IOSR Journal of Mechanical and Civil Engineering (IOSR-JMCE)
e-ISSN: 2278-1684,p-ISSN: 2320-334X, Volume 18, Issue 2 Ser. IV (Mar. – Apr. 2021), PP 14-20
www.iosrjournals.org

Computational and Experimental Study of Frequency Dependent Impedance Response of Epoxy-Tin Composite

Fazle Rabbi[1], Sadman Hassan Labib[1]
[1]*(Department of Mechanical & Production Engineering, Ahsanullah University of Science & Technology, Bangladesh)*

Abstract:
Frequency dependent impedance response can give insight to the charge transport mechanism of a composite material. Charge transport mechanism is influenced by individual constituent properties and interphase between the constituents. Computational study can be used to predict these behaviors. To conduct computational study of the charge transport on the composite material, these frequency dependent properties were taken into account. A computational FEA model was developed and verified by using experimental data. Composite of study was epoxy-tin material. Computational calculation was conducted by using frequency dependent single phase material properties of the domain representing insulating epoxy and conductive tin.
Key Word: *Maxwell's equation, FEA, Dielectric Spectroscopy, Impedance*

Date of Submission: 15-04-2021 Date of Acceptance: 29-04-2021

I. Introduction

Composite materials consist of multiple unique materials as individual phases and their accompanying interfaces. Properties absent in these different material phases arise from the interaction between these distinct phases and their interfaces. The global material properties, functional behavior and system efficiency of these composite materials might often depend on the specific morphology and geometric scale at the micro or nano level. Advances in the development of these composite materials have influenced the field of conversion and storage of energy, chemical and fuel processing system based on ceramic membrane systems. Ion conducting ceramic membranes has been used in a wide range of energy conversion devices. Of these systems, oxygen separation and permeation membranes [1], natural gas processing reactors by partial oxidation [2], solid oxide fuel cells [3] and high temperature electrolysis cells [4] has now moved from research and development to commercial applications.

Broadband dielectric spectroscopy (BbDS) is a useful technique that can be used to study different charge transport processes at the local level of these composite functional materials. BbDS is the interaction between electromagnetic waves and matter in the frequency range from 10^{-6} Hz to a frequency of 10^{12} Hz. Impedance behavior within this frequency zone comprises of information of the system about charge transport, molecular and collective dipolar fluctuation and polarization effects occurring at the vicinity of the boundary between each phase. And these characteristics are represented in the form of different dielectric properties of the material [5]. To analyze the internal charge transport mechanisms in these materials, measurement of the dielectric response over a broad frequency and temperature limit can be made. At higher frequency, the charge carriers are driven over distances corresponding to atomic scales by exterior electric field while in the lower frequency region the charge carriers propagate from one side of the sample to the other on some percolation path. Thus, a length scale is involved with decreasing frequency changing from microscopic to macroscopic scale. These responses are represented in the complex dielectric behavior and are found to be similar for a wide range of material, such as electron conducting conjugated polymers [6], ionic glasses [7], ion conducting polymers [8], and electron conducting carbon black composites [9]. For all of these materials, conductivity and permittivity change occurs with changing frequency. The different complex dielectric functions representing these materials are equivalent but they emphasize different aspects of the underlying mechanisms.

Computational BbDS study can facilitate comprehensive study of the mechanisms and their dependence on not only material properties but also material structures and their interaction with each other. A computational model is developed based on quasi-static Maxwell's equation [10], capable of measuring impedance response of a given domain over a wide frequency range. But dielectric properties of materials i.e. conductivity, permittivity, are also frequency and temperature dependent. And for calculation, the model requires material properties. So, a constant value of conductivity or permittivity for the whole frequency and temperature range will not yield the actual impedance response for the given domain. So, frequency and temperature dependent material properties of individual single phase material properties are needed to be measured and should be used.

NOVOCONTROL, a high precision impedance measurement device was used to measure the material properties. For model verification an ideal composite of regular geometric shape was prepared. And both computational and experimental data comparison were made for the given structure

II. Method of Approach

First step of this study was to measure the frequency dependent electrical properties i.e., electrical conductivity and permittivity of the individual constituents. For this study our material of choice was a composite of epoxy-tin. To get frequency dependent dielectric properties of individual materials we prepared a solid cubic sample of epoxy and tin and measured their conductivity and permittivity. A high precision (Phase accuracy of 0.002˚ or tanδ accuracy of $3x10^{-5}$) impedance measurement device, NOVOCONTROL was used to measure the properties. It has a frequency range from 3μHz-20MHz. But For our experiment we choose a range from 3Hz-1MHz. Figure 1 and Figure 2 shows conductivity and permittivity is changing with increasing frequency. For tin its conductivity and permittivity was constant throughout the frequency range at $9.17x10^{6}$ S/m and 1.

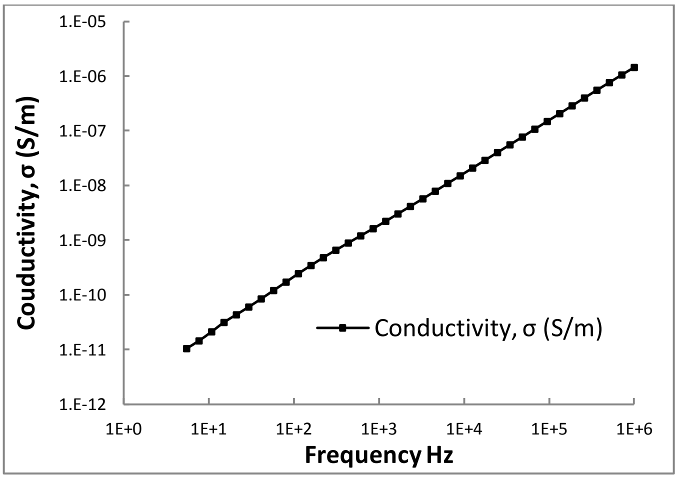

Fig 1. Single phase epoxy Conductivity increasing with increasing frequency, obtained using NOVOCONTROL

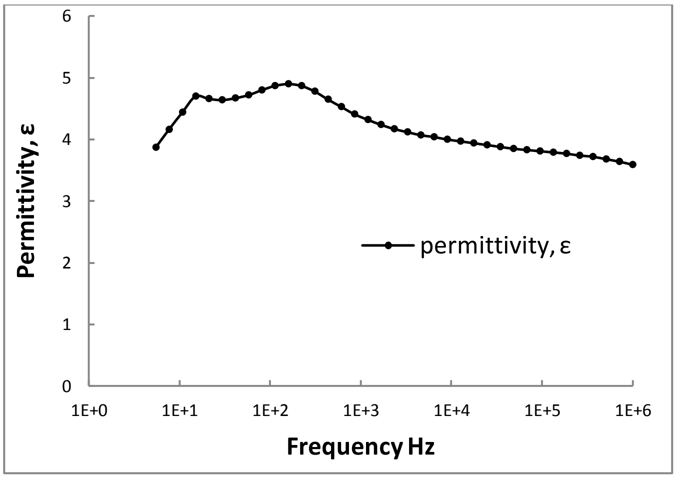

Fig 2. Single phase epoxy permittivity changing with frequency, obtained using NOVOCONTROL

For our experiment we made a composite sample of high and low conductive material tin and epoxy (Figure 1). Dielectric permittivity of epoxy was higher than tin. The tin inclusion in the sample was prepared by using a die. Then the surface of the tin inclusion was machined to obtain smooth surface. The Lenticular shape can be described as the intersecting region between two cylinders. Then the tin inclusion was submerged in a pool of liquid epoxy inside a cubic die. After the epoxy solidified the combination structure was machined to obtain the shape shown in Figure 3.

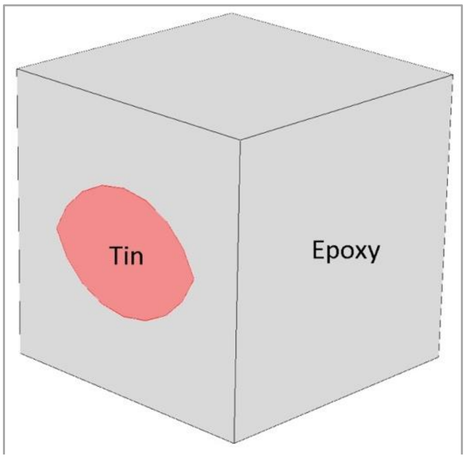

Fig 3. **Epoxy-tin composite sample**

To apply potential to the sample for NOVOCONTROL measurement, a spring-loaded fixture was used to maintain constant pressure. Copper plates were used as electrode for the samples shown in Figure 4.

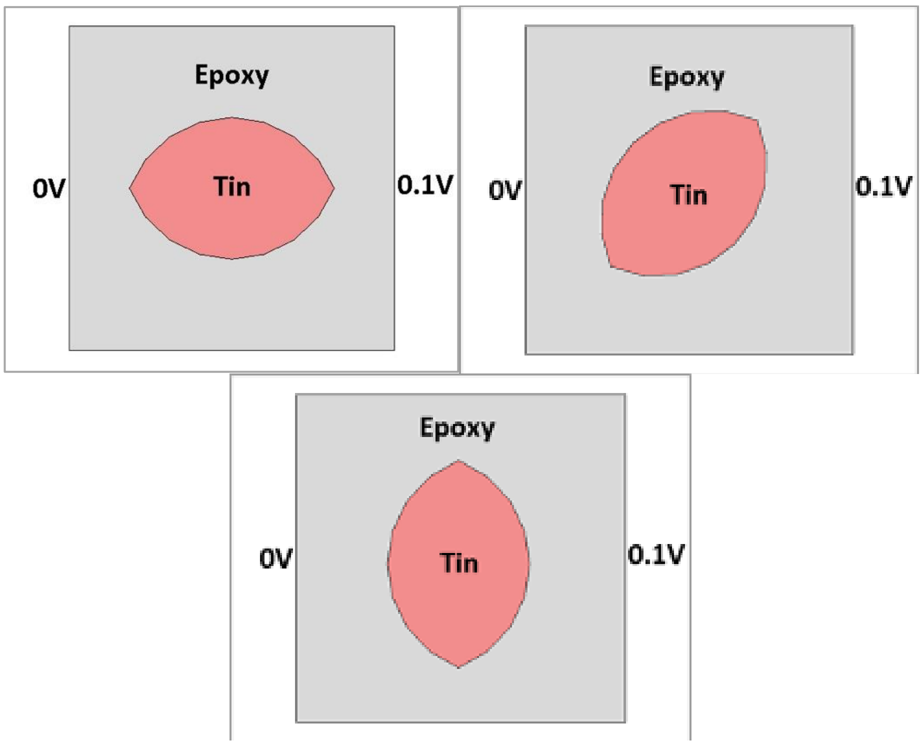

Fig 4. Boundary condition for measurement and

Three different types of sample were made by arranging the orientation of the lenticular tin inclusion. The orientations of tin inclusions were 0˚, 45˚ and 90˚ with respect to the applied field direction. For the measurement 0.1 volt was applied shown in figure 4.

We developed a model to computationally calculate impedance response of a volume for a broadband frequency spectrum. For experiment these samples were created using conventional machining techniques. Our first goal was to create some regular structured material and measure their impedance response and verify our computational model with the data. For this we used epoxy as the matrix material with low conductivity and tin with higher conductivity.

For the finite element analysis to generate a model capable of calculating impedance response for a given microstructure domain with different materials we used modelled Maxwell's equation. Similar models have been used to analyze dielectric response of water absorbance in organic coatings [11]. This application is obtained when inductive effects are negligible.

The time-harmonic, quasi-static, Maxwell's equations in vector form for the electric potentials are:

$$-\nabla.\boldsymbol{d}\big((\boldsymbol{\sigma} + \boldsymbol{j\omega\varepsilon_0})\nabla\boldsymbol{V} - (\mathbf{J}^{\mathrm{c}} + \boldsymbol{j\omega}\mathbf{P})\big) = \boldsymbol{dQ_j} \text{ -------------------------(1)}$$

ω	Angular frequency (rad/s)
Q_j	Current (A/m^3)
$\mathbf{J}_s$	Current density (surface) (A/m)
$\mathbf{J}$	Current density (volume) (A/m^2)
V	Electric potential (V)
σ	Electric conductivity (S/m)
ν	Frequency (Hz)
Z, η	Impedance (Ω)
ε	Permittivity (F/m)
$\mathbf{P}$	Polarization (C/m^2)

d Material thickness

The constitutive equations for linear, isotropic materials have been applied for the electric displacement vector, D:

$$\vec{\boldsymbol{D}} = \boldsymbol{\varepsilon_o\varepsilon_r}.\vec{\boldsymbol{E}} \text{ -------------------------(2)}$$

$\mathbf{D}$	Electric displacement (C/m^2)
$\mathbf{E}$	Electric field (V/m)
ε_r	Relative permittivity
ε_0	Relative permittivity of vaccum

Due to the absence of current source and external current density, equation (1) becomes

$$-\nabla.\boldsymbol{d}\big((\boldsymbol{\sigma} + \boldsymbol{j\omega\varepsilon_0})\nabla\boldsymbol{V}\big) = \mathbf{0} \text{ -------------------------(3)}$$

Equation (1-3) describes the Impedance response. The measured frequency range was from 3Hz-1x10^6 Hz. Applied voltage was 0.1 Volt.

The two input boundary conditions of 0V and 0.1 volt used in the model are shown in figure 4. All the other boundaries of the domain were insulated. Frequency dependent single phase material properties, i.e., conductivity and permittivity obtained from the experimental results were applied in the model and the tin properties were kept constant for all the calculation.

III. Results and Discussions

Experimental data in Figure 5 for three different orientations of lenticular sample is shown. Global impedance of the samples measured between the two-boundary shown in Figure 4 shows no variation for the three different orientation of the lenticular inclusion indicating that impedance is independent of the internal phase orientation. The gradual decrease of impedance of the sample with increasing frequency also in agreement with low conductive material behavior [5].

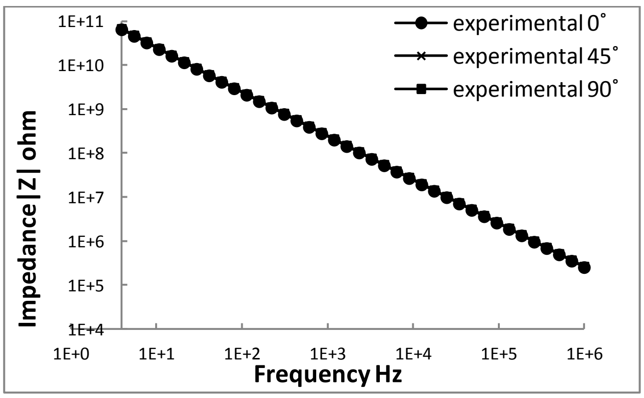

Fig 5. Experimental impedance response of the ideal sample for 0˚, 45˚ and 90˚ lenticular tin inclusion orientation

Figure 6 shows computational model calculated impedance of the samples with three orientation. Where some small variations are seen between impedance for three different orientations but follow the same pattern as the experimental data.

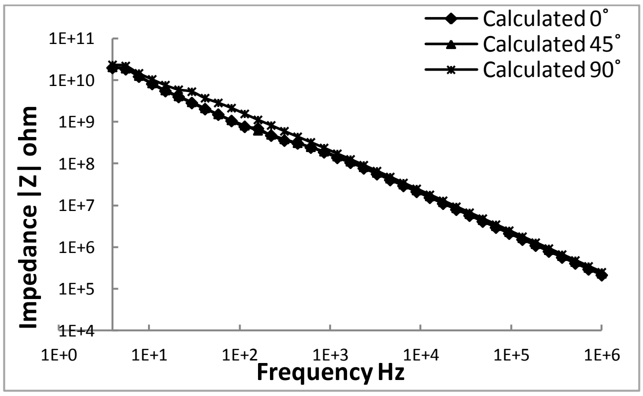

Fig 6. Computational impedance response of the ideal sample for 0˚, 45˚ and 90˚ lenticular tin inclusion orientation

Figure 7 shows voltage distribution inside the sample where a gradual decrease of voltage from high potential side near the input to the grounded surface. In the middle, a non-uniform potential distribution is visible, caused by the presence of high conductive tin inside the epoxy matrix.

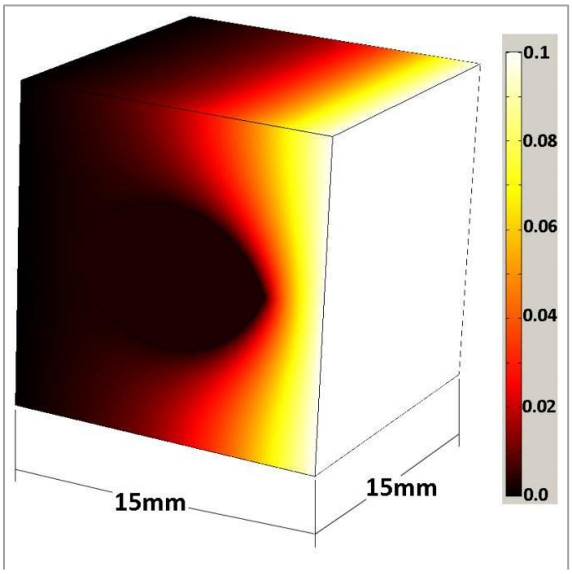

Fig 7. Voltage distribution (0.1V input voltage) in epoxy-tin composite material

Figure 8 shows comparison of experimental and computational value of impedance for three different orientations. For all the orientations, impedance gradually decrease with increasing frequency. For each case, at higher frequency the data look to be in good agreement and at lower frequency the values have a very small difference with experimental values have slightly higher value than the calculated values. This may be due to the fact that for the experiments the impedance might include the impedance originating from the contact between the sample and electrode. Although the surface as well as the electrode was polished to attain a good surface connectivity there still might be where the electrode was not touching properly. But for the computation we directly applied direct potential to the surface without any electrode. As a result, polarization occurring from electrode contact was ignored during computational calculation. Hence lower impedance was measured for calculation. To avoid this in future a metallic conductive paste might be added on the input surface of the sample. To simulate contribution from the electrode to the global impedance behavior, an electrode domain with contact surface impedance can also be added to the model.

So, from the compared data it can be said that if a static material property value free of influence of frequency variation were to be applied for the epoxy material domain to calculate computationally the impedance response of the sample, value for the higher frequency would not have matched the experimentally measured data. From the impedance response, other global material properties i.e., permittivity, conductivity of the composite material can be calculated using Maxwell's equation [12]. This model is also capable of calculating voltage distribution, polarization, charge distribution and charge flow inside the material domain. Thus, this model can be utilized to study the charge transport behavior inside a composite material and the factors that impact these phenomena. This model can also be utilized to study interfacial charge transport between two phases in a composite material. For this study, we have generated the computational domain using CAD software. This model is capable of measuring impedance of material domain obtained from X-ray tomography representing real physical system micro structure [13]. In that case, individual constituent material property of the physical system is needed to be measured to apply to the model domain phases.

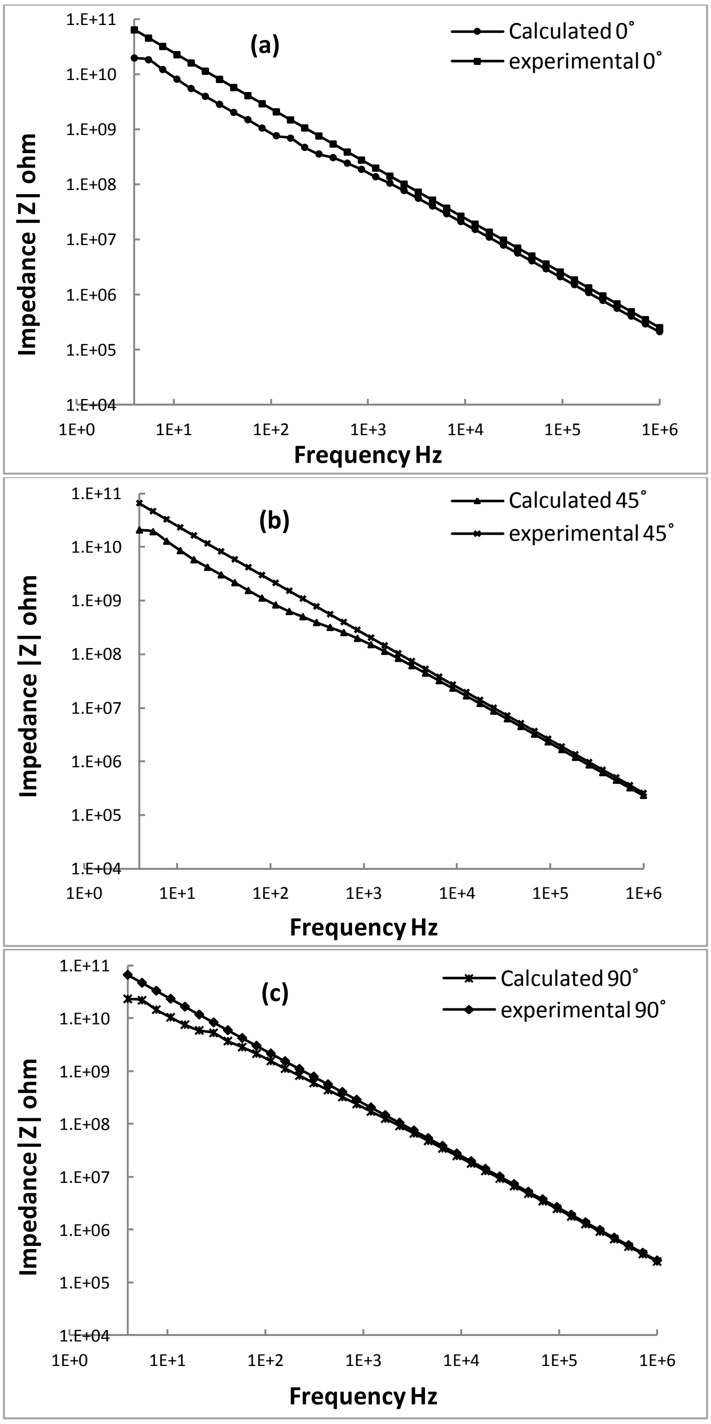

Fig 8. Measured impedance response vs computed impedance response of the epoxy-tin composite material (a) 0˚ orientation of the lenticular tin inclusion with the applied field, (b) 45˚ orientation of the lenticular tin inclusion with the applied field and (c) 90˚ orientation of the lenticular tin inclusion with the applied field.

IV. Conclusion

A computational model to measure impedance response of a material over a wide range of frequency was constructed using Finite Element Analysis Method. This model is capable of calculating impedance response for a given material, while taking into account the dependence of material dielectric properties i.e., conductivity and permittivity on changing frequency. These frequency dependence material properties of a single-phase material were obtained using NOVOCONTROL. NOVOCONTROL can also be used to measure dielectric property variation for changing temperature. For model verification a composite sample of epoxy-tin with regular geometric shape was used. First the impedance response of the composite was experimentally measured by using NOVOCONTROL. Then an identical volume domain was created for the finite element analysis using standard modeling tools available. Then each domain of epoxy and tin were assigned with material properties that were frequency dependent and obtained by using NOVOCONTROL. Then the FEA model solve for each frequency points taking into account varying dielectric properties corresponding to that certain frequency value. And three different orientation of tin inclusion relative to applied field was used. And for each case, calculated and experimentally measured data showed good agreement. In future, this model can be utilized to study charge transport behavior between phases and interfaces in composite materials.

References

[1]. Pal, U. B., Woolley, D. E. & Kenney, G. B. Emerging SOM technology for the green synthesis of metals from oxides. *Jom* **53**, 32–35 (2001).

[2]. Takamura, H., Ogawa, M., Suehiro, K., Takahashi, H. & Okada, M. Fabrication and characteristics of planar-type methane reformer using ceria-based oxygen permeable membrane. *Solid State Ionics* **179**, 1354–1359 (2008).

[3]. O'hayre, R., Cha, S.-W., Colella, W. & Prinz, F. B. *Fuel cell fundamentals*. (John Wiley & Sons, 2016).

[4]. Yang, C., Coffin, A. & Chen, F. High temperature solid oxide electrolysis cell employing porous structured (La0.75Sr0.25)0.95MnO3 with enhanced oxygen electrode performance. *Int. J. Hydrogen Energy* **35**, 3221–3226 (2010).

[5]. Kremer, F. & Schönhals, A. *Broadband dielectric spectroscopy*. (Springer Science & Business Media, 2002).

[6]. EPSTEIN, A. J. AC conductivity of polyacetylene: distinguishing mechanism of charge transport. *Handb. Conduct. Polym*. 1041–1097 (1986).

[7]. Angell, C. A. Fast ion motion in glassy and amorphous materials. *Solid State Ionics* **9–10**, 3–16 (1983).

[8]. Shklovskii, B. I. & Efros, A. L. *Electronic properties of doped semiconductors*. vol. 45 (Springer Science & Business Media, 2013).

[9]. Pelster, R. & Simon, U. Nanodispersions of conducting particles: Preparation, microstructure and dielectric properties. *Colloid Polym. Sci.* **277**, 2–14 (1999).

[10]. Fleisch, D. *A student's guide to Maxwell's equations*. (Cambridge University Press, 2008).

[11]. Stafford, O. A., Hinderliter, B. R. & Croll, S. G. Electrochemical impedance spectroscopy response of water uptake in organic coatings by finite element methods. *Electrochim. Acta* **52**, 1339–1348 (2006).

[12]. Monk, P. *Finite element methods for Maxwell's equations*. (Oxford University Press, 2003).

[13]. Nelson, G. J. *et al.* Comparison of SOFC cathode microstructure quantified using X-ray nanotomography and focused ion beam-scanning electron microscopy. *Electrochem. commun*. **13**, 586–589 (2011).

Fazle Rabbi, et. al. "Computational and Experimental Study of Frequency Dependent Impedance Response of Epoxy-Tin Composite." *IOSR Journal of Mechanical and Civil Engineering (IOSR-JMCE)*, 18(2), 2021, pp. 14-20.