

Title:

APPLICATION OF ULTRASONIC SPRAYED ZIRCONIUM OXIDE DIELECTRIC IN
ZINC TIN OXIDE-BASED THIN FILM TRANSISTOR

Authors:

Abayomi T. Oluwabi^{1*}, Atanas Katerski¹, Emanuel Carlos², Rita Branquinho², Arvo Mere¹,
Malle Krunk¹, Elvira Fortunato², Luis Pereira², Ilona Oja Acik^{1*}.

Affiliations

¹Laboratory of Thin Film Chemical Technologies, Department of Materials and
Environmental Technology, Tallinn University of Technology, Ehitajate tee 5, 19086 Tallinn,
Estonia.

²CENIMAT/i3N, Departamento de Ciência dos Materiais, Faculdade de Ciências e
Tecnologia–Universidade NOVA de Lisboa and CEMOP/Uninova, Campus de Caparica,
2829-516 Caparica, Portugal.

The supporting information contains data related to the production of ZTO semiconductor thin film by spin coating techniques and its application in TFTs. Figure S1 shows the X-ray diffractogram of ZrO_x dielectric thin films. Figure S2 shows the TG-DSC curve for both the ZnO_x and SnO_x precursor solution. Figure S3 shows the thermal behaviour of ZTO semiconductor precursor solution up to 540 °C, determined by conducting thermogravimetric (TGA) and differential scanning calorimetry (DSC) analysis. Figure S4 shows the AFM 3D and 2D surface morphology of ZTO semiconductor thin film deposited at 350 °C. Figure S5 shows the ATR-FTIR spectral of the ZTO precursor solution. Figure S6 shows the typical transfer and the output curve of the ZTO/SiO₂ TFT at 250 °C. Table S1 shows the statistical summary of all the important electrical parameters extracted from both the MIS and TFT device studies.

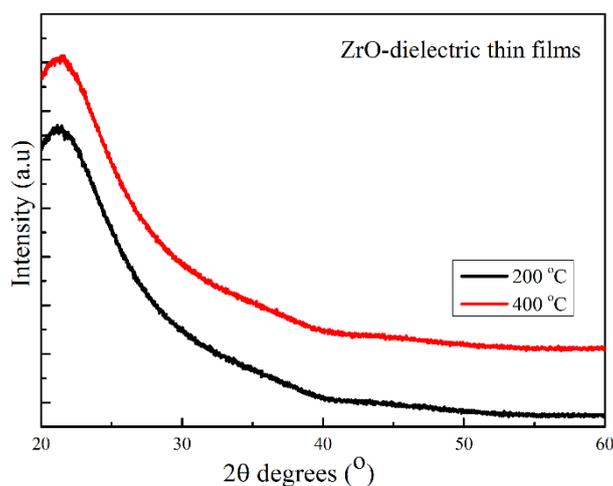


Figure S1. X-ray diffractogram of the deposited ZrO_x dielectric films at 200 and 400 °C.

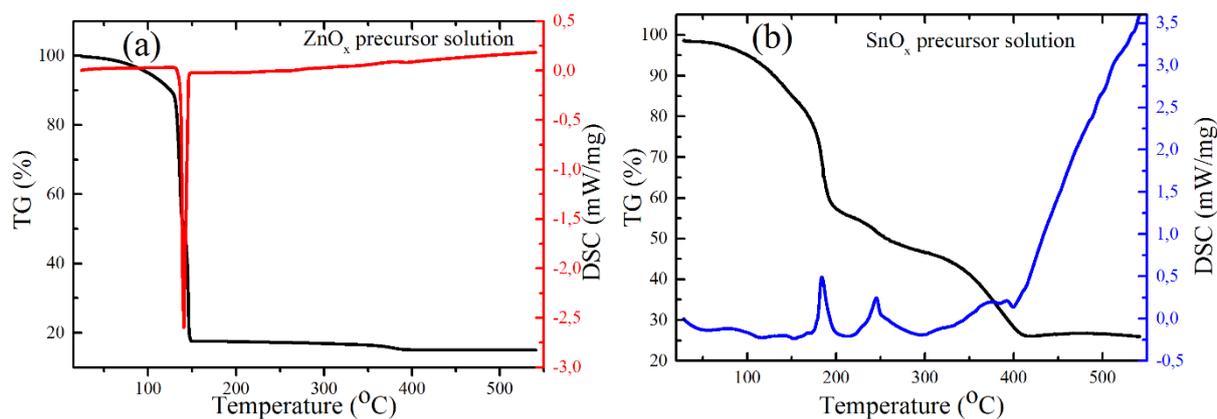


Figure S2. TG–DSC analysis of both (a) ZnO_x -based precursor solution, and (b) SnO_x -based precursor solution.

Figure S2 (a) shows the TG/DSC of the ZnO_x precursor solution and (b) shows the TG/DSC of the SnO_x precursor solution. Also, it is worth mentioning that different chemical reactions occur in the corresponding SnO_x and ZnO_x precursor solutions; the former shows an endothermic reaction, which is evidently seen by the endothermic peaks at 184 and 245 °C on the DSC plot, while the later exhibited an exothermic reaction and can be seen by an intense exothermic peak on the DSC plot

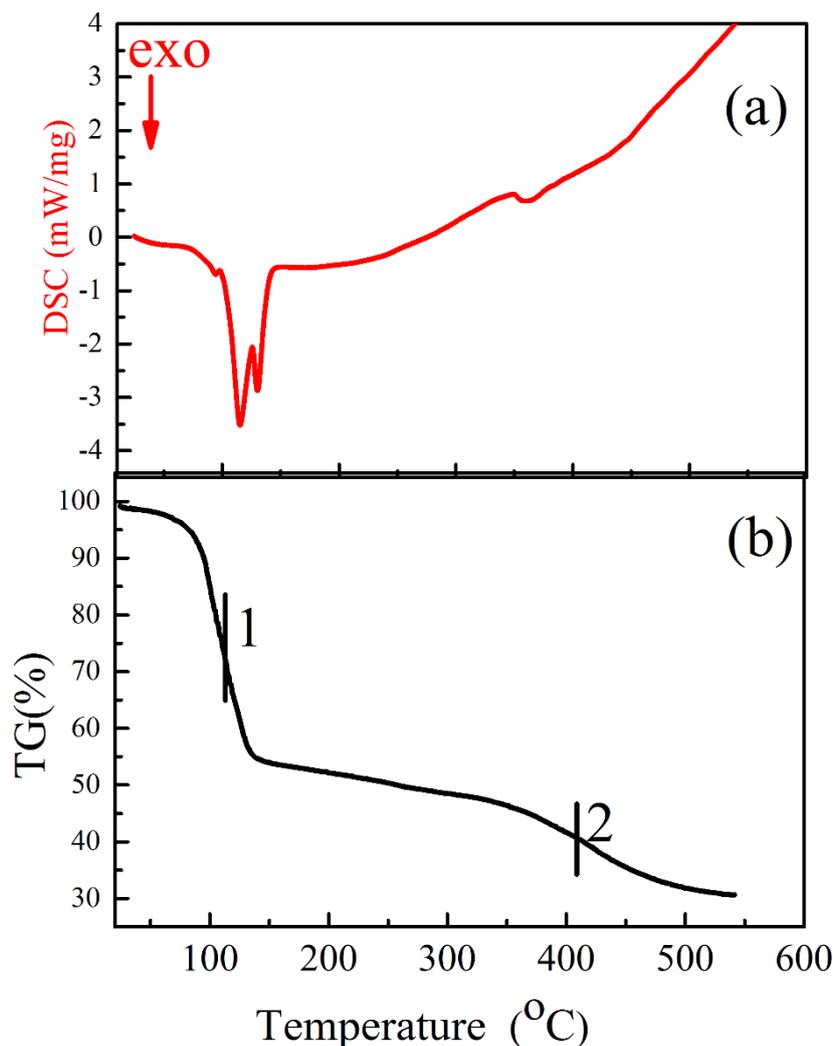


Figure S3. Thermal analysis of the ZTO precursor solution: (a) DSC, and (b) TG curves. Note the mole ratio between the constituent Zn and Sn atoms is 2:1.

Figure S3 shows the thermal behaviour of ZTO semiconductor precursor solution from 20 °C up to 540 °C, determined by conducting thermogravimetric (TGA) and differential scanning calorimetry (DSC) analysis. As shown in Fig. S3a, the DSC curve shows a weak endothermic peak at 79 °C and a strong exothermic peak at 118 °C, resulting from the evaporation of water and solvent from the ZTO-gel. The gravimetric weight loss in the ZTO precursor solution is shown in Fig. S3b. The weight loss started slowly below 79 °C and it could be attributed to evaporation of organic additive in the ZTO precursor. The second stage occurred very rapidly at about 116 °C and it is attributed to the evaporation of solvent, which has a boiling point between 110–120 °C from the ZTO-gel. The third stage in the weight loss process occurred above 150 °C and it can be attributed to the decomposition of nitrate ligands from the organic group, which is associated with the Zn-source. Final stage of the degradation process occurred above 350 °C and it could be attributed to the decomposition of the chloride ligands related Sn-precursor. This process continues until about 450 °C, which suggested that full degradation of organic moiety from the ZTO could be achieved at this point. The total mass loss during the decomposition process from 20 – 540 °C is 70.28%. According to the TG/DSC result of

Salgueiro et.al [1], 2-methoxyethanol has the possibility to serve as a fuel during the sol-gel synthesis of ZTO precursor solution. So, the same could be possible for 2-methoxyethanol solvent in assuming the role of a fuel or stabilizer to initiate the formation of complex ligands during the reaction between Zn and Sn-precursors. This is totally different from its initial solvation duty; thus, lowering the decomposition temperature of ZTO-gel. Although, this has not been carefully studied, but it is expected that when Zn-precursor and Sn-precursor are mixed to form the ZTO solution: NO_3^- , and Cl^- ions from both Zn and Sn sources respectively are free to react as an oxidizing agent due to their combustive nature [1] [2].

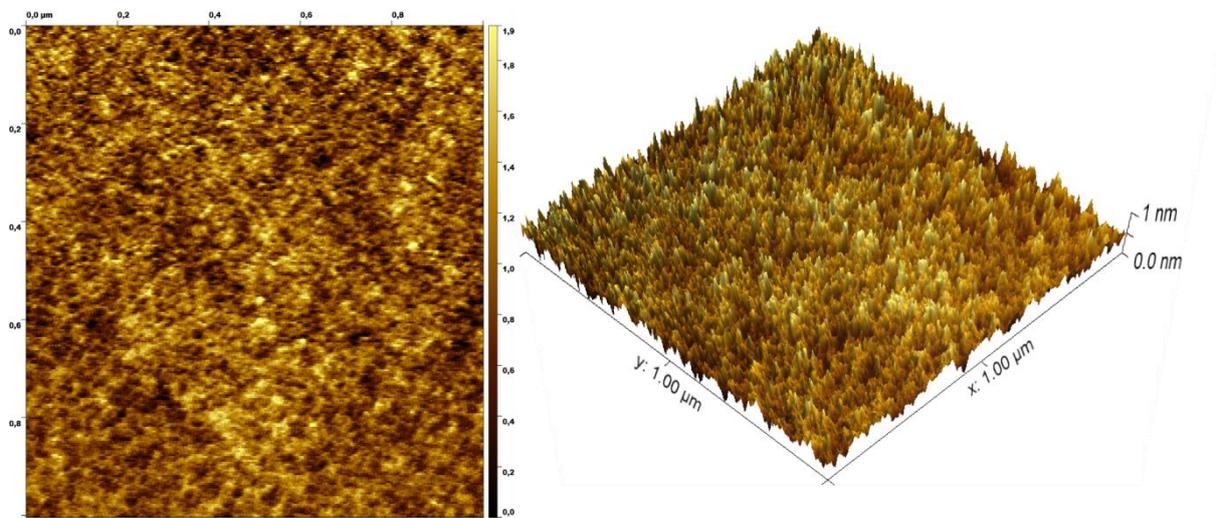


Figure S4. Surface morphology of the deposited ZTO semiconductor film after annealing at 350 °C.

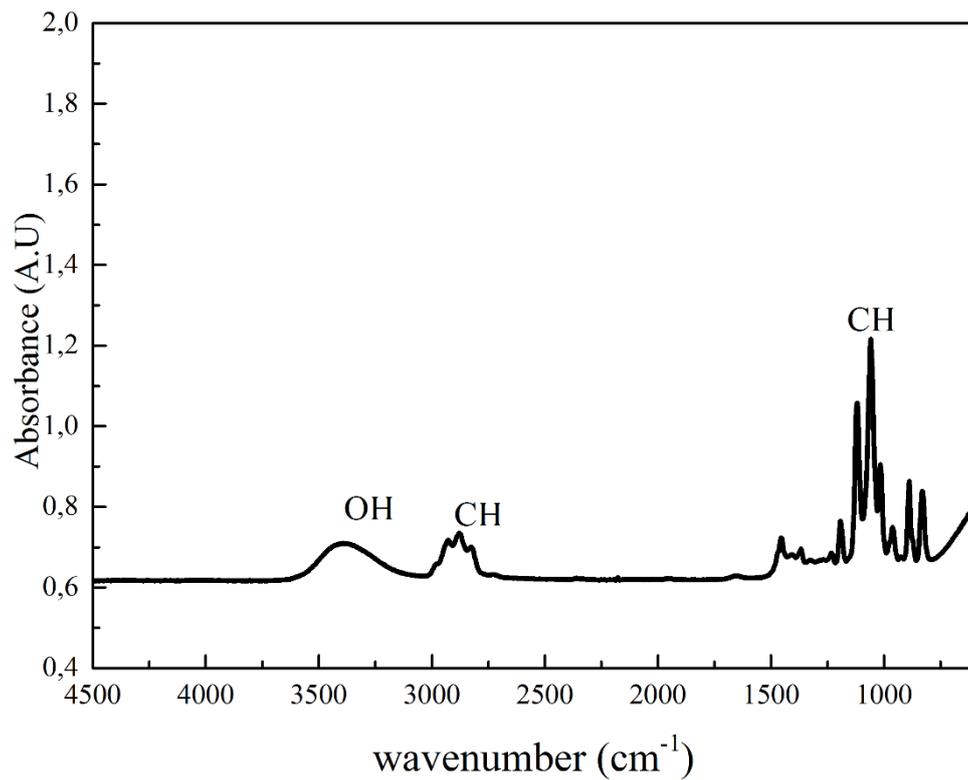


Figure S5. ATR-FTIR spectrum for the ZTO precursor solution at room ambient temperature.

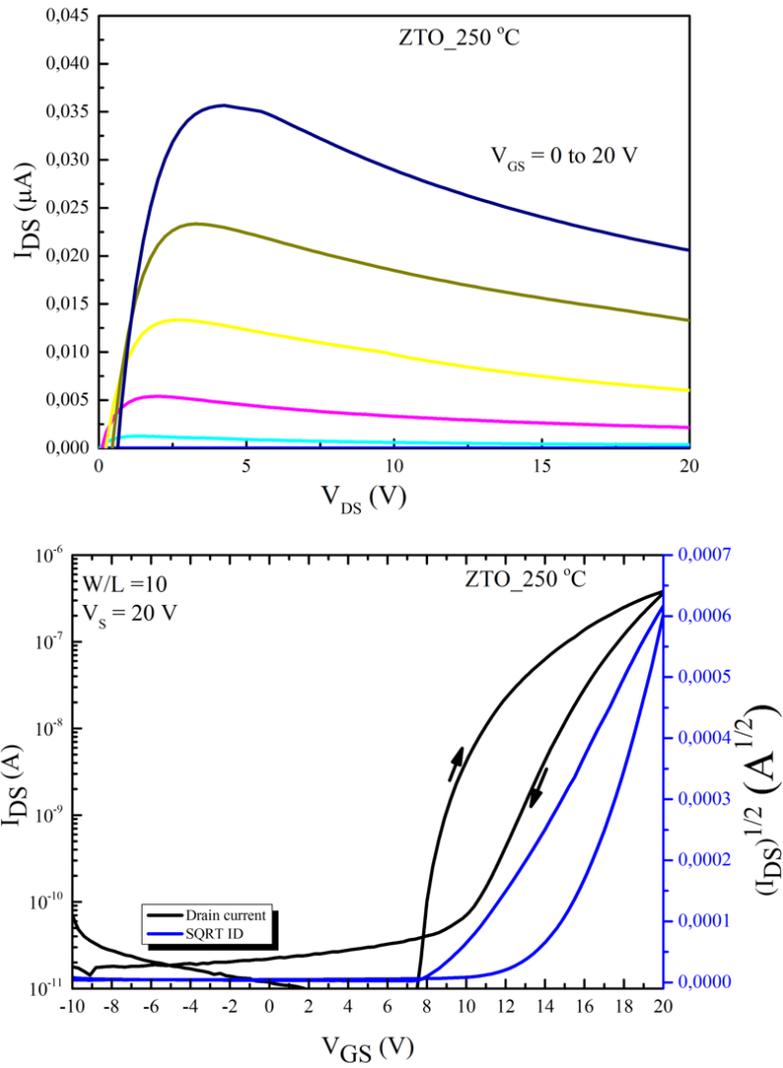


Figure S6. Electrical performance of the ZTO-250/SiO₂ TFT device.

Table S1. Statistical summary of all the important electrical parameters extracted from both the MIS and TFT device studies.

Note: Ave = average, S.D = standard deviation; number of samples analysis: 10 MIS devices, and 5 TFT-Devices

MIS-Device					TFT-Device												
Dielectric k	capacitance (nF)	leakage current (A/cm ²)	Sat. Mob. (cm ² V ⁻¹ s ⁻¹)	Von (V)	Vth (V)	SS (V.dec ⁻¹)	Hysteresis (V)										
Ave.	S.D	Ave.	S.D	Ave.	S.D	Ave.	S.D	Ave.	S.D								
8.5	0.3	0.36	0.03	4.24E-04	1.26E-05	ZTO-350/ZrO _x -400	4.61	0.06	-0.89	0.06	0.03	0.02	0.25	0.01	-0.18	0.06	
400	22.6	0.1	0.67	0.17	3.45E-06	1.06E-07	ZTO-350/SiOx	0.47	0.01	-0.3	0.5	1.32	0.08	0.76	0.01	-0.37	0.08

References

- [1] D Salgueiro, A Kiazadeh, R Branquinho, L Santos, Pedro Barquinha, R Martins and E Fortunato, "Solution based zinc tin oxide TFTs: the dual role of the organic solvent," *J. Phys. D: Appl. Phys.* , vol. 50, p. 065106, 2017.
- [2] A.K. Rai, L. T. Anh, J. Gim, V. Mathew, and J. Kim , "Electrochimica acta low temperature synthesis of porous tin oxide anode for high-performance lithium-ion battery Homog enously mixed solution stirred at 250 °C gel formation," *Electrochim. Acta* , vol. 109, p. 461, 2013.