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In situ resistance measurement during FCVA deposition of ZnO thin films

S.M. Sim, B.J. Chen, X.W. Sun*

School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore

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Abstract

In situ resistance measurement of zinc oxide (ZnO) thin films deposited by filtered cathodic vacuum arc (FCVA) technique was performed using a four/two-point probe method. Large oscillations were observed during ZnO thin films resistance measurements. The resistance dramatically increased during the arc plasma was on, and decreased when the arc plasma was off. During the arc plasma was on, a space charge region of positive and negative plasma ions is gathered on the substrate surface, which acted as a potential energy barrier preventing electrons to move across, hence increasing the resistance measured by the voltmeter. The initial chamber pressure strongly affects the deposition rate of ZnO. A capacitive effect is also observed when four-point probe configuration is adopted. In situ resistance measurements will be useful to monitor deposition processing and also may help to optimize and understand the properties of thin films.

Keywords: In situ resistance; Filter cathodic vacuum arc; Four-point probe; ZnO thin films

The initial growth dynamics of thin films is important in determining the ultimate properties of the thin films [1–3]. To monitor and study the initial growth mode of zinc oxide (ZnO) thin films deposited by filtered cathodic vacuum arc (FCVA) technology, we used a direct technique based on in situ resistance measurement. Such studies are possible because of the interdependence of resistivity with the oxygen content or the crystalline structure of the ZnO thin films.

The FCVA technology has been widely used to produce good quality dense carbon coatings [4]. FCVA allows the use of low temperature for deposition of thin-films while achieving high deposition rate. The coatings are also of a much superior quality as compared to other sputtering techniques, as they are generally homogenous, dense, hard, corrosion and reproducible [5].

The schematic of the FCVA system is shown in Fig. 1. It utilizes high-energy electric and magnetic field to filter off unwanted micro particles and neutrals. The impact energy of the depositing ions at the growth surface can be readily controlled using electric field by means of ionization of the cathode materials in the plasma. The high-energy plasma plume will ionize readily with most of the background gases. Only the ions that are within the well-defined energy range will be able to reach the substrate and be deposited onto the surface of the substrate. This will ensure that the high quality films are coated.

A cylindrical target, which comprised of 99.95% of zinc and 0.05% of gallium, was mounted onto a water-cooled stainless-steel block. A thermocouple is placed under the substrate in the chamber to monitor the deposition temperature. The arc current of 60A was used to ignite the plasma. Nitrogen (N_2) and oxygen (O_2) was supplied into the chamber with a constant flow rate of 5 sccm for each gas. A toroidal magnetic field of about 40 mT was employed to produce the axial and curvilinear fields to steer the plasma. The initial conditions in the vacuum arc need to be optimized so only ions with energy that are within the specific range are able to successfully reach the substrate.

The substrate used in this experiment is ITO glass with sheet resistance of 50 kΩ. The center part of the ITO substrate had been etched off leaving only the glass. The ITO served as the conductor between the substrate and the wires, which were connected to the current source and voltage multimeter.

The resistance of the film was measured using a four/two-point probe technique during deposition as shown in Fig. 2. The four-point setup as shown in Fig. 2(a) has

* Corresponding author. Tel.: +65-6790-5369; fax: +65-6793-3318.
E-mail address: exwsun@ntu.edu.sg (X.W. Sun).

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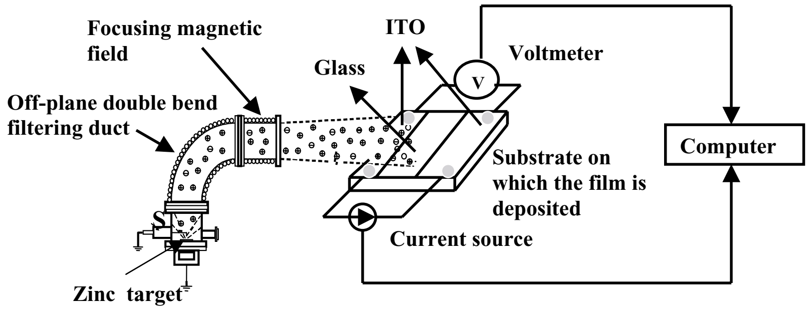

Fig. 1. Experimental setup for in situ resistance measurements using in the FCVA system.

three strips of ITO etched-off exposing the substrate. The remaining strips of ITO acted as electrodes where copper probes of the resistance measurement setup were connected. The two-point probe setup is shown in Fig. 2(b). A current source was used to bias the substrate and the voltage across the film was measured and recorded by interfacing the setup with a LabView program used for resistance measurement on a computer. The substrate was placed in the vacuum chamber in the FCVA system and the wires that were used to connect the system to the measurement equipment were attached to two contacts outside the system and then in turn connected to the substrate.

The in situ resistance readings obtained from a sample with four strips of ITO and a sample with two strips of ITO during FCVA deposition were plotted in a resistance against time as shown in Fig. 3a and b, respectively. Tables 1 and 2 show the experimental readings of the four and two-point probe configuration, respectively.

The four peaks in Fig. 3a correspond to the instance of switching the plasma 'on' or the ignition of the arc current. It has been believed that when the arc current is ignited, the resistance measured should be very low. This can be attributed to the fact that the 'sea' of ions and electrons would provide excellent conduction between the probes, across the sample. However, from the plots obtained from FCVA deposition using both configurations as shown in Fig. 3a and b, contrary to the above mentioned belief, it was observed that the resistance measured when arc current is ignited was very

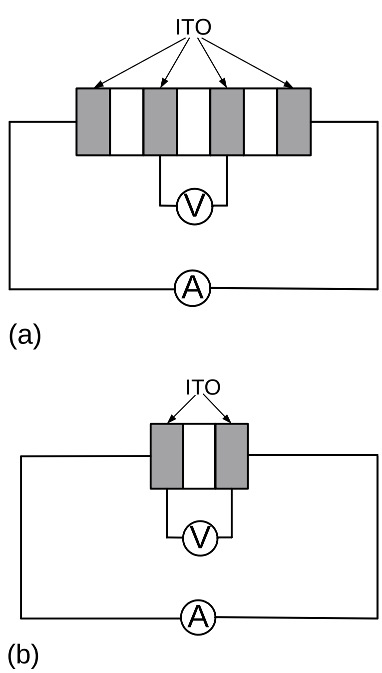

Fig. 2. Two-point probe configuration (a), and four-point probe configuration (b) used.

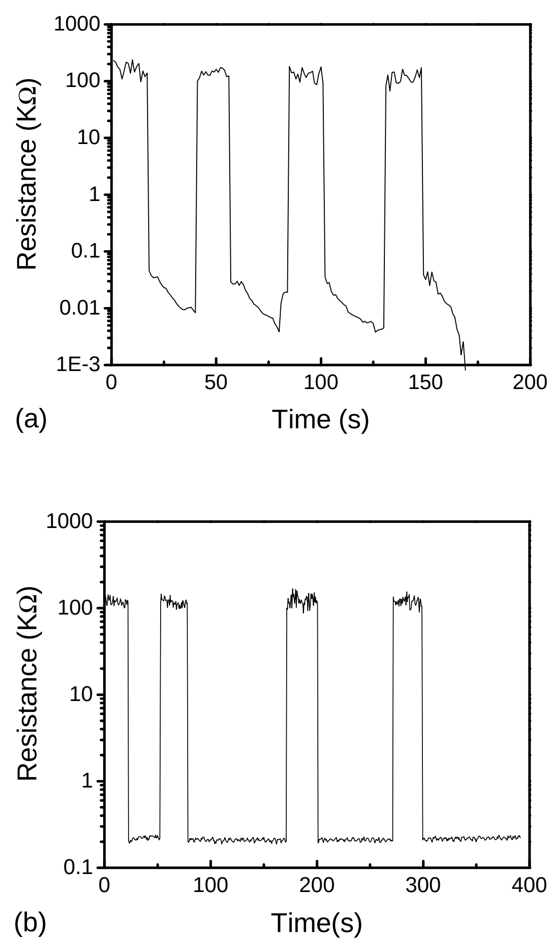

Fig. 3. Plot of resistance vs. time for two-point probe configuration (a), and four-point probe configuration (b).

Table 1
Experimental reading of resistance vs. time for four-point probe configuration

No.	Plasma	Resistance (kΩ)	Pressure (Torr)
1	On	161	6.5×10^{-5}
2	Off	0.045	7.5×10^{-5}
3	On	101	6.0×10^{-5}
4	Off	0.02911	7.5×10^{-5}
5	On	180.6	5.5×10^{-5}
6	Off	0.0137	7.0×10^{-5}
7	On	83	5.5×10^{-5}
8	Off	0.0388	7.5×10^{-5}

The serial number indicates the sequence of the plasma being turned on and off.

high, reaching values of as high as 100 kΩ. Conversely, the resistance drops when the plasma is in the 'off' state. Each of plasma 'off' state lasts approximately 1 min each.

A probable explanation for this phenomenon was put forward. When the chamber is under a plasma condition, the sample is coated with a layer of ions. During the injection of plasma, a space charge region of positive and negative plasma ions is gathered in random orientation in fixed space on the glass surface etched from the ITO substrate. Generally, the positive and negative ions in the plasma gathered on the glass surface are without specific charge orientation because they are immobile due to their relatively heavier mass and larger size as compared to free moving electrons. The positive and negative ions on the whole in this randomly orientated space charge region will neutralize each other and will not give rise to an electrostatic potential difference. This built-in electrostatic potential difference will arise if the positive ions and negative ions aligned themselves in the direction of the negative and positive terminal of the applied field respectively. This randomly orientated space charge region basically acts as a physical barrier obstructing the flow of the 'sea' of electrons across it, hence increasing the resistance measured by the voltmeter during plasma injection. When the plasma condition is off, upon deposition of ZnO film on the center portion of the substrate initially etched to expose a glass surface, resistance will decrease to values of about 0.1 kΩ as shown in Fig. 3a and b.

Table 2
Experimental reading of resistance vs. time for two-point probe configuration

No.	Plasma	Resistance (kΩ)	Pressure (Torr)
1	On	142.0	6.5×10^{-5}
2	Off	0.206	7.5×10^{-5}
3	On	114.1	7.0×10^{-5}
4	Off	0.206	7.5×10^{-5}
5	On	99.0	7.0×10^{-5}
6	Off	0.193	7.5×10^{-5}
7	On	134.9	7.0×10^{-5}
8	Off	0.206	7.5×10^{-5}

The serial number indicates the sequence of the plasma being turned on and off.

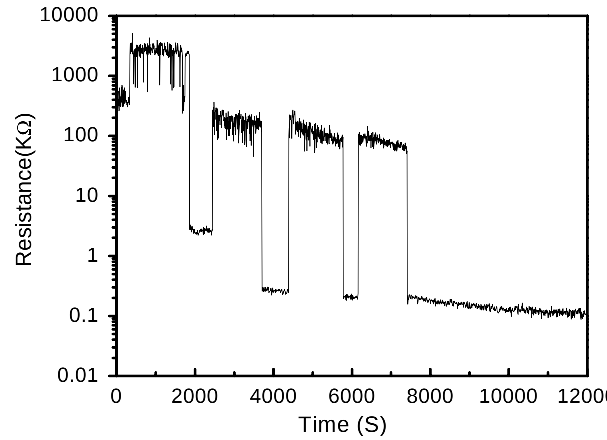

Fig. 4. Plot of resistance vs. time for FCVA deposition at initial chamber pressure of lower than 4.0×10^{-5} Torr.

By keeping the initial conditions stated above constant, a similar sample with two strips of ITO was used for FCVA deposition at an initial chamber pressure lower than 4.0×10^{-5} Torr. The plot of the corresponding in situ resistance of this FCVA deposition is shown in Fig. 4. Table 3 shows the experimental readings of this FCVA deposition. Comparing to the resistance plot of the FCVA deposition shown in Fig. 3b, which has a higher initial chamber pressure of 5.5×10^{-4} Torr, the resistance plot in Fig. 4 has a significant change in resistance for subsequent plasma on–off. By relating the resistance measured to the deposition rate of the ZnO thin films, we can deduced that initial chamber pressure strongly affect the rate of depositing ZnO thin films in subsequent plasma on–off. We deduce that once the resistance measured reached a constant value after subsequent plasma pumping process, the equilibrium is being reached between the deposition rate of ZnO on the substrate and the etching rate of ZnO deposited on the substrate by subsequent plasma pumping processes. Hence, we concluded that the initial chamber pressure strongly affects the rate at which this equilibrium between the deposition rate and etching rate of ZnO is reached.

Unlike using two-point probe configuration, it is also noted that when adopting four-point probe configuration,

Table 3
Experimental readings of resistance vs. time for FCVA deposition at initial chamber pressure of lower than 4.0×10^{-5} Torr

No.	Plasma	Resistance	Pressure (Torr)
1	On	2 MΩ	9.0×10^{-5}
2	Off	2.5 kΩ	1.4×10^{-4}
3	On	100–250 kΩ	4.6×10^{-5}
4	Off	250	1.5×10^{-4}
5	On	100–200 kΩ	5.5×10^{-5}
6	Off	200	1.7×10^{-4}
7	On	70–100 kΩ	7.0×10^{-5}
8	Off	200	1.8×10^{-4}

The serial number indicates the sequence of the plasma being turned on and off.

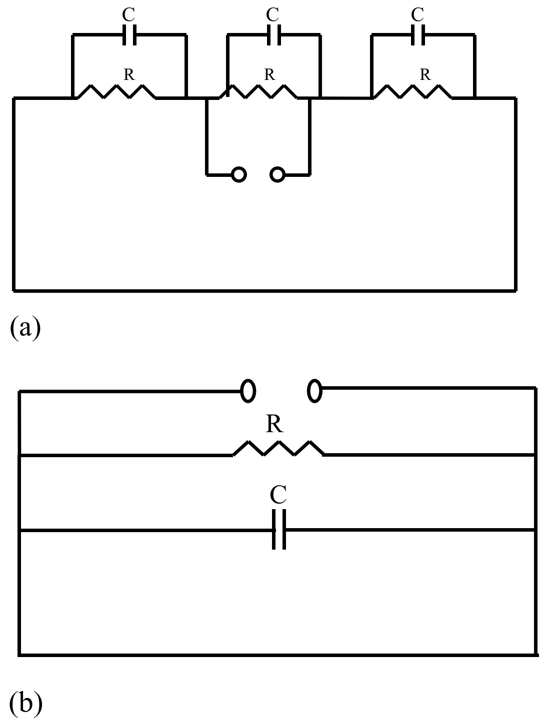

Fig. 5. Model of two-point probe configuration (a), and four-point probe configuration (b).

when the arc current is switched off, the resistance decreases with time. A possible explanation lies with the configuration used in this experiment. This might be better understood by looking at the Fig. 5a.

By modeling the setup in Fig. 2a as a simple circuit diagram in Fig. 5a, the three areas of the exposed substrate are modeled as simple resistors in parallel with capacitors. The voltmeter is connected in series with the center resistor. Since a voltmeter has ideally high internal impedance, it is modeled as an open circuit. The ammeter is connected in series with all three resistors and has ideally zero internal impedance and therefore is modeled as short circuit. It then becomes clear that there is a sort of charging and discharging effect due to the presence of the open circuit in series with the center resistor. Hence, when the arc current is switched off, there is a delay as the circuit discharges any stored charge, which results in the decreasing resistance that is observed during the plasma ‘off’ period. This phenomenon is absent in the other experiment, which adopts a two-point probe configuration.

The phenomenon of decreasing resistance is absent when we connect the voltmeter and ammeter onto two strips of ITO. Again, as shown in Fig. 2b, we model the setup shown in Fig. 5b as a simple circuit diagram so as to gain an insight for the constant resistance obtained in this case. The center portion of the substrate being etched away is modeled as a simple resistor in parallel with a capacitor. There is an absence of the charging and discharging effect using this configuration unlike the case using the four-point probe configuration. This is because any charges stored during the plasma ‘on’ period will be instantaneously discharged through the short circuit path modeled to represent the ammeter which has an ideally zero internal impedance. Hence, the resistance measured during the plasma ‘off’ period was relatively constant.

In summary, in situ resistance measurement was employed to study the onset of plasma ‘on’ and plasma ‘off’ effects during the deposition of ZnO using the FCVA technique. It is found that the resistance of the substrate measured during the plasma ‘on’ and plasma ‘off’ conditions was high ($\sim$100 kΩ) and low ($\sim$0.1 kΩ), respectively. We concluded that this is because during plasma ‘on’ condition, the space charge region formed is randomly orientated and acts as a physical barrier obstructing the flow of the ‘sea’ of electrons across it, which contributes to the increase in resistance measured. The initial chamber pressure strongly affects the deposition rate of ZnO. A capacitive effect is also observed when four-point probe configuration is adopted, and simple circuit diagrams are used to illustrate this phenomenon.

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