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Humidity Sensing Properties of Transparent Sputter-Coated Indium–Tin Oxide and Printed Polymer Structures

Jack R. McGhee, *Member, IEEE*, Jagdeep S. Sagu, Darren J. Southee, and K. G. U. Wijayantha

***Abstract*— The humidity sensing properties of sputter-coated indium–tin oxide (ITO) and printed dielectric structures were tested for samples with sheet resistances ranging from 10 to 50 Ω/sq. ITO/Polymer composite sensors were fabricated to form a parallel-plate capacitive-based humidity sensor that could detect relative humidity within a tested range of 5%–95%. The sensors were most stable and gave a linear response between 5% and 75% relative humidity. The capacitive sensors were characterized, using a range of techniques, to establish their capability and performance as humidity monitors. Response time of the humidity sensors was measured to be an average of 31.5 s and the recovery time was measured at an average of 31 s in the capacitive mode. Complex impedance spectroscopy was used to determine the mechanism of action for the sensors, which was found to be both the diffusion of water molecules into the dielectric layer and an increase of ionic conductivity within the dielectric layer. Stability of the humidity sensors was tested at three different humidity levels over seven days and sensors were found to be stable or follow a predictable change for this time span.**

***Index Terms*— Physical and chemical sensors, humidity sensors, transparent conducting oxides, capacitive sensor, indium tin oxide.**

I. Introduction

HUMIDITY is an important factor in many electrical, industrial and environmental systems and therefore it is as essential as temperature to monitor with ease and accuracy [1], [2]. Environmental monitoring, agriculture and manufacturing of electronics require wide range, accurate and high response humidity sensors to ensure data is reliable [2]. Most personal electronic devices in recent times are now equipped with humidity sensors and domestic applications such as smart buildings for human comfort are becoming more popular.

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Typically, humidity sensors measure via two methods. The first is resistive sensing where chemisorption or physisorption of water molecules change the resistivity of the bulk material. The second method is capacitive sensing where the capacitance of the sensor changes upon interaction with water vapor [3]. The mechanism of sensing usually depends on the type of material used to fabricate the sensor. Three material types are used in the fabrication of humidity sensors, the first set of materials are ceramic metal oxides, with semiconducting perovskite structures being a popular subgroup [4]. Organic polymer films are also commonly used materials and detect humidity either via impedance change in a conducting polymer or through use as a dielectric for capacitive type sensors [2].

Although semiconductor materials typically make good humidity sensors and see widespread commercial use, little work has been done on using transparent humidity sensors. Therefore, the stability of these materials is an important factor and the ability to embed a sensor into the electronics to monitor degradation is of interest and is currently being researched [5]–[7].

Indium tin oxide (ITO) is one of the most commonly used and available transparent conducting oxides (TCOs), much research has been performed into gas sensing using ITO [8]–[14]. However, TCOs tend to lack the selectivity needed to become a viable gas sensor [2], [15]. Due to this, work has been conducted into the use of the humidity sensing properties of ITO as it also has an affinity for water vapor. Research has been conducted into the effect of annealing temperature on spin coated ITO films, showing that a higher annealing temperature increases humidity sensing performance [16]. The same group has investigated how morphology and crystal structure of the spin coated films affects performance of the sensor [17]. It has also been recently demonstrated that nano-structuring the ITO films can also affect the materials humidity sensing performance [18].

In the present, sputter coated, transparent, flexible ITO films are investigated for their humidity sensing properties. A screen printed dielectric polymer was incorporated into the sensor structure to develop a wide range of detectable relative humidity. The sensor structure incorporates both a metal oxide and polymer to give wide ranging capacitive measurements and highly sensitive resistive measurements. The tensile strength and sensing stability of the sensors are investigated, and the

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mechanism of sensing is determined using complex impedance measurements. Investigating the humidity sensing properties of ITO gives insight into the ability of the material to withstand humidity and helps determine potential failure points for future transparent electronics.

II. Materials and Methods

A. Materials

Indium tin oxide (ITO) films were prepared by magnetron sputter coating onto Dupont Teijin Films Melinex O (125μm, 89.5% transmittance) polyethylene terephthalate film (PET) at Diamond Coatings LTD. ITO films where prepared at varying sheet resistances (Ω/sq or Ω/□), sputter coated PET was prepared at 10, 20 and 50Ω/sq. Gwent Electronic Materials supplied UV-curable, screen printable transparent dielectric ink D2150901D1 which is polymer based.

B. Sensor Fabrication

Dielectric ink was printed onto a 7.5cm^2 area of a 10cm^2 indium tin oxide film using a DEK1202 screen printer. The screen used has a polyester thread with a thread count of 305 threads per inch (tpi). A second 10cm^2 indium tin oxide film with 1cm^2 removed was then placed on top of the wet dielectric layer leaving both sections of the ITO film and dielectric layer exposed while forming a parallel plate capacitor. The sensor was then cured for five minutes under a UV lamp (365nm) and removed. Copper tape was placed on the exposed ITO to create sturdier electrical contacts for measurements to be taken. The total area of the parallel plate capacitor after fabrication where ITO/Dielectric/ITO layers directly overlap is 5cm^2. Five sensors were fabricated with 10Ω/sq ITO, five more prepared with 20Ω/sq ITO and a final five prepared with 50Ω/sq ITO. The end contacts of the ITO film were exposed alongside a central dielectric square. This is to allow for the characterization of how the water molecules interact with the dielectric and the ITO film. These samples were also compared to sensors with the ITO film encapsulated, leaving only the dielectric exposed and compared to pure ITO film. The contribution of each part of the structure can then be determined.

C. Measurement Equipment

Capacitance measurements were taken using a UNI-T digital LCR meter (UT612), these measurements were taken using the LCR meter connected to a PC using the UNI-T digital multimeter software provided. Measurements of capacitance were taken with the LCR meter set to a frequency of 1KHz. Humidity measurements were taken using a Rotronic Instruments HygroGen humidity and temperature measurement chamber. The software controller for the chamber was the Eurotherm 2704 temperature and humidity controller.

Scanning electron microscope (SEM) images were taken using a Leo 1530 VP field emission gun scanning electron microscope (FEG-SEM). The accelerating voltage used for all SEM images was 5kV and the working distance ranges between 5.00mm and 6.1mm. Sheet resistance measurements were taken using a Jandel HM21 four-point probe. Transmittance data for the humidity sensors were taken using a Perkins Elmer Lambda 35 UV/VIS spectrophotometer with a 100% reflectance standard and the software package used was UV Winlab.

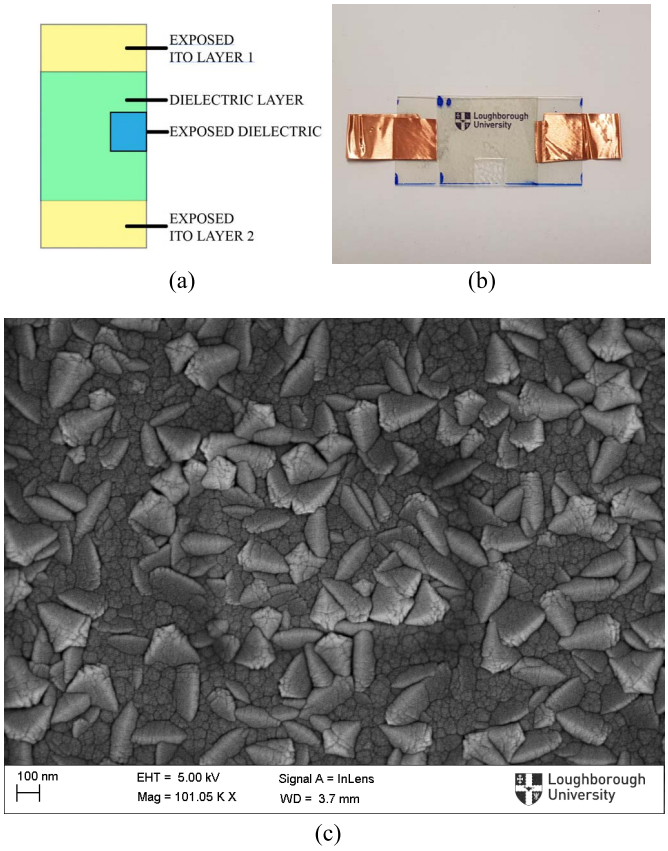


Fig. 1. Humidity sensor structure with: (a) humidity sensor structure showing top down view of parallel plate capacitive sensor, (b) prototype sensor, and (c) scanning electron microscope image of indium tin oxide structure.

Impedance spectroscopy measurements were taken using a Metrohm Autolab Instruments multi-channel potentiostat / galvanostat (PGSTAT12) using Metrohm's Frequency Response Analyzer software (v4.7.900). Data analysis of complex impedance data was performed using Autolab Nova (v2.0). Impedance measurements were taken using 50 measuring steps between 100kHz and 0.1Hz with an amplitude of 0.01V.

III. Results and Discussion

A. Determination of Dielectric Layer thickness

Typically, printed layer thickness is determined by the screen with the caveat that the printed ink will shrink. As UV inks do not contain evaporating solvents which are normally required for curing, no loss of mass occurs. To determine the layer thickness of dielectric polymer after printing, the polymer was printed onto soda lime glass (NSG, cleaned in ultrasonic bath with acetone, deionized H_2O and ethanol). These prints were then cured via UV (365nm) and cut with a diamond tipped blade for cross-sectional SEM.

The cross-sectional SEM (Fig.2a-b) shows that the printed dielectric layers had uniform layer thicknesses of 20.98μm and

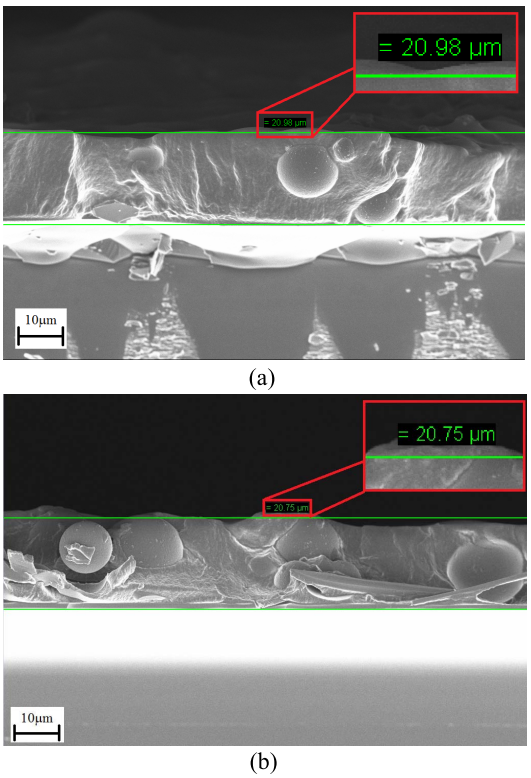


Fig. 2. Cross-sectional SEM of screen printed dielectric polymer: (a) sample 1 and (b) sample 2.

20.75μm, respectively. To ensure the accuracy, the average of these two values are used to calculate the dielectric constant of the ink.

B. Determination of Dielectric Constant

In parallel plate capacitors, the capacitance (C / units F) measured in Farads is determined using the permittivity of free space (ε_0 / units Fm^{-1}) multiplied by the relative permittivity of the dielectric material used (k) and the area of the electrode (A / units m^2). Capacitance is then calculated by dividing the permittivity and area by the distance of separation (d / units m) between the two electrodes

$$C = \frac{k\varepsilon_0 A}{d} \tag{1}$$

This equation is rearranged to find the dielectric constant (k):

$$k = \frac{Cd}{\varepsilon_0 A} \tag{2}$$

For sensors using 10Ω/sq indium tin oxide films, distance of separation was an average of 20.87μm, average capacitance was 522pF and area of the coated electrodes was 7.5cm^2. This gives an average dielectric constant of 1.63 for the polymer. Typically, standard polymers have a dielectric constant between 1.5 and 3 [19].

C. Transmittance of the Sensors

Humidity sensors fabricated using the thickest ITO films (10Ω/sq) were found to have a minimum transmittance of 75% in the visible spectrum (Fig.3a). Sensors fabricated using the thinnest ITO films (50Ω/sq) had a minimum transmittance of 82% in the visible spectrum (Fig.3b).

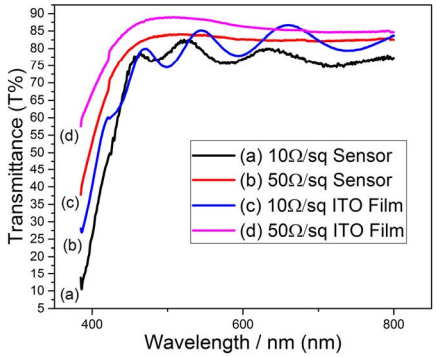


Fig. 3. UV/VIS transmission data for humidity sensors at 10Ω/sq and 50Ω/sq and pure indium tin oxide sputter coated films at 10Ω/sq and 50Ω/sq.

Due to the UV absorbing nature of the dielectric polymer ink, UV/VIS spectrophotometry measurements contained noise below 385nm. There is a 7% difference in transmittance between the thickest and thinnest films used for sensing. For comparison, the literature value given for the Melinex O PET film used is 89.2% transmittance [20].

D. Sensing Characteristics

Capacitance versus relative humidity was measured at three different frequencies, 100Hz, 1kHz and 100kHz. (Fig.4a-c). Capacitance increases with an increase in relative humidity for all frequencies. Measurements at 100Hz show a 152% increase in capacitance between 5 - 95% relative humidity. At 1kHz a more linear 116% increase in shown and for measurements at 100kHz, the increase for the full range was 105%. For all sensors, capacitance versus relative humidity measurements were all linear up to 75% relative humidity. These large changes in capacitance suggest the interactions with the water have a larger effect on either the dielectric permittivity of the polymer or conductivity of the electrode between 75 – 100% RH.

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This shows that if measuring humidity above the cutoff frequency of the capacitor, a different trend should be expected. As can be seen in Fig. 5, three sensors were measured simultaneously with the general trend that the lower the sheet resistance, the higher the capacitance. Capacitors generally have a wide tolerance and the sensors are no exception. Capacitance measurements between 10Ω/sq and 20Ω/sq often overlapped in terms of values. However, 10Ω/sq sensors gave larger changes in capacitance compared to 20Ω/sq sensors of a similar base value. This suggests a thicker and more conductive ITO film will give more sensitive readings. For all

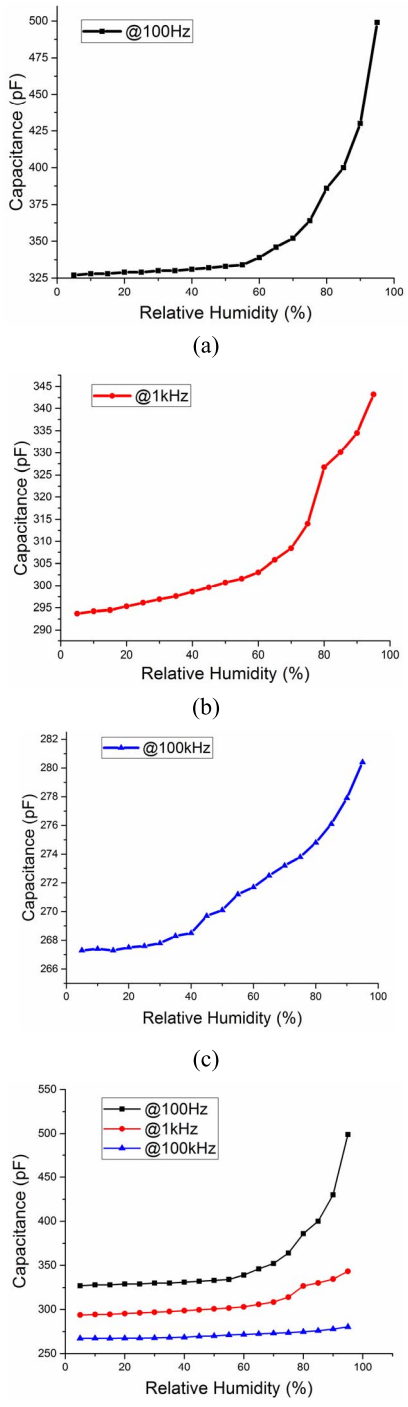


Fig. 4. Humidity measurements for 10Ω/sq sensors, Capacitance VS Relative Humidity for humidity sensor @ (a) 100Hz, (b) 1kHz, (c) 100kHz, and (d) data overlay of (a – c).

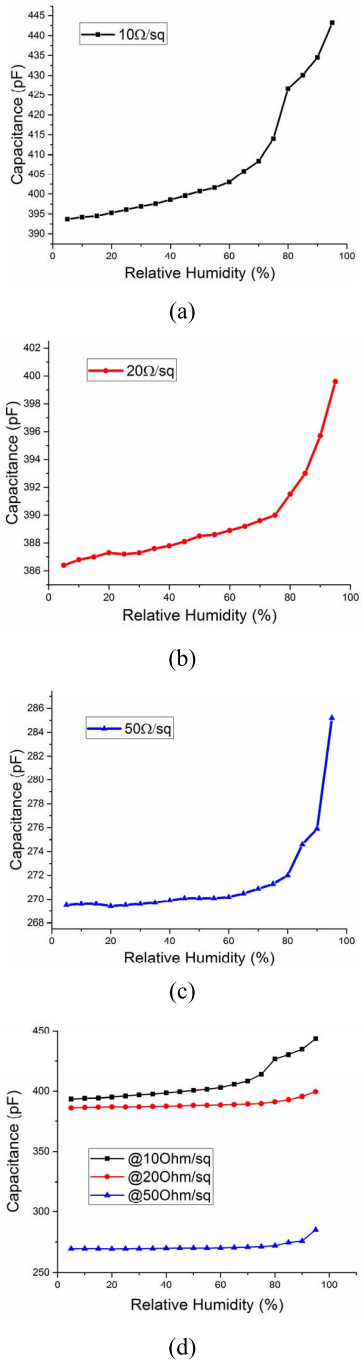


Fig. 5. Capacitance VS Humidity of the sensor at varying sheet resistances, (a) 10Ω/sq sensor, (b) 20Ω/sq, (c) 50Ω/sq measured @ 1kHz, and (d) data overlay of (a – c).

film thicknesses, sensors give a linear response up to 75% relative humidity. The 50Ω/sq sensors had minimal 0.1 – 0.2pF changes until a relative humidity of 20% had been achieved. The lower conductivity of the film affects the overall capacitance creating a less sensitive sensor.

To measure dynamic response and hysteresis effects, a sweep was performed between 60% and 80% relative humidity. The sweep would stabilize at 60% RH, hold for 30 seconds, move up to 80% RH, hold for another 30 seconds and then repeat the cycle. As can be seen in Fig. 6, capacitance of the sensor vs time is plotted against relative humidity vs time. The 10Ω/sq ITO sensor shows an average hysteresis effect of 0.8pF per cycle. Response times were measured as the time for the capacitance to peak after the humidity level reaches 80%. The 10Ω/sq ITO sensor shows an average response time of 31.5 seconds and average recovery time of 31 seconds as seen in Figure 6.

The hysteresis in the sensors can be explained using both indium doped tin oxide and the dielectric polymer.

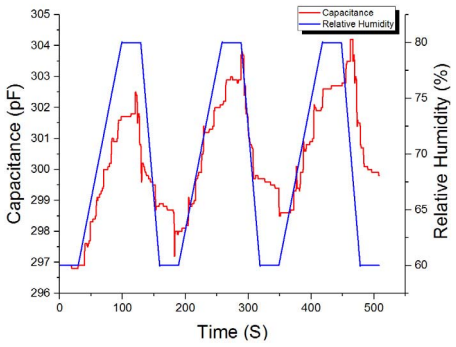


Fig. 6. Capacitance VS Time against Relative Humidity VS Time for a range of 60% – 80% relative humidity.

Doped oxides contain oxygen vacancies in the crystal structure and contact with open air can lead to surface alteration via oxygen containing species. When taking capacitive measurements, the dielectric polymer introduces hysteresis to the capacitance. When a water molecule absorbs onto the surface of the polymer, it can either diffuse into the polymer or desorb. Hysteresis is affected by a failure of water molecules to desorb back into the atmosphere. Another potential contribution to the hysteresis is capillary condensation where water can absorb into the pores of materials and is not always reversible, trapping the water [21].

Stability measurements were taken over seven days for 10Ω/sq, 20Ω/sq and 50Ω/sq sensors and can be seen in Fig 7(a-c). At 20% relative humidity sensors were at their most stable with day to day changes in capacitance kept in a small range. At 60% relative humidity the average trend over the seven days was also linear with any changes in capacitance staying within a certain range. However, for sensors at 95% relative humidity, a general rising trend can be seen over the seven days with larger fluctuations in humidity. This suggests that the high levels of moisture create an ongoing hysteresis effect at this humidity and the changes in capacitance match the data seen in Fig 4(a-c).

As can be seen in Figures 4(d) and 5(d), after 80% relative humidity, a non-linear increase in capacitance is recorded. Combining this with the data from Figure 7(a-c) suggests that a point in relative humidity has been reached were water is more likely to be trapped into the dielectric layer, changing the dielectric constant with greater effect leading to a jump in capacitance. In figure 7(b) for the most conductive and highest capacitance sample a variation can be seen at the 4 – 5 days region. In figure 7(c) this can also be seen on all the samples, suggesting this is an artefact of increased humidity and sensitivity. In figures 4 and 5, capacitance changes become larger and the sensor becomes more susceptible to humidity changes at higher humidity's and higher ITO conductivities. Therefore, this variation is most likely due to a combination of higher sensor sensitivity and this sensitivity giving the hysteresis a more prominent effect on measurements. This data suggests the sensors are more stable within a 5 – 75% relative humidity range and that the most transparent, least conductive sensors are the most reliable.

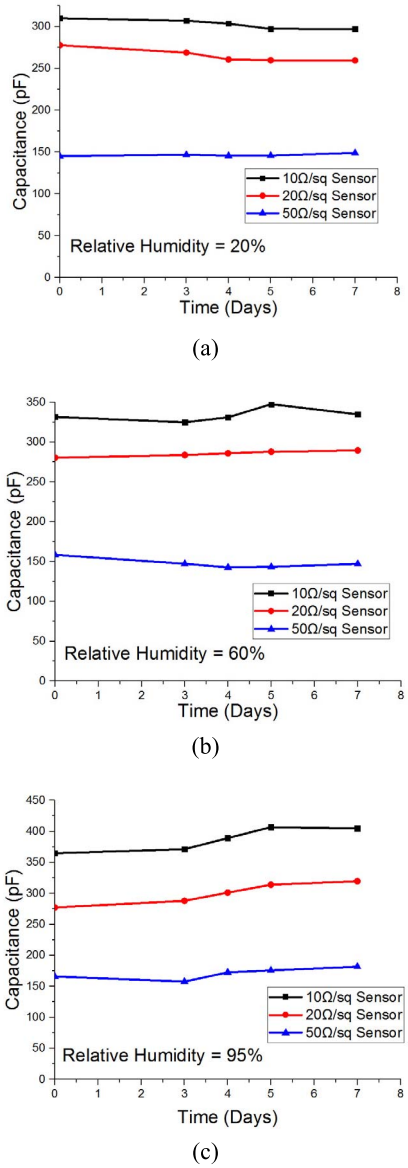


Fig. 7. Stability of sensors over 7 days taken at (a) 20% relative humidity, (b) 60% relative humidity and (c) 95% relative humidity.

E. Complex Impedance and Sensing Mechanism Analysis

Impedance measurements were taken at six different relative humidity levels, ranging from 5% to 95% relative humidity. The sensor response was measured from 0.1Hz to 100kHz. As seen in Fig 8(a) the impedance spectra are altered with increasing humidity. An increase in relative humidity is associated with an observed shrinking of the high frequency semi-circle on the Nyquist plot (Fig. 8(a)). This semi-circle corresponds to the bulk resistance of the dielectric layer and is seen to decease as the humidity level increases. The semicircle can be modelled seen in Fig 8(a-b), using a parallel Resistor (R) – Constant Phase Element (CPE) circuit (R-CPE circuit) and this typically represents adsorption levels in humidity sensors [22]–[23]. The CPE in this section of the circuit is a near ideal capacitor where if n = 1 is an ideal capacitor, modelling demonstrates a consistent $1 > n > 0.972$ for these sensors.

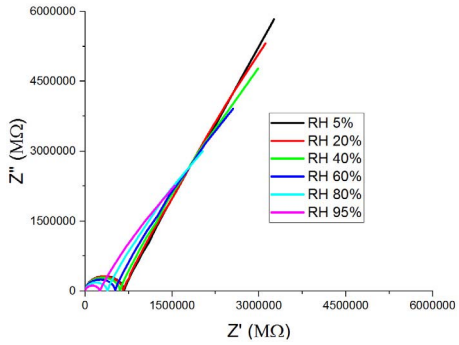


Fig. 8. Nyquist plots for 10Ω/sq sensors at 5%, 20%, 40%, 60%, 80% and 95% relative humidity.

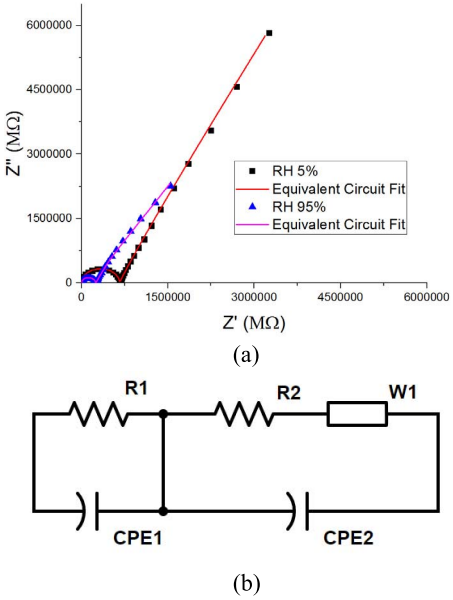


Fig. 9. Equivalent circuit diagrams with fitted plots: (a) 5% and 95% relative humidity equivalent circuit fit and (b) humidity sensor equivalent circuit diagram.

At lower frequencies, another feature is seen in the Nyquist plot, after the high frequency semi-circle. A straight line is seen (at low humidity) which seems to transform I to a second semi-circle at higher humidity. This can be attributed to a double layer capacitance forming on the exposed dielectric, this occurs from the diffusion of the water into the polymer, changing the dielectric permittivity and therefore capacitance of the sensor. The largest change in both the straight line and the semi-circle also is confirmed by the capacitance vs humidity measurements in Fig 4(a-c). Polymer dielectric based humidity sensors tend to have a greater response to relative humidity changes between 75% and 100% [1], [24].

Typically, capacitors in complex impedance spectroscopy rarely behave ideally, instead acting as a constant phase element (CPE) or non-ideal capacitor. In a Nyquist plot this generally represents a charge transfer reaction and in a humidity sensor can be attributed to both ionic diffusion and double layer capacitance. As seen in Fig 9(a-b) the change in dielectric permittivity of the dielectric can be modeled as a parallel CPE – RW circuit. In the CPE – RW section represents both ionic diffusion into the dielectric of the sensor and a double layer capacitance forming via a layer of adsorbed water molecules onto the water surface.

At a higher relative humidity, the straight-line is shown to curve which indicates it is the start of a larger semi-circle, rather than a straight line. This can also be modelled with Warburg impedance which can be associated with a charge transfer element in the sensor. This suggests that at higher humidity levels, ionic diffusion begins to occur. At a higher relative humidity, the ionic conductivity increases as more H+ and OH− ions complex with the surface, lowering the bulk resistance of the dielectric and increasing the capacitance [24], [25].

IV. Conclusions

The humidity sensing properties of sputter coated indium tin oxide were investigated by the design and fabrication of parallel plate capacitors. Sensors were transparent with a range of 75 – 82% transmittance in the visible region. The relative humidity range tested was between 5% and 95% RH. Response and recovery times were measured for both modes. In capacitive mode response times were on average 31.5 seconds with an average recovery time of 31 seconds. Complex impedance spectroscopy was performed on the sensors at different humidity levels to determine the sensing mechanism. The sensing mechanism was found to be a combination of ionic diffusion and ionic conductivity increasing in the dielectric layer, a double layer capacitance forming on the dielectric layer altering dielectric permittivity. These results provide insight into the use of transparent conducting oxides as electrodes for dielectric based capacitive transparent humidity sensors.

Acknowledgments

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