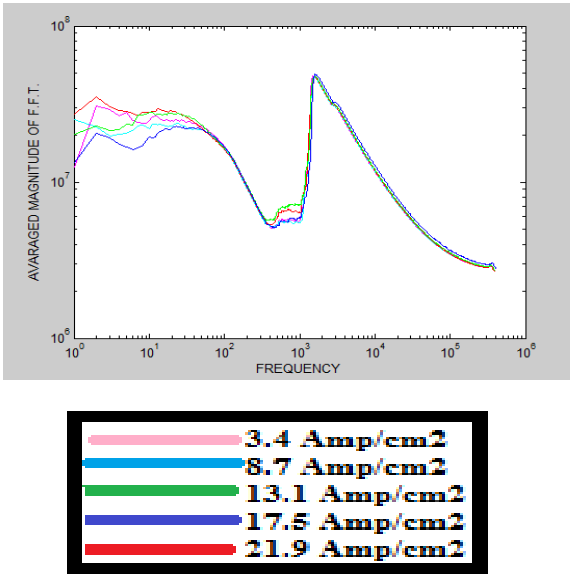


***Fig.5** Variation of 1/f noise spectrum with annealing temperature of 300 ^{0}C, 350 ^{0}C, 400 ^{0}C, and 450 ^{0}C.*

S.No	Annealing temperature of ITO/TiO_2 thin film (^{0}C)	Surface roughness (μm)	Kurtosis	Average slope of 1/f noise
1	300	1.31	2.69	0.560
2	350	1.44	3.78	0.561
3	400	1.68	5.03	0.579
4	450	1.70	9.01	0.581

***Table 5.** Different Parameters of 1/f noise images*

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III. MAIN SIGNIFICANCE OF PRESENT WORK

1. Titanium dioxide (TiO_2) (Sigma Aldrich; > 99%) films are coated on ITO glass substrate by spray pyrolysis technique; it was clearly shown in Fig. 1.
2. The formation of interfacial phases after increasing annealing temperatures at 300 °C to 450 °C at the ITO/TiO_2 interfaces and it's demonstrated that the ITO/TiO_2 thin films strongly formed on glass substrate identified amorphous and crystalline natures as shown in fig.2.
3. Fig.3. shows the AFM images are obviously shown that the surface roughness and thickness of TiO_2 thin films are strongly dependent on the annealing temperatures. As the grain size becomes larger, indirectly it develops the crystalline quality of the TiO_2 films.
4. These EDAX analyses showed that TiO_2 thin films were highly stoichiometric. The elemental Ti/O atomic ratios at all the annealing structures are given in tables. 1 to 4, here the results showed the constituent element percentages. The increase in particle size agrees well with both AFM and SEM with EDAX results are shown in fig. 4.
5. The Now 1/f noise analysis was studied at various annealing temperatures was shown in Table 5. Finally, the careful analyses of the 1/f noise plots are achieve the impartiality at all the annealed samples it was shown in fig.5.

IV. CONCLUSION

The ITO/TiO_2 based thin films on glass substrate were prepared by Chemical spray pyrolysis method for different annealing temperatures 300 ^{0}C, 350 ^{0}C, 400 ^{0}C and 450 ^{0}C. The crystallite size, structural, morphological and i/f noise properties of the prepared samples were investigated using AFM, XRD, SEM with EDAX methods and 1/f noise. The XRD analysis of ITO/TiO_2 films confirmed the presence of crystalline natures. When the amount of temperature is increased, the crystallite size of the sample is also increased gradually. This has been observed in AFM and SEM. EDAX analysis showed that ITO/TiO_2 films were highly stoichiometric. The elemental composition determination by EDAX showed that all the synthesized ITO/TiO_2 films were tin deficient. The analyses of the 1/f noise plots are achieve the impartiality at all the annealed samples, In dye-sensitized solar cells (DSSCs) the presence of a TiO_2 is essential to avoid short circuits between electrodes, this layer determines by fact that compared with surface having in flat, the DSSCs absorbent surface area is increase the absorption and hence conduct to the rise of solar cells performance, it is concluded that these paper work is useful for solar cell applications.

V. ACKNOWLEDGEMENTS

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AUTHORS PROFILE

P. Ananda received his B.Ed degrees in 2006 and M.Sc (Physics) in 2008 from Sri Venkateswara University, Tirupati. And is currently a Ph.D. candidate in JNTUCEP pulivendula and MITS Madanapalle, department of physics. He was published five research articles in refereed journals, and two conference proceedings. Attended, two conferences for present his research work. His Research interest focus on fabrication and characterization of thin films, here he was studied electrical/optical, structural, surface morphological and noise properties of the thin films technology at varies applications like optoelectronics and solar cells

S. Victor Vedanayakam is an Assistant Professor from the Department of Physics, Madanapalle Institute of Technology and Science, Madanapalle, Chittoor (Dt), A.P. India. He has obtained his Ph.D from the Sri Krishna Devaraya University, Anantapur. He has almost 17 years of experience in teaching with a rigorous zeal and aspiration to introduce the concepts of Physics in an interesting and student friendly atmosphere. His research area includes 1/f noise studies in thin films and semiconductor devices, theoretical study and development of solar cells. For his credit he has eighteen papers published in national and international journals. He has published a book in the field of electronics "1/f noise measurement system". He has attended number of conferences and workshops to share his knowledge and to enrich his teaching and research abilities.

K. Thyagarajan received the M.Sc, M.Phil and Ph.D degrees in Physics from Sri Venkateswara University, Tirupati. He is presently Professor and head in the Department of Physics, JNTUA college of Engineering (Autonomous), Pulivendula, A.P, India. Prof. Thyagarajan has published more than 56 articles in referred journals, and has attended number of national/international conferences to present his research work. He was awarded 9 Ph.D.'s to their research students and 8 research students are pursuing Ph.D under the guidance of Prof. Thyagarajan. His most experienced in academic events also, and finally his current research interests are preparation of Nanomaterials and thin films for studying there electrical, structural, morphological and optical properties of the films.

V. Manjunath received his M.Sc (Physics) and B.Ed degrees from SriVenkateswara University, Tirupati in 2009 and 2013. He was awarded Ph.D form Sri Venkateswara University in 2018 where his Ph.D thesis covers **''Studies on Electrical Properties of GaN Based Metal-Insulator Semiconductor (MIS) Structures''**. Qualified CSIR-NET in 2015, also awardee of BSR Research fellowship in science for meritorious students in 2015, and selected National Fellowship for Higher Education (NFHE) from UGC for the year of 2016-17. He was published many research articles in refereed journals, and attended number of conferences to present his research work. Research interest is the fabrication and characterization of high quality Ohmic and Schottky contacts to III-V compound semiconductors. Also fabricate and analysis of thin films on ITO coated glass/quartz glass substrates. He is currently working on teaching faculty (on contract) in Sri Padmavati Mahila visvavidyalayam, (Women's University) Tirupati, India.

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