Numerical Methods for the Calculation of the Cole-Cole Parameters

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The methods of evaluating the numerical values of the parameters needed for the calculation of the dielectric constant in the Cole-Cole model are considered. A distinction has been made between the cases when the experimental data include and do not include the maximum value of the imaginary part of the dielectric constant. The calculations have been performed for two ferrite samples. The results obtained gave a reasonable quantitative agreement with the experimental data.

Introduction

The Cole – Cole model [1] has been used successfully to describe the experimental data for the dielectric constant of many materials as a function of frequency [2 to 6]. In this model the dielectric constant depends mainly on four parameters, the static dielectric constant ε_o, the dielectric constant at infinite frequency ε_∞ , the relaxation time τ_o and an exponent factor α. In principle, ε_o and ε_∞ can be experimentally measured and the other two parameters τ_o and α have to be treated as fitting parameters whose values can be retrieved from the best fit to the experimental data. In most of the cases, however, ε_o and ε_∞ cannot be obtained directly from the experimental data since it is difficult to perform the measurements at very low and very high frequencies and to detect the saturated values in the two limits. The present work is concerned with presenting a simple method to extract ε_o and ε_∞ from the available measurements and to obtain subsequently the other two parameters.

The work is arranged in the following way. In ⇓2 a brief summary is given of the Cole – Cole model. The present method is displayed in ⇓3. Some applications are finally considered in ⇓4.

Summary of the Cole – Cole model:

According to this model

$$\varepsilon^* - \varepsilon_\infty = (\varepsilon_o - \varepsilon_\infty) / [1 + (i\, \omega\, \tau_o)^{1-\alpha}] \quad , \qquad (1)$$

where

$$\varepsilon^* = \varepsilon` - i\, \varepsilon`` \quad , \qquad \omega = 2\, \pi\, f \quad , \qquad (2)$$

f is the frequency, $\varepsilon`$ and $\varepsilon``$ are the real and imaginary parts of the dielectric constant. The above equation represents an arc of a circle of radius r and center (a,-b) in the $\varepsilon`$, $\varepsilon``$ complex plane as shown in Figure (1). In this figure ($-\varepsilon``$) is plotted as positive and

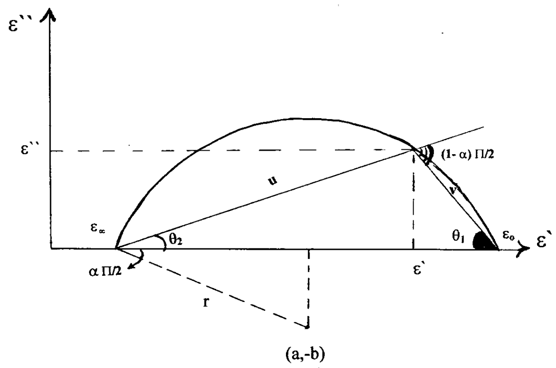

Fig. (1)
Complex plane (Argand diagram) of the dielectric constant as given by the Cole-Cole model.

$$u = \varepsilon^* - \varepsilon_\infty \ , \quad v = \varepsilon_o - \varepsilon^* = u\,(\,i\,\omega\,\tau_o)^{1-\alpha}. \qquad (3)$$

Hence,

$$|\,v/u\,| = (\omega\,\tau_o)^{1-\alpha} \ , \quad \mathit{Arg}\, v - \mathit{Arg}\, u = (1-\alpha)\,\pi/2. \quad (4)$$

Evaluation of ε_o, ε_∞ from the experimental data:

In order to obtain ε_o, ε_∞ directly, the measurements have to be carried out at very low and very high frequencies so that ε`` vanishes and ε` tends to ε_o and ε_∞ respectively. As has been mentioned above the measurements in these two frequency limits are not always possible with reasonable accuracy. We thus need to extract ε_o and ε_∞ from the measurements in the intermediate frequency range.

3.1 The data include the maximum value of ε`` :

We first consider the case when the measurements are performed on both sides of the maximum of ε``. The maximum measured value of ε``will be taken for simplicity to be the exact maximum value $\varepsilon``_m$. We also denote the corresponding measured real part by $\varepsilon`_m$. It is readily shown from Figures (2a,b) that

$$\varepsilon_o + \varepsilon_\infty \approx 2\,\varepsilon`_m\,. \qquad (5)$$

For each measured point ($\varepsilon`_1$, $\varepsilon``_1$) on the left of ($\varepsilon`_m$, $\varepsilon``_m$) we determine a point ($\varepsilon`_2$, $\varepsilon``_1$) on the right of ($\varepsilon`_m$, $\varepsilon``_m$) by using a linear fitting between the two successive measured points ($\varepsilon`_i$, $\varepsilon``_i$) , ($\varepsilon`_j$, $\varepsilon``_j$), as is illustrated in Figure (2a). Thus

$$\varepsilon`_2 = \varepsilon`_i + (\varepsilon``_1 - \varepsilon``_I)\,(\varepsilon`_j - \varepsilon`_i) \,/\, (\varepsilon``_j - \varepsilon``_i). \qquad (6)$$

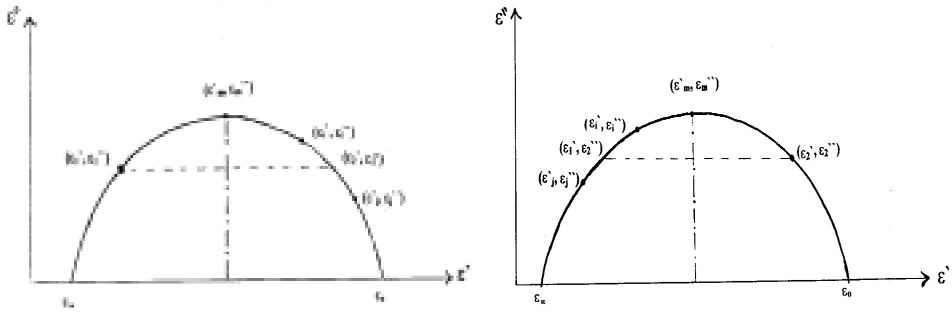

Fig. (2): (a) Calculation of $\varepsilon`_2$ from $\varepsilon`_1$(Eq. 6).
(b) Calculation of $\varepsilon`_1$ from $\varepsilon`_2$ (Eq. 7).

This procedure necessitates that the data points are very close to each other. Moreover, if accidentally $(\varepsilon`_2,\varepsilon``_1)$ is a measured point say $(\varepsilon`_i, \varepsilon``_i)$ then we do not need to use equation (6) since $\varepsilon`_2$ is already given from the measurements.

Also, for each data point $(\varepsilon`_2,\varepsilon``_2)$ on the right of $(\varepsilon`_m ,\varepsilon``_m)$ we determine $\varepsilon`_1$, as shown in Figure (2b), so that

$$\varepsilon`_1 = \varepsilon`_i + (\varepsilon``_2 - \varepsilon``_i) (\varepsilon`_j - \varepsilon`_i) / (\varepsilon``_j - \varepsilon``_i). \qquad (7)$$

It is then clear from Figures (2a,b) that

$$\varepsilon_o + \varepsilon_\infty \approx \varepsilon`_1 + \varepsilon`_2 . \qquad (8)$$

We thus take

$$\varepsilon_o + \varepsilon_\infty = \textit{Average}\ (\varepsilon`_1 + \varepsilon`_2), \qquad (9)$$

where the value $2\varepsilon`_m$ should be included in the average on the RHS of equation (9).

We still need another equation to determine ε_o, ε_∞. It can be shown by taking the two points $(\varepsilon`_1,\varepsilon``)$ and $(\varepsilon`_2 ,\varepsilon``)$ to lie on the circle that

$$[(\varepsilon`_2-\varepsilon`_1) / 2]^2 + \varepsilon``^2 = - 2\ \varepsilon``b + [(\varepsilon_o - \varepsilon_\infty) / 2]^2 \qquad (10)$$

We can consequently use the least squares method to obtain the best straight line which passes through the data points (x, y) where

$$y = [(\varepsilon`_2 - \varepsilon`_1) / 2]^2 + \varepsilon``^2 , \quad x = \varepsilon``. \qquad (11)$$

If “m” is the slope of the obtained line and “c” is its intersection with the y-axis, then

$$b = - m / 2, \qquad (12a)$$

and

$$\varepsilon_o - \varepsilon_\infty = 2\sqrt{c} \ . \qquad (12b)$$

Now ε_o, ε_∞ can be obtained from equations (9) and (12b). If either ε_o or ε_∞ are measured experimentally then it is enough to use equation (9) to obtain the other unknown value.

After knowing ε_o, ε_∞ the values of α and τ_o can be obtained by using one of the following procedures:

i - According to equation (4) and Fig.(1)

$$\alpha = 1- \frac{2}{\pi} (\hat{\theta}_1 + \hat{\theta}_2)$$
$$= 1- \frac{2}{\pi} [tan^{-1} \frac{\varepsilon``}{\varepsilon_o - \varepsilon`} + tan^{-1} \frac{\varepsilon``}{\varepsilon` - \varepsilon_\infty}] , \qquad (13)$$

$$\tau_o = \frac{1}{\omega} [\frac{(\varepsilon_o - \varepsilon`)^2 + \varepsilon``^2}{(\varepsilon` - \varepsilon_\infty)^2 + \varepsilon``^2}]^{\frac{1}{2(1-\alpha)}} . \qquad (14)$$

The value of α and τ_o can then be calculated from the above two equations for each data point. An average value of α and τ_o will finally be obtained.

ii – Also from Fig.(1) α is given by

$$\alpha = \frac{2}{\pi} \ tan^{-1} [2b / (\varepsilon_o - \varepsilon_\infty)] , \qquad (15)$$

where b is obtained from (12a).

iii – It follows from equation (4) that

$$ln| v/u | = (1 - \alpha) \ ln\ \omega + (1 - \alpha) \ ln\ \tau_o . \qquad (16)$$

The above equation implies that the relation between the data points $y` = ln|v/u|$ and $x` = ln\,\omega$ is linear. We thus determine the best straight line which passes through the experimental data (x`, y`) and find α, τ_o from the slope and intersection with the y`-axis of this line.

3.2 The data do not include the maximum of ε``:

In some cases the measured values of ε`` increases or decreases continuously with the frequency. In other words the data do not include the maximum point of ε``. Also, in some other cases the number of points on one side of the maximum is not enough to perform the above procedure. In these cases more complicated approaches have to be utilized. Here we apply the following two methods:

In the first method the data points are divided into three groups. We then choose the first point from each group and find the circle which passes through the three points. The center (a, -b) of the circle and its radius r are given by

$$a = \frac{1}{2D}\ [S_{12}\ (\varepsilon``_1 - \varepsilon``_3) - S_{13}\ (\varepsilon``_1 - \varepsilon``_2)]\ , \qquad (17a)$$

$$b = \frac{1}{2D}\ [S_{12}\ (\varepsilon`_1 - \varepsilon`_3) - S_{13}\ (\varepsilon`_1 - \varepsilon`_2)] \qquad (17b)$$

and $$r = [\ (\varepsilon`_1 - a\ )^2 + (\varepsilon``_1 + b\ )^2\ ]^{1/2}\ , \qquad (17c)$$

where $$S_{ij} = \varepsilon`_i{}^2 + \varepsilon``_i{}^2 - (\varepsilon`_j{}^2 + \varepsilon``_j{}^2) \qquad (18a)$$

and $$D = (\varepsilon`_1 - \varepsilon`_2)\ (\varepsilon``_1 - \varepsilon``_3) - (\varepsilon`_1 - \varepsilon`_3)\ (\varepsilon``_1 - \varepsilon``_2). \qquad (18b)$$

We repeat the procedure by taking the next point in each group until all the points are used. We then take an average value for the three parameters a, b, r. The values of ε_o, ε_∞ are consequently given by

$$\varepsilon_o = a + \sqrt{r^2 - b^2}\ ,\ \ \varepsilon_\infty = a - \sqrt{r^2 - b^2}\ . \qquad (19)$$

Also α can be evaluated from equation (15) or equivalently from

$$\alpha = \frac{2}{\pi} \sin^{-1} (b / r). \qquad (20)$$

As regards the relaxation time τ_o, it can be obtained by using one of the approaches (i) or (iii) considered in ⇓3.1. The value of α can be checked by using these approaches.

In the second method we fit the whole set of data by using the equation of a circle and applying the least squares method. The equation of the circle can be taken as

$$\varepsilon`^2 + \varepsilon``^2 = 2a\, \varepsilon` - 2b\, \varepsilon`` + C, \qquad (21a)$$

where

$$C = r^2 - a^2 - b^2. \qquad (21b)$$

The three parameters a, b, C can then be obtained from the experimental data by solving the three equations

$$\left.\begin{aligned} &\sum (\varepsilon`^2 + \varepsilon``^2) = NC + 2a \sum \varepsilon` - 2b \sum \varepsilon``, \\ &\sum \varepsilon` (\varepsilon`^2 + \varepsilon``^2) = C \sum \varepsilon` + 2a \sum \varepsilon`^2 - 2b \sum \varepsilon`\varepsilon``, \\ &\varepsilon``^2 . \sum \varepsilon`\varepsilon`` - 2b \sum \varepsilon`` + 2a \sum \varepsilon`` (\varepsilon`^2 + \varepsilon``^2) = C \sum \end{aligned}\right\} \qquad (22)$$

Here N is the number of the data points. Consequently, r can be found from (21b). Also, α and τ_o can be deduced by using the same procedures applied in the first method.

The two methods considered in this subsection seem to be more complicated than the method given in ⇓3.1. However, they are more general and can be used in any case.

4. Applications:

A computer program has been prepared to perform the above procedures. The calculations have been carried out for the two ferrite samples $Li_{0.5-0.5x}Cd_xFe_{2.5-.5x}O_4$, x=0.3 and $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45. For the first sample, the experimental values of ε`, ε`` at T=300K, 470K are taken from Refs. [7, 8] and are shown here in Figs. (3, 4). The results obtained are given in Table (1). Also, the representation of the input experimental data together with the obtained output circle on the Argand complex plane are displayed in Figs. (5,

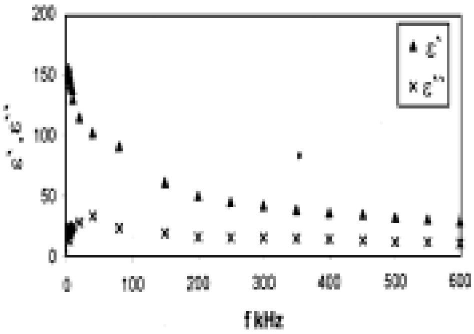

Fig. (3)

Experimental values of ε`, ε`` against frequency for $Li_{0.5-0.5x}$ $Cd_xFe_{2.5-.5x}O_4$, x=0.3, at T=300K. They are reproduced from Refs. [7,8]. The measurements were performed on a specimen of thickness 1.5mm and diameter 8.54mm.

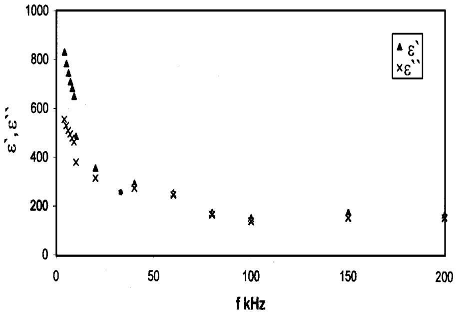

Fig. (4)

Experimental values of ε`, ε`` against frequency for $Li_{0.5-0.5x}Cd_xFe_{2.5-.5x}O_4$, x=0.3, at T=470K.

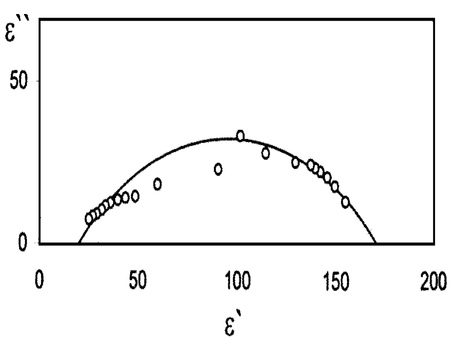

Fig. (5)

Argand diagram of the dielectric constant of $Li_{0.5-0.5x}Cd_xFe_{2.5-.5x}O_4$, x=0.3, T=300K. The experimental data are represented by the open circles o while the theoretical results (method II 2) are represented by the solid circular arc.

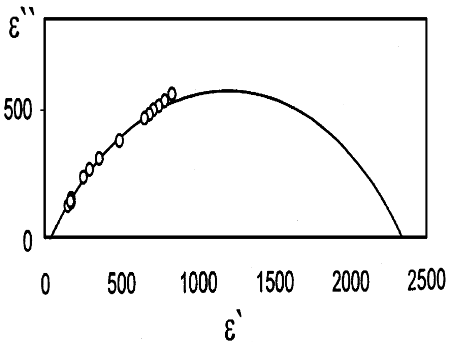

Fig. (6)

Argand diagram of the dielectric constant of $Li_{0.5-0.5x}Cd_xFe_{2.5-.5x}O_4$, x=0.3, T=470K. The theoretical results are obtained from method II 1.

6). The agreement between the input and output results seems to be quite reasonable, in particular for T=470K.

Table (1)

The results obtained for $Li_{0.5-0.5x}Cd_xFe_{2.5-.5x}O_4$, x=0.3, at T=300, 470K. The labels I, II 1, II 2 refer respectively to the method given in ⇓3.1 and to the two methods given in ⇓3.2

T(K)	Method	ε_o	$\varepsilon\infty$	α	τ_o (μ sec)	A	B	r
300	I	175	23.1	0.464	8.57	98.8	67.7	102
	II 2	171	20.0	0.484	11.4	95.6	71.8	104
470	II 1	2340	23.7	0.423	60.1	1180	908	1470
	II 2	2440	31.0	0.417	66.4	1230	925	1520

As regards the second sample, the experimental values of ε`, ε`` are depicted in Figs. (7, 8) for T=305K, 600K respectively. They are reproduced from the results of Ref.[9]. The output results obtained are shown in table (2). The Argand diagrams given in Figs. (9, 10) exhibit a good quantitative agreement between the experimental and theoretical results.

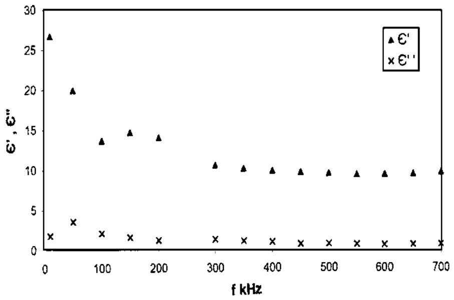

Fig. (7)

Experimental values of ε`, ε`` against frequency for $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45, at T=305K. They are reproduced from Ref. [9]. The measurements were performed on a specimen of thickness 1.94mm and diameter 10.0mm

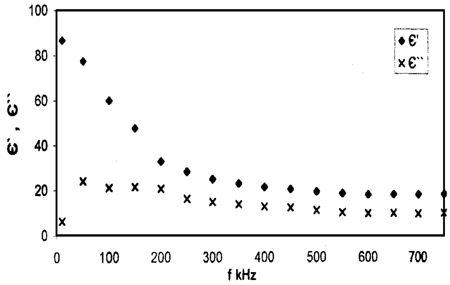

Fig. (8)

Experimental values of ε`, ε`` against frequency for $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45, at T=600K.

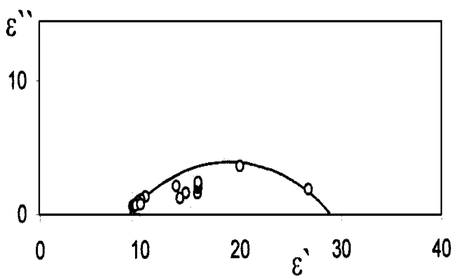

Fig. (9)

Argand diagram of the dielectric constant of $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45, T=305. The experimental data are represented by the open circles o while the theoretical results (method II 2) are represented by the solid circular arc.

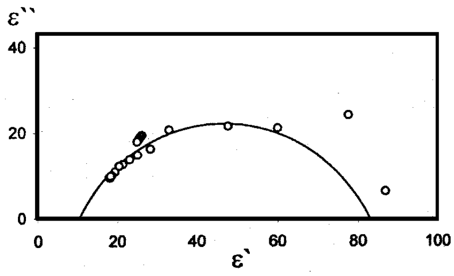

Fig. (10)

Argand diagram of the dielectric constant of $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45, T=600. The theoretical results are obtained from method II 1.

Table (2)

The results obtained for $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45, at T=305, 600K.

T(k)	Method	ε_o	ε_∞	α	τ_o (μ sec)	A	b	R
305	I	30.8	9.29	0.491	63.7	20.0	10.4	15.0
	II 2	28.9	8.93	0.529	47.9	18.9	10.9	14.8
600	I	97.5	15.0	0.238	2.51	56.2	16.2	44.3
	II 1	82.9	10.3	0.306	2.38	46.6	18.7	40.9
	II 2	92.5	12.7	0.284	2.59	52.6	19.1	44.2

5. Conclusion:

The results obtained for $Li_{0.5-0.5x} Cd_xFe_{2.5-.5x} O_4$, x=0.3 and $Mg_{1-x}Ti_xFe_{2-2x}O_4$, x=0.45 agree reasonably with the experimental data. Also, the numerical values of the parameters that calculated by using different methods are consistent. The study confirms, that the Cole-Cole model is an adequate approach for the calculation of the dielectric constant of complicated ferrite samples.

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