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RESEARCH ARTICLE

ELECTRICAL AND MORPHOLOGICAL CHARACTERIZATION OF SOLUTION-PROCESSED GEL/PZN-PT PEROVSKITE NANOPARTICLES THIN FILM FOR TRANSISTOR DIELECTRICS

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Abstract

Thin film filed effect transistors (TFTs) have drawn significant attention from scientific community as one of the most attractive for low-cost, large area and flexible electronics.In this context, the development of high-k dielectric materials with additional functional properties is essential for low voltage operating and enhancement of device performance. This work presents the investigation of solution processed $Pb(Zn_{1/3}Nb_{2/3})O_3$ 4.5$PbTiO_3$(PZN PT) perovskite nanoparticles thin films, with and without Mn doping, as gate dielectrics for thin film transistor applications. PZN-PT with 1% Mn doped and undoped PZN-PT films were deposited by spin coating on nanostructured doped n-type silicon substrate to form metal-oxide-semiconductor (MOS) structures with top silver contact. SEM analysis shows Mn doping PZN-PT presents a denser and more homogeneous microstructure compared to undoped. Electrical measurements confirmed transistor operating mode and highlighted the effect of ferroelectric polarization on device behavior. The results show that device from PZN-PT Mn doped films exhibit higher values of current ~$2.5x10^{-1}$ A and more electrical stability than the undoped device.

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Introduction:-

Flexible and thin film electronics has emerged as a promising technology for low-cost, large area microelectronic applications RFID tags and smart labels. Organic–inorganic perovskite materials are currently attracting considerable attention as an absorber for highly efficient solar cells[1,2].For several decades, silicon oxide SiO_2 has been used as the gate dielectric material in Si-based MOSFETs and organic field effect transistors OFETs due to the high quality of the Si/SiO_2 interface and its mature processing technology. However, the low dielectric constant value of SiO_2[3] limits further voltage scaling. As a result, several materials like HfO_2[4] and Al_2O_3[5] and

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biopolymer Gum [6–9] have been investigated as dielectric material to enhance gate insulator capacitance and reduce leakage currents. Recent researches have focused attention on using perovskite materials in field-effect transistors because perovskites promise the processability and flexibility inherent to thin film transistors as both dielectric and semiconductor materials [10–14]. Perovskite oxides with high dielectric constants offer a unique opportunity to serve as gate dielectric and to introduce ferroelectric functionality. The ferroelectric functionality opens pathways toward ferroelectric field effect transistors (FeFETs) and non-volatile memory devices[15–17].When placed in the context with conventional SiO_2 dielectric[18], $Pb(Zn_{1/3}Nb_{2/3})O_3$-4.5$PbTiO_3$(PZN-PT)and 1% Mn-doped PZN-PT materials offer a distinct advantage due to its high dielectric constant in bulk [19,20] and inthin film [21]. AmongPZN-PT solution processed thin films represent a promising pathway toward future thin film electronic devices. However, the performance of solution processed thin films materials depends highly on film microstructure, nanoparticle dispersion and defect density. Structural defects, grain boundaries and poor film densification can lead to increase leakage currents and charge trapping. Therefore, an investigation of the effect between film morphology and electrical behavior is necessary to optimize PZN-PT thin film based dielectric in transistors architectures.In this work, we investigate the applicability of solution processed$Pb(Zn_{1/3}Nb_{2/3})O_3$-4.5$PbTiO_3$(PZN-PT) PZN-PT nanoparticle thin films, with and without Mn doping, as gate dielectrics for transistors in top gate configuration.

Experimental Procedure:-

$Pb(Zn_{1/3}Nb_{2/3})O_3$-4.5$PbTiO_3$(PZN-PT) nanoparticles thin film wasgrown on nanostructured silicon doped ntype as substrate to form a p-MOS channel structure. PZN-PT perovskite material is previously grinded and dispersed in a gel to obtained homogeneous solution. Substrates are firstly cleaned with acetone and alcohol with ultrasonic baht. PZN-PT nanoparticles thin film is deposited by spin coating as dielectric material on the nanostructured silicon.To ensure ferroelectric properties effect on electrical behaviour of the transistors, we processed two different devices in top contact structure. Device 1 was made from non-doped PZN-PT nanoparticles thin film. In contrast, device 2 was made with 1% Mn doped PZN-PT film.The Gate, Source and Drain contact were fabricated bylayering silver over several millimeters to form a top contacts structure of film effect transistor as illustratedin Fig. 1. SEM analysis were performed by using a MERLIN FEG of Zeiss scanning electronmicroscopy (SEM) model to carried out the surface morphology of the film but also to observe the dispersion of the nanoparticles in the deposited film. Electrical characterization was performed at room temperaturewith a KEITHLEY 2612B source-meter to observe output characteristics drain current. As we couldn't estimate the capacitance of the dielectric, we didn't evaluate the field effect mobility of free charges carries of the active material.

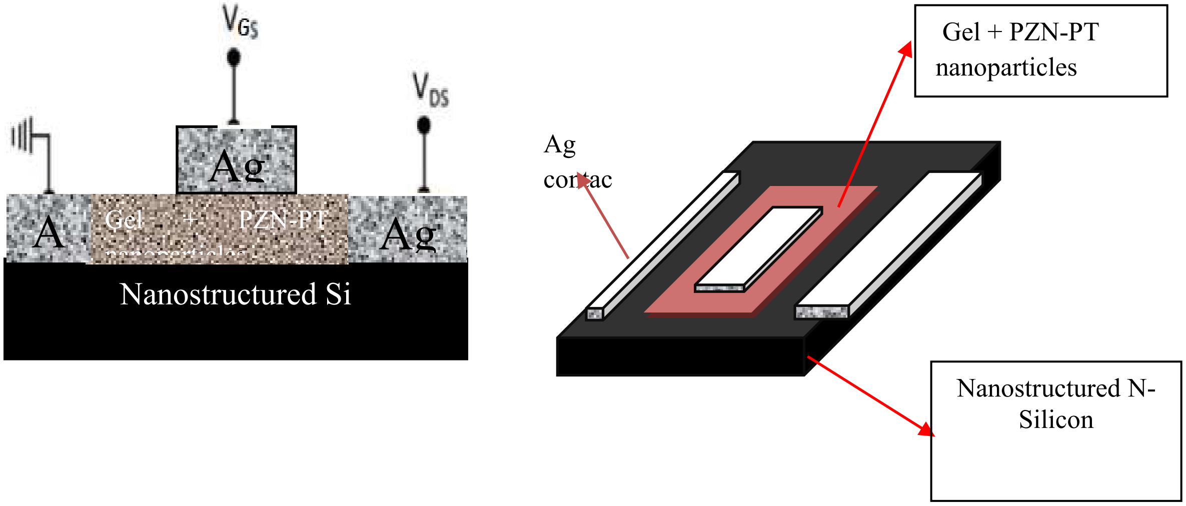

Fig. 1: Schematic structure PZN-PT nanoparticles thin film as gate dielectric

Results and Discussion:-

The morphological and electrical characterization of the PZN-PT thin films provides critical insights into the role of Mn doping and the intrinsic ferroelectric nature of PZN-PT material when applied as a high-k dielectric.

SEM Morphological Analysis:-

SEM analysis clearly distinguished the differences between the undoped and Mn-doped thin films.TheMn-doped PZN-PT films (image a) exhibited a compact microstructure with uniformly distributed grains, which implies a stable crystallization pathway.TheMn-doped films exhibited denser structures and uniform nanoparticle distribution, with average grain sizes lower than those undoped ones.On the other hand,the undoped films in imageb shows grain agglomeration and rough surfaces, with visible pores and cracks.The observed structural differences are attributed to the acceleratedcrystallization kinetics induced by Mn doping, which acts as a sintering aid. Accelerated the kinetics process of PZT thin film leads to better crystallization and avoids or minimises the interfacial defect [22]. Meanwhile the grain growth observed in undoped film introduces structural defects that could compromise dielectric reliability. Ndioukaneet al. found colossal relative dielectrics constant at 1kHz of PZN-PT and its 1% Mn doped thin film respectively about 2.76×10^4 and 17.7×10^4[21].This difference values also shows the effect of Mn doping in the structure ofthe formed thin film on silicon substrate. Daus et al. shows in their study on methyl-ammonium lead iodide (MAPbI3) a giant value of dielectric constant >1000 as gate dielectric material for thin film transistors [23]. These different studies should encourage the use of perovskite materials as high k dielectric material in field effect transistors.

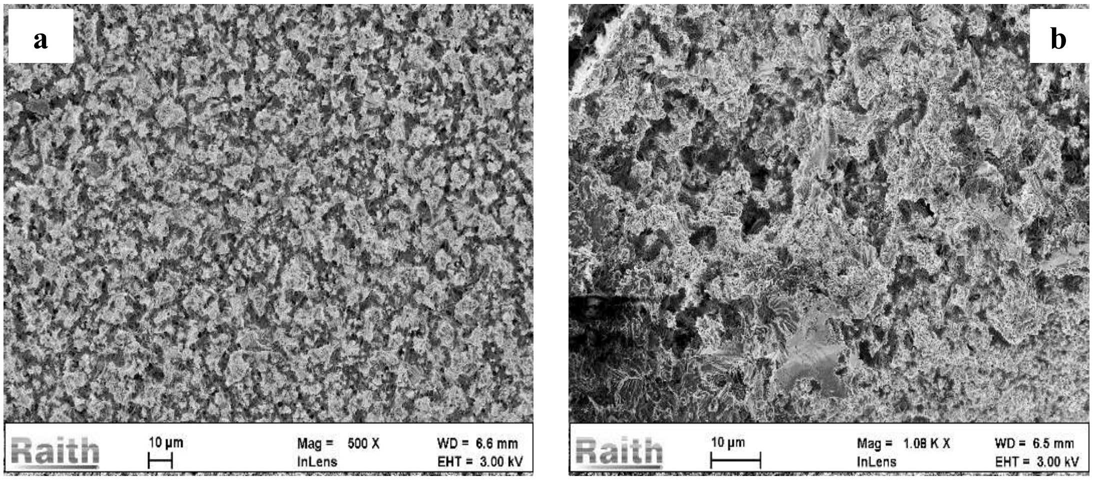

Fig. 2: SEM images of PZN-PTthin film: a) 1% Mn doped; b) Undoped

Electrical Measurements:-

The electrical output characteristics shown in Fig.3a&b presents difference in drain current magnitude. Both Mn-doped and undoped electrical characteristics demonstrated transistor-like behavior. And shows that the devices operate in p-type transistor. Which could confirm the feasibility of integrating PZN-PT perovskite material into thin film field effect transistor architectures.TheMn-doped films displayed more stable and higher drain current responsesFig.3a. The highest drain current obtained in p-type channel transistor is over -250 mA at V_G = -10 V. SEM image of Mn-doped with better crystallization and dense structure could explain the high values of drain current and lower presence of leakage current. Ndioukane et al. showed enhanced open circuit voltage on Mn-doped PZN-PT perovskite thin film solar cell over undoped, where they explained it to the lattice-free crystallization induced by the Mn doping [2]. Mn^{2+} ions replaced B-site cations in the perovskite lattice. This replacement leads to improve lattice symmetry and defects passivation. Therefore, the Mn doping reduced trap states and enhanced the interfacial quality. One could also suggest that the Mn-doping enhanced polarization of PZN-PT thin film and reduced the leakage current which made the dielectric more effective for low voltage operation.

In contrast, the undoped devices present lower drain current less than -100 mA at the same gate voltage Fig.3b. As it is shown previously in SEM image, the dependence of electrical performance and film structure is clearly observed. The undoped devices suffered leakage current observed in Fig.3b. The structural defects observed in undoped films could compromise the positive impactof dielectric proprieties and increase the observed leakage current. The lower values of drain current and instability of electrical parameters of undoped devices are similar of previous results in electrical dependencywith the dielectric interface quality in field effect transistors[18,24–26]. The poor quality of the dielectric interface due toundoped film defects could induce charge trapping and carrier scattering. Moreover, the gate and dielectric contact resistance should be high because of the poor quality of the undoped film microstructure. And then, the injection and/or charges collection would be lower. Since this perovskite material should be proposed as gate dielectric, better interface quality is necessary to improve and stabilize the electrical characteristics of devices. However, Ndioukane et al. presented Mn doping as stabilizer of the electric field dependence of PZN-PT film[21].Moreover, the absence of dielectric capacitance measurements limited quantitatively the benchmarking of this study. The FETs keys parameters such like field effect mobility was not calculated.In comparison with others perovskites oxides as gate dielectric, one could find some similarity in the low voltage operating (below 10 V). Previous studies on thin film transistors using quasi-2D perovskite oxide dielectricand strontium titanate (STO) as a gatedielectrics showed devices operating at low voltage ~ 5 V [27,28].

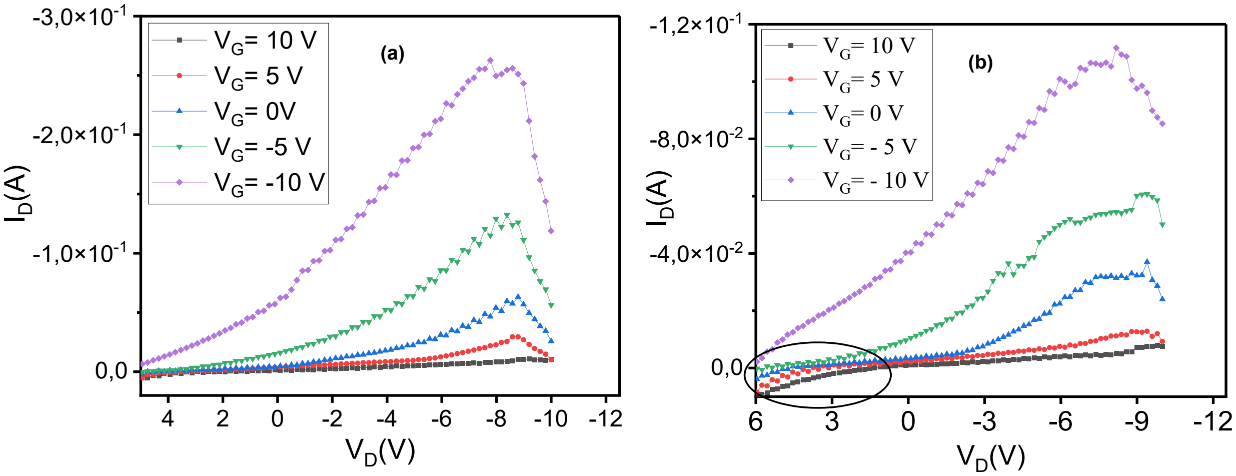

Fig. 3: Output electrical characteristics of PZN-PT as dielectric transistorat room temperature:a) 1% Mn doped and b) non-doped.

Ferroelectric effect in devices operating:-

A particularly observation in Fig.4a&b is the manifestation of ferroelectric polarization in the PZN-PT dielectric at the values of V_G=± 20 V. The polarization leads to hysteresis in the output curves.This behavior indicates the dual role of the material as both high-k and ferroelectric proprieties. This ferroelectric behavior is in strong agreement with the growing studies in ferroelectric field effect transistors (FeFETs), where polarization switching enhances non-volatile behavior [17,29]. The coercive field E_c (Fig4.b) is about 20 V/cm proportionally to our devices dimensions for the undoped and Mn-doped one. The doping has no effect on the coercive field as the ferroelectricity propriety is typically based on the PZN-PT perovskite material. This value is in accordance with the coercive field values given by Ndioukane et al. [21]. Ferroelectricity effects in FeFETs are more often observed in transfer characteristics as it permits to identify the stability of the FETs [30–33].In this study the ferroelectricity was observed in output characteristics at high value of V_G in the saturated mode of the transistor. This behavior for variable values of gate voltage showed the impact of dielectric control on the transistor operating phase and dielectric material nature. Puebla et al. showed that there is an interplay between the ferroelectric polarization and interfacial phenomenaobserved by the clockwise hysteresisin the transfer curve [31].To ensure the electrical stability by adjusting the memory window and to exploit our PZN-PT FeFETs as non-volatile memory [32,34,35], the transfer characteristics is primordial.

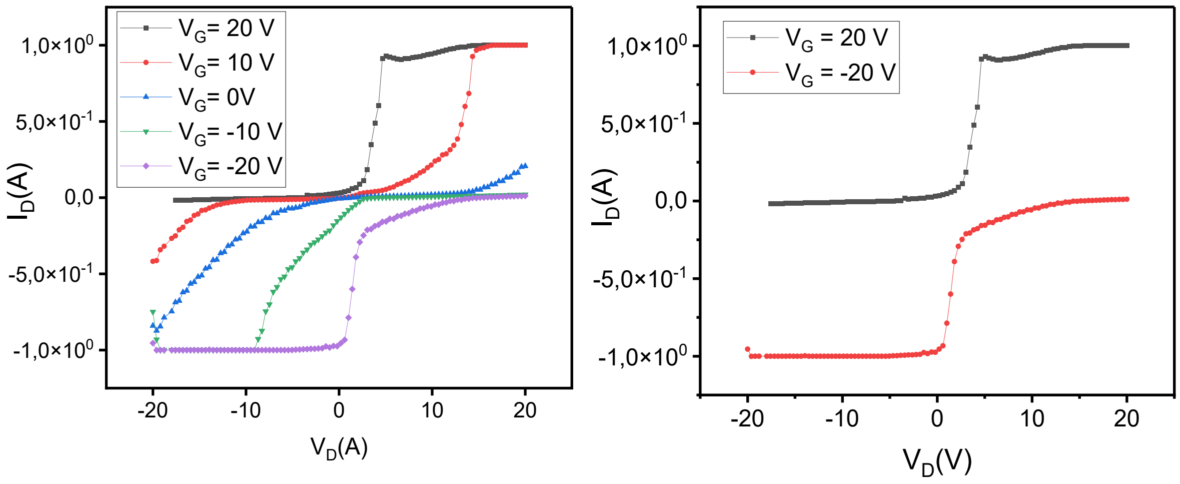

Fig. 4: Ferroelectric effect over transistor Output characteristics a) from variable voltage of V_G and b) V_G fixed at -20V and 20V.

Conclusion:-

This work demonstrates the successful fabrication and characterization of PZN-PT thin film as dielectric for transistors applications. Mn-doped films exhibit compact and dense microstructure and morphology. Devices from the doping PZN-PT material present superior electrical stability with drain current over 250 mA. Whereas the undoped PZN-PT films, even developing grain growth, present defects that decrease device performance. The ferroelectric nature of the PZN-PT perovskite material is showed in hysteresis effect, which confirming its potential application in ferroelectric field effect transistors (FeFETs). The unknown of the capacitance of ours devices limits this study. So it will be interesting to evaluate the insulator capacitance andto perform transfer curves characterisation to evaluate the stability of our devices and optimize the material dielectric and ferroelectric proprieties.

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Conflicts of Interest:-

The authors declare no conflicts of interest regarding the publication of this paper.

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