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# Fabrication of Custom-Designed and Cost-Effective Spin-Spray Pyrolysis Unit

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***Abstract: In this paper, an efficient, spin-spray pyrolysis unit is designed and constructed for the deposition of oxide thin films. The system is an effective combination of two independent techniques such as spin and spray methods. The rotation of the substrate by keeping the spray nozzle stationary makes the system unique. The system allows the decomposition of the solution before reaching the substrate. Spin-spray pyrolysis unit is capable of coating thin films on different substrates like float glass, FTO coated glass, and Aluminum coated glass, Teflon, and kepton, of dimensions up to 4-inch diameter, any contour and scalable to industrial applications. Metal oxides like $Al_2O_3$, GZO, ZnO etc., can be coated for many applications such as solar cells, thin-film transistors, sensors, etc. The elements required for construction of spray pyrolysis units are a heater, spray nozzle, thermocouple, solution feeding unit, airflow assembly, substrate rotator, and exhaust assembly. The elements of the system are discussed in accordance with the cost estimation. The working principle of each element of the system is explained in a separate block diagram. The system is optimized for deposition of ZnO thin films on a glass substrate and is characterized. The thin-films can be used for development of TFT's, heaters, thermistors, piezo-electronics devices, sensors, antireflection coatings and solar cell.***

***Keyword: Spray pyrolysis unit; thin films; instrumentation;***

## I. INTRODUCTION

A predominant step in fabrication technique is the deposition of the material on the different substrates. The deposition technique is the process of depositing a layer by controlled transfer of atoms from a source to a substrate located at a distance. Various techniques such as chemical vapour deposition[1], plasma arc enhanced chemical vapour deposition, sol-gel, reactive evaporation, RF magnetron sputtering, spin coating[2], spray pyrolysis[3], molecular beam epitaxy, pulsed laser deposition, etc., have been used for the deposition of thin films and each method has its merits and demerits[4]. The challenges in modern engineering are to improve the deposition technique by, reducing the temperature of the substrate, designing of more economical system, uniform deposition, repeatability in results, simplifying masking and coating of large areas[5][6]. Spray pyrolysis is getting attention in the scientific community due to its cost-effectiveness and ease of integration. This method can be used for the deposition of thin-film and crystal growth[7].

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Spray pyrolysis has been developed as a powerful tool in the preparation of thin-film for various kinds of technological materials such as metals[8], metal oxides[9][10][11], superconducting materials, Nanophase materials and carbon nanotubes (CNT) [12][13][14]. The thin films deposited in this method have several applications in various fields of electronics such as Thin Film Transistors (TFTs)[15], sensors, photo-detectors, solar cells[8], thermistor[16] and other devices [17]. The most common problem addressed by many researchers is uniformity of the films due to blockage of the nozzle on one side or due to variation in chamber pressure caused due to waste gases evacuation. In this paper, we have combined two deposition methods, i.e., spin and spray pyrolysis and constructed the system. The advantages of spin-spray pyrolysis process help to deposit thin films, this process crosses the barrier of overcoming the failures of the two systems such as the uniformity of spray and masking problems of spin coating. The system is optimized for repeatability and uniformity of the ZnO thin films deposition.

## II. DESIGN

**Schematic Diagram**

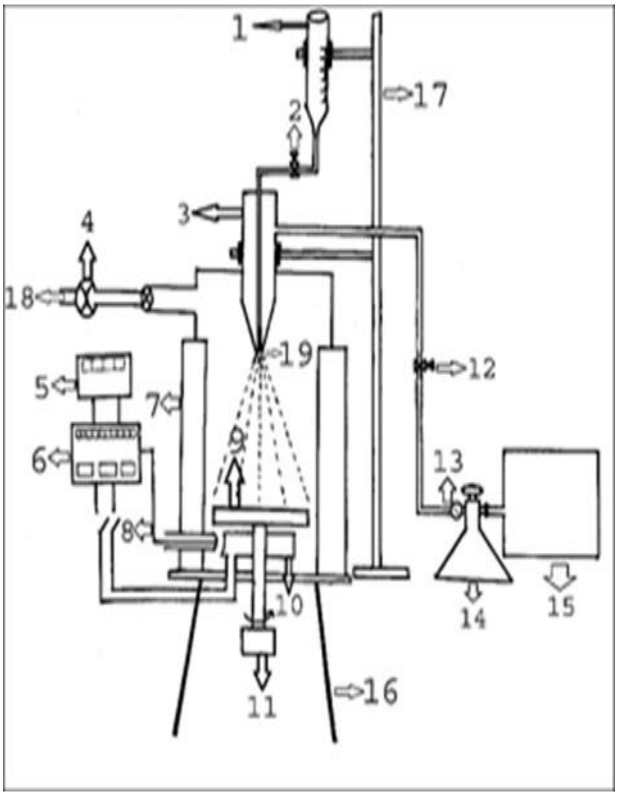


**Figure 1. Schematic diagram of spin spray pyrolysis**

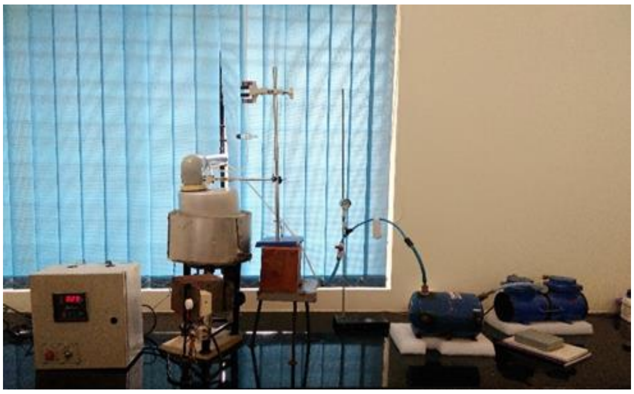

**Figure 2. The spin-spray pyrolysis equipment**

**Component List**

1) Burette 2) Solution feeder valve 3) Sprayer 4) Exhaust fan 5) Temperature display 6) Temperature controller unit 7) Chamber 8) Thermocouple 9) Substrate holder 10) Heater 11) Substrate rotator 12) Air control valve 13) Pressure gauge 14) Air cylinder 15) Compressor pump 16) Equipment stand 17) Burette holder 18) Air exhaust 19) Spray nozzle

## III. COMPONENT DESCRIPTION

### A. Heater And Control Unit

This unit includes an embedded heater, K type thermocouple, and temperature control assembly. The heater is made of nichrome wire with a capacity of half a kilowatt. The heater is embedded in alumina cement to get the uniform temperature and to avoid shorting. The heater is capable of attaining a temperature of $400^0$C. The thermocouple is a K type thermocouple which has the temperature sensing capacity of $1000^0$C with $\pm1^0$C accuracy. Control unit assembly consists of the temperature display unit and relay. The temperature display unit consists of a switch and knob to set the temperature, as shown in Figure 3. The relay is used to maintain the temperature with $\pm5^0$C accuracy. The heater, chamber, and control unit is fabricated in accordance with the design by Ravi Thermal engineering private limited Bangalore.

### B. Spray Head

The spray head consists of two inlets for feeding the solution and feeding the air. We have used a spray nozzle made of quartz with an inner diameter of 0.3mm. The spray nozzle is also capable of withstanding a higher temperature. The spray nozzle diagram is, as shown in Figure 4 and Figure 5.

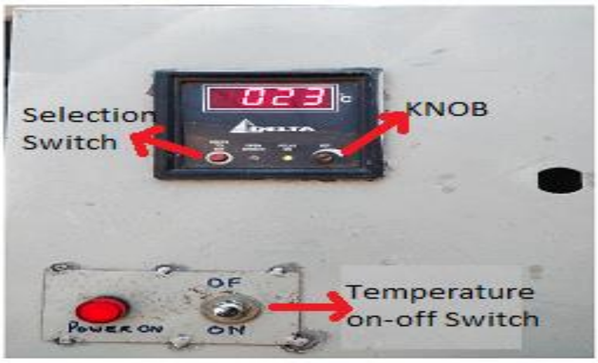


**Figure 3. Temperature Display Unit**

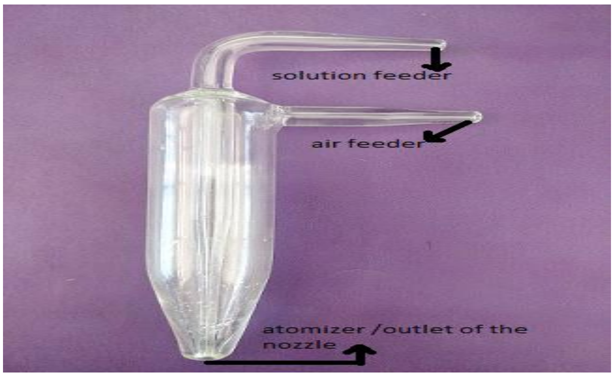


**Figure 4. Quartz Sprayer**

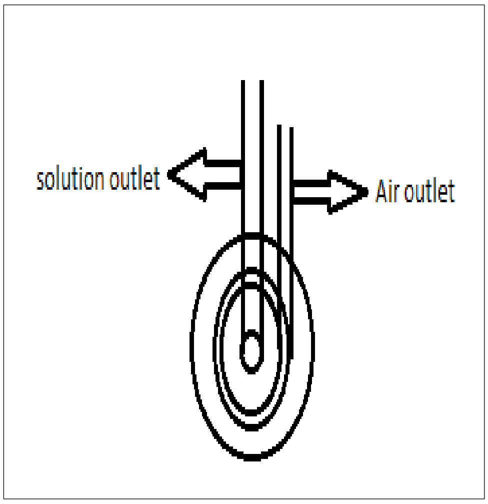


**Figure 5. Sprayer head**

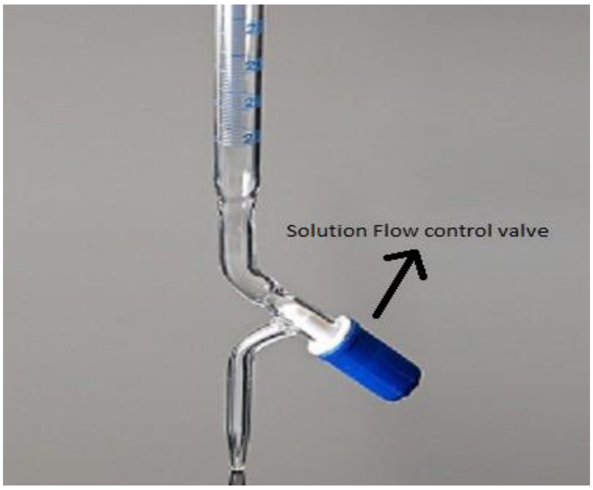


**Figure 6. Solution Feeder**

### C. Solution Feeder

Solution feeder consists of a burette which feeds the solution to the nozzle inlet using a pipe. The solution flow is controlled by the valve of the burette. The flow can be adjusted to 3ml/min to 0.5ml/min, as shown in Figure 6.

### D. Air Exhaust Assembly

Air exhaust assembly consists of a duct and exhausts fan of 4" diameter 2600-2800 rpm rotation speed 96-105 CFM airflow and is used to remove unwanted gases from the chamber. It also helps in maintaining uniform pressure in the chamber.

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If unwanted gasses in the chamber is not removed the possibility of catching fire is higher due to the presence of alcohol in the solution.

### E. Airflow Assembly

Airflow assembly consists of a compressor pump, cylinder, pressure gauge, and valve. The valve used is to control the airflow which is fed to the air inlet of the sprayer. As the compressor is not capable of maintaining constant air pressure, the air is fed to the cylinder, and then the uniform pressure is maintained by the valve and gauge. The cylinder and the valve assembly help to maintain the pressure and also to alter the pressure if required so that the droplet size can be altered.

### F. Chamber

The chamber consists of the two concentric hollow circular tubes of diameter 6.5" and 8". The inner tube is made of stainless steel, which is chemically inert. The outer is made of the aluminium sheet. Glass wool is incorporated between, the two cylinders to provide an electrical and thermal insulator. The chamber consists of a heater assembly with a hole at the centre to connect the job holder with the rotary shaft for rotation of the job holder.

### G. Substrate Rotation

The chamber resides a rotary unit with rotating speed in the range of 50-100rpm. The shaft of the motor is connected to the substrate holder through the heater. The substrate rotator is used for making the uniform coating on the substrate. As the speed of the rotator increases, the uniformity of the film also increases.

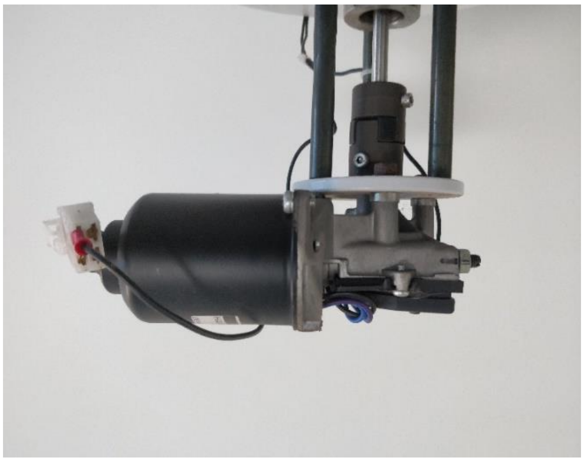

**Figure 7 Substrate rotator**

## IV. WORKING OF THE SYSTEM

The working of the system is classified into three steps as mentioned below

1. Atomization of a precursor solution in the nozzle to form tiny droplets.
2. Spraying the droplets onto a preheated substrate.
3. Decomposition and adhesion of the material into a thin film onto the substrate.

The substrates are loaded into the system. The heater is set to the required temperature and allowed to heat. The system is made to attain an equilibrium state, during this time, the solution is fed to the solution feeding unit (burette). The pressure is set and maintained at 1 bar constantly. Consequently, the substrate rotator is switched on and set to the required RPM (rotation per minute). The sprayer is ensured for non-blockage. The valve of the solution feeder is adjusted to a flow rate of 1ml/min. The pressure is set, and the later spray pyrolysis unit is ready for coating. After the coating is completed the airflow and solution feeding valve are closed. Substrate rotation is stopped. The sample is allowed to cool, and then the substrate is removed.

## V. MERITS OF THE SPIN SPRAY PYROLYSIS UNIT

The spin spray pyrolysis unit is a simple and quick method for the deposit of metal oxide[18][19], CNT, polymers thin films. It is one of the best choices for deposition of low cost, large area scalable thin films. The system is capable of utilizing material effective for thin film deposition. The system is equipped with the rotation mechanism to avoid the movement of the sprayer, which is not available in the existing system. When compared to the existing system a nozzle can be used instead of an ultrasonic atomizer. The diameter of the spray drops depends on the size of the nozzle head, the surface tension of the precursor solution, its viscosity and the pressure difference before and after the spraying. Alternatively, ultrafine dispersive powders[20] could be produced by the swift rise of the temperature inside the chamber. The rotation enables uniform film deposition throughout the area. The weight and size of the system are lighter and smaller. The system can be assembled and disassembled at any point in time, which makes it easy to handle and portable. The system is fabricated with the cost of 60,000/- in Indian Rupees (INR), which is lower than the existing systems which are not less than 10,00,000/- INR.

## VI. MATERIALS AND METHODS

### A. Materials

Zinc Acetate Dehydrated (LOBA Chemie), Multiwall carbon nanotubes (United Nanotech Innovation Pvt. Ltd) and Gallium Nitrate Hydrated (Alfa Aesar), plain glass, Aluminum coated glass, FTO glass procured from HHV Private Limited Bangalore, and Teflon sheets.

### B. SYNTHESIS OF Zno

Zinc acetate dehydrated precursor is dissolved in a mixture of method and deionized water (2:1) to form a solution of 0.3M. While stirring the solution, a few drops of acetic acid are added to form a transparent solution.

### C. Synthesis Of Gzo

Gallium nitrate hydrated precursor separately was dissolved in methanol form 0.3M gallium oxide solution. Zinc acetate dehydrated precursor has used to synthesis the ZnO solution of 0.3M. The volumetric addition of gallium oxide solution is added to ZnO solution. The doping of Ga to ZnO is varied from 1% to 10%. The coating is performed at 5% doping.

### D. Deposition Process

The substrates are loaded onto a job holder and kept in the chamber. The distance between the spray head to the substrate is set to 25 cm. The solution is fed to the solution feeder unit. The air pressure is set to 1 bar. The heater is switched on set to 250℃. After attaining the temp 250℃, it maintained for 15 minutes to attain equilibrium inside the chamber. The exhaust is switched on. The substrate rotation is kept to maximum speed. The air admittance value & solution feeder value is set to open. The solution flow valve is set to 2-5ml/min. After completing the coating, the solution feeder value and air admittance value is also closed. The substrate rotation is turned off. The exhaust and heater are switched off. The substrate is allowed to cool to the room temperature and then the job holder is removed.

### E. Characterization

The characterization of ZnO thin films deposited in the system was characterized for thickness and roughness by optical profilometer CENSE Laboratory IISc, Bangalore and AFM at Materials Research Center IISc, Bangalore. The SEM and EDAX were performed at PURSE Laboratory Mangalore University.

## VII. TESTED RESULTS

The ZnO[21][22] thin film is deposited using this spin-spray pyrolysis unit on substrates like FTO coated glass; Aluminum coated glass, Teflon[23] and Float glass as shown in Figure 8. The experiment was also carried with different coating material like CNT, GZO, and ZnO is carried on the glass substrate as shown in Figure 9.

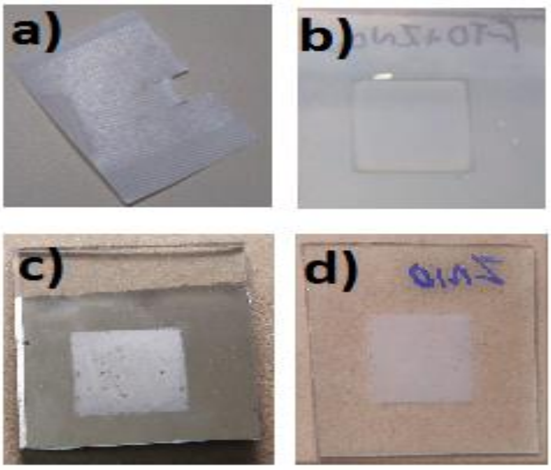


**Figure 8. Zinc Oxide on a) Teflon b) FTO coated glass c) Aluminum coated glass d) Glass**

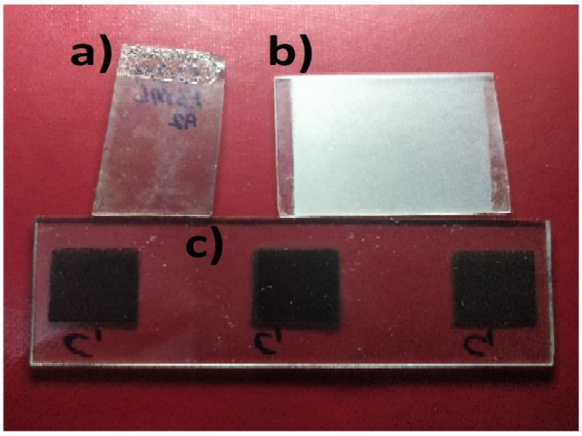


**Figure 9. a) ZnO b) GZO c) CNT thin films coated on a glass substrate**

In this system, trials are carried out with different parameter variations such as rotation speed, flow-rate of the solution, the concentration of the solution, substrate temperature, spraying distance and air pressure for the sprayer. The coating experiment was repeated with different configurations of parameters. The trails were conducted to find the relation between thickness with the amount of solution sprayed and are graphically represented in Figure 10.

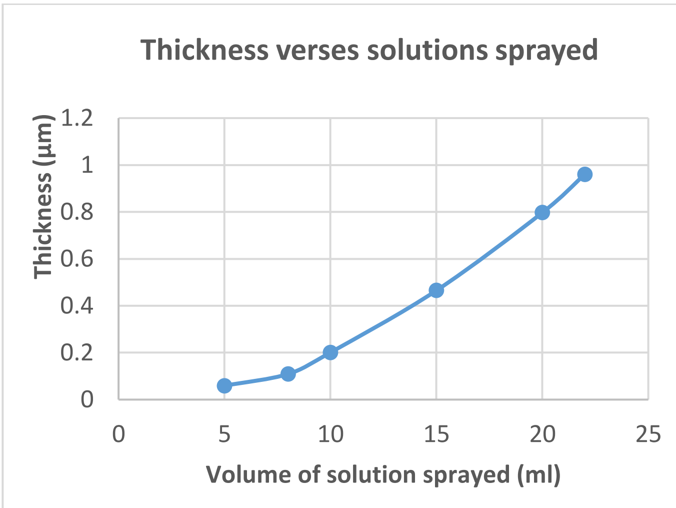


**Figure 10. Thickness versus solutions sprayed**

The variation of the temperature of the substrate is carried out in the range of 150℃ to 300℃ and is as shown in Figure 11.

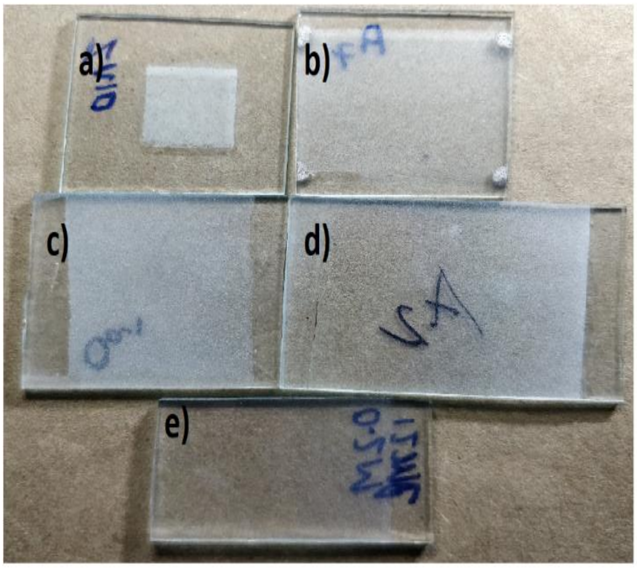


**Figure 11. ZnO thin films coated at a)150°C b)200°C c)250°C d)270°C e) 300°C**

Figure 12 and Figure 13 illustrates thickness which is found to be 250 nm, and the roughness is 75 nm of the ZnO films examined by using Optical Profilometer. Figure 14 shows the AFM images of ZnO deposited at 270°C. The thickness is in the order of 109 nm and roughness in order of 20nm as calculated from Figure 15.

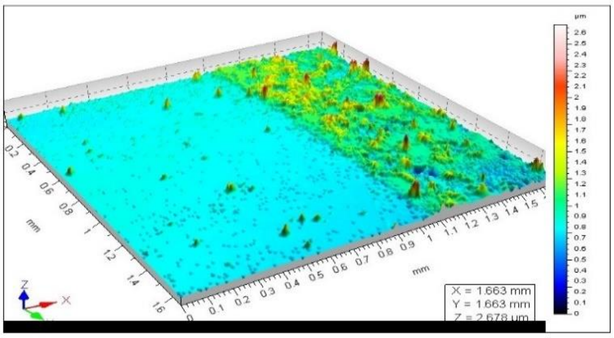


**Figure 12. Thickness of the ZnO thin film deposited at 200°C.**

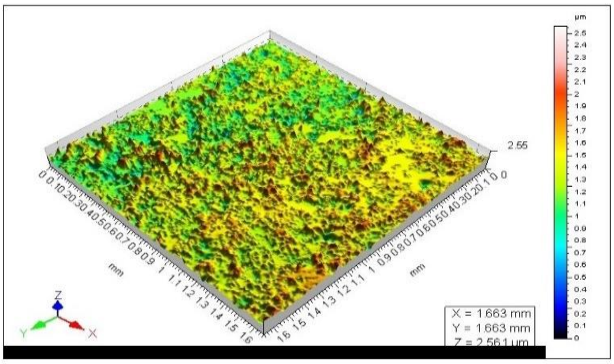


**Figure 13. Roughness of the ZnO thin film deposited at 200°C.**

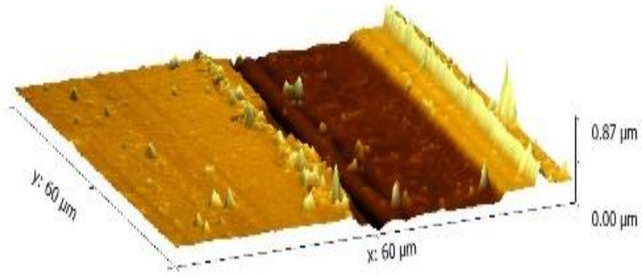


**Figure 14. AFM image of ZnO thin film deposited at 270°C**

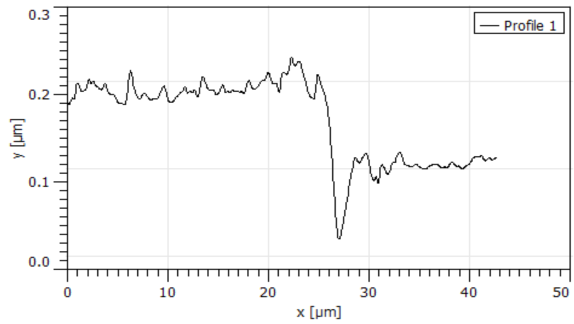


**Figure 15. Graph of AFM to measure thickness and roughness.**

The SEM and EDAX are carried out and the images of ZnO deposited at various temperature shows the variation in the morphology of the ZnO thin film. The lower temperature up to 220°C results to formation of the amorphous state. Whereas the temperature above 270°C will result in the creation of different structures like leaves, flower and rods as shown in Figure 16. EDAX confirms the presence of Zinc oxide in the films Figure 17.

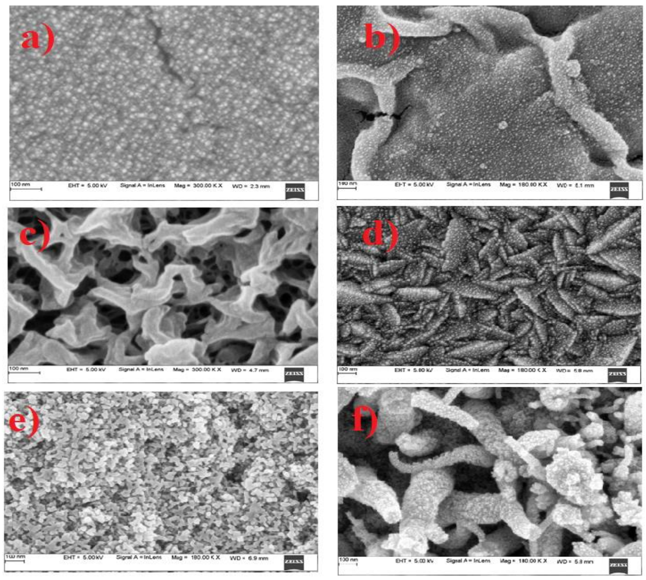


**Figure 16. ZnO thin films deposited at a different temperature a) 150°C b) 170°C c)200°C d) 250°C e)270°C f)300°C**

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| Elem... | Weight% | Atomic% | Compd% | Formula |
|---|---|---|---|---|
| O K | 33.34 | 44.94 | 45.86 | O2C |
| Si K | 11.35 | 8.71 | 0.00 | |
| Zn L | 72.40 | 23.88 | 0.00 | |
| C | 12.52 | 22.47 | | |
| Totals | 129.61 | | | |

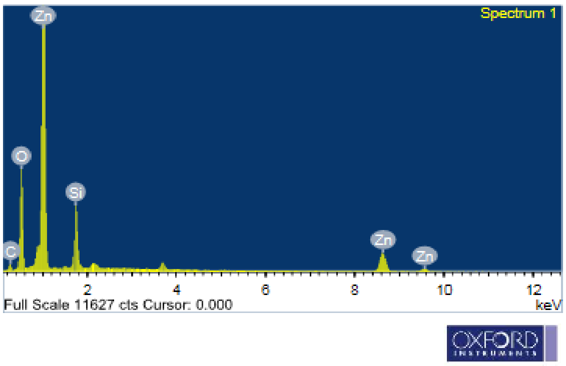


**Figure 17. EDAX of the ZnO thin film.**

## VIII. CONCLUSION

An economical, simple spin-spray pyrolysis unit is designed, constructed, and optimized. The cost of the developed system is 60,000/- INR which is much cheaper than existing systems. The novelty of the system is the integration of substrate rotation to improve the uniformity in thin-film formation. The specifications and working principles of each component of the system are explained. Using the system, numerous samples are coated on float glass, FTO coated glass, aluminium coated glass, and Teflon. The coating is performed with varying temperature, varying thickness and different active materials are carried out. The uniformity, roughness, and morphology of coated samples are studied. The temperature variation resulted in the formation of different structures as revealed by SEM images. The change in volume of the active material resulted in variation of thickness. The roughness of the film is dependent on the temperature.

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He is currently working toward the PhD degree in the field of fabrication Thin Film Transistor for microelectronic circuit design in Mangalore University, Mangalore, India. He has presented 10 international and 1 national research papers and published 5 international journals and 1 meeting abstract on his ongoing work.

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