

Short Communication

## Development of a continuous impedance monitoring system: A study to monitor d-glucose concentration

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The present study describes the development of a continuous impedance monitoring system. Zinc oxide (ZnO) thin film electrode array was developed over a glass substrate. Two consecutively placed electrodes were connected with copper wires and the impedance properties of the electrode- D-glucose solutions were studied in-depth. Based on the information collected from the electrochemical impedance studies, an in-house impedance measurement system was fabricated and tested for its capability for continuous impedance monitoring.

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**Keywords:** Zinc oxide, thin film, electrodes, impedance analyzer, D-glucose

### 1. INTRODUCTION

The detection of the biological analytes plays a very important role in the improving of diagnosis and monitoring of health and diseases [1]. The development of the monitoring platforms plays an equally major role in achieving the target of personalized point-of-care technology. Hence, there is a need for the development of monitoring platforms at low costs. In this regard, there has been an increase on the monitoring of the analytes using impedance measuring devices. This may be accounted to the fast and reliable results obtained measuring the impedances of the analytes. The cost of the impedance measurement devices is relatively low. Apart from the above, the impedance measurement allows both qualitative and quantitative analyses of the analytes.

Zinc oxide (ZnO) is a versatile material with wide ranges of properties (e.g. electro-optical, piezoelectric, acousto-optical and luminescence characteristics) [2-3]. It has emerged as one of the

materials of choice for the fabrication of the electrode system for the impedance based measurement system. This may be attributed to the excellent electrical properties of ZnO. Apart from the excellent electrical properties, ZnO has been reported to be non-toxic, biocompatible, and can be easily prepared by various methods. Due to its above-mentioned properties, ZnO has found extensive research applications in designing different kinds of biosensors.

The measurement of the impedance of the ZnO thin film sensors for estimating the concentration of the analytes is a commonly used technique [4]. Unfortunately, most of the study uses costly impedance measurement devices which are bulky and costly [5]. This hinders the use of these devices for day-to-day measurement of the analyte for the various industrial processes. Also, most of the devices do not provide the facility to continuously record the impedances, which is required for the online monitoring of the analytes. Keeping the afore-mentioned facts in mind, in the current study, a low-cost two-terminal impedance monitoring system was developed. D-glucose solutions were taken as the representative analyte solutions [6]. The electrode-analyte solution system was thoroughly characterized using commercially available impedance analyzer. Taking information from the electrode-analyte solution analysis, the parameters of the developed impedance monitoring system was altered. Subsequently, the developed device was tested for its efficacy for continuous monitoring of two-terminal devices. A program in MATLAB® was developed for acquisition, monitoring and saving the acquired signals in a text.

## 2. METHODS

### 2.1. Deposition of ZnO Thin Films

Thin films of ZnO were deposited over a glass substrate (25 mm x 25 mm), maintained at 27 °C, using metallic zinc (99.995% purity) as the target material. The deposition was carried out by radio frequency (RF) sputter coating as per the reported literature with slight modifications [7-8]. RF magnetron core (Model: 12-3, Make: Excel-SPCH) sputtering system was used for ZnO coating. The distance between the target and the substrate was 10 cm. A vacuum of  $4 \times 10^{-3}$  mbar was applied to the reaction chamber. Subsequently a mixture of argon and oxygen was filled in the reaction chamber such that the total deposition pressure was 13.33 Pa. The target bias, the rf-power (100 W) and the rf-frequency (13.56 MHz) were kept constant during the deposition process. The deposition of the film was carried out for a period of 30 min.

### 2.2. Electrical characterization

50 µl of either distilled water (D1), 0.5 % D-glucose solution in distilled water (D2) or 2 % D-glucose solution in distilled water (D3) was put in between two electrodes and the electrical characteristics were measured [9]. The electrical characterization of the sensor was done using an impedance analyzer (3532-50 LCR HiTester, HIOKI, Japan) set within the frequency range of 100 Hz-1 MHz. The input voltage for the analysis was 1 V. The I-V characteristics of the electrode-analyte were studied using an in-house developed I-V measurement system. The study was conducted in the

current range of 0-50 mA. The frequency of the input current was varied in the order of 100 Hz, 200 Hz, 500 Hz, 1 kHz, 5 kHz and 10 kHz.

### 2.3. Designing of impedance monitoring system

An impedance monitoring system was developed as per the previous literature [10]. The schematics for the monitoring system have been provided in Figure 1. The system used IC 8038 as the signal generator. The signal generator generated a sinusoidal waveform having amplitude of 4 V<sub>pp</sub> and frequency of 10 kHz. The output of the signal generator was fed into a voltage buffer [11]. The output of the voltage buffer was fed into an OPAMP based inverting amplifier. The input resistance of the amplifier was 100 kΩ (R5; Figure 1). The electrode cell formed the feedback impedance. The voltage across the electrode cell was acquired in a computer using a data acquisition system (DAQ) of Arduino UNO microcontroller (Figure 2). A user-interface based impedance monitoring and display software was also developed. The software was capable of saving the information in a text file.

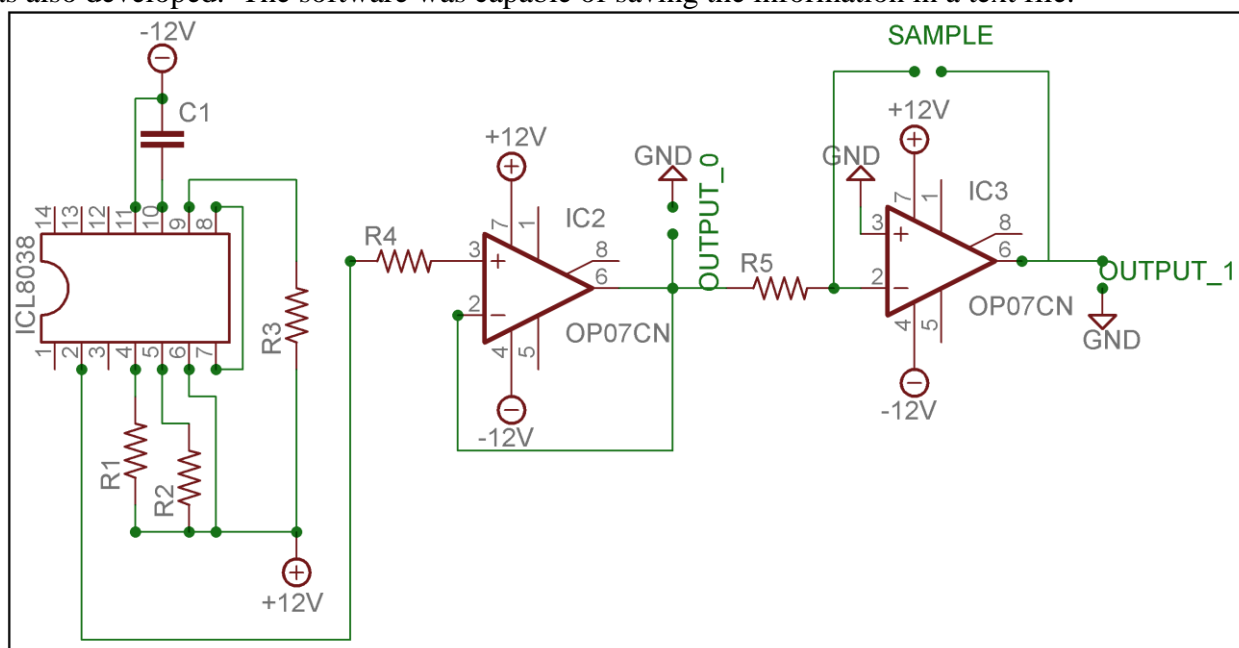


Figure 1. Schematic representation of Impedance Measuring Circuit

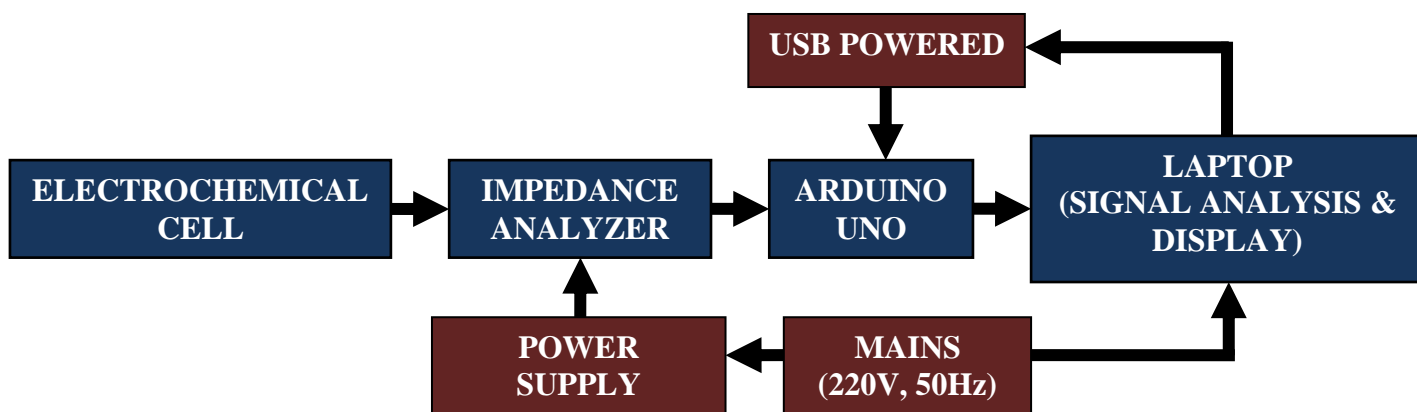


Figure 2. Block diagram of the developed setup

### 3. RESULTS AND DISCUSSION

#### 3.1. Development of ZnO thin films

A thin layer (95 nm) of ZnO was deposited over the glass substrate. An array mask was used for the development of the thin film electrode array. The electrodes were having a diameter of 1mm and were 1 mm apart from each other. The schematic of the electrode array has been shown in Figure 3. The electrodes appeared as equally spaced silver-colored circular elements throughout the glass substrate. There was no deposition of ZnO in between the electrodes, which remained as transparent.



**Figure 3.** Pictorial representation of the electrode array

#### 3.2. Electrical characterization

The analysis and model fitting using Z Simp Win software of the Nyquist plot ( $Z'$  vs.  $Z''$ ) has been shown in Figure 5a. Because of its simplicity and also as it can provide a clear and comprehensive picture of the whole system, electrical equivalent circuit is generally used for the analysis of the impedance spectrum [12]. Fitting of the experimental data are shown by the solid red lines. This fitting shows that there is general consensus between the experimental data and the proposed equivalent circuit shown in Figure 5e. The obtained error percentages in the fitting were within the limits for all the samples analyzed (Table 1). The semi-circular arc at higher frequencies as well as the spikes in the lower frequencies was present in almost all the test cases. The higher frequency region semicircle can be associated with the bulk properties of the electrode-analyte electrochemical system arises from the parallel combination of constant phase element (CPE1) and the bulk resistance ( $R_b$ ) [13]. The spikes found in the the low frequency region may be explained by the polarization occurring at the electrode and analyte interface and is represented by the series constant phase element (CPE2) [14]. The bulk resistance of the system ( $R_b$ ) can be easily deduced from the information provided by the real axis ( $Z'$ ) intercept of the semicircle in the lower frequency regions [15]. It was found that with the increase in the D-glucose concentration the bulk resistance decreases and was in the order of  $D1 > D2 > D3$ . The flattening of the semicircle and the tilting of the spike can be explained by the introduction of a constant phase element (CPE) in the equivalent circuit [16]. The CPE that replaces the capacitors in the circuit can be considered as a leaky capacitor, which has intermediate characteristics between a resistor and a capacitor.

**Table 1.** Parameters obtained from the fitting of impedance data

Samples	R1 (Ohms X 10 <sup>5</sup> )	CPE1 (S-sec <sup>n</sup> X 10 <sup>-12</sup> )	CPE2 (S-sec <sup>n</sup> X 10 <sup>-7</sup> )
D1	5.425	2.29	4.44
D2	5.069	2.278	1.365
D3	3.617	1.975	1.338

The tangential loss ( $\tan \delta$ ) profiles of the electrode- analyte have been shown in Figure 5b. The results showed that there was a shift in the  $\tan \delta$  peak towards higher frequency as the concentration of the D-glucose was increased. The peak value of the water and 0.5% D-glucose solution was obtained within the frequency range of 2 kHz to 4 kHz while the 2 % D-glucose solution was showed a peak at 10 kHz. Also, as the concentration of D-glucose was increased to 2 % there was a corresponding decrease in the amplitude of the  $\tan \delta$  profile. The combined effect of shifting of the peak in the higher frequency region along with a decrease in the peak amplitude suggested that the stability of the electrochemical cell-analyte was improved with the increase in the concentration of the D-glucose.

The conductivity of the electrode-analyte system has been shown in Figure 5c. The plots exhibited near frequency independent behavior under the experimental variables. The conductivity corresponding to the frequency independent plateau is regarded as d.c. conductivity ( $\sigma_0$ ). The a.c. conductivity spectra obeys Jonscher power law in upper frequency region of the measurement and is given by the equation [17]:

$$\Sigma_{ac} = \sigma_0 + A\omega^s$$

where,  $\sigma_{ac}$  is the ac conductivity;  $\sigma_0$  is the d.c. conductivity; A is a pre-exponential constant;  $\omega = 2\pi f$  is the angular frequency; and s is the power law exponent, where  $0 < s < 1$  [14].

The conductivity of the electrode-analyte system increased with the increase in the concentration of D-glucose. It was in the order D1 < D2 < D3. The d.c. conductivity of the systems was calculated using the formula:

$$\sigma_0 = (1/R_b) * (l/A)$$

where, A is the area of the sample and l is the thickness.

The d.c. conductivity of the systems was in the same order as the a.c. conductivity (S1 < S2 < S3) (Table 2).

**Table 2.** DC conductivity of the samples

Samples	DC conductivity( $\sigma_0$ ) (x 10 <sup>-5</sup> ) (Scm <sup>-1</sup> )
D1	4.01771
D2	4.5171
D3	5.69604

Impedance profile (log impedance vs. log frequency) of the electrode-analyte system is shown in Figure 5d. It helps in understanding the capacitive effects of the electrochemical systems. The impedance setup is shown in Figure 4.

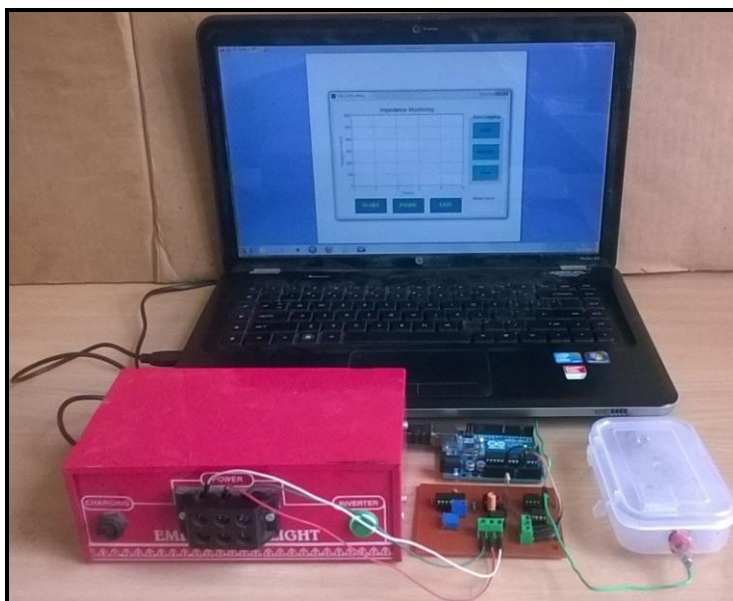


Figure 4. Pictorial representation of the in-house developed impedance monitoring system,

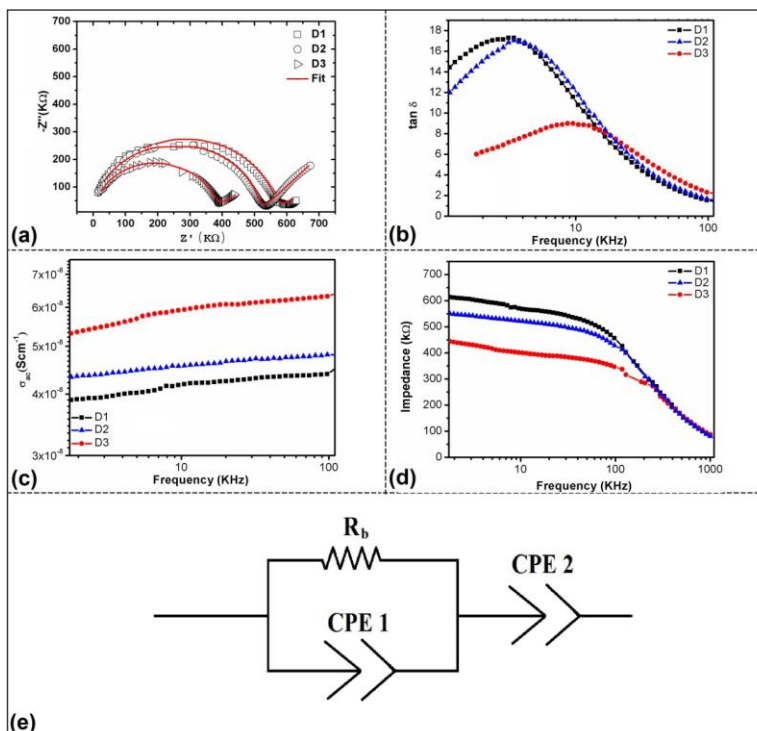
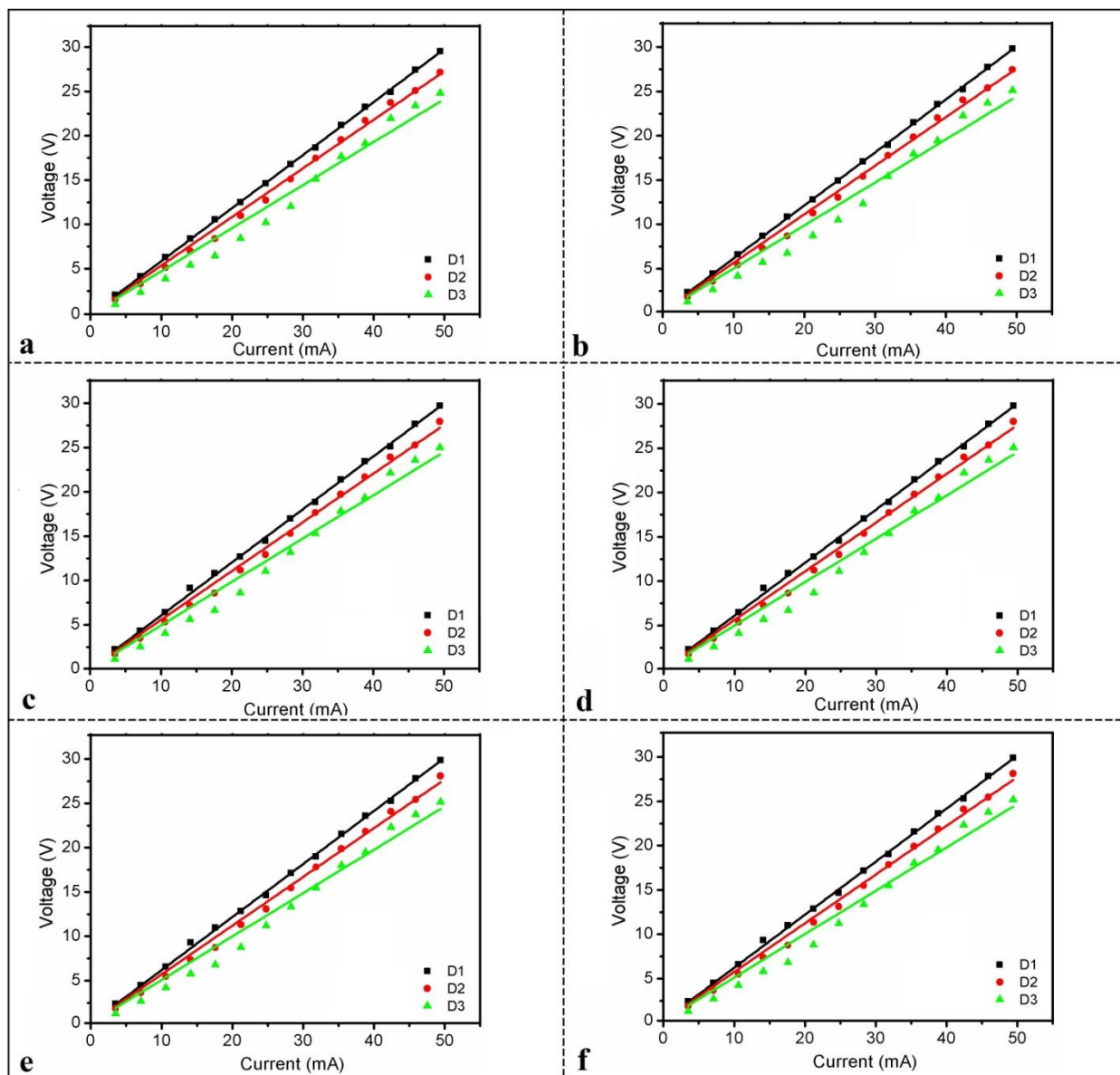


Figure 5. Electrochemical analysis and model fitting (a) Nyquist Plot, (b) frequency dependent tan delta plot (c) a.c. conductivity spectra, (d) frequency dependent impedance spectra, and (e) equivalent circuit diagram.

The impedance profile of the electrode-analyte systems showed mainly two regions which correspond to the presence of two types of elements in the equivalent circuit (Figure 5e) [18]. In the frequency range of 1 kHz to 100 kHz, a plateau was obtained with negligible changes in the impedance [19]. In the high frequency range (> 100 kHz), a steep decrease in the impedance was observed.

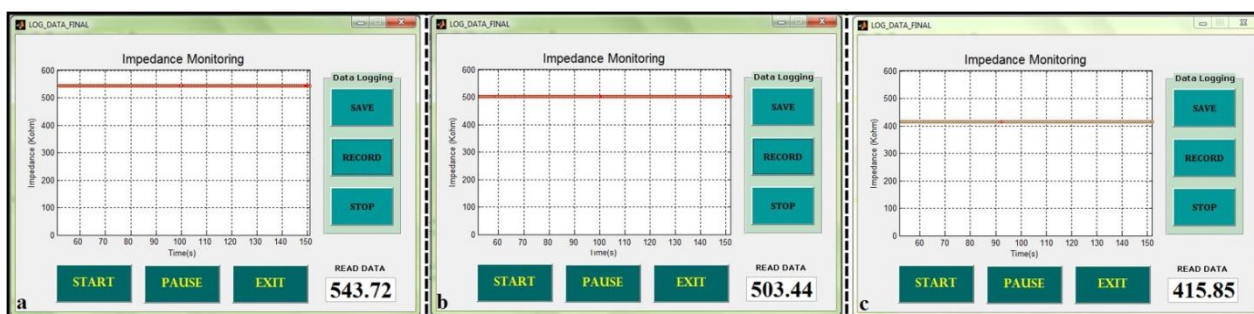
The I-V characteristics of the electrode-analyte system were measured using in-house developed I-V measurement device (Figure 6). The I-V curves were found to be linearly increasing with the increase in the current. The changes in the impedance were found to be negligible within the experimental frequency range. Similar results were also obtained from the impedance profile, which showed a plateau phase within the frequency range of 1 kHz – 100 kHz.



**Figure 6.** I-V Characteristics measured using in-house measurement device at different frequencies (a) 100 Hz (b) 200 Hz (c) 500 Hz (d) 1 kHz (e) 5 kHz and (f) 10 kHz

### 3.3. Impedance monitoring system

The electrochemical analysis of the electrode-analyte solutions suggested that the impedances of the analytes examined had sufficiently different impedances at 10 kHz frequency to be measured properly. Taking a note on this, the impedance monitoring system was developed which measured the modulus of impedance at 10 kHz frequency (Figure 7) [20]. The system injected a current of 14  $\mu\text{A}$  ( $I_{\text{rms}}$ ) into the electrochemical cell and the corresponding voltage drop across the electrochemical cell was acquired in a laptop using an Arduino microcontroller. The  $V_{\text{rms}}$  of the voltage drop across the electrochemical cell was calculated and the corresponding impedance was plotted as a function of time for a period of 5 min. The results indicated that impedance values recorded was stable for 5 min without much disturbance (Figure 7). The impedance values were in accordance with the impedances obtained from the electrical characterization of the electrochemical cell. The results indicated that the changes in the D-glucose concentration can be efficiently monitored using the ZnO thin film electrodes and the in-house developed electrochemical cell impedance analyzer.



**Figure 7.** User display of the Impedance Monitoring System Software, Impedance Monitoring for (a) D1, (b) D2, and (c) D3

## 4. CONCLUSION

The current study discusses about the successful development of a low cost impedance monitoring device. To test the efficacy of the system ZnO thin film based microelectrodes were prepared. D-glucose solutions (in different concentrations) were used as the representative model analyte. The electrode-analyte combination was thoroughly characterized using a commercially available impedance analyzer and in-house developed I-V measurement device. The results suggested that the polarization of the electrode analyte interface was minimal at 10 kHz frequency. Hence, the in-house developed impedance monitoring device (with data logging software) was made to function at 10 kHz frequency. The preliminary results suggest that the developed device may have a great potential in impedance monitoring of biological analytes and also may be used for designing impedance based biosensors.



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