STUDIES OF THE CLAUSIUS–MOSSOTTI FACTOR

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This paper introduces a dielectrophoretic system for the analysis Clausius–Mossotti factor. The literature in this domain has shown that the real part contains information calculated by the crossover frequency. We validated in this study that the imaginary part also contains the information calculated by the relaxation frequency. Our findings confirm a relationship between relaxation frequency and crossover frequency, and also the variation of the imaginary part as a function of real part, being a semicircle, and finally we applied our model of polystyrene in deionised water.

Key words: dielectrophoresis (DEP), polystyrene, deionised water, Clausius–Mossotti factor, crossover frequency.

I. INTRODUCTION

Dielectrophoresis occurs when a polarizable particle is exposed to an electric field. The electric field causes a dipole to form within the material. If the field is uniform, the forces on the dipole charges are equal and act in opposite directions, so that the net force is zero. In a non-uniform field, however, the force on one charge will be different from the force on the other charge resulting in a net force on the particle. The force is referred to as the dielectroforetic force and the phenomena are known as dielectrophorsis [1]. The magnitude and direction of the force is related to the dielectric properties of the particle and the suspending medium. The dielectrophoretic force is frequency-dependent. The frequency dependance is given by [2].

The Clausius–Mossotti relation is named after the Italian physicist Ottaviano-Fabrizio Mossotti, whose 1850 book [3] analyzed the relationship between the dielectric constants of two different media, and the German physicist Rudolf Clausius, who gave the formula explicitly in his 1879 book [4] in the background not of dielectric constants but of indices of refraction [5].

In this paper, in the first section, we briefly present the theoretical background of The Clausius–Mossotti function and we establish a link between the real part and imaginary part. In the second section, we find the relationship between the crossover frequency and the relaxation frequency. In the third section, we present an application example of the polystyrene in deionised water.The DEP force is generated through the interaction of an induced dipole and the non uniform field. The magnitude and direction of the force is related to the dielectric properties of the particle and the suspending medium.

II. THEORY

For a fluid between two electrodes, an alternating electric field between two electrodes can be applied. The non-uniform field creates a so-called dielectrophoretic force (DEP). It is important that the particle is polarizable, e.g. that an electric dipole moment is induced by the field. Modeling a cell as a sphere, the DEP force can be expressed as [6].

$$\mathbf{F}_{\mathrm{DEP}} = 2\pi\varepsilon_0\varepsilon_m a^3\Re[\mathrm{CM}(\omega)]\nabla E_{\mathrm{rms}}^2$$

Where ε_m is the relative permittivity of the surrounding medium, the radius of the particle, $\nabla E_{\mathrm{rms}}^2$ the gradient of the root-mean-square of the applied electric field squared. The frequency (ω) dependence of the Clausius–Mossotti factor, $\mathrm{CM}(\omega)$ was determined using the complex dielectric permittivity of the medium and particle, the Clausius–Mossotti factor which is defined in the following equation:

$$\mathrm{CM}(\omega) = \frac{\varepsilon_p^* - \varepsilon_m^*}{\varepsilon_p^* + 2\varepsilon_m^*} \tag{1}$$

Where ε_m^* and ε_p^* are the complex permittivities of the medium and the particle with $\varepsilon_m^* = \varepsilon_m - j\frac{\sigma_m}{\omega}$ and $\varepsilon_p^* = \varepsilon_p - j\frac{\sigma_p}{\omega}$ where ε_m, ε_p are the permittivities of the medium and the particle, respectively, σ_p and σ_m are the conductivities of the medium and the particle, respectively, and ω is the angular frequency. Separating the real and imaginary parts of the complex Clausius–Mossotti factor $\mathrm{CM}(\omega)$ in equation (1), we finally obtain:

$$\Re[\mathrm{CM}(\omega)] = \frac{\omega^2(\varepsilon_p - \varepsilon_m)(\varepsilon_p + 2\varepsilon_m) + (\sigma_p - \sigma_m)(\sigma_p + 2\sigma_m)}{\omega^2(\varepsilon_p + 2\varepsilon_m)^2 + (\sigma_p + 2\sigma_m)^2}, \tag{2}$$

$$\Im[\mathrm{CM}(\omega)] = \frac{\omega(\sigma_m - \sigma_p)(\varepsilon_p + 2\varepsilon_m) - (\varepsilon_p - \varepsilon_m)(\sigma_p + 2\sigma_m)}{\omega^2(\varepsilon_p + 2\varepsilon_m)^2 + (\sigma_p + 2\sigma_m)^2}. \tag{3}$$

A. The limiting cases of $\Re \mathbf{CM}(\omega)$

The first limit is considered at low frequencies, giving the ionic contribution to the permittivity. It is the limit $\omega \to 0$ of expression (2)

$$\Re[\mathrm{CM}(\omega \to 0)] = \frac{(\sigma_p - \sigma_m)}{(\sigma_p + 2\sigma_m)}. \tag{4}$$

It is easy to verify that this limit can be positive if $\sigma_p > \sigma_m$ or negative if $\sigma_p < \sigma_m$ The second limit is considered at higher frequencies, giving the ionic contribution to the permittivity. It is the limit $\omega \to \infty$ in expression (2)

$$\Re[\mathrm{CM}(\omega \to \infty)] = \frac{(\varepsilon_p - \varepsilon_m)}{(\varepsilon_p + 2\varepsilon_m)}. \tag{5}$$

It is easy to verify that this limit can be negative if $\varepsilon_p > \varepsilon_m$ or positive where $\varepsilon_p < \varepsilon_m$.

B. Crossover frequency

One can identify a crossover frequency for which the Clausius–Mossotti factor, and therefore the strength of DEP vanishes from a system of positive DEP to negative or vice versa.

$$\Re[\mathrm{CM}(\omega_c)] = 0$$

From the previous equation the following expression can be derived:

$$\omega_c = \sqrt{\frac{(\sigma_m - \sigma_p)\sigma_p + 2\sigma_m)}{(\varepsilon_p - \varepsilon_m)(\varepsilon_p + 2\varepsilon_m)}}. \tag{6}$$

C. The Imaginary part of the Clausius–Mossotti factor

The analysis of the imaginary part of the Clausius-Mossotti factor $\Im[\mathrm{CM}(\omega)] = \mathrm{CM}''(\omega)$ also includes some information, it is shown that this function passes by a maximum corresponding to the relaxation frequency ω_R, therefore, the relaxation frequency was found by derivation of the imaginary part of the Clausius–Mossotti factor

$$\frac{d}{d\omega}\Im[\mathrm{CM}(\omega)] = 0.$$

The frequency ω_R (corresponding to CM'' maximum) gives the most probable relaxation time τ_R from the condition $\omega_R \tau_R = 1$.

The relaxation frequency given by

$$\omega_R = \frac{\sigma_p + 2\sigma_p}{\varepsilon_p + 2\varepsilon_m}. \tag{7}$$

The relaxation time is directly related to the relaxation frequency, which we defined as the peak position on the imaginary part of the complex Clausius–Mossotti factor, it is given by:

$$\tau_R = \frac{\varepsilon_p + 2\varepsilon_m}{\sigma_p + 2\sigma_m}. \tag{8}$$

The analysis of the $\mathrm{CM}(\omega)$ imaginary part, allows us to find the crossover frequency as a function of the relaxation frequency.

$$\omega_c = \sqrt{\frac{\sigma_m - \sigma_p}{\varepsilon_p - \varepsilon_m}\omega_R}. \tag{9}$$

D. Representation of the complex Clausius-Mossotti factor

From the expression of the real part (2), we obtain:

$$\Re[\mathrm{CM}(\omega)] = \frac{\varepsilon_p + \varepsilon_m}{\varepsilon_p + 2\varepsilon_m} - \frac{k}{1 + (\tau\omega)^2}, \tag{10}$$

where

$$k = \frac{\varepsilon_p + \varepsilon_m}{\varepsilon_p + 2\varepsilon_m} - \frac{\sigma_p + \sigma_m}{\sigma_p + 2\sigma_m}.$$

We posed

$$\mathrm{CM}'[\omega] = \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m} - \Re[\mathrm{CM}(\omega)] = \frac{k}{1 + (\tau\omega)^2}. \tag{11}$$

One of the commonly used phenomenological models is the Debye model, which was originally formulated for the complex dielectric constant [7]:

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + j\omega\tau}, \tag{12}$$

where ε_0 and ε_∞ are the low-frequency and the high-frequency dielectric constants values, respectively, τ is the central relaxation.

Based on the analogy, it is easy to reformulate Eq. (12) for the complex Clausius–Mossotti factor:

$$\mathrm{CM}^* = \mathrm{CM}_\infty + \frac{\mathrm{CM}_0 - \mathrm{CM}_\infty}{1 + j\omega\tau} \tag{13}$$

From the expression of the imaginary part (3), we obtain:

$$\mathrm{CM}''(\omega) = \frac{\omega\tau k}{1 + (\omega\tau)^2}. \tag{14}$$

From equation (11) and equation (12) we obtain:

$$(CM''(\omega))^2 + (CM'(\omega))^2 - kCM'(\omega) = 0 \qquad (15)$$

or

$$(CM''(\omega))^2 + \left(CM'(\omega) - \frac{1}{k}\right)^2 = \left(\frac{1}{k}\right)^2. \qquad (16)$$

This is the equation of a semicircle centered at $(k/2, 0)$ with the radius $k/2$ (diameter k). Equation (1) shows that the expression of the Clausius–Mossotti factor $CM^*(\omega)$ is a complex number. It consists of a real part $CM'(\omega)$ and an imaginary part $CM''(\omega)$. If the real part $CM'(\omega)$ is plotted on the abscissa (X) and the imaginary part $(CM''(\omega)$ on the ordinate axis (Y) we obtain a semicircle of centered at $(k/2, 0)$.

III. APPLICATION

The Clausius–Mossotti factor is shown in Figures 1, 2, and 3 for the parameters of a particle and medium; it is indicated in Table 1.
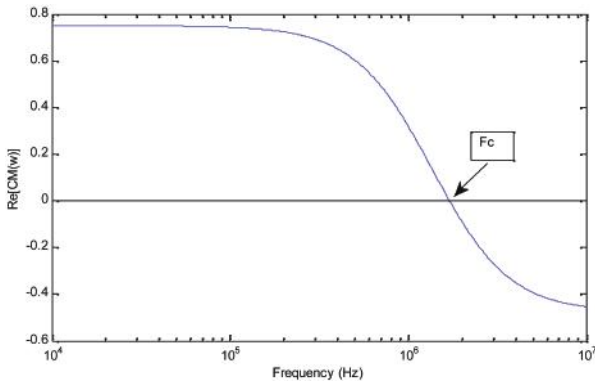

Fig. 1. Real part as a function of frequency of the Clausius–Mossotti factor for polystyrene in deionised water.
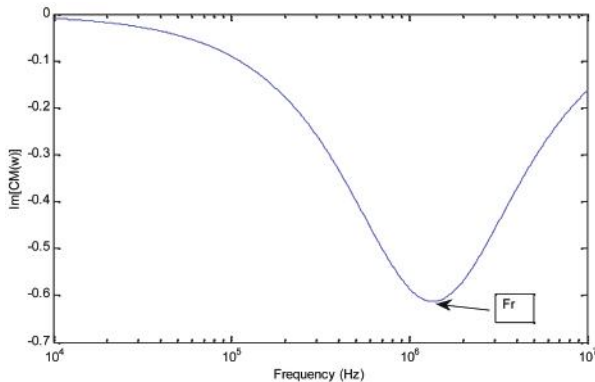

Fig. 2. Imaginary part as a function of frequency of the Clausius–Mossotti factor for polystyrene in deionised water.
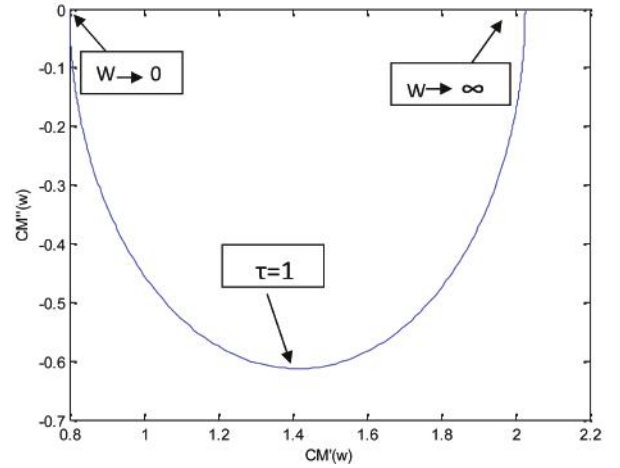

Fig. 3. Imaginary part as a function of real part shifted, of Clausius–Mossotti factor for polystyrene in deionised water.

	Dielectric permittivity	Electrical conductivity
Polystyrene (p)	2.5ε	$1E^{-2}$
DI water (m)	78ε	$1.00E^{-3}$

Table 1. Parameters of a particle and medium [8].

The complex Clausius–Mossotti factor (real part CM' and imaginary part CM'') spectra of polystyrene in deionised water, are shown in Figs. 1 and 2. This spectra confirm the real part of Clausius–Mossotti factor, it can be seen that for frequencies below approximately 1.76 MHz the real part is positive and for the frequencies above that the real part is negative. The frequency where $\Re[CM(\omega)] = 0$ is called the cross-over frequency. At that frequency a dielectrophoretic force does not act on the particles and the traveling wave dielctrophoresis becomes most prominent [9]. The dependence of the imaginary part as a function of frequency (Fig. 2) shows that CM is negative throughout the frequency range, which meant the rotation of dipoles is done in the same direction as the field [10]. The relaxation frequency of the system subjected to an electric field, denoted as "Fr", is the frequency at which this system changes its rate regime: for frequencies lower than Fr the particles constituting the material are polarized under the influence of the impressed current and dissipate the energy received in the electric form while for higher frequencies, the signal is too fast and to maintain a state of equilibrium, the material must dissipate the stored electrical energy in another form.

Figure 3 shows the evolution of the imaginary part CM'' as a function of the real CM' in the frequency range $1\,\mathrm{Hz} - 10^7\,\mathrm{Hz}$ and at room temperature for polystyrene in deionised water. The value for the constant (k) is equal to the intersection of the corresponding semicircle with the x-axis. The relaxation time can be calculated using the frequency of maxima $(f_{\max})$ of the semicircle from the equation $\tau \times \omega_R = 1$.

IV. CONCLUSIONS

In this paper, a study was developed, based on the work of K. Khoshmanesh *et al.* [8]. We tried to prove in this study that the imaginary part also contains some information calculated by the relaxation frequency. If the Clausius–Mossotti is taken seriously, we should be able to calculate the relaxation frequency and the crossover frequency, from the Bode graph or the complex diagram.

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ВИВЧЕННЯ ФАКТОРА КЛАУЗІУСА–MOSSOTTI

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У статті використано діелектрофоретичну систему для аналізу фактора Клаузіуса–Mossotti. Літературні джерела в цій ділянці показують, що дійсна частина містить інформацію про частоту переходу. Ми встановили в цьому дослідженні, що уявна частина містить інформацію про частоту релаксації. Наші результати підтверджують зв'язок між частотою релаксації та частотою переходу і те, що уявна частина як функція від дійсної частини має вигляд півкола. Ми також застосували нашу модель до полістиролу в дейонізованій воді.