

## Influence of Structure on Curie Weiss Constant of [(Pb<sub>1-x</sub> Sr<sub>x</sub>)<sub>1-1.5z</sub> La<sub>z</sub>]TiO<sub>3</sub> Ceramics

G. A. Gamal, M. K. Gergs and M. A. Massaud

*Physics Department, Faculty of Science (Qena), South Valley  
University, Egypt.*

*Theoretical studies have been carried out to reveal the proper concentration of Sr and La-content which provide the samples of [(Pb<sub>1-x</sub> Sr<sub>x</sub>)<sub>1-1.5z</sub> La<sub>z</sub>] TiO<sub>3</sub> ceramics with optimum ferroelectric properties and to find an interpretation for the behavior of the Curie Weiss constants ( $C$ ,  $C^+$ ). In this work, three types of samples were considered according to the concentration of La-content. The frequency of electric field used in dielectric measurements equal 1KHz. From the relation between Curie Weiss constants and Sr content, the samples of first type ( $z = 3$  mole % La) exhibited two minimum values for ( $C$ ,  $C^+$ ) at  $x = 20$  and  $40$  mole %Sr. The samples of second type ( $z = 6$  mole %La) showed a similar behavior but the minimum values for ( $C$ ,  $C^+$ ) at  $x = 25$  and  $35$  mole %Sr. The samples of third type ( $z = 9$  mole %La) were characterized with only one minimum value for ( $C$ ,  $C^+$ ) at  $x = 30$  mole %Sr. In this paper, we concluded that at a constant concentration of Sr content (26mol%) or Pb content (60.5mol%) both values of Curie- Weiss constants ( $C$ ,  $C^+$ ) and  $\epsilon_{max}$  increase with increasing of La content. This remark is a unique phenomenon for the behavior of Curie-Weiss constants ( $C$ ,  $C^+$ ). The interpretation of the behavior of Curie- Weiss constants ( $C$ ,  $C^+$ ) for the samples is attributed to the types of unit cells which are dominated in structure.*

### 1. Introduction:

A very important group of ferroelectric class is that known as ABO<sub>3</sub> perovskites. The ferroelectric properties in perovskites structure have been the subject of many experimental and theoretical investigations. In recent years [1]PbTiO<sub>3</sub> ceramics have attracted much attention due to their high Curie temperature (763 K) and low dielectric constant which make them more attractive for high- temperature and high-frequency transducer applications. It is also known that pure lead titanate ceramics is too fragile [2], since its crystal anisotropy generates high internal stresses which make it break during cooling.

However, dense modified  $\text{PbTiO}_3$  ceramics were obtained by lead substitution with small amounts of dopants which enhance their mechanical, dielectric and piezoelectric properties. Strontium titanate  $\text{SrTiO}_3$  has been one of the most popular materials [3] since the discovery of its dielectric properties. They have been widely used to fabricate some electronic ceramic components, such as grain boundary layer capacitors, sensors and so on. The Curie temperature ( $T_c$ ) of pure  $\text{SrTiO}_3$  is about 44 K, which can shift to the higher temperature by  $\text{Ba}^{2+}$  or  $\text{Pb}^{2+}$  substituting for  $\text{Sr}^{2+}$ . In lanthanum containing lead titanate [4], the  $\text{La}^{3+}$  ions occupy  $\text{Pb}^{2+}$  sites and generate vacancies ( ) in the cation lattice of  $(\text{Pb}_{1-1.5x} \text{La}_x \text{ }_{0.5x})\text{TiO}_3$  ceramics. With increasing  $\text{La}^{3+}$  content the transition temperature  $T_c$  decreases linearly. Experimental and theoretical studies on some ferroelectric properties of ceramics with previous chemical formula have been already published [5]. It was observed that the behaviour of dielectric peak  $\epsilon_{\text{max}}$  is similar to the behaviour of single vacancies at the same La-content. The highest value of  $\epsilon_{\text{max}}$  at  $x=0.2$  corresponds the maximum number of single vacancies i.e. both the dielectric peak  $\epsilon_{\text{max}}$  and the number of single vacancies are proportional to La content until the value 20 mol %. L. Wu *et al.* [6] reported some data about the perovskite structure with general formula  $\text{ABO}_3$  where the A-site of vacancies reduces the local stress in the domains that undergo domain switching. Tura *et al.* [7] showed in his studies that the grain size increases with decreasing the internal stress. Also, the domain width, as reported by King *et al.* [8], is proportional to the grain size. It is observed in many literatures [9-15] that increasing of the grain size is associated with increasing the dielectric peak  $\epsilon_{\text{max}}$ . Binomial distribution function is an appropriate equation for the theoretical basis of the present paper. It was examined by FTIR-spectroscopy on two systems of ceramics, namely  $(\text{Pb}_{1-1.5x} \text{La}_x \text{ }_{0.5x})\text{TiO}_3$  [16] and  $(\text{Pb}_{1-x} \text{Ca}_x)\text{TiO}_3$  [17].

The aim of present investigation is summarized in two points. The first one is related to the study of the Curie-Weiss constant and their related features, such as the quality of ceramics, taking into consideration the suitable content of Sr- and La-ions which provide the samples with the optimum ferroelectric properties. The second is related to binomial distribution model of nine stoichiometric which was realized for the incorporation of Ca-ions at Pb-site in  $(\text{Pb,Ca})\text{TiO}_3$  system and the attempt to apply it on samples of this paper. This is realized through the calculation of phase transition temperature on the basis of the former model and comparing the results with the experimental values.

## 2. Experimental Procedures:

Specimens of the general formula  $(\text{Pb}_{1-x} \text{Sr}_x)_{1-1.5z} \text{La}_z \text{ }_{0.5z})\text{TiO}_3$  were prepared according to the procedures of the usual firing technique. The starting

materials (99.9% purity PbO, SrCO<sub>3</sub>, TiO<sub>2</sub> and La<sub>2</sub>O<sub>3</sub>) in corresponding stoichiometric ratios were homogenized and pressed into discs. The discs were then calcined at the range of temperature between 700 and 850 °C. This was done on the basis of the calibration line between the two calcined temperatures where one of them for PbTiO<sub>3</sub> - and the other for SrTiO<sub>3</sub> ceramics, for two hours. The calcined powder was pressed into discs. The discs were thereafter sintered at the temperature ranging between 1210 and 1250 °C, according to the prementioned technique but for two sintered temperatures, for 4 hours in an oxygen atmosphere. The samples are classified into three types according to the concentrations of La-contents. The values of z for the three types equal to 0.03, 0.06 and 0.09, respectively. The cell employed in the present work for the dielectric measurements is described in reference [18].

### 3. Results and Discussion:

#### 3.1. Dielectric Constant and Curie Weiss Constant:

The results of specimens used in data which were presently obtained by the authors in reference [17]. Fig. (1) shows the temperature dependence of the inverse dielectric constant  $\epsilon^{-1}$  for (Pb<sub>1-x</sub> Sr<sub>x</sub>)<sub>0.955</sub> La<sub>0.03</sub> TiO<sub>3</sub>-ceramics as a function of the Sr-content at frequency equals to 1kHz. This figure contains a set of curves corresponding to six Sr contents (x = 10, 20, 30, 40, 50 and 60 mole%). The inverse of the dielectric constant peak  $\epsilon_{\max}^{-1}$  shifts to lower temperatures on increasing the Sr content. The minimum values of  $\epsilon_{\max}^{-1}$  are found at x = 0.2 and 0.4 mol. The previous results are important for calculation of the Curie-Weiss constant which is, in turn, necessary for studying the quality of ceramics for ferroelectric properties. This was based on the fact that the relation between  $\epsilon^{-1}$  and T (K) is linear near the phase transition temperature and the inverse of its slop gives value of Curie- Weiss constant.

Figure (2) represents the relationship between the Curie-Weiss constant (C<sup>-</sup>, C<sup>+</sup>) and the variation in Sr content for (Pb<sub>1-x</sub>Sr<sub>x</sub>)<sub>0.955</sub>La<sub>0.03</sub> TiO<sub>3</sub>-ceramics. The Curie-Weiss constant decreases with increasing of Sr content to its minimum value at Sr content 20mol% Sr, but, when the Sr content increases more the value of Sr content which was mentioned before, the Curie-Weiss constant increases to the critical point on the curve at Sr content 30 mol%. With more increase of Sr content, the Curie-Weiss constant decreases to its minimum value at Sr content 40 mol%, then, the Curie-Weiss constant increases again with increasing of Sr content.

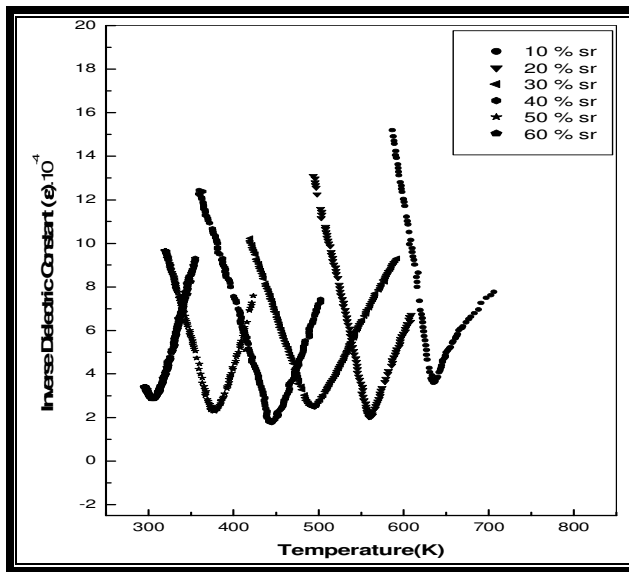


Fig.(1): The temperature dependence of the inverse dielectric constant  $\epsilon^{-1}$  for samples with structure formula  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.955}\text{La}_{0.03}\text{TiO}_3$  (the frequency of electric field equal to 1 kHz).

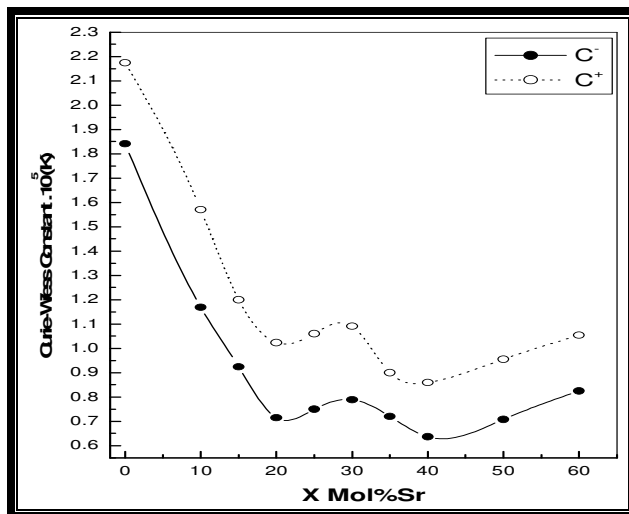
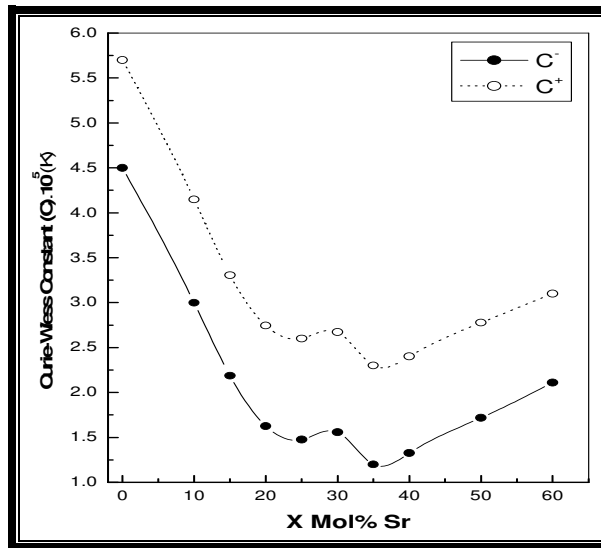


Fig.(2): A plot of Curie-Weiss constant  $C^-$  and  $C^+$  versus the Sr content (x) for samples with formula  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.955}\text{La}_{0.03}\text{TiO}_3$ .

The behaviour of  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.91}\text{La}_{0.06}\text{TiO}_3$ -ceramics about the temperature dependence of the inverse dielectric constant  $\epsilon^{-1}$  as a function of the Sr-content at 1KHz electric field is found to be similar to the behavior which is represented in Fig.(1). The minimum values of  $\epsilon_{\text{max}}^{-1}$  are existed at  $x = 0.25$  and  $0.35$  mole.

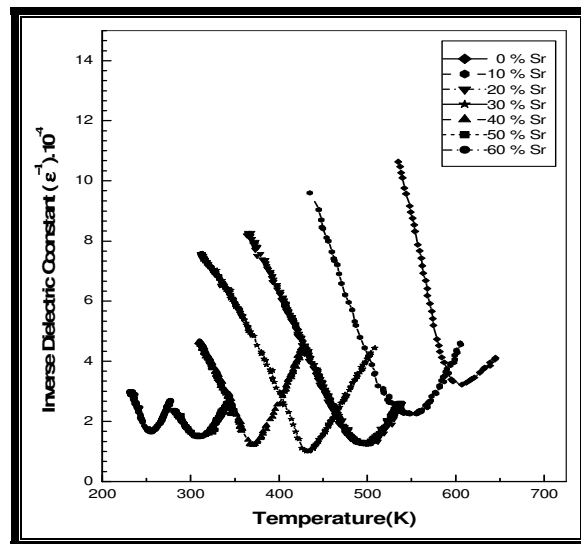
Figure (3) illustrates the relationship between the Curie-Weiss constant ( $C^-$ ,  $C^+$ ) and the variation in Sr content for  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.91}\text{La}_{0.06}\text{TiO}_3$ -ceramics. The two minimums of Curie Weiss constant are existed at Sr content 25 and 35 mol%.



**Fig.(3):** A plot of Curie –Weiss constant  $C^-$  and  $C^+$  verses the Sr content ( $x$ ) for samples with formula  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.91}\text{La}_{0.06}\text{TiO}_3$ .

Figure (4) shows the temperature dependence of the inverse dielectric constant  $\epsilon^{-1}$  and Fig.(5) depicts the relationship between the Curie-Weiss constant ( $C^-$ ,  $C^+$ ) and the variation in Sr-content for  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.865}\text{La}_{0.09}\text{TiO}_3$ -ceramics as a function of the Sr-content at frequency equals to 1 kHz. The inverse of the dielectric constant peak  $\epsilon_{\text{max}}^{-1}$  shifts to lower temperatures on increasing the Sr-content. The minimum value of  $\epsilon_{\text{max}}^{-1}$  is exist at  $x = 0.3$ . The Curie Weiss constant decreases to its minimum value at Sr-content 30 mol%. So, one can conclude the followings:

- 1- In the whole range for the variation of Sr-content the value of ( $C^+$ ) is higher than that of ( $C^-$ ) and the rate of decreasing for both values is more than the rate of increasing.
- 2- The variation of the value ( $C^+$ ) as function to Sr-content is similar to that for ( $C^-$ ).
- 3- The samples with the Sr-content at  $x= 0.2$  and  $0.4$  mole are characterized by the lowest value of Curie-Weiss constants ( $C^-, C^+$ ), which are ( $0.7 \times 10^5$ ,  $1.2 \times 10^5$  K) and ( $5 \times 10^4$ ,  $9 \times 10^4$  K), respectively,
- 3- In the case of the second type ,the samples with the Sr content at  $x = 0.25$  and  $0.35$  mole are characterized with the lowest value of Curie-Weiss constants ( $C^-, C^+$ ) which are ( $1.3 \times 10^5$ ,  $1.5 \times 10^5$  K) and ( $1.1 \times 10^5$ ,  $1.44 \times 10^5$  K) respectively.
- 4- As to the samples of the third type, the sample with the Sr content at  $x= 0.3$  mole is characterized by the lowest value of Curie-Weiss constants ( $C^-, C^+$ ), which are equal to  $2.25 \times 10^5$  and  $2.63 \times 10^5$  K, respectively.



**Fig.(4):** The temperature dependence of the inverse dielectric constant  $\epsilon^{-1}$  for samples with structure formula  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.865}\text{La}_{0.09}\text{TiO}_3$  (the frequency of electric field equal to 1 kHz).

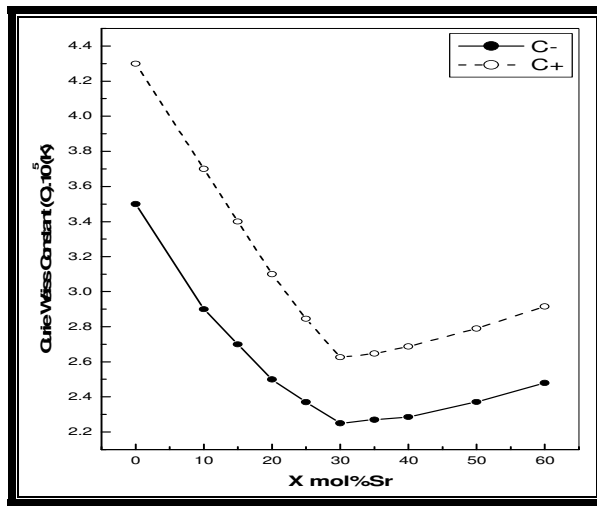


Fig.(5): A plot of Curie –Weiss constant  $C^-$  and  $C^+$  versus the Sr content (x) for samples with formula  $(Pb_{1-x}Sr_x)_{0.865}La_{0.09}TiO_3$ .

From the fitting relations between Curie-Weiss constants and the real concentration of Sr-ion or Pb-ion which are presented in Fig. (6) and (7) one can conclude that both contents of Sr- and Pb-ions for the minimum in the third type of samples are existed in the other type of samples (first and second minimum for Curie-Weiss constants respectively).

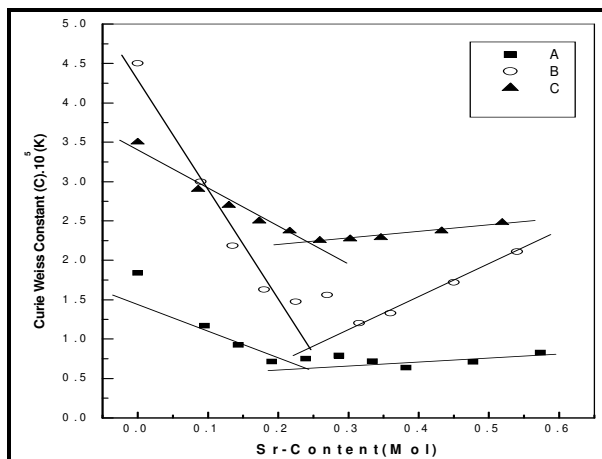
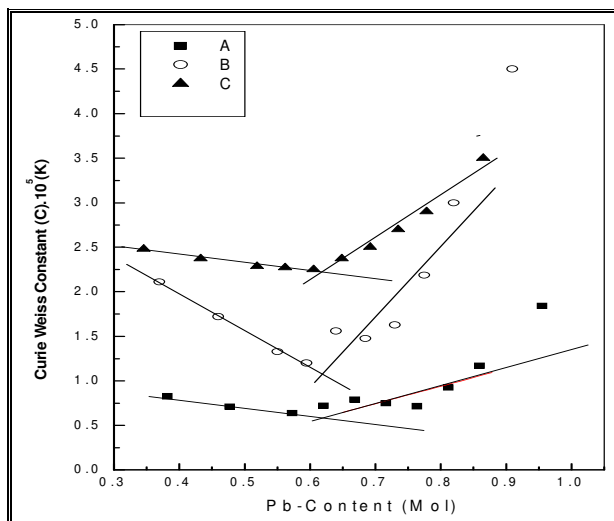


Fig.(6): A plot of Curie –Weiss constant  $C^-$  versus the Sr content(Y)for samples with formula  $(Pb_{1-x}Sr_x)_{1-1.5z}La_zTiO_3$ , where A,B and C for z equals to 0.03, 0.06 and 0.09, respectively, N.B:  $Y = x \cdot (1-1.5z)$ .



**Fig.(7):** A plot of Curie –Weiss constant  $C^-$  versus the Pb content(1-Y)for samples with formula  $(Pb_{1-x}Sr_x)_{1-1.5z}La_zTiO_3$ , where A,B and C for z equals to 0.03 , 0.06 and 0.09 ,respectively, **N.B:**  $Y= x \cdot (1-1.5z)$ .

Hence, the Sr content 26 mol % and Pb content 60.5mol % are responsible for the first and second anomaly behaviors, respectively.

The previous results which are related to dielectric peak  $\epsilon_{max}$  and Curie-Weiss constants which in turn corresponding to the anomalies peak in the three types of samples can be summarized in Fig. (8) and (9). Those figures represented the relationships between the La content and the both values of  $\epsilon_{max}$  and Curie-Weiss constant ( $C^-$ ) for the Sr-content 26mol % and Pb content 60.5 mol % respectively. The value of dielectric constant peak  $\epsilon_{max}$  is proportional to the number of single vacancies per unit volume according to the reference [5]. From the figures one can conclude that both values of Curie-Weiss constants ( $C^-$ ) and  $\epsilon_{max}$  increases with increasing of La content. This result is unusual behavior for the Curie Weiss constants with respect to the dielectric peak  $\epsilon_{max}$ . The following discussion is a complementary part to shed some light on this unique phenomenon.



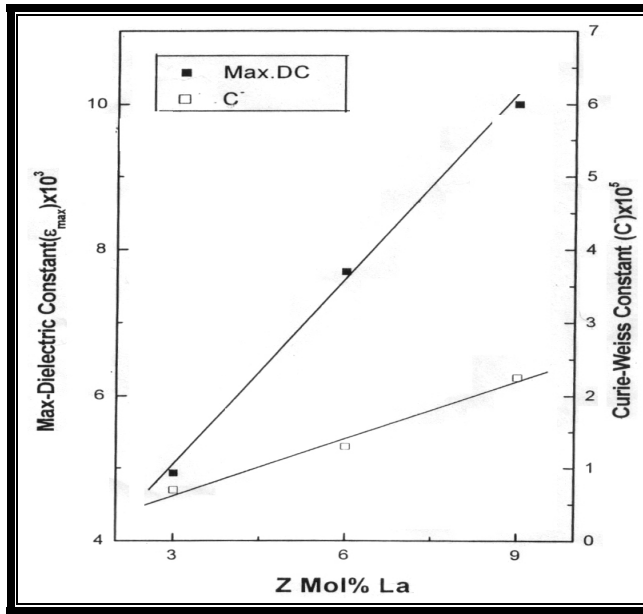


Fig.(8): A plot of both  $\epsilon_{max}$  and Curie -Weiss constant  $C$  versus La content at Sr content equals to 26mol % .

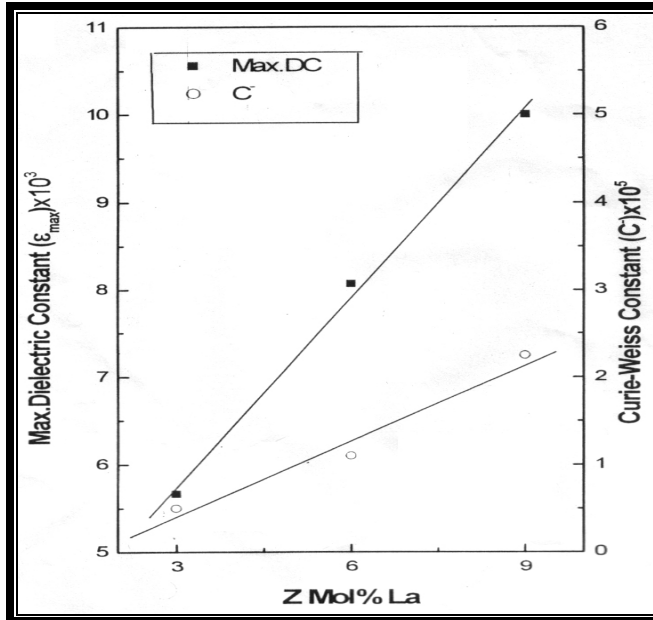


Fig.(9): A plot of both  $\epsilon_{max}$  and Curie -Weiss constant  $C$  versus La content at Pb content equals to 60.5mol % .

#### 4. Theoretical Estimation of the Phase Transition Temperature:

The following equation is used for calculation of the phase transition temperature. It is an empirical model and can be given in the following form:

$$T_c(x) = \sum_{\alpha=1}^{\alpha=3} \Delta T_{ca}^{(x)} \quad (1)$$

$$T_c(x) = \Delta T_{c1}(x) + \Delta T_{c2}(x) + \Delta T_{c3}(x)$$

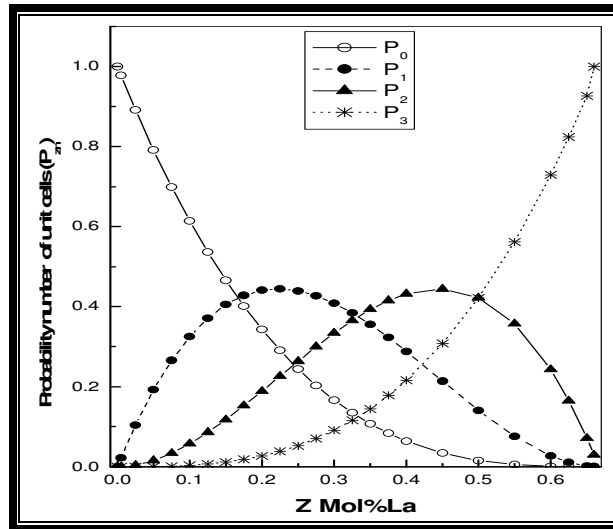
where:

$$\Delta T_{ca}(x) = R_{\alpha} P_{na} \{ [T_c(\text{PbTiO}_3) \cdot (M-N) + T_c(\text{SrTiO}_3) \cdot N] / 8 \} \quad (2)$$

$$P_{na} = x^N (1-x)^{M-N} \binom{M}{N} \quad (3)$$

$$\binom{M}{N} = M! / [(M-N)! (N)!] \quad (4)$$

$P_{na}$  is the probability number which is described by binomial distribution a function in equation (3), the probability number  $R_{\alpha}$  is function of La-content and types of unit cell which represented in Fig.(10), where  $R_1$ ,  $R_2$  and  $R_3$  are corresponding to the type of unit cells  $[(8-N)\text{Pb} + \text{NSr}]$ ,  $[(5-N)\text{Pb} + \text{NSr} + 2\text{La}^+]$  and  $[(2-N)\text{Pb} + \text{NSr} + 4\text{La}^+ + 2]$ , respectively.

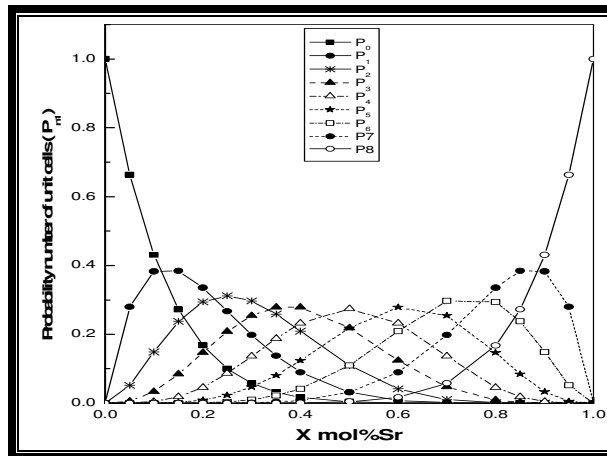


**Fig.(10):** Represent the probability number for the different types of unit cells ( $P_{zn}$ ) versus the La content ( $z$ ) for the sample with formula  $(\text{Pb}_{1-x}\text{Sr}_x)_{1-1.5z}\text{La}_z\text{TiO}_3$ . Where  $P_{zn}$  with  $n=0,1,2$  and 3 represents, respectively, the unit cell which contain  $[(8-N)\text{Pb} + \text{NSr}]$ ,  $[(5-N)\text{Pb} + \text{NSr} + 2\text{La}^+]$ ,  $[(2-N)\text{Pb} + \text{NSr} + 4\text{La}^+ + 2]$  and  $8\text{La}$ .

So, the conditions for  $\Delta T_{c1}(x)$  are:  $M=8$ ,  $N=0, 1, 2, \dots, 8$  and  $P_{n1} = P_{01}, P_{11}, P_{21}, P_{31}, P_{41}, P_{51}, P_{61}, P_{71}, P_{81}$  corresponding the following unit cells, (8Pb), (7Pb, 1Sr), (6Pb, 2Sr), (5Pb, 3Sr), (4Pb, 4Sr), (3Pb, 5Sr), (2Pb, 6Sr), (1Pb, 7Sr), (8Sr) respectively which are represented in Fig.(11).

The conditions for  $\Delta T_{c2}(x)$  are  $M = 5$ ,  $N = 0, 1, 2, 3, 4$  and  $5$  and  $P_{n2} = P_{02}, P_{12}, P_{22}, P_{32}, P_{42}, P_{52}$  corresponding the following unit cells, (5Pb, 2La, ), (4Pb, 1Sr, 2La, ), (3Pb, 2Sr, 2La, ), (2Pb, 3Sr, 2La, ), (1Pb, 4Sr, 2La, ), (5Sr, 2La, ) as indicated in Fig.(12).

The conditions for  $\Delta T_{c3}(x)$  are  $M = 2$ ,  $N = 0, 1, 2$ . and  $P_{n3} = P_{03}, P_{13}, P_{23}$  corresponding the following unit cells (2Pb, 4La, 2 ), (1Pb, 1Sr, 4La, 2 ), (2Sr, 4La, 2 ) as plotted in Fig(13). The values of  $T_c$  for  $\text{PbTiO}_3$  and  $\text{SrTiO}_3$  are 763 and 44K, respectively, according to the references (1, 3).



**Fig.(11):** Represent the probability number for the different types of unit cells ( $P_{n1}$ ) versus the Sr content ( $x$ ) for the sample with formula  $(\text{Pb}_{1-x}\text{Sr}_x)\text{TiO}_3$ . Where  $P_{n1}$  with  $n= 0,1,2, 3, 4, 5, 6, 7$  and  $8$  represents, respectively, the unit cell which contain [8Pb], [7Pb,Sr], [6Pb, 2Sr], [5Pb,3Sr], [4Pb,4Sr], [3Pb,5Sr], [2Pb,6Sr], [Pb,7Sr] and 8Sr.

##### 5. Comparison between theoretical and experimental values of phase transition temperature for $(\text{Pb}_{1-x}\text{Sr}_x)_{1-1.5z}\text{La}_z\text{TiO}_3$ -system :

For  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.955}\text{La}_{0.03}\text{TiO}_3$ -system, the experimental values of  $T_c$  (exp.) and the corresponding calculated values of  $T_c$  (cal.) by the Eq.(1) are represented in Fig.(14) while in Fig.(15) it is for  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.865}\text{La}_{0.09}\text{TiO}_3$ -system. In the two figures, the both values  $T_c$  (exp.) and  $T_c$  (cal.) of the phase transition temperature are decreasing linearly, in accordance behavior, with increasing the Sr content. This leads to the following:

- 1- The results support the binomial distribution function, which are presented in Fig. (10) for  $(\text{Pb}_{1-x}\text{Sr}_x)_{1-1.5z}\text{La}_{z-0.5z}\text{TiO}_3$ –system.
- 2- The results clearly confirm that the binomial distribution function represented in Fig (10) for the pervious formula is not enough for describing the positions of Pb-, Sr-, La-ions and the vacancies in unit cells but another distributions are needed. This is exactly presented graphically in Figs. (11), (12) and (13).

#### 6. The interpretation of the Curie Weiss constant on the basis of binomial distribution function for (Pb, Sr, La, )TiO<sub>3</sub>– systems:

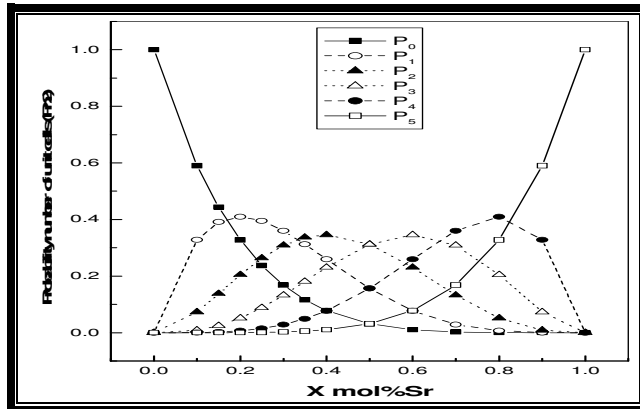
The previous results as reported in Fig. (14) and (15) confirm the agreement between the experimental values of the phase transition temperature  $T_c$  (exp.) and the values of  $T_c$  (cal.) which were calculated from eq.(1). Since, the basis of the eq.(1) is the binomial distribution function which are represented in Figs. (10), (11), (12) and (13). Hence, it is essential to interpret the anomalies which are manifested in the behaviors of Curie-Weiss constants for  $[(\text{Pb}_{1-x}\text{Sr}_x)_{1-1.5z}\text{La}_{z-0.5z}]\text{TiO}_3$  ceramics in terms of binomial distribution function.

The comparison between the behaviours of Curie-Weiss constants and the unit cells in binomial distribution is a helpful method for this purpose. Fig. (16) is a plot of both Curie–Weiss constant ( $C^*$ ) and ( $P_1, P_2$ ) versus the Sr-content ( $x$ ) for  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.955}\text{La}_{0.03-0.015}\text{TiO}_3$  –systems, where  $P_1$  is the summation of the probability number for the average unit cells of the types  $\{[(7\text{Pb}, 1\text{Sr})+(6\text{Pb}, 2\text{Sr})] / 2\}$  and unit cell of type  $(4\text{Pb}, 1\text{Sr}, 2\text{La}, )$ . Also  $P_2$  is the summation probability number for the two types of unit cell  $[(5\text{Pb}, 3\text{Sr})+(3\text{Pb}, 2\text{Sr}, 2\text{La}, )]$ .

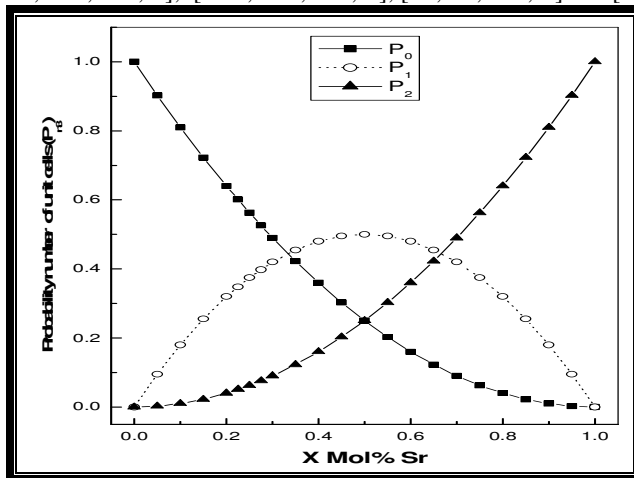
It is clear from this figure that the curve which represents the variation of the value of  $C^*$  has two minimums. The first one lies at Sr-content 20 mol% and the second at Sr-content 40 mol%. It is also evident that the curves which represent  $P_1$  and  $P_2$  show dissimilar behavior as happened in the curve which represented  $C^*$ . The maximum value of  $P_1$  corresponds to minimum value of  $C^*$  at Sr-content 20 mol% and the maximum value of  $P_2$  corresponds to minimum value of  $C^*$  at Sr-content 40 mol%. The point for the two curves ( $P_1, P_2$ ) corresponds to the critical point on the curve of  $C^*$  at at Sr-content 30 mol%.

Figure (17) represents a plot of both  $C^*$  and  $P_3$  versus the Sr-content ( $x$ ) for  $(\text{Pb}_{1-x}\text{Sr}_x)_{0.865}\text{La}_{0.09-0.045}\text{TiO}_3$  – systems, where  $P_3$  is the summation of ( $P_1, P_2$ ) i.e. the summation of probability number for the average unit cells of types  $\{[(7\text{Pb}, 1\text{Sr})+(6\text{Pb}, 2\text{Sr})] / 2\}$ , unit cell of type  $(4\text{Pb}, 1\text{Sr}, 2\text{La}, )$  and the

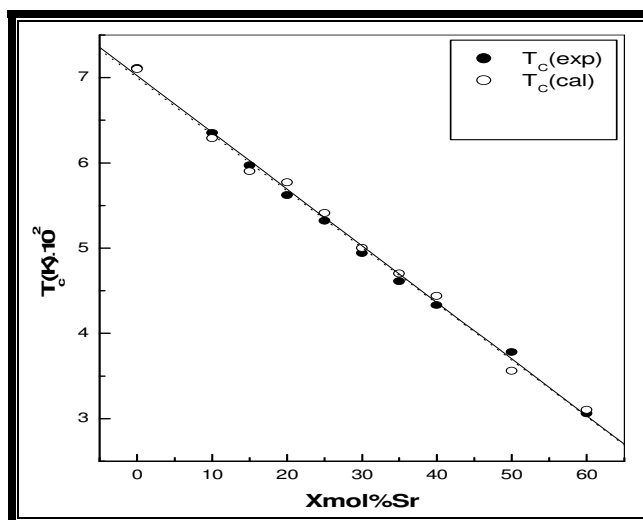
two unit cells of types  $[(5\text{Pb},3\text{Sr})+(3\text{Pb},2\text{Sr},2\text{La})]$ . It can also be seen that the curve which represents  $P_3$  takes the dissimilar behavior as in the curve which represented  $C$ . The maximum value of  $P_3$  corresponds to minimum value of  $C$  at Sr-content 30 mol%.



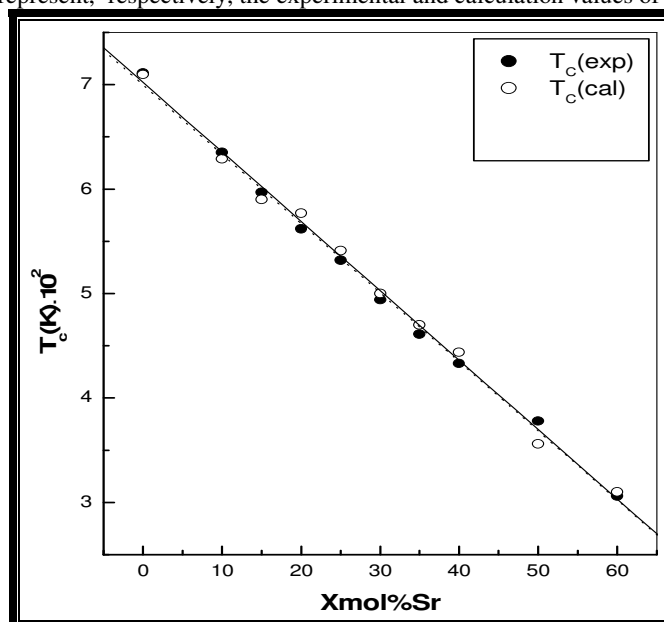
**Fig.(12):** Represent the probability number for the different types of unit cells ( $P_{n2}$ ) versus the Sr content ( $x$ ) for the sample with formula  $[(\text{Pb}_{1-x}\text{Sr}_x)_{0.625}\text{La}_{0.25}\text{TiO}_3]$ . Where  $P_{n2}$  with  $n= 0,1,2,3,4$  and  $5$  represents, respectively, the unit cell which contain  $[5\text{Pb},2\text{La}]$ ,  $[4\text{Pb},\text{Sr},2\text{La}]$ ,  $[3\text{Pb},2\text{Sr},2\text{La}]$ ,  $[2\text{Pb},3\text{Sr},2\text{La}]$ ,  $[\text{Pb},4\text{Sr},2\text{La}]$  and  $[5\text{Sr},2\text{La}]$ .



**Fig.(13):** Represent the probability number for the different types of unit cells ( $P_{n3}$ ) versus Sr content ( $x$ ) for the sample with formula  $[(\text{Pb}_{1-x}\text{Sr}_x)_{0.25}\text{La}_{0.5}\text{TiO}_3]$ , where  $P_{n3}$  with  $n= 0,1,2$  and  $3$  represents, respectively, the unit cell which contain  $[2\text{Pb},4\text{La}]$ ,  $[\text{Pb},\text{Sr},4\text{La},2]$ ,  $[2\text{Sr},4\text{La},2]$ .



**Fig.(14):** A plot of the phase Transition Temperature  $T_C$  versus the Sr content ( $x$ ) for samples with formula  $(Pb_{1-x}Sr_x)_{0.955}La_{0.03}TiO_3$ , since  $T_C$ (exp.) and  $T_C$ (cal.), represent, respectively, the experimental and calculation values of  $T_C$ .



**Fig.(15):** A plot of the phase Transition Temperature  $T_C$  versus the Sr content ( $x$ ) for samples with formula  $(Pb_{1-x}Sr_x)_{0.865}La_{0.09}TiO_3$ , since  $T_C$ (exp.) and  $T_C$ (cal.), represent, respectively, the experimental and calculation values of  $T_C$ .

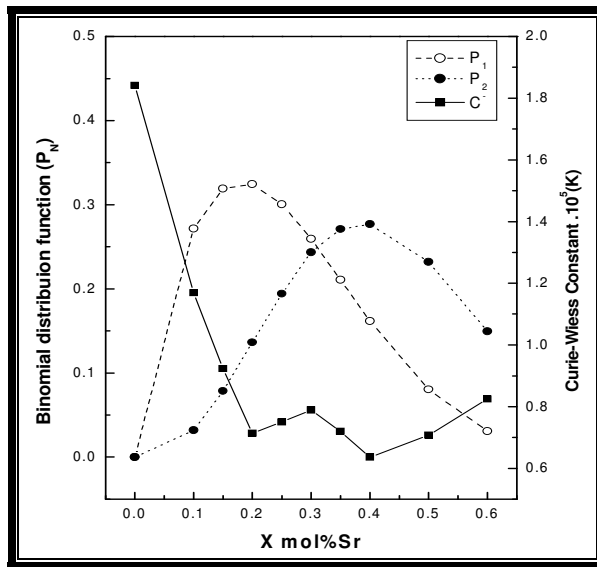


Fig.(16): A plot of both Curie –Weiss constant  $C^-$  and  $(P_1, P_2)$  verses the Sr content (x) for samples with formula  $(Pb_{1-x}Sr_x)_{0.955}La_{0.03}TiO_{3.015}$ .

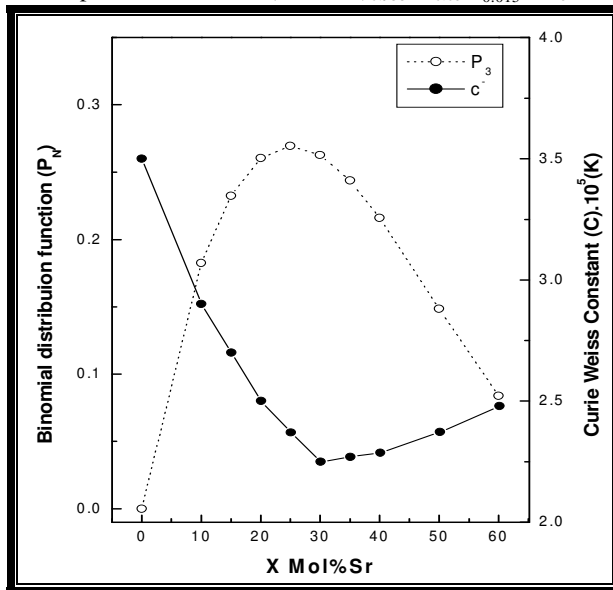


Fig.(17): A plot of both Curie –Weiss constant  $C^-$  and  $P_3$  verses the Sr content (x) for samples with formula  $(Pb_{1-x}Sr_x)_{0.865}La_{0.09}TiO_{3.045}$ .

For better understanding of the relationship between Curie-Weiss constants and the types of unit cells, it is necessary to consider the effect of unit cells of the types (4Pb,1Sr,2La, ) and (3Pb,2Sr,2La, ), which is resulting from the concentration of La-ions equals to 3mol % is negligible. This discussion means that the unit cells of types (7Pb+1Sr), (6Pb+2Sr) and (5Pb+3Sr) are only effective for the behavior of Curie –Weiss constants in pervious of La content but with increasing the La content to 9 mol % the opposite case must be exist and the influence of unit cells of types (4Pb,1Sr,2La, ) and (3Pb,2Sr,2La, ) is not negligible.

## 7. Conclusions:

- 1- The investigation reveals the existence of five distinguished samples. The formula of these samples are  $(\text{Pb}_{0.695} \text{Sr}_{0.26} \text{La}_{0.03})\text{TiO}_3$ ,  $(\text{Pb}_{0.605} \text{Sr}_{0.35} \text{La}_{0.03})\text{TiO}_3$ ,  $(\text{Pb}_{0.65} \text{Sr}_{0.26} \text{La}_{0.06})\text{TiO}_3$ ,  $(\text{Pb}_{0.605} \text{Sr}_{0.305} \text{La}_{0.06})\text{TiO}_3$  and  $(\text{Pb}_{0.605} \text{Sr}_{0.26} \text{La}_{0.09})\text{TiO}_3$ .
- 2- The number of single vacancies which are created from the substitution of La –ions in Pb- or Sr- site is effective for increasing the anomalies dielectric peak in the five distinguished samples.
- 3- According to the value of Curie Weiss constants the sample of formula  $\text{Pb}_{0.605} \text{Sr}_{0.35} \text{La}_{0.03} \text{TiO}_3$  possesses, relatively, the highest value of quality for ferroelectric properties and applications. It is evident that the unit cells of type (5Pb+3Sr) are only dominated in the sample of former formula and this type of unit cell acts as the role of single domain in ferroelectric crystals. The opposite behavior can be seen in the sample of formula  $(\text{Pb}_{0.605} \text{Sr}_{0.26} \text{La}_{0.09} \text{TiO}_3)$ , since, the unit cell of types, (7Pb+1Sr), (6Pb+2Sr), (5Pb+3Sr), (4Pb+1Sr+2La+ ) and (3Pb+2Sr+4La+2 ) are dominated in its structure and act as the role of many domains in ferroelectric crystals.

## Acknowledgment

The author is greatly indebted to Prof. Dr. M. Amin, Physics Department, Cairo University, Egypt, for his interest during the preparation of the manuscript.

## References

1. S. Y. Chu and T. Chen, *Sensors and Actuators*, **A116**, 10 (2004).
2. L. Amaranda, C. Miclea and C. Tanasoiu, *J. European Ceramic Soc.*, **22**, 1269 (2002).



3. L. Li, J. Zhao and Z. Gui, *Ceramic International* **30**, 1073 (2004).
4. W. Windsch, M. K. Gerges, D. Michel, H. Schlemmbach, A. Salzer and P. Reich, *Ferroelectrics*, **109**, 119 (1990).
5. M. K. Gerges, *Egypt. J. Solids* **19**, 2, 305 (1996).
6. L. Wu, C. C. Wei, T. S. Wu and C. C. Teng, *J. Phys. C: Solid State Phys.* **16**, 2803 (1983).
7. V. Tura, L. Mitoseriu, C. Papusoi, C. Harnagea and D. Ricinschi, Anaele Stintifice Ale Universitatii "AL. I. CUZA" Din Iasi Tomul XLIII- XLIV, s.I.b.fasc.2 Fizica Solidelor – Fizica Teoretica ,103 (1997-1998).
8. G. King and E. K. Goo, *J. American Ceramics Society* **73** (6), 1534 (1990).
9. C. Chan, Y. Hsieh, C. Yang and P. Cheng, *Ceramics International* **29**, 495 (2003).
10. S. L. Swartz, T. R. Shrout, W. A. Schulze and L. E. Cross, *J. American Ceramics Society* **67** (5), 311 (1984).
11. M. Chen, X. Yao and L. Zhang, *Ceramics International* **28**, 201 (2002).
12. B. S. Kang and S. K. Choi, *Solid State Communications* **121**, 441 (2002).
13. S. Garcia, J. Portelles, F. Martinez, R. Fount, J. R. Quinones, *Revista Mexicana De Fisica*, **49** (1), 15 (2003).
14. L. Szymczak, Z. Ujma, J. Handere and J. Kapusta, *Ceramics International* **30**, 1003 (2004).
15. M. P. McNeal, S. J. Jang and R. E. Newnham, *J. Applied Physics* **83** (6), 3288 (1998).
16. M. K. Gerges, *Egypt J. Solids* **19** (1), 129 (1996).
17. A. K. Hesham, *Ph.D. Thesis* (physics). Faculty of Science (Qena), South Valley University, Egypt (2004).
18. M. K. Gerges, *Egypt J. Solids* **29** (2), 371 (2006).