

Characterization of dielectric mixtures by the time domain reflectometry (TDR)

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In this paper we present some results concerning the study of mixed dielectrics; two types of materials were studied: a compound of titanate and epoxy resin and mixtures of pentanol-cyclohexane in different concentrations. For the measurement of permittivity the Time Domain Reflectometry (TDR) method was used; this method enables to determine the reflection coefficient and next by applying a Fourier transformation one obtains the complex dielectric permittivity in a large range of frequencies.

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1. Introduction

Direct experimental determination of dielectric permittivity turns out to be difficult for the solid and liquid form substances in a large range of frequencies. In this paper we propose a method that makes it possible to characterize the dielectric response for solid and liquid substances in a large range of microwave frequencies. For solid materials the technique is based on a previous characterization of binary composites prepared by mixing the dielectric under study with a polymeric substrate, in different proportions. Also, using this technique can be characterized the binary mixture of liquid dielectrics in different concentrations. Several mixture laws that can be found in the literature predict the dielectric properties of binary substances once the constituents' characteristics and proportions are known [1]. Here, such a process is reversed, and the dielectric properties of one component are found once the other constituent and the mixture have been characterized.

Effective permittivity modeling of mixtures constituted of particles embedded in a low-loss dielectric substrate requires assumptions about the particle shape and about its distribution along the substrate [2]. Approximate analytical mixture laws for the effective dielectric permittivity may be possible assuming factors like homogeneous distribution and restricted possible particle shapes and sizes, but as a consequence the validity of such laws is limited to specific kinds of mixture.

Numerical techniques may be used to evaluate the influence of inhomogeneities of particle distribution on the validity of mixture laws. It was found that the direct and inverse Wiener laws [3] constitute theoretical limits for the effective permittivity of a binary mixture, but unless the samples are strongly anisotropic the effective permittivity is limited by the Hashin-Shtrikman conditions [4]. On the other hand, analytical treatment has led to a general mixture law [5], based on the Lichteneker and incorporating many other mixture laws found in the literature.

As well as permittivity measurements of several composites of titanate-epoxy resin and pentanol-cyclohexane in different proportions (including one sample of pure epoxy resin and pure pentanol), it is shown here that not only is mixture characterization possible, but even the determination of titanate and cyclohexane permittivity is possible. To date such a law had been used for only static permittivity, but here it is shown that its validity may be extended to the gigahertz range, and that both real and imaginary parts of the permittivity components obey to the general mixture law.

2. Theoretical foundation

The time domain reflectometry (T.D.R.) is a known method for detecting and modeling of the discontinuities in of transmission line.

Each sample was washer-shaped, in order to fit the coaxial guide geometry used in the measurements. The effective complex dielectric permittivity of each sample was determined in the range from direct current to 25GHz by time domain reflectometry using the well-known adapted line method.

In the experimental setup (Fig. 1) is used a generator unit (*HP 54121A*) that gives a 200mV step pulse with a rise time of 35 ps. This was propagated along a standard APC-7mm precision coaxial guide (characteristic impedance 50 Ω) to a 25GHz digitizing oscilloscope (*HP54120B*). The sample manufacturing process is broadly described in [6], as well is the calculation procedure employed. In particular, it is explained in [6] how this method may also be applied to obtain the static conductivity σ_s . In order to reduce the signal-noise ratio, the signal reflected is obtained from 512 sample after an average of 128 runs each record.

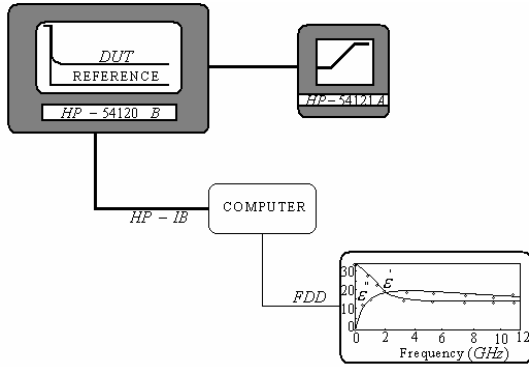


Fig. 1. Installation setup for the measurements in temporal domain.

3. Experimental

In this section we present several experimental results that were obtained by characterization of the dielectric medium, as the polar-non-polar system constituted by pentanol-1 and cyclohexane liquids in concentrations of: 20%, 40%, 60%, 80%, 100% and the mixture of titanate of calcium with resin-epoxy solids in concentrations of: 10%, 15%, 20%, 25%, 30% .

The experimental values obtained directly in temporal domain permit the determination of the real ϵ' and imaginary ϵ'' components of the complex dielectric permittivity and, also, of the relaxation parameters for non-homogeneous mixtures.

The dielectric constants ϵ_S (static permittivity), ϵ_∞ (very high frequencies permittivity) and the relaxation time τ_0 can be obtained using the Cole linear diagram [10]:

$$\epsilon' = \epsilon_S - \tau_0(\omega\epsilon'') \quad \epsilon' = \epsilon_\infty + \frac{1}{\tau_0} \left(\frac{\epsilon''}{\omega} \right) \quad (1)$$

The ensemble of results for pentanol-1 (PT) mixture in different concentrations is presented in Table 1.

Table 1. Results for pentanol-1 (PT) mixture in different concentrations.

PT	100%	80%	60%	40%	20%
ϵ_S (exp.)	15.526	11.592	7.854	4.513	2.606
ϵ_S (C - C)	15.576	11.638	7.824	4.504	2.610
ϵ_∞ (exp.)	3.231	3.069	2.870	2.553	2.298
ϵ_∞ (C - C)	3.479	3.289	2.890	2.804	2.348
τ_0 (ps) (exp.)	683	660	622	508	249
τ_0 (ps) (C - C)	672	656	600	471	230
f_0 (MHz) (exp.)	233	241	256	313	643
f_0 (MHz) (C - C)	237	243	265	336	663
ϵ''_{max} (exp.)	6.180	4.288	2.580	0.860	0.124

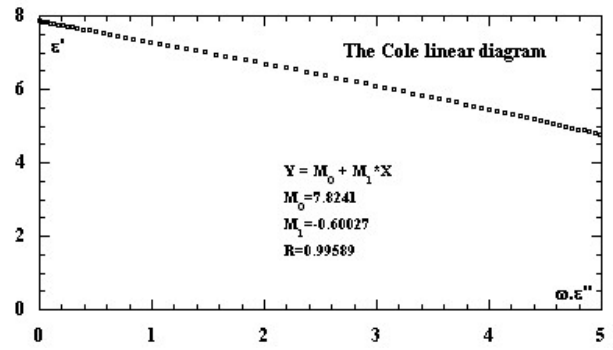


Fig. 2. The Cole linear diagram (pentanol-1+cyclohexane, 60%).

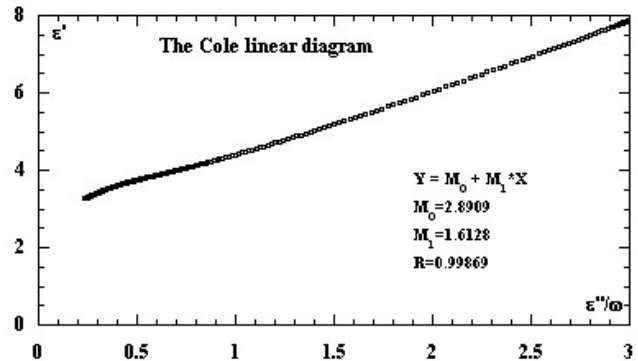


Fig. 3. The Cole linear diagram (pentanol-1+cyclohexane, 60%).

In Fig. 4 are presented the variations of the relaxation time τ_0 , which is gave by the straight line slope, for each concentration pentanol-1.

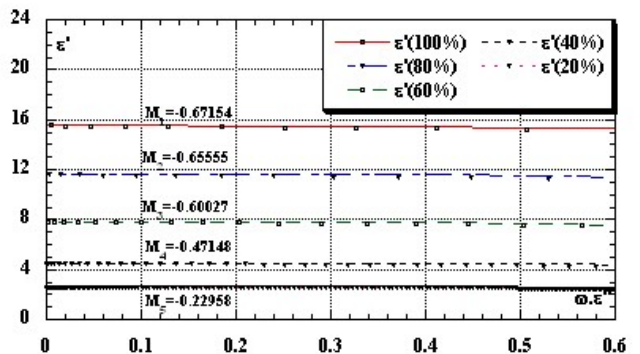


Fig. 4. The concentration dependence of the Cole linear diagram for pentanol-1.

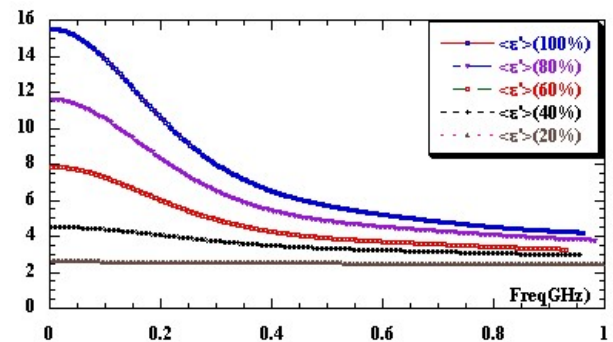


Fig. 5. The frequency variation of ϵ' .

In the Figs. 5, 6, 7 are presented the frequency variations of ϵ' and ϵ'' and also the diagram Argand for multiply concentration in study.

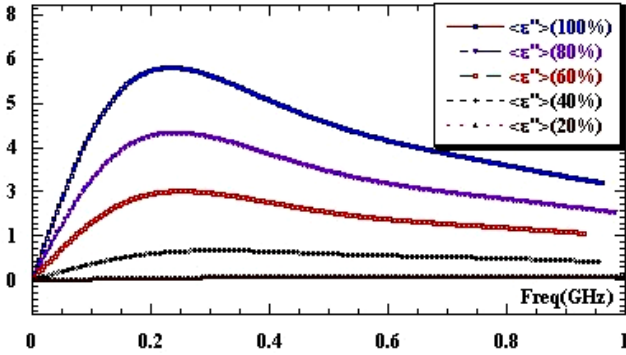


Fig. 6. The frequency variation of ϵ'' .

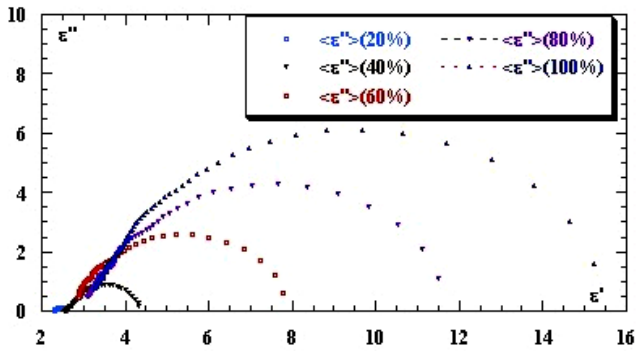


Fig. 7. The Argand diagram (pentanol 1+cyclohexane).

Supplementary we consider that an analysis of the variation of losses tangent is opportune

$$tg\delta = \frac{\epsilon''(\omega)}{\epsilon'(\omega)} \quad (2)$$

and also of the dielectric increment $\Delta\epsilon = \epsilon_S - \epsilon_\infty$. In the Fig. 8 is presented the frequency dependence of the loss tangent $tg\delta$ and in Fig. 9 it is shown the concentration variation of the dielectric increment $\Delta\epsilon$ for the polar component. Here we estimate that the variation of the dielectric increment is sensible linear with concentration above the concentrations of 40% for the polar component. We consider two regions: first one is localize at low concentration under 30% and the second one is found above the concentrations of 30% for the polar component (PT).

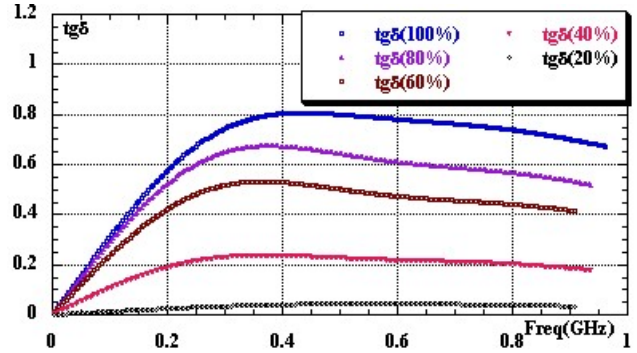


Fig. 8. The frequency variation of the losses tangent.

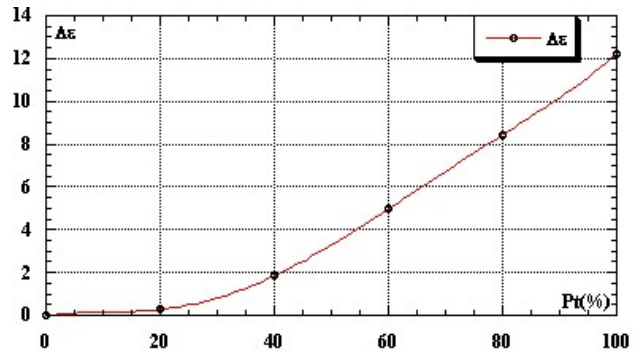


Fig. 9. The concentration variation of the dielectric increment ($\epsilon_S - \epsilon_\infty$).

The ensemble of results obtained allows the realization of a comparative study for the dielectric mixture usually laws. Next are verified some theoretical models: direct and inverse Wiener laws, Lichtenecker law and polar-non-polar model presented in the Table 2.

The direct Wiener (W.U.) model:

$$(\epsilon_S)_{W.U} = (\epsilon_S)_{PT} \cdot x + (1-x)(\epsilon_S)_C \quad (4)$$

$$\Delta\epsilon_S^E = (\epsilon_S)_{exp} - (\epsilon_S)_{theor} \quad (5)$$

The inverse Wiener (W.L.) Model:

$$\frac{1}{(\epsilon_S)_{W.L}} = \frac{1}{\frac{x}{(\epsilon_S)_{PT}} + \frac{1-x}{(\epsilon_S)_C}} \quad (6)$$

The Lichtenecker (L.) model:

$$(\epsilon_S)_{teor} = (\epsilon_S)_{PT}^x \cdot (\epsilon_S)_C^{1-x} \quad (7)$$

The polar-non-polar (p-np) model:

$$(\epsilon)_{teor} = x \cdot (\epsilon_S)_{PT} \quad (8)$$

where ϵ_S is the dielectric constant for these models; x - is the concentration of the pentanol and $\Delta\epsilon_S^E$ represents the dielectric excess constant.

Table 2. The comparative study for the dielectric mixture usually laws.

PT(%)	$(\epsilon_S)_{exp}$	$(\epsilon_S)_t$				$(\Delta\epsilon_S)^E$			
		W.U.	W.L.	L.	p-np	W.U.	W.L.	L.	p-np
100	15.52	15.52	15.52	15.51	15.52	0.00	0.00	0.01	0.00
80	11.59	12.82	6.63	10.31	12.42	-1.23	4.95	1.28	-0.82
60	7.85	10.12	4.21	6.85	9.31	-2.26	3.63	0.99	-1.46
40	4.51	7.42	3.09	4.56	6.12	-2.90	1.42	-0.04	-1.69
20	2.06	4.71	2.44	3.03	3.10	-2.11	0.16	-0.42	-0.49
0	2.01	2.01	2.01	2.01	0.00	0.00	0.00	0.001	2.016

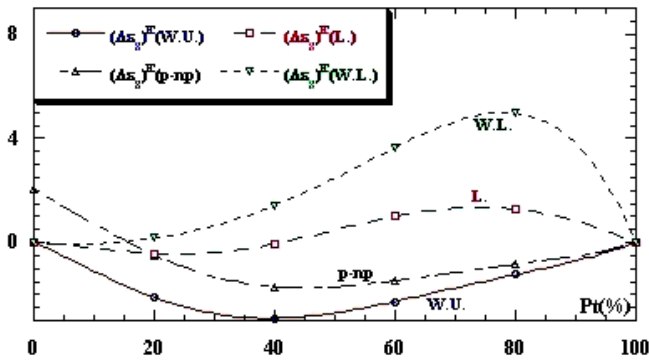
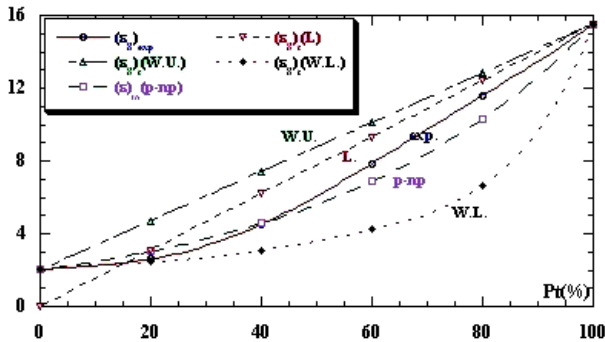

 Fig. 10. The concentration dependence of the dielectric increment $(\epsilon_S - \epsilon_\infty)$.

 Fig. 11. The concentration dependence of the static permittivity ϵ_S .

Fig. 10 resumes the dielectric behavior of the excess constant for different models. In this graphic we estimate two analogies: in one hand is present a significant correlation between the results deduced with the Lichtenecker law and with the inverse Wiener law and on the other hand the direct Wiener model and the polar-non-polar model present a similarity for big concentrations of the polar component. We observe that for a concentration of 40% (PT) the minimum of the (W.U.) and (p-np) dependences coincides with the modulation point for the (L) and (W.L.) models.

The application of the multiply reflections method in the study of the dielectric mixtures as titanate calcium-

epoxy-resin (TCA/RE) permits the analysis of the dielectric behavior at low and high frequencies.

At low frequency (L.F), the scope of the work is to present the study of the ohmic behavior of the mixture.

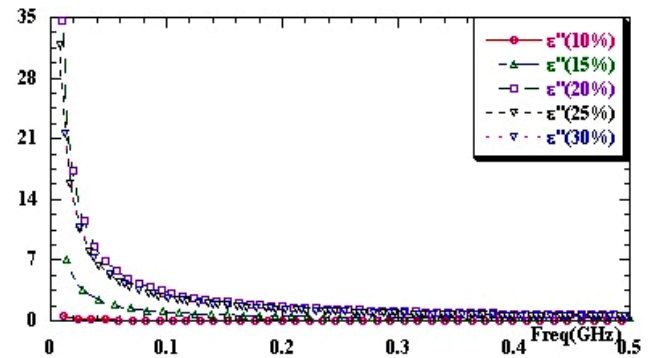
The complex dielectric constant is given by [11]:

$$\epsilon^* = \epsilon_\infty + \frac{\epsilon_S - \epsilon_\infty}{1 + j\omega\tau} - j \frac{\sigma_S}{\omega\epsilon_0} \quad (9)$$

where the influence of the third term is decisive in low frequency region

$$\epsilon'' = \frac{\sigma_S}{\omega\epsilon_0} \quad (10)$$

The last relations proves that the dielectric mixture has conductive behavior at low frequencies.


 Fig. 12. The frequency dependence of ϵ'' for the TCA/RE (L.F) mixture.

In Fig. 12 we present a comparative form of the ϵ''_{LF} profile for different concentrations. We can observe that if the titanate concentration increases result an evident increasing of the static conductivity contribution in the imaginary component of the dielectric permittivity.

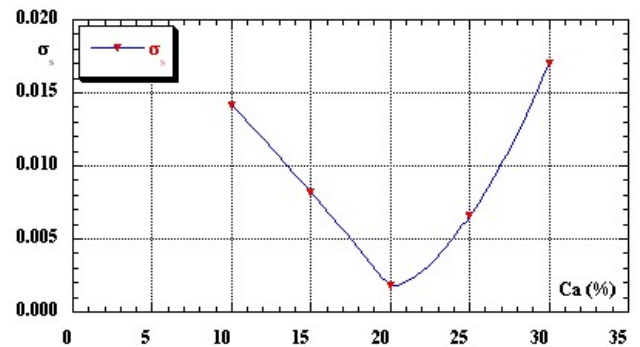


Fig. 13. The concentration dependence of the static conductivity for TCA/RE.

In Fig. 13 we observe that the minimum of the static conductivity $\sigma_S = 0.370 \times 10^{-2} (\Omega \cdot m)^{-1}$ is obtained for an approximately concentration of 20% of Calcium titanate. This minimum indicates that two different ohmic behaviors are presented.

In dispersion spectrum region at high frequencies (H.F) is possible the observation of the resonance phenomenon due to the effect length of the sample or due to the own resonance effect of the dielectric which is express by [12]

$$(f_R)_T = p \frac{c}{\sqrt{\epsilon}} \cdot \frac{1}{2e} \quad (11)$$

where p is an entire number, c - is the light velocity, e - is the diameter of the sample and ϵ - is the dielectric constant.

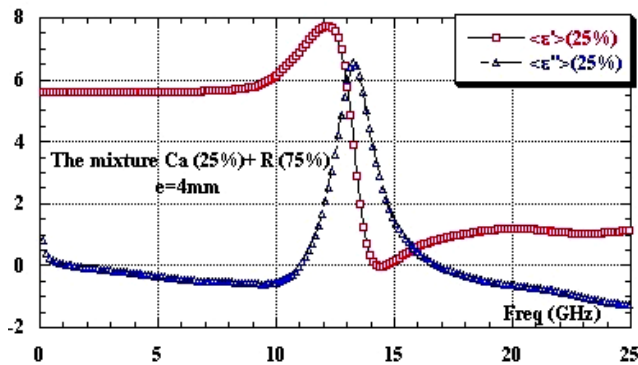


Fig. 14. The frequency dependence of the total complex permittivity ϵ^* for the mixture TCA/RE(25%).

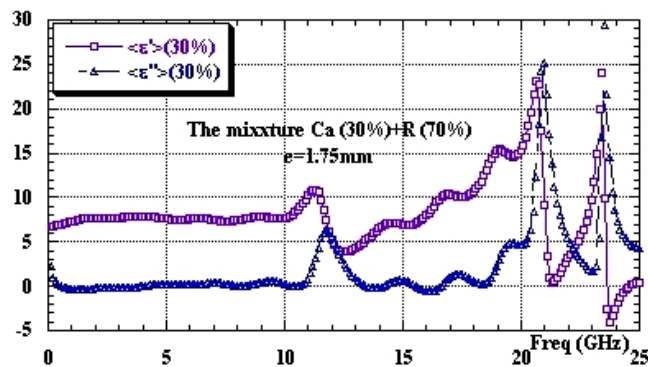


Fig. 15. The frequency dependence of the total complex permittivity ϵ^* for the mixture TCA/RE(30%).

Table 3. The experimental results the TCA/RE mixtures at high frequencies.

TCa (%)	E (mm)	ϵ_S	$(f_R)_{exp}$ (GHz)	$(f_R)_t$ (GHz)
10	3.25	3.925	19.02	20.17
15	3.35	5.504	15.83	18.65
20	3.85	5.984	14.01	15.75
25	4.00	7.192	12.55	13.46
30	1.75	8.804	11.64	-

In Fig. 15 we observe that the resonance first order frequency is 11.64GHz and that the second order

resonance is presented at 21GHz. For Ca titanate (30% concentration) we ignored the theoretic value of the resonance frequency because the sample diameter is very small and so is introduced an appreciable error which do not permit to effectuate an adequate correlation. The extrapolation of results confirms the coincidence of the values for the concentration of 30%.

As a final result we presented in Table 3 the numerical values of the static constant ϵ_S , of the resonance frequency f_R and also the plot representations of these

values (Fig. 16).

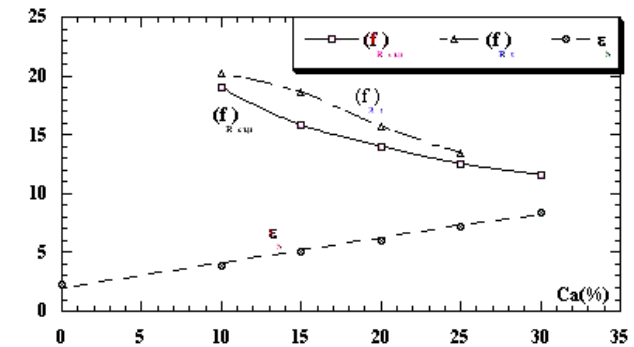


Fig. 16. The concentration variation of ϵ_S and f_R for the mixture TCA/RE(HF).

It is necessary to take in account that the samples must have the same order diameters in order to facilitate the results treatment and also to provide a very good correlation between the theoretical dates and the experimental dates.

Also it is necessary to observe the linear evolution of the static constant when the titanate concentration increases and the decreasing of the resonance frequency.

4. Conclusions

1. The complex dielectric permittivity of several pentanol-1+cyclohexane and calcium titanate+epoxy-resin composites for different concentrations has been measured.

2. We estimate two analogies: in one hand is present a significant correlation between the results deduced with the Lichtenecker law and with the inverse Wiener law and on the other hand the direct Wiener model and the polar-non-polar model present a similarity for big concentrations of the polar component. We observe that for a concentration of 40% (PT) the minimum of the (W.U.) and (p-np) dependences coincides with the modulation point for the (L) and (W.L.) models.

3. The Time Domain Reflectometry technique also makes possible to determine the static electric conductivity of each sample (titanate+ epoxy-resin) for low frequency. From this it is possible to estimate calcium titanate static conductivity and, thereafter, to separate the dielectric and the ohmic contributions to the imaginary component of the complex titanate calcium permittivity.

4. For high frequencies appear relaxations or resonance processes.

5. It is necessary to take in account that the samples must have the same order diameters in order to facilitate the results treatment and also to provide a very good correlation between the theoretical dates and the experimental dates.

6. This method is limited by the instrumental conditions due to the of the bandwidth used, by the presence of noise a high frequency and also by the presence of the errors due to the fact that the polar-non-polar mixture is very instable, having the tendency evaporation which modify the homogeneous state of the mixture.

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