

BIOLOGICAL DIELECTRICS- THEORY, INSTRUMENT AND ANALYSIS**Mohammed Ibraheem Altaf and Adeel Ahmad**

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Abstract: The paper reports theory, instrument and analysis of Biological Dielectrics. Dielectric relaxation time and frequency dependence of dielectrics is explained. Instruments and the lead capacitance of the conductivity measuring cell is a frequency dependent parameter which adversely influences the dielectric measurements. Theoretical aspects of Cole – Cole plots for the calculation of relaxation time are presented. Living matter, having conductive regions separated by barrier layers, can appear to have enormous dielectric constants. In biological systems, interfacial polarization effects are prominent at radio frequencies. At low frequencies dielectric constant is high and electrical conductivity is low. At high frequencies, dielectric constant is low and conductivity is high. The physiology of blood and its constituents (plasma and erythrocytes) is altered significantly and is mirrored in its dielectric behaviour.

Key words: Dielectric properties, Digital meter (LCZ), Dielectric behaviour, Biological systems

1. Theoretical Aspects:**1.1. Theories of dielectrics****1.1.1. Homogeneous theory of dielectrics**

The conductivity σ of a material and electric permittivity ϵ are the current densities and charges induced considerably. The current densities and the charge induce in return to applied electric fields of unit amplitude. It can be shown by considering an ideal parallel plate capacitor, with area of plate A and distance d, which contains the material under investigation (Fig. 1.1.).

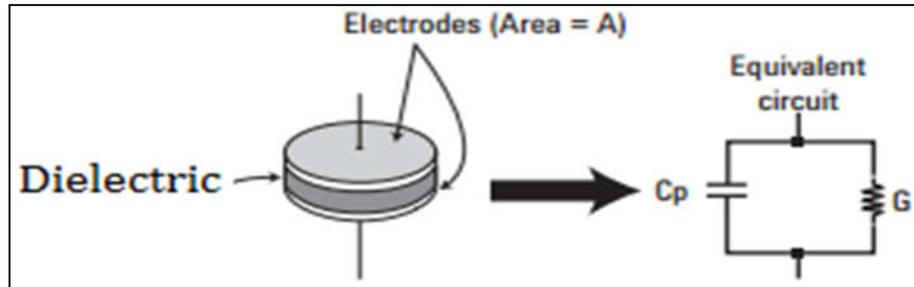


Fig. 1.1. Equivalent circuit of an ideal parallel-plate capacitor occupied by the material

Primarily, let the area between the plates be vacuum. A charge density D is induced, where ϵ_0 is free space permittivity and V be the potential difference between the plates related by

$$D = \epsilon_0 \frac{V}{d} \quad (1)$$

Where ϵ_0 value is equal to 8.85×10^{-12} F/m. The capacitance C is the ratio of the charge induced to the applied voltage. Ignoring the plates from the fringing fields is among the plates, the capacitance is related by

$$C = \frac{\epsilon_0 A}{d} \quad (2)$$

If an element is now placed among the plates, the charge density P_0 will develop on the plates due to the polarization of charges within the element. For adequately low voltages and this charge density is comparative to E can be written as

$$D = \epsilon_0 E + P_0 = \epsilon_s \epsilon_0 E \quad (3)$$

The capacitance is characterized by

$$C = \frac{\epsilon_s \epsilon_0 A}{d} \quad (4)$$

Where, ϵ_s resembles static permittivity of the element. The circuit consists of a conductance G in parallel with the capacitance can be written as,

$$G = \sigma_s \frac{A}{d} \quad (5)$$

Where, σ_s elements static conductivity

For sinusoidal oscillating fields, the electric properties of the circuit can be specified in several ways:

Complex admittance at angular frequency ω , can be given as

$$Y^* = G + j\omega\varepsilon$$

$$Y^* = \frac{A}{D} (\sigma + j\omega\varepsilon_0\varepsilon_s) \quad (6)$$

Where σ and ε are conductivity and permittivity, respectively, at the angular frequency ω . then the complex conductivity σ^* of the element

$$\sigma^* = \sigma + j\omega\varepsilon_0\varepsilon \quad (7)$$

Complex capacitance c^* can be written as

$$c^* = \frac{Y}{j\omega} \quad (8)$$

And complex relative permittivity ε^*

$$\varepsilon^* = \varepsilon - j\varepsilon''$$

$$\varepsilon^* = \varepsilon' - j\frac{\sigma}{\omega\varepsilon_0} \quad (9)$$

The permittivity and complex conductivity is given by

$$\sigma^* = j\omega\varepsilon^*\varepsilon_0 \quad (10)$$

Lastly, the series equivalent impedance z^* can be inscribed as,

$$z^* = \frac{1}{Y^*}$$

$$Z^* = R + jX = \frac{G - j\omega c}{G^2 + (\omega c)^2} \quad (11)$$

The complex specific impedance of the element Z_1^* is given as

$$Z_1^* = \frac{1}{\sigma_0} = \frac{\sigma - j\omega\varepsilon_0\varepsilon}{\sigma^2 + (\omega\varepsilon_0\varepsilon)^2} \quad (12)$$

When a homogeneous element of permittivity occupies a parallel plate capacitor of gap d and conductivity σ and the potential difference between the plates is V , the charge per unit area is,

$$Q = \frac{\epsilon_0 \epsilon V}{4\pi d} \quad (13)$$

And the current density is,

$$I = \frac{dQ}{dt} + \frac{\sigma V}{d} \quad (14)$$

If V is the alternative potential $V = V_0 \exp(j\omega t)$

$$I = \frac{(j\omega\epsilon_0\epsilon + \sigma) V}{4\pi d} \quad (15)$$

$$I = \frac{j\omega\epsilon_0}{4} \left(\epsilon - \frac{j4\pi\sigma}{\omega\epsilon_0} \right) \frac{V}{d} \quad (16)$$

From this equation one could evidently consider the medium as having a complex conductivity,

$$\sigma^* = \sigma + \frac{j\omega\epsilon_0}{4\pi} \quad (17)$$

or a complex permittivity,

$$\epsilon^* = \epsilon - \frac{j4\pi\sigma}{\omega\epsilon_0} \quad (18)$$

is inversely proportional to the frequency, becoming infinite at zero frequency. Conductivity will, thus, lead to a dielectric loss.

1.1.2. Theory of heterogeneous dielectrics

Consider a dielectric made up of two parallel slabs of material of thickness d_1 and d_2 between the capacitor plates, the first having permittivity ϵ_1 , conductivity σ , the second permittivity ϵ_2 and zero conductivity. This is equivalent to the capacitor C_1 and C_2 , in series, the capacitances per unit area being,

$$C_1 = \frac{[\epsilon_1 - \frac{j4\pi\sigma}{\omega\epsilon_0}] \epsilon}{4\pi d_1} \quad (19)$$

$$C_2 = \frac{\epsilon_2 \epsilon}{4\pi d_2} \quad (20)$$

Hence, ϵ is the permittivity of the heterogeneous element, measured at the angular frequency ω . The total capacitance per unit area,

$$C_T = \frac{C_1 C_2}{C_1 + C_2} = \frac{\epsilon}{4\pi} \cdot \frac{\{(\epsilon_1 - j4\pi\sigma/\omega\epsilon)\epsilon_2 / d_1 d_2\}}{\{(\epsilon_1 - j4\pi\sigma/\omega\epsilon)^2 / d_1\} + (\epsilon_2 / d_2)} \quad (21)$$

So that the apparent permittivity is

$$\varepsilon_T = \frac{\varepsilon_2(\varepsilon_1 - j4\pi\sigma / \omega\varepsilon)}{(\varepsilon_1 - j4\pi\sigma / \omega\varepsilon)f_2 + \varepsilon_2f_1} \quad (22)$$

Where, $f_1 = d_1 / (d_1 + d_2)$, $f_2 = d_2 / (d_1 + d_2)$

When $\omega \rightarrow 0$, $\varepsilon_T \rightarrow \varepsilon_0 = \varepsilon_2 / f_2$

When $\omega \rightarrow \infty$, $\varepsilon_\infty \rightarrow \varepsilon_1\varepsilon_2 / (\varepsilon_1f_2 + \varepsilon_2f_1)$

It follows

$$\frac{\varepsilon_T - \varepsilon_\infty}{\varepsilon_0 - \varepsilon_\infty} = \frac{-j4\pi\sigma f_2 / \omega\varepsilon}{\varepsilon_1f_2 + \varepsilon_2f_1 - j4\pi\sigma f_2 / \omega\varepsilon} = \frac{1}{1 + j\omega\tau} \quad (23)$$

Where,

$$\tau = \frac{\varepsilon_1f_2 + \varepsilon_2f_1}{f_2\sigma} \cdot \frac{\varepsilon}{4\pi} \quad (24)$$

The arrangement behaves accurately like a polar dielectric, with 'τ' as relaxation time which turns out to be greater as σ becomes smaller.

Such a composite arrangement as considered here consisting of two parallel – sided slabs of material with different properties, is very unlikely to occur in practice, but a similar effect will be produced in a much more general inhomogeneous medium, for example, a dispersion of conducting spheres in a non-conducting medium, the case considered by Wagner. The possibility of spurious results due to this effect (Maxwell – Wagner effect) must be considered, whenever dielectric is not known for certain to be homogeneous.

2. Types of Polarization

It is known that the blocker or the restricted motion of the charges in matter shows up as a polarization. The continuous motion of such charges shows up as a conductive response, the two processes are linked over the entire frequency range by the Kramer's –Kronig relations. In general, five types of polarization exist in biological matter in a wide range of frequency are:

1. Electronic polarization
2. Atomic polarization
3. Orientation or polar polarization
4. Interfacial polarization
5. Electronic double layer and counter ion polarization

2.2.1. Electronic Polarization

The most common polarization response is that which arises from the slight distortion of the centres of positive and negative charge of the atoms. The electronic distribution about the nuclei is distorted by an imposed electric field. The distortion is usually very small. Because the electric fields within the atoms are already very large compared to imposed electric field (about 10^{11}Vm^{-1} within the atom and $0 - 10^8 \text{Vm}^{-1}$ for the external fields), the electronic shifts with respect to the nucleus is not large. An approximate value of the shift of the centre of the electronic charge in the atom of the centre of the external fields is about 10\AA or 10^{-8}m . Electronic polarization normally contributes only rather to the polarizability of solids. In most organic solids, electronic polarization leads to a value for the relative dielectric constant of about 1.8 to 4. Inorganic solids, especially elements of high atomic number which have high electronic density, can exhibit somewhat higher dielectric constant values than those of carbon compounds.

2.2.2. Atomic Polarization

Atomic polarization arises from the shifts of contrarily charged atoms relatively to the matter. In maximum organic solids, if ions are absent, this polarization makes negligible contribution to the whole polarization. This contribution is determined by comparing the dielectric constant measured at less (radio) frequencies and at high (optical) frequencies. Typically, the atomic polarization in such compounds contributes about 15% to the total. In the case of inorganic compounds, on the other hand, the atomic polarization can be quite larger.

2.2.3. Orientational or Polar Polarization

Orientational polarization arises from the orientational response of molecules or parts of molecules. The molecules that have permanent dipoles incline to retort to a peripheral electric field by realignment to decrease their potential energy. The possibilities of rotational polarization exist for pendent polar groups hung flexibly on molecules. This type of polar response by intermolecular dipoles plays a significant part in the low, medium and high frequency polarization of macromolecular solids. The orientation polarization of solids is rather larger. Rotational of

dipolar molecules or pendant groups in the solid state is well known, and occurs in solids such as polymers containing polar groups such as $-\text{OH}$, $-\text{CN}_2$ $-\text{Cl}$ and NO_2

2.2.4. Interfacial Polarization

Interfacial polarization arises by the action of charge distributions occurring at the interfaces of the phases making an inhomogeneous dielectric. Interfacial polarization occurs because of a migration of charged carriers through the several phases of the dielectric in such a way to produce differing charge accumulations at the interfaces. At low field strengths interfacial polarization in materials is low on the other hand, in materials that comprise of more than one phase of a combination of materials; It is large. Living matter, having conductive regions separated by barrier layers, can appear to have enormous dielectric constants. In biological systems, interfacial polarization effects are prominent at radio frequencies.

A different diverse model that can be considered in many biological situations that are real is that of a permeable dielectric. For such a diverse system with $\sigma_2 \gg \sigma_1$, Van Seeke showed that the dielectric dispersion is shown as

$$\varepsilon' = (1 - v)\varepsilon_1 + v\varepsilon_2 \quad (25)$$

and
$$\varepsilon'' = \frac{\sigma}{\sigma_0 \omega} \quad (26)$$

Where 'v' is the fraction of volume occupied by the water filled pores in the total system.

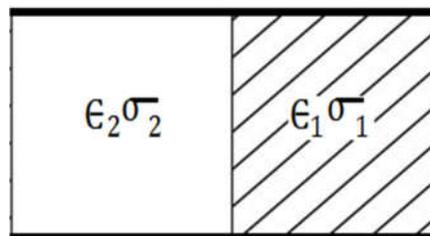


Fig. 2.2. A heterogeneous system

The simplest example of interfacial polarization is that of the classical Maxwell – Wagner two-layer capacitor (Fig. 2.2.), the dielectric constant of which is ε_i ; conductivity is σ_i and thickness is d_i .

When a.c. is field is applied to the capacitor, then the a.c. voltage $v(t) = v^0 e^{j\omega t}$ can be obtained using the impedance z_i of the equivalent circuit of the Maxwell - Wagner capacitor.

From the circuit theory,

$$Z = Z_1 + Z_2 = \frac{V}{I} \quad (27)$$

Where Z_1 and Z_2 are the impedance of the two materials 1 and 2, while I is the current flowing through the circuit and V is the applied voltage

$$\text{But } Z_i = \frac{R_i}{1 + j\omega R_i C_i} = \frac{R_i}{1 + j\omega \tau_i} \quad (28)$$

Therefore, total impedance, z may be written as

$$Z = \frac{(R_1 + R_2)(1 + j\omega \tau)}{(1 + j\omega \tau_1)(1 + j\omega \tau_2)} \quad (29)$$

The total current in the circuit, I is shown as

$$I = \frac{V}{Z} \quad (30)$$

Where $C_0 = \epsilon_0$

$$\frac{1}{Z} = j\omega \frac{\epsilon}{\epsilon_0}; C_0 = j\omega K C_0 \quad (31)$$

Then the relative dielectric constant,

$$K = \frac{1}{j\omega C_0 Z} = \frac{(1 + j\omega \tau_1)(1 + j\omega \tau_2)}{j\omega \epsilon_0 (R_1 + R_2)(1 + j\omega \tau)} = K' - jK'' \quad (32)$$

The in phase and out of phase components are:

$$K' = \frac{\tau_1 + \tau_2 - \tau + \omega^2 \tau_1 \tau_2 \tau}{C_0 (R_1 + R_2)(1 + \omega^2 \tau^2)} \quad (33)$$

$$K'' = \frac{1 - \omega^2 \tau_1 \tau_2 + \omega^2 \tau (\tau_1 + \tau_2)}{\omega C_0 (R_1 + R_2)(1 + \omega^2 \tau^2)} \quad (34)$$

At very low frequency increases ($\omega \rightarrow \infty$) to the optical value as

$$K_{\infty} = \frac{\tau_1 + \tau_2}{C_0(R_1 + R_2)\tau} \quad (35)$$

The term K will be written in the form

$$K' = \frac{K_s - K_{\infty}}{1 + \omega^2\tau} + K_{\infty} \quad (36)$$

On the other hand, the out of phase term k'' , which may be written as

$$K'' = \frac{K_s - K_{\infty}}{1 + \omega^2\tau^2} \cdot \omega\tau + \frac{1}{\omega C_0(R_1 + R_2)} \quad (37)$$

When compared to that for a simple Debye dielectric is seen to constant an extra term,

$\frac{1}{\omega C_0(R_1 + R_2)}$ this corresponds to the conductivity, σ ,

Where $\sigma = \frac{\epsilon_0}{C_0(R_1 + R_2)}$ and is the result of the action of the series resistors, $R_1 + R_2$.

2.2.5. Electrical Double Layer and Counter Ion Polarization

When an electrically neutral object is immersed in a liquid medium the surface of the object acquires a net electrical charge. This surface charge can arise from several possible processes. The most basic one is that arising from differences in thermodynamic or electrochemical potential between the solid and solution. Basically, one can say that two material phases in contact will have different affinities for electrons, and that the thermodynamic equilibrium exists because of the flow of electrons or ions from one phase to another. Another effect occurs for the particles, which attains a net charge because of the replacing of one of its constituent ions with one of the lower valency. This situation is most relevant for biological particles in weak electrolytes, where the surface charge is influenced by the absorption of ionisable chemicals or by the presence of constituent ionisable surface chemical groups such as COOH or NH_2 . Because of the build-up of such charges, an electrostatic potential will be present in the locality of the particles whose values decrease to that of the bulk liquid medium with increasing distance from the particles. Because of the electrostatic potential, opposite

charge ions are attracted with that of the surface charge, near the particle, whereas like charge ions will be deterred into the bulk liquid medium. The presence of negatively charged particles surrounded by an atmosphere of positive charge counter ions, gives concept of an electrical double layer.

A major class of dielectric phenomenon rises from ions dispersion in the electrical double layers next to charge surfaces. The time constant of such effects is of the form D is diffusion co-efficient and L^2/D , where L is the length over which diffusion occurs. At audio frequencies, counter ion polarization effects are pronounced in biological system. At low frequencies, Counter ion effects contribute to the α dispersion in biological tissues.

To explain dielectrics polarization effects associated with counter ion polarization, Schwarz considered the case of a macroscopic sphere with radius 'a' with counter ion surface charge density ε_0 (in units of m^{-2}), in which the electrical double layer thickness is much less than the particle diameter.

Effective permittivity of a particle ε_p^* can be obtained, by solving the equation of electro diffusion, as

$$\varepsilon_p^* = \varepsilon_b + \frac{1}{1 + j\omega\tau} \frac{e_0^2 \delta_0 a}{\varepsilon_0 K \tau} \quad (38)$$

Where ε_b is the permittivity of the bulk material of the particle, e_0 is the charge of the counter ion; and τ (relaxation time).

As τ (relaxation time) is proportional to square of the radius of the particle, hence can be written as

$$\tau = \frac{\varepsilon_0 a^2}{2\mu K \tau} \quad (39)$$

Where μ is the surface mobility of the counter ions.

2.5.6 Dielectric Loss and Dissipation Factor

Current leads the applied voltage by 90° when an alternating current voltage is subjected to a perfect insulator. When the same a.c. voltage is applied to a dielectric, the electrical energy is dissipated in the form of heat. This dissipation of energy is termed as dielectric loss.

In commercial dielectrics, voltage lags by current by an angle θ . This phase angle θ is less than 90° . Complementary angle $\delta = 90 - \theta$ is known as dielectric loss angle. Power dissipation can be measured by the loss angle for each cycle.

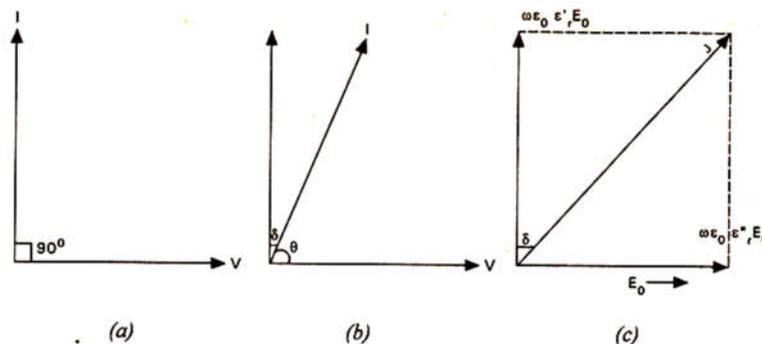


Fig: 2.3. (a) Applied voltage and charging current phase relation of a perfect insulator.
 (b) Applied voltage and charging current phase relation of commercial dielectric material.
 (c) Field direction and Current density components in a lossy dielectric material.

An oscillating electric field $E = E_0 e^{j\omega t}$ is subjected across the plates. Displacement field $D = D_0 e^{j(\omega t - \delta)}$

$$\frac{D_0}{E_0} e^{-j\delta} = \epsilon'_r - j\epsilon''_r = \epsilon_r^* \quad (40)$$

Dielectric constant $\epsilon'_r = \frac{D_0}{E_0} \cos \delta$, dielectric loss $\epsilon''_r = \frac{D_0}{E_0} \sin \delta$ dissipation factor

$$\tan \delta = \frac{\epsilon''_r}{\epsilon'_r}$$

2.5.7. Frequency dependence of dielectrics

A dielectric is basically treated as an insulator. The existence of the dielectric enhances the capacity of a condenser. The remarkable property of a dielectric is its ability to get polarized in the presence of an electric field. This ability is measured in terms of relative permittivity (ϵ_r)

or dielectric constant. Microscopically the dielectric constant is related to the electric moment induced by charged particles in a material under the influence of an applied electric field.

The influence of frequency on the dielectric constant of an ionic crystal is related to the distinction of polarizability. At low frequencies, the dielectric constant possesses space charge, electronic, atomic polarization. At higher frequencies such as in MHz region, the dielectric constant reduces and then becomes independent of frequency. At high frequency the value remains constant until the strong resonance absorption frequency is advanced in the infrared region.

Electronic polarization alone exists, beyond the resonance frequency since the ions cannot follow in the field. In this region dielectric constant is termed as high frequency dielectric constant (ϵ_{∞}).

In addition to the loss due to relaxation effects, there may be loss due to conduction. The relation between the conductivity and the loss is given by

$$\sigma = \frac{\omega\epsilon}{4\pi} \quad \text{or} \quad \sigma = \epsilon_0\epsilon''\omega \tan \delta \quad (41)$$

Where ϵ_0 is the free space permittivity

$$\text{Hence} \quad \sigma = \epsilon_0\epsilon''\omega \tan \delta \quad \text{Or} \quad \tan \delta = \sigma/(\epsilon_0\epsilon''\omega) \quad (42)$$

The entire contribution to the $\tan \delta$ (dielectric loss) is given by equation (43) due to free valencies, dipolar Debye loss is

$$\tan \delta = \frac{\sigma}{\epsilon_0\epsilon''\omega} + \frac{(\epsilon_s - \epsilon_{\infty})\omega\tau}{\epsilon_s + \epsilon_{\infty}\omega^2\tau^2} \quad (43)$$

The frequency dependence of the loss because of conduction (1 term) in equation (22) and dipolar loss given by the second term are different. Unlike the loss due to conduction which displays a linear log $\tan \delta$ against frequency plot, the loss due to relaxation effect displays maximum at a certain frequency.

At low frequencies and normal temperature, the second term in equation (43) is negligible. The term d.c. conductivity becomes predominant and conductivity is usually independent of frequency. The loss due to conduction may be defined clearly due to motion of free vacancies, whereas the Debye loss due to relaxation effects is not well-defined. The loss may be due to rotating polar compounds. The response of a dielectric to the state field is instantaneous. The quantities involved are real numbers. When an alternating field is applied the shift cannot follow the field. This is due to inertial effects and specially oriented defects. As a result, there will be a loss in the material. The dielectric constant and the related quantities are to be treated as complex quantities.

$$\bar{\epsilon}(\omega) = \epsilon' - i \epsilon'' \quad (44)$$

$$\epsilon'(\omega) = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{1 + \omega^2 \tau^2} \quad (45)$$

$$\epsilon''(\omega) = \frac{(\epsilon_s - \epsilon_{\infty})\omega\tau}{1 + \omega^2 \tau^2} \quad (46)$$

Where τ is the relaxation time and ω the angular frequency. The imaginary part ϵ'' is related to the dielectric power loss. The loss can be represented in terms of $\tan \delta$ as

$$\tan \delta = \frac{\epsilon''}{\epsilon'} \quad (47)$$

Combining equations (45) and (46), we get

$$\tan \delta = \frac{\epsilon''}{\epsilon'} = \frac{\epsilon_s - \epsilon_{\infty}}{\epsilon_s + \epsilon_{\infty} \omega^2 \tau^2} \quad (48)$$

The loss ($\tan \delta$) is associated with the dipolar impurities, thermally generated charge carriers like electrons, holes, ions, or defects which can be specially oriented entities, electrode polarization due to space charge and even due to presence of air gap.

2.5.8. Temperature dependence of Dielectrics

The properties of a dielectric material are a function of many experimentally controlled parameters. In this regard, the main concern is the temperature. The difference of dielectric constant with temperature depends on the variation in polarizabilities. Havingand Bosman (1965) have shown that the increase in temperature results in three effects (1) a decrease in the

number of polarizable particles per unit volume (2) an increase of polarizability of a fixed number of polarizable particles per unit volume and (3) a dependence of polarizability itself on temperature. Above room temperature the formation and transport of lattice defects affect the dielectric constant. The values are highly frequency dependent. Large variations are observed due to phase transformation and its order-disorder changes are present (Zhehedev).

The temperature dependence of loss ($\tan \delta$) depends on the nature of loss. As discussed earlier the loss due to free carriers i.e., the loss represented by the first term in equation (27) increases with temperature in an exponential manner. If the relaxation effects are strong the loss displays a maximum at certain temperature. The temperature at which maximum is obtained is frequency dependent. The analysis is made complicated if the crystals have more than one relaxation effect. When the dielectric exhibits both D.C. conductivity and relaxation losses, explicit analysis is possible only if temperature and frequency dependence is measured over a wide range.

The temperature dependence of conductivity in crystals can be represented as

$$\sigma = A e^{-E/kT} \quad (49)$$

Where A is the function of number of ions variable for conduction and their mobility. E is the activation energy required to liberate the ions. At low temperature region or the extrinsic region, the conductivity is due to impurities and defects. The conductivity shows frequency dependence. At higher temperature (termed as intrinsic conductivity region) conduction is due to motion of ions from lattice positions. The conduction above a certain temperature becomes frequency independent.

In the crystals with ionic bonded structures, the conduction is expected to be by motion of ions in the lattice or the defects (Berry et al.) At very low frequencies less than 1 KHz the conductivity is a function of temperature. Since the dielectric loss associated with relaxation effects can be neglected, DC conductivity solely represents the conductivity. At higher frequencies the conductivity will have components due to AC and DC conductivity.

Hence the net conductivity is

$$\sigma_{\text{total}} = \sigma_{\text{DC}} + \omega \epsilon_0 \epsilon' \tan \delta \quad (50)$$

At high temperatures and low frequencies, the II term in equation (50) can be neglected and the a.c. conductivity approached d.c. conductivity at high temperatures. Below few KHz, the d.c. conductivity is the dominant and a.c. conductivity may be insignificant. At higher frequencies the a.c. Conductivity is dominant and the contribution due to d.c. conductivity becomes insignificant.

2.5.9. Dielectric Relaxation

All polarization mechanisms respond to an electric field. When the frequency of the external electric field is enhanced, some polarizations will not reach their low frequency. Orientation of dipole moments is not enough to keep in phase with the applied field and hence the whole polarizability reduces from α_T to $(\alpha_T - \alpha_0)$. This reduction of polarization reduces its permittivity, and the manifestation of energy absorption, is termed as dielectric relaxation.

From Laplace transformation, the dielectric response of a first order system in the frequency domain is given by.

$$\epsilon^* = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{1 + j\omega\tau} \quad (51)$$

Where ϵ_{∞} = Dielectric constant measured at high frequency.

ϵ_s = Limiting low frequency dielectric constant.

$\omega = 2\pi\nu$ Angular frequency of the applied electric field.

τ = relaxation time

$$j = \sqrt{-1}$$

In contrast the imaginary and real components of complex permittivity is given by

$$\epsilon^* = \epsilon' - j \epsilon'' \quad (52)$$

The real part ϵ' (dielectric constant)

$$\epsilon' = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{1 + \omega^2\tau^2} \quad (53)$$

The imaginary factor ϵ'' represents dielectric loss can be given by

$$\epsilon'' = \frac{(\epsilon_s - \epsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \quad (54)$$

Both the parameters, ϵ' and ϵ'' are frequency dependent. Their dissimilarities with the frequency of external electric field are revealed in figure 2.5. The equations (53) and (54) are generally called as Debye Dispersion formulae, which are stated, especially in a state where equilibrium is achieved exponentially with respect to time, when a persistent external electric field is applied to a dielectric.

The association between the frequency of extreme loss, the magnitude of extreme loss and the relaxation time τ , can be evaluated by differentiating equation (33) with respect to ω and equated to zero.

$$\frac{d\epsilon''}{d\omega} = 0 \text{ at } \omega_{max} = \frac{1}{\tau} \quad (55)$$

And
$$\epsilon''_{max} = \frac{\epsilon_s - \epsilon_\infty}{2} \quad (56)$$

Equations (34), (35) clearly shows that the loss peak ϵ''_{max} resemble the relaxation time τ and usually arises at half the length of dielectric dispersion.

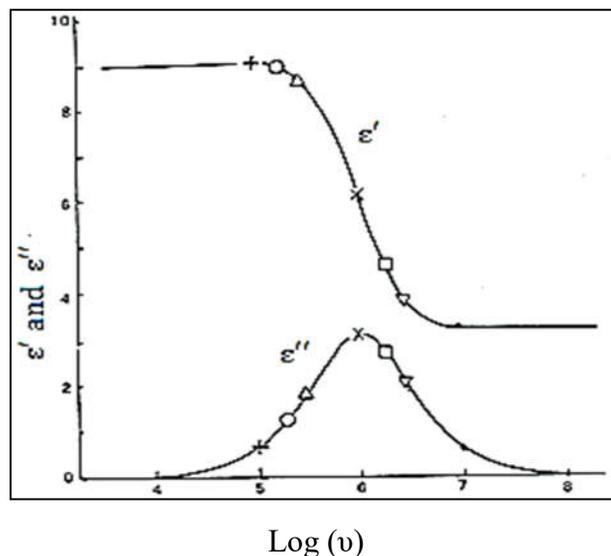


Fig. 2.4. Variation of the dielectric parameters ϵ' and ϵ'' with frequency for a Debye-type relaxation process

3. Cole-Cole Plots

An ideal Debye type single relaxation time can be found by reorganizing equations (53) and (54) to eliminate $\omega \tau$ gives

$$\left[\epsilon' - \frac{(\epsilon_s - \epsilon_\infty)}{2} \right]^2 + [\epsilon'']^2 = \left[\frac{(\epsilon_s - \epsilon_\infty)}{2} \right]^2 \quad (57)$$

The above equation is in the form $x^2 + y^2 = a^2$ (equation of circle of radius 'a'). Plot ϵ' with respect to ϵ'' give rise to a semi-circle of radius $\frac{(\epsilon_s - \epsilon_\infty)}{2}$ with the centre at $\left[\frac{(\epsilon_s - \epsilon_\infty)}{2}, 0 \right]$ such a plot of ϵ' and ϵ'' is referred as a Cole-Cole plot. (Fig. 2.5.).

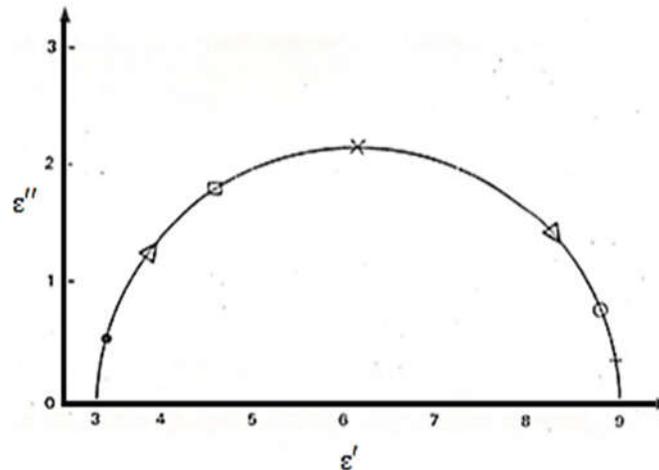


Fig. 2.5. Cole-Cole plot (semi-circle)

If a regular dispersal of relaxation time occurs in line with average relaxation time, semicircle (depressed) is observed in the Cole-Cole plot of ϵ' with respect to ϵ'' (Fig 2.6.) to account for such asymmetrical dispersal of relaxation times, Cole and Cole revised equation (51) to the form.

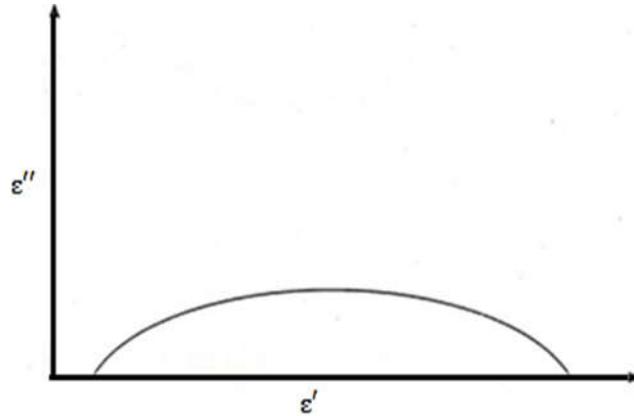


Fig. 2. 6. Cole-Cole plot semi-circle(depressed)

$$\epsilon^* = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{(1 + j\omega\tau)^\beta} \quad (58)$$

β is a parameter $0 < \beta \leq 1$, which resembles single relaxation and $\beta = 1$ also β tends to zero as the distribution approaches to infinity.

Cole and Davidson later suggested another modification to equation (58).

$$\epsilon^* = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{(1 + j\omega\tau)^\gamma} \quad 0 < \gamma < 1 \quad (59)$$

Which resembles a skewed arc shaped curve ϵ', ϵ'' which is distinctive of a non-uniform dispersal of relaxation process, where the dispersals on the higher frequency side of the principal relaxation time moves fast than those on the lower frequency region. (Fig.2.7.)

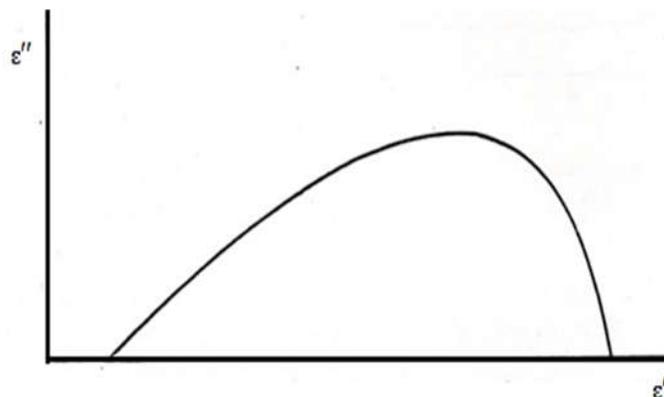


Fig. 2.7. Cole-Cole plot (skewed arc)

Dielectric behaviour of majority of materials is more complicated compared to single relaxation system. Since, if there are multiple relaxations with different relaxation time. Thus,

the dielectric response to a step change in fieldstrength can be characterized as the superposition of first order processes.

$$\varepsilon^*(\omega) = \varepsilon_\infty + \frac{\Delta\varepsilon_1}{1 + j\omega\tau_1} + \frac{\Delta\varepsilon_2}{1 + j\omega\tau_2} + \frac{\Delta\varepsilon_3}{1 + j\omega\tau_3} \quad (60)$$

If the relaxation times are detached i.e., if $\tau_1 \ll \tau_2 \ll \tau_3$, a plot (2.8) of conductivity and permittivity versus frequency will clearly show resolved dispersions, divided by plateaus at intermediate frequencies. A plot between ϵ' and ϵ'' in the complex permittivity plane will then appear as a series of semicircles each with a centre on the real permittivity axis and they will traverse the real axis at the points.

$$\varepsilon_\infty, \varepsilon_\infty + \Delta\varepsilon_1, \varepsilon_\infty + \Delta\varepsilon_1 + \Delta\varepsilon_2, \varepsilon_\infty + \Delta\varepsilon_1 + \Delta\varepsilon_2 + \Delta\varepsilon_3 \quad (61)$$

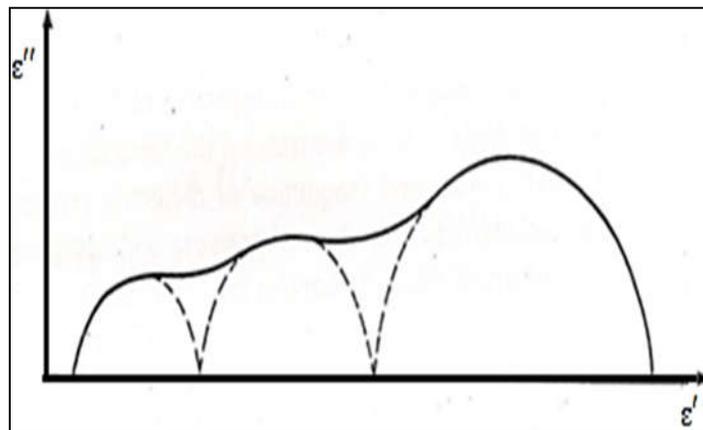


Fig. 2.8. Cole-Cole plot showing complex plane loci (multiple relaxations)

The rates with which polarization can occur are limited, so that as the frequency of the applied electric field is increased some polarization will no longer be able to attain their low frequency or d.c. values. The dipole moments are just not able to orient fast enough to keep in alignment with the applied field and the total polarizability falls from α_T to $(\alpha_T - \alpha_0)$. This fall, with its related reduction of permittivity, and the occurrence of energy absorption, is referred to as dielectric relaxation or dispersions.

The long – chain molecule and polymers show a broader dispersion curve and lower maximum loss than would be expected from the Debye relationships. In such cases the ϵ'' - ϵ' curve falls inside the Debye Semicircle. K.S.Cole and R.H. Cole suggested that in this case the permittivity might follow the empirical equation.

$$\epsilon^* - n^2 = \frac{\epsilon_0^2 - n^2}{1 + (j\omega\tau)^{1-h}} \tag{62}$$

Where h is a constant, $0 \leq h \leq 1$.

Rationalizing this expression and using $j^{(1-h)} = \exp(j \pi (1-h)/2)$, we obtain

$$\frac{\epsilon' - n^2}{\epsilon_0 - n^2} = \frac{1 + (\omega\tau)^{(1-h)} \text{Sin}(\pi h / 2)}{1 + (\omega\tau)^{2(1-h)} + 2(\omega\tau)^{1-h} \text{Sin}(\pi h / 2)} \tag{63}$$

$$\frac{\epsilon''}{\epsilon_0 - n^2} = \frac{(\omega\tau)^{(1-h)} \text{Cos}(\pi h / 2)}{1 + (\omega\tau)^{2(1-h)} + 2(\omega\tau)^{1-h} \text{Sin}(\pi h / 2)} \tag{64}$$

To find the locus in the complex plane, of which these are the parametric equations, we must eliminate $\omega\tau$. We can do this by solving for $\omega\tau$ and substituting its value back into the equation. We write the equations

$$\frac{\epsilon' - n^2}{\epsilon_0 - n^2} = \frac{1}{2} \left[1 - \frac{\frac{1}{2} \{ (\omega\tau)^{2(1-h)} - 1 \}}{\frac{1}{2} \{ 1 + (\omega\tau)^{2(1-h)} \} + (\omega\tau)^{1-h} \text{Sin}(\pi h / 2)} \right] \tag{65}$$

$$\begin{aligned} \frac{\epsilon' - n^2}{\epsilon_0 - n^2} &= \frac{1}{2} \left[1 - \frac{\frac{1}{2} \{ (\omega\tau)^{(1-h)} - (\omega\tau)^{-(1-h)} \}}{\frac{1}{2} \{ 1 + (\omega\tau)^{(1-h)} + (\omega\tau)^{-(1-h)} \} + \text{Sin}(\pi h / 2)} \right] \\ &= \frac{1}{2} \left[1 - \frac{\text{Sinh } \theta}{\text{Cosh } \theta + \text{Sin}(\pi h / 2)} \right] \end{aligned} \tag{66}$$

Where, $(\omega\tau)^{1-h} = \exp \theta$, or $\theta = (1-h) \ln (\omega\tau)$

Similarly,

$$\frac{\varepsilon''}{\varepsilon_0 - n^2} = \frac{1}{2} \left[\frac{\cos(\pi h / 2)}{\cosh \theta + \sin(\pi h / 2)} \right] \tag{67}$$

Solving these equations for $\sinh \theta$ and $\cosh \theta$ given

$$\sinh \theta = \frac{\varepsilon_0 + n^2 - 2\varepsilon'}{2\varepsilon''} \cos(\pi h / 2)$$

$$\cosh \theta = \frac{\varepsilon_0 - n^2}{2\varepsilon''} \cos(\pi h / 2) - \sin(\pi h / 2)$$

We can now eliminate θ (hence $\omega\tau$) by using

$$\cosh^2 \theta - \sinh^2 \theta = 1$$

The equation obtained in this way can be arranged in the form

$$\left\{ \frac{1}{2}(\varepsilon_0 + n^2) - \varepsilon' \right\}^2 + \left\{ \varepsilon'' + \frac{1}{2}(\varepsilon_0 - n^2) \tan(\pi h / 2) \right\}^2 = \frac{1}{4}(\varepsilon_0 - n^2)^2 \sec^2(\pi h / 2)$$

This is the equation of a circle with its centre at

$$\left\{ \frac{1}{4}(\varepsilon_0 + n^2), -\frac{1}{2}(\varepsilon_0 - n^2) \tan(\pi h / 2) \right\} \text{ and radius } \frac{1}{2}(\varepsilon_0 - n^2) \sec(\pi h / 2).$$

The relaxation time can be found from the Cole-Cole arc by using,

$$\omega\tau = \left(\frac{V}{U} \right)^{(1-h)} \tag{68}$$

U and V are the geometrical lengths, for an angular frequency ω obtained from Cole-Cole plot. The parameter h can be calculated by using the relation $\alpha = \pi h / 2$, Where α is the angle made by the line joining the centre of arc and the point at which the arc cuts the ε' axis at high frequency and, with the ε' axis.

The above relation may be proved, by writing

$$\frac{V^2 (\varepsilon_0 - \varepsilon')^2 + \varepsilon''^2}{U^2 (\varepsilon' - n^2)^2 + \varepsilon''^2} = \frac{\{\varepsilon_0 - n^2 - (\varepsilon' - n^2)\}^2 + \varepsilon''^2}{(\varepsilon' - n^2)^2 + \varepsilon''^2} \tag{69}$$

and using the expressions for $(\varepsilon' - n^2)$ and ε'' as found above. If $h = 0$ the Cole – Cole curve reduces to the Debye semicircle.

3.1. Instrumentation: The following instruments are used for evaluation of different dielectric parameters



Fig. 3.1. LCZ meter



Fig. 3.2. Conductivity Cell

To study dielectric behaviour of blood and its constituents of healthy subjects and subjects suffering from different diseases, the dielectric parameters such as dielectric constant, dielectric loss were determined at audible frequencies ranging from 100Hz to 25k Hz, using digital LCZ meter (Fig. 3.1.). A standard conductivity cell (Fig. 3.2.) was used for dielectric measurements of blood and its constituents.

The given formulae calculated the dielectric constant, dielectric loss and conductivity,

$$\text{Dielectric constant, } \varepsilon' = \frac{C_s}{C_a} = \frac{(C'_s - C_L)}{(C'_a - C_L)}$$

Where C_s : Actual capacitance of the cell with sample; C_L : Lead capacitance; C_a : Actual capacitance of the cell with air; C'_s : Measured capacitance of the cell with sample; C'_a : Measured capacitance of the cell with air i.e., without sample.

$$\text{Dielectric loss, } \varepsilon'' = \varepsilon' \tan \delta ,$$

Where ε' is the dielectric constant and δ is the dissipation factor.

$$\text{Electrical Conductivity, } K = \frac{GL}{A}$$

Where G: Conductance; L: Distance between the plates of the cell; A: Area of the plates.

The lead capacitance of the conductivity measuring cell is a frequency dependent parameter which unpleasantly effects the dielectric extents. Hence, one ought to regulate the lead capacitance of the cell before making dielectric measurements of a sample. The lead capacitance acts in parallel amalgamation with the capacitance of the measuring cell. It should be deducted from the measured capacitance, to get actual capacitance. In the present study the capacitance of the parallel plate capacitor, when the space between two plates is filled with the sample was referred as sample capacitance (C_s). If the dielectric medium in the parallel plate capacitor is air, then its capacitance is called air capacitance (C_a). The air capacitance of the measuring cell was determined by considering the liquids of known dielectric constant such as water, acetone, or alcohol. The experimental measured capacitance C'_s of a parallel plate capacitor when filled with a liquid whose dielectric constant is known is,

$$C'_s = C_s + C_L$$

Where, C_s = Actual sample capacitance

C_L = Lead capacitance

$$C_s = C'_s - C_L$$

Similarly, when air is the medium between two plates of the capacitor, the measured capacitance,

$$C'_a = C_a + C_L$$

$$C_a = C'_a - C_L$$

The dielectric constant (ϵ') defined as the ratio of capacitance (C_s), of the cell with the liquid as dielectric medium to the capacitance (C_a) when air is the dielectric medium.

$$\text{Therefore, } \epsilon' = \frac{C_s}{C_a} = \frac{C'_s - C_L}{C'_a - C_L}$$

$$\epsilon'(C'_a - C_L) = C'_s - C_L$$

$$C_L = \frac{\epsilon' C'_a - C'_s}{(\epsilon' - 1)}$$

C'_a and C'_s are measured capacitances of the conductivity cell without and with liquid of known dielectric constant at 1kHz, respectively and ϵ' is the dielectric constant of distilled water. Knowing C'_a , C'_s and ϵ' , lead capacitance was calculated at the frequencies ranging from 100 to 25 kHz.

The dielectric relaxation time of normal and diseased blood was calculated knowing ϵ' and ϵ'' at different frequencies in audible frequency range and drawing Cole- Cole plots (Fig. 3.3).

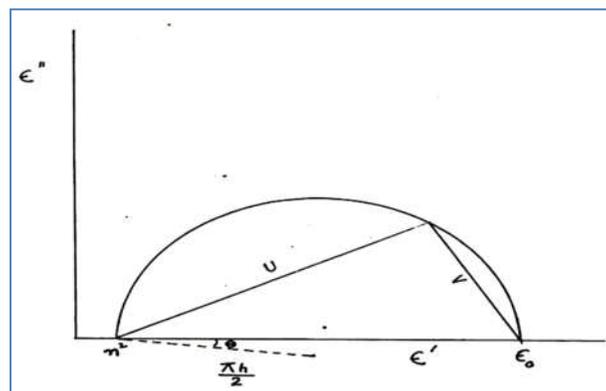


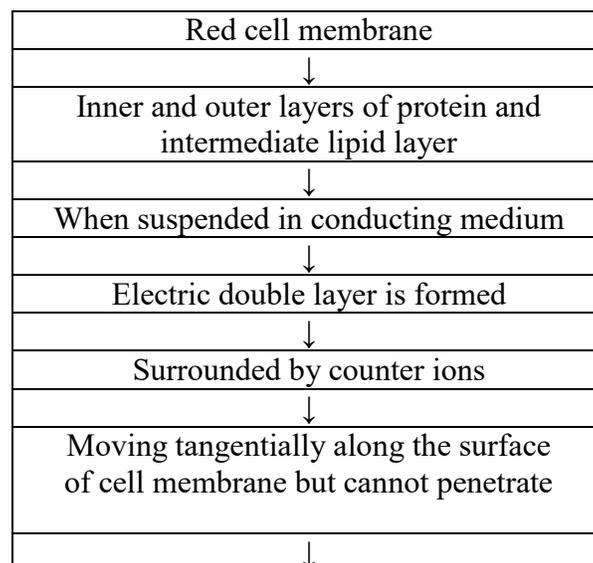
Fig. 3.3. A typical Cole – Cole Plot

Analysis:

The present study is an attempt to understand dielectric behaviour of normal and diseased blood and its constituents (plasma and erythrocytes) to have an insight into physiological alterations and adaptations and is confined to low frequency region (100 Hz to 25 kHz). Since, blood is heterogeneous dielectric medium. The α -dispersion at low frequencies, therefore, arises from the inhomogeneous nature of the blood tissue and is associated with interfacial

polarizations occurring at erythrocyte membrane. With these features and phenomena of diseased blood, one can say that in any disease, whether it is genetic or tissue or organ level, the physiology of blood and its constituents (plasma and erythrocytes) is altered significantly and is mirrored in its dielectric behaviour.

The erythrocytes, the major content of the blood, consist of a denser outer envelope resembling cell membrane and a stroma containing hemoglobin. The stroma contains 50% proteins and 10% lipids. As mentioned, red cell membrane consists of inner and outer layers of proteins (glycol proteins) and an intermediate lipid layer. Such cells are well known to possess many fixed charges at their surfaces. When the cells are suspended in the conducting medium, electric double layer is formed, surrounded themselves with counter ions of opposite sign to attain overall electrical neutrality. These counter ions can move tangentially along the surface of the cell membrane. They cannot penetrate due to the isolating properties of the cell. Such a tangential movement may be induced by low frequency alternating fields. This phenomenon is summarised in the form of flow chart (Fig. 3.4.). Therefore, polarization of the cell is achieved. This polarization mechanism is highly frequency dependent triggering the dispersive behavior of electric bulk properties. The theory of the counter ion relaxation effect has been developed by Schwarz (1962), making assumption valid for colloidal particles. This may also be valid for blood and its constituents of the present investigation, with their high dielectric constant at the lower end of the α -dispersion (



Such a movement may be induced by low frequency Alternating fields. Hence, the polarization of cell is achieved

Fig. 3.4 Polarisation of erythrocytes at audio frequencies

In the electrical behavior of blood tissue, erythrocyte membrane plays important role. The capacitance or dielectric constant of blood or erythrocytes can be attributed to the erythrocyte membrane alone, because of the fact that the interior and exterior of erythrocytes are purely conductive. Hence the electrical properties of the cell membrane can be modeled with equivalent circuit of resistor and capacitor connected in parallel (Fig. 3. 5.) and to express membrane impedance in terms of conductance and capacitance of the hypothetical circuit elements. The resistance of the cell membrane (R_m) is very high compared to the resistance of extracellular or intracellular media. The capacitance of the membrane is also high.

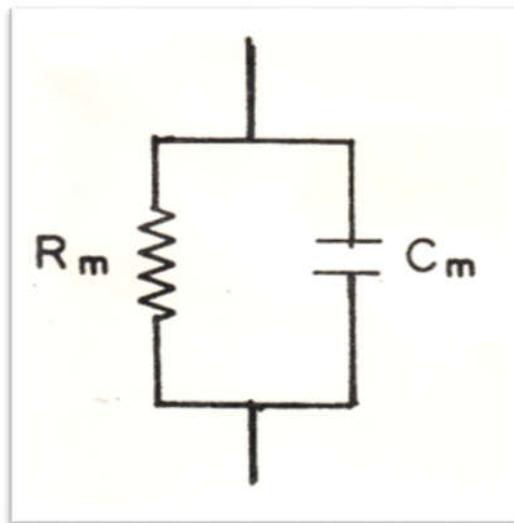


Fig. 3.5. Electrical circuit modeling of biological membrane

For the explanation of dielectric dispersion in α -region, when erythrocytes are suspended in plasma, an equivalent circuit (Fig. 5.5.) is considered. It comprises of a parallel combination of a series and parallel RC combination. In the circuit, C_m = membrane capacitance; R_m =

membrane resistance; R_i = resistance of the cell interior; R_o = resistance of the cell exterior; C_o = capacitance of the cell exterior.

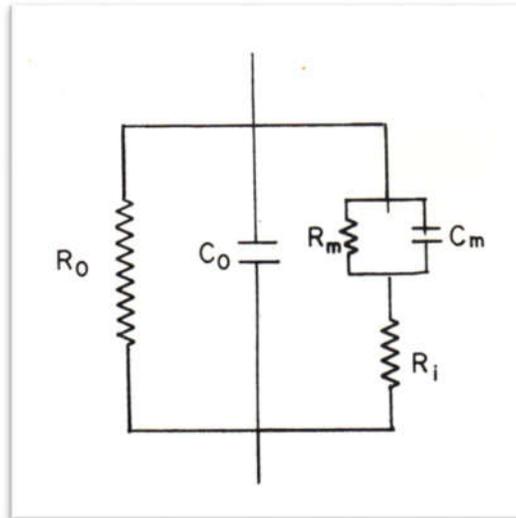


Fig. 3.6. Electrical circuit modelling of spherical cells in suspension

<u>At low frequencies</u>	<u>At high frequencies</u>
<ul style="list-style-type: none"> ➤ Current is blocked by C_m from entering the cell ➤ Membrane becomes charged ➤ Induced dipole moment for the entire cell is high ➤ This gives high dielectric constant and low conductivity. 	<ul style="list-style-type: none"> ➤ C_m is short circuited, ➤ Cell exterior the plasma fully participates in the conduction of electricity through the cell suspension ➤ Dipole moment is low. ➤ This gives low dielectric constant and high conductivity

The parallel branch represents the dielectric constant and conductivity of the path conducting electricity by passing the cell i.e., the extracellular current path and series branch consists of a capacitor representing the membrane capacitor and a resistance representing the cell interior. This branch determines the part of the total current entering the cell. At low frequencies the membrane capacitance (C_m) blocks this current from entering the cell and membrane becomes charges, resulting in a high induced dipole moment for the entire cell and in turn high dielectric constant and low conductivity. At high frequencies, the total interior participates in the conduction mechanism and the dipole moment is low. This gives low dielectric constant and high conductivity to the cell.

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