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Investigation of frequency-dependent Dielectric properties for recycled local domestic hen eggshell powder, a prelude for eco-friendly medical application

Moustafa Hussein Moustafa

Biophysics Department, Medical Research Institute, Alexandria University, Egypt

Email: Moustafa-hm@alexu.edu.eg

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Abstract

Biomaterials have received considerable attention in many medical applications; at both diagnostic and therapeutic levels. Hen eggs are one of the most daily consumed biomaterials. Millions of hen eggshells (ES) are produced annually. The use of these eco-friendly waste resources is highly attractive in a positive impact worldwide. Morphological and physical properties of ES have made them a good candidate for medical uses. However, to date, there are no sufficient studies on this topic. The dielectric constant $\epsilon'(\omega)$ and loss $\epsilon''(\omega)$ are studied as a function of the temperature and frequency. Furthermore, the dielectric data is fitted and analyzed according to the Cole-Cole model.

Keywords: Dielectric properties, hen eggshell, eco-friendly

Introduction

Biomaterials have received considerable attention in many medical applications, leading to new developments in various medical fields at both the diagnostic and therapeutic levels. One of the most daily consumed biomaterials is hen eggs [1]. Millions of hen eggshells (ES) are produced annually. The burden of food processing industry waste is still a great challenge. The principles of (reduce, reuse, and recycle) are the basis of the "circular economy. As a result, industrial waste can be converted into valuable and useful resources [2,3].

Eggshell (ES) is a typical example of a food processing waste product. As eggshells are considered useless, most of this waste is commonly disposed of in landfills without any transformation into useful materials. ES waste is among the most abundant biomaterial waste coming from food

processing technologies. At least 60 million tons of hen eggs are produced annually. The utilization of this waste resource is highly attractive in positively impacting sustainability worldwide [4].

At a glance at the morphology and physical properties of ES, to date, no sufficient studies have demonstrated nor reported for purpose. Hereby the current work aims to study frequency-dependent Dielectric properties for recycled local domestic hen eggshell powder as a prelude to eco-friendly medical application

Dielectric studies of biomaterials have been conducted for a long time, as how tissues interact with electromagnetic waves (EMW) [5]. On this basis, the mechanism of EMW interaction has the striking property that energy is conserved or "stored" in addition to being dissipated and that the ratio of the average energy stored to the energy dissipated per

cycle is independent of the frequency [6]. It is of great interest in electrophysiology and biophysics. The dielectric properties of biological materials are particularly remarkable. These frequency-dependent properties allow for the identification and investigation of various underlying mechanisms. One way to measure these electrical characteristics is through conductivity measurement, which is the inverse of electrical resistivity and represents the resistance to electric current flow [7,8].

By passing a known current of constant voltage through a known volume of the material and determining the resistance, the conductivity of the material can be determined. Dielectric properties refer to the electrical properties that measure the interaction of materials with electromagnetic fields. The dielectric permittivity of a material is a physical quantity that can be measured, with the relative static permittivity (ϵ') or dielectric constant for static electric fields. This is done by first measuring the capacitance of a test capacitor (C_0) with vacuum between its plates and then measuring the capacitance (C_s) with a dielectric between the plates. Dielectric constant (ϵ') can then be calculated and expressed relative to the value of vacuum (ϵ_0) in modern usage Equation 2 [8,9]

$$\epsilon_r = \frac{\epsilon_s}{\epsilon_0} = \frac{C_s}{C_0} \text{Equation 1}$$

The relative permittivity is a complex frequency-dependent quantity that is expressed with real (ϵ') and imaginary (ϵ'') components. The Debye equation in its simplest form of the complex dielectric constant (ϵ^*) is shown in Equation 3. The real part (ϵ') is known as the dielectric constant and is related to the capacitance of a substance and its ability to store electric energy. The imaginary part (ϵ''), also known as the loss factor, represents the energy loss of the external electric field when applied to the tested material, such as in dielectric relaxation and ionic conduction Equation 7.

The loss factor is always positive, has smaller values than the dielectric constant, and is related to various absorption mechanisms of energy dissipation. A substance with a dielectric loss factor of 0 is considered lossless [5,7,10].

$$\epsilon^* = \epsilon_r + j\epsilon_i = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega\tau_0} \text{Equation 2}$$

$$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + \omega^2\tau_0^2} \text{Equation 3}$$

$$\epsilon_i = \frac{(\epsilon_s + \epsilon_\infty)\omega\tau}{1 + \omega^2\tau_0^2} \text{Equation 4}$$

Where ϵ_s is the low frequency of ϵ' (the static dielectric constant) and ϵ_∞ is the high frequency of ϵ' (the optical dielectric constant), ω is the applied angular frequency ($\omega=2\pi f$) and τ is the average relaxation time.

Another descriptive dielectric parameter is the dissipation factor or loss tangent ($\tan\theta$), which reflects the ratio of the dielectric loss to the loss factor. This parameter is often used as an index to describe the ability of a material to generate heat.

As a simple evaluation of the Debye equations, Cole introduced a cole-cole plot for studying the nature of frequency dependence of dielectric materials. A plot of the imaginary part of the dielectric constant ϵ'' against the real part ϵ' for a given temperature, with frequency, almost always results in a curve which is the arc of a circle [9,10]

Cole and Cole's plot found for a considerable number of liquids and solids that the values of ϵ'' fell below the semicircle arc intersecting the real axis at the values of ϵ' and ϵ'' . The center of the circle of which this arc was part lay below the real axis and the diameter drawn through the center from the ϵ point made an angle $\alpha < 2$ with the real axis. Cole and Cole suggested that in this case, the complex dielectric constant might follow the empirical relation in Equation 3.[10,11]

Material and method

Preparation of Eggshell Powder

Household chicken eggshells were collected and washed twice with tape and deionized water. It was

boiled for 30 minutes and then dried in a hot air oven at 80°C for 2 hours. The dried ES was first ground with a mortar and then pestle to reduce in size. The yield ES powder was sterilized by a UV lamp and then stored for further use.



Figure 1: Eggshell compacted disk

Dielectric Measurement

Dielectric properties of samples were measured using a four-probe automatic programmable LCR Meter (FLUKE, PM 6306) in the frequency range (1000Hz - 1 MHz) at room temperature (≈ 298 K). Prior characterizations, the ES powder was compacted to a disk-shaped pellet (diameter 0.5 mm, thickness ≈ 1 mm) (0.5 g) Figure 1 [12].

The pellets were then placed in the spectrometer sample holder and isolated from the metallic plates by two insulating barriers. Then, both sides of the glass samples were coated with the gold electrodes using Sputter Coater SC7620 to ensure the electrical connection to eliminate electrode interfering impedance, the present current was applied and probe attendant voltage was measured using external pair of the electrode [13].

A rectangle conductivity cell from plexiglass was used in all impedance measurements. Two disk-shaped planar parallel silver electrodes measuring the same dimension of pellet were used to pass current through samples. The cell constant $k = (\sigma z)^{-1}$ was determined by placing saline solution (0.9%) in the conductivity cell. Since the saline solution is

purely resistive; the stray capacitance and admittance of the solution is directly related to this capacitance. Both electrodes of the conductivity cell were connected to LCR meter terminals [14].

The $\epsilon'(f, \omega)$ and $\epsilon''(f, \omega)$ were recorded from $f = \omega/2\pi$ at T 298 K. The corresponding values of $\epsilon'(\omega)$ and $\epsilon''(\omega)$ are calculated according to Equation 5 and Equation 6:

$$\epsilon_r(\omega) = \frac{d \cdot C(\omega)}{\epsilon_0 \cdot A} \text{Equation 5}$$

$$\epsilon_i(\omega) = \frac{d}{\epsilon_0 \cdot A \cdot R(\omega)} \text{Equation 6}$$

Conductivity is calculated by measuring the electric current flowing through a material of a unit via the cross-sectional area (A), unit length (L), and resistance (R) Equation 1.

$$\sigma(\omega) = \frac{L}{R(\omega) \cdot A} \text{Equation 7}$$

RESULTS

SEM photographs reveal a rough crystalline structure, it may be due to calcium carbonate also monocarboaluminate. The porous structure was also noticed in **Figure 2**.

Dielectric properties

The frequency dependence of the dielectric permittivity (ϵ') and loss tangent ($\tan\delta$) of ES have been measured at room temperature as in Figure 3 and Figure 4. It is seen that the value of ϵ' decreases with an increase in frequency, which is a typical characteristic of the polar dielectrics. The trend begins with a relative increment followed by a

sudden decrease till the plateau stage [15]. The conductivity curve behaves in the opposite behavior.

The imaginary part of complex permittivity $\epsilon''(f, \omega)$ was recorded according Equation 6. Conductivity is calculated by measuring the electric current flowing through a material as shown in Figure 5.

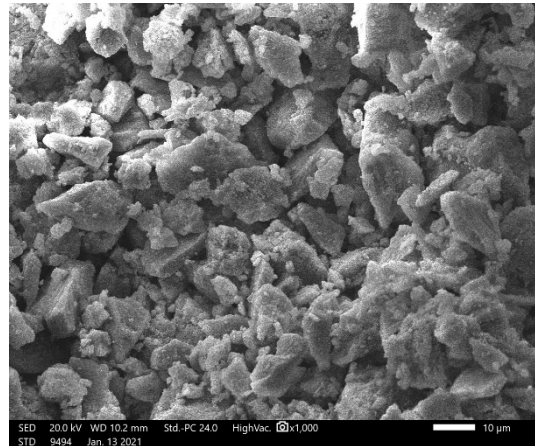


Figure 2: Photography of ES powder showing ultrasture

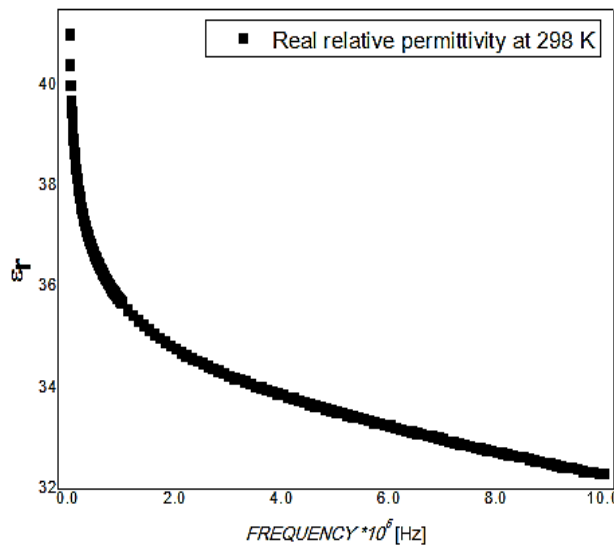


Figure 3: Variation of the real part (ϵ') of complex dielectric permittivity as a function of frequency.

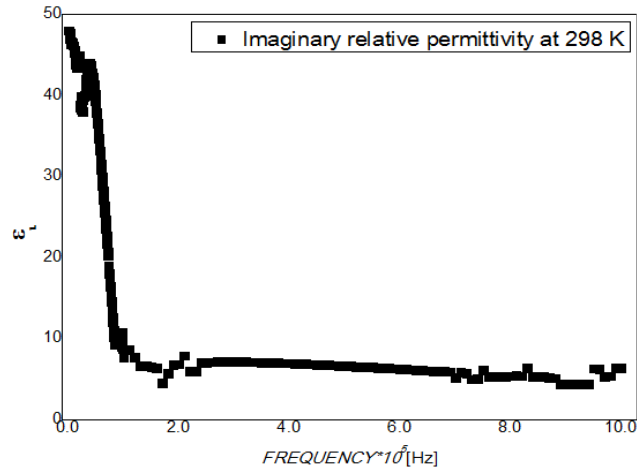


Figure 4: Variation of imaginary part (ε'') of complex dielectric permittivity as a function of frequency

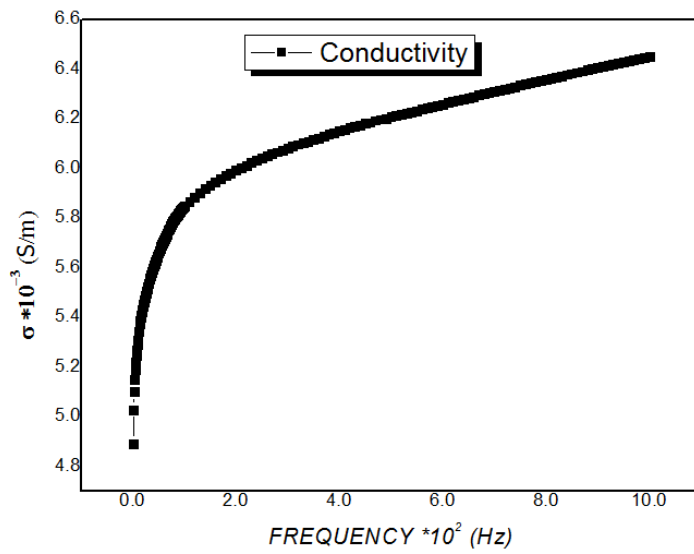


Figure 5: Behavior of electrical conductivity of ES as a function of frequency

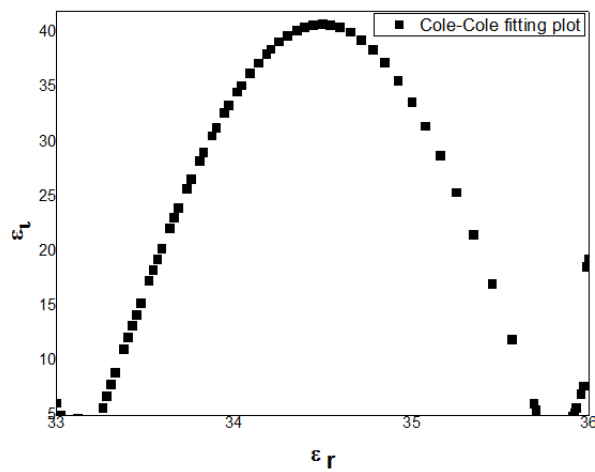


Figure 6: Cole-cole fitting plot of ES showing real and imaginary x-axis intersection

Discussion

The relative penetration that an electromagnetic signal in a material is determined by its dielectric constant. The speed of the waves decreases by a factor nearly equal to the square root of the dielectric constant when microwaves enter a dielectric material. Commonly, four types of polarizations contribute to dielectric permittivity: interfacial, dipolar, atomic, and electronic [16].

In the current work, the dielectric relaxation peaks appear when the externally applied alternating current (AC) electric field becomes equal to that of the jumping frequency of localized electric charge carriers. Dipolar and interfacial polarizations are strongly temperature-dependent, at the low-frequency region, so permittivity increases faster leading to a high dielectric constant [13,17,18].

The dielectric constant and the loss factor of ES were computed using the Debye relaxation method. The higher value of dielectric constant ϵ' at low frequencies may be due to the effect of ionic conductivity which varies inversely proportional to the frequency [11]. At the current frequency range, only β -dispersion of dipole relaxation may be mainly concerned during the permittivity response, α -dispersion (ionic relaxation) could not occur as it is related to a lower frequency range. Cole-cole fitting plot was deduced according to Debye equations. The plot shows a semicircle pattern with its center lying below (ϵ'') axis as seen in Figure 6. [6,15,18]

Conclusion

Various dielectric and related parameters such as dielectric constant, dielectric loss, and conductivity in the frequency range (1000Hz - 1 MHz) at room temperature (≈ 25 °C). Overall low dielectric parameters were obtained, that suggest the potential utilization of these materials for different biotechnological applications of ES waste, and summarize applications for biomedical, chemical, engineering, and environmental technologies.

Biomedical technologies include the production of calcium lactate, calcium phosphate, and health-promoting products

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