Impedance analysis of Sodium alginate : Graphene oxide composite

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Abstract. Sodium alginate (SA) and graphene oxide (GO) composite pellets are prepared by one step simple procedure. The composites are found to be thermally stable up to 220°C in nitrogen atmosphere. The composite was studied for impedance analysis with varying temperature ranging from room temperature to 170°C. Dielectric constant of the samples was measured using the impedance data using standard formulae. It was found that the dielectric constant of the composites is high at low frequencies which may be due to the contribution from all the dipoles responding to the applied signal. However, at high frequencies, may be due to non-participating dipoles, the dielectric constant of the composites is lower.

Keywords: Impedance, Pellets, Graphene oxide, Sodium alginate.

1. Introduction
Polymers have found their applications in various fields including consumer goods, electronics, medical industry, etc. Due to their vastly variable nature with possible additives, it has become easy for one to see their uses in many applications. Applications of polymers in the field of electronics are due to their conducting as well as non-conducting nature. Polymers are used as non-conductive covering for insulating applications and as conductors in the field of thin film conductors. However, their usage as dielectric materials for enhancing the capacitance of the capacitors is also an area where people have shown interest in [1]. Many dielectric materials used in capacitor manufacturing have two-phases, one being the host polymer and the other being an additive. The additive used with the host polymer plays an important role in varying the nature of the host polymer [2]. Many fillers have found to enhance the dielectric nature of the host polymer [2].

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Fabrication of new generation, low-cost composite dielectric materials are receiving increased attention in the recent times [3]. Sodium alginate (SA) is a natural polymer obtained from a particular variety of marine brown algae. SA is a nontoxic polysaccharide with a varying sequence of D-mannuronic acid and L-guluronic acid units attached to form a copolymer. SA is a versatile material which finds application as an additive for food, drug delivery medium to being a stabilizing agent for the synthesis of nanoparticles [4,5]. Graphene oxide (GO) is a monoatomic thin sheet with oxygen functional groups attached on to the basal planes and edges. GO is receiving great attention in the recent years due to its capacity to significantly influence the properties of other materials.

S Tsubaki et al. have reported the effect of acidic functional groups of SA on the dielectric behaviour [6]. S Bekin et al. studied the dielectric response of calcium ion crosslinked SA films [7].

In this study, we are reporting the preliminary results of impedance analysis of SA-GO composites. The impedance analysis is used as a tool to study the dielectric nature of the composites to be used for capacitor applications.

2. Experimental

2.1. Materials

Analytical grade sodium alginate was purchased from S.D. Fine-chem Limited. Graphene oxide (98% purity, 1-2 number of layers) is a gift sample supplied by United Nanotech Ltd., Bangalore, India. Both the chemicals are used as such without further purification.

2.2. Sample Preparation

SA and GO are ground together into fine powder with pestle and mortar in the following proportions. 5mg of GO is added into 1g of SA and named as SG-5 and SG-10 are prepared accordingly. Pure SA and GO also ground into fine powder. A known weight of each composition is made into a pellet using a pellet maker by applying a pressure of 25MPa. The pellets had a thickness of 1.8 mm and diameter of 12 mm each. Four samples are prepared.

2.3. Characterization

Mass change and thermal degradation pattern of the samples were analyzed using TA instruments (SDT Q600) thermal analyzer in nitrogen atmosphere with 100 ml/min flow rate. The samples were heated from 30°C to 600°C with a heating gradient of 10°C per minute. Impedance analysis of the composites was carried out using CH Instruments electrochemical workstation in the frequency range of 100Hz to 1MHz. The composites were sandwiched between two ion blocking silver electrodes and varying frequency AC signal with strength of 0.5V was used for the study. Impedance of the composites was measured with varying temperature for which a home-made variable temperature setup was used.

3. Results and discussion

3.1. Thermogravimetric analysis

Mass change and thermal degradation pattern of the samples were analyzed using TA instruments (SDT Q600) thermal analyzer in nitrogen atmosphere with 100 ml/min flow rate. The samples were heated from 30°C to 600°C with a heating gradient of 10°C per minute. Thermal stability and
degradation patterns recorded are shown in the figure 1. GO being thermally unstable starts losing mass much before 100°C due to the decomposition of oxygen containing functional groups getting liberated as CO and CO\textsubscript{2}. Mass loss to an extent of 33% has been observed between 100°C to 200°C. SA has exhibited two stages of thermal decomposition with a mass loss of 11% from 30°C – 220°C which corresponds to dehydration and another rapid degradation that occurred to an extent of 11% in the range of 220°C – 265°C due to the decomposition of SA polymeric structure. The SA-GO composites followed the degradation behavior of SA which indicates that the presence of GO did not play any significant role in altering the degradation pattern of the composites due to its lower concentration.

![Figure 1. TGA curves of SA, GO, SG-5 and SG-10.](image)

3.2. Impedance Measurements
Impedance measurements as a function of temperature was carried out for SA-GO composites using CH Instruments between the frequency 100Hz to 1MHz with signal strength of 0.5V. Cole-Cole plots obtained for different samples are given below in figure 2. It can be seen from figure 2 that the bulk resistance of the composites varies with temperature. At higher temperature composites show higher bulk resistance representing a conductor like behavior. This kind of a behavior may be due to the presence of conducting GO in the composites. We have also observed that the bulk resistance of the composite is higher than that of the individual hosts at room temperatures. Although there is a small concentration of GO in the composite it plays a major role in altering the composites’ resistance and hence is expected to play a major role in dielectrics of the material.
Impedance data was used to calculate the dielectric constant and loss of the composite with the formula:

\[ \varepsilon' = \frac{-Z''}{\omega C_0 \left( Z''^2 + Z'''^2 \right)} \quad \& \quad \varepsilon'' = \frac{Z'}{\omega C_0 \left( Z''^2 + Z'''^2 \right)} \]

As seen from figure 3, dielectric constant of the composites is high at low frequencies and low at higher frequencies. This may be due to the following reason: At higher frequencies of applied signal, response may be given by lesser number of dipole sites as their relaxation time compared to the time required for them to respond to the signal, however, at low frequencies, all the available dipole sites respond to the applied signal producing a higher dielectric constant in both the samples. Also, it can be observed that the dielectric constant of the composites does not vary much at higher temperatures as a function of frequencies. This may be due to the following reason: At higher temperatures, due to the thermal energy provided to the system, the orientation of dipoles may become random making the net dipole moment less thereby reducing the dielectric constant of the system. It can also be observed that the dielectric constant of the composites is high at low frequencies and lower temperature which may be also due to the reason discussed earlier.

4. Conclusions

Impedance analysis of SA-GO composites was carried out for two compositions. The composites are found to be stable up to 220°C. Thermal stability and degradation patterns of the composites are not found to be markedly different from that of SA. It was observed that the impedance of the composites was proportional to temperature representing metal-like behaviors which may be due to the presence...
of conducting GO in the samples. It was also observed that the dielectric constant was almost constant at higher frequencies which may be due to non-participating dipoles at higher frequencies of the applied signal. However, at low frequencies, dielectric constant was high for low temperatures and low for higher temperature.

5. References
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