

## The effect of temperature on the dielectric properties of Lead Titanate $PbTiO_3$

Dr. Jassim Mohammed Yaseen

Department of Science

College of Basic Education / University of Mosul

Received: 25/4/2011 ; Accepted: 15/9/2011

### Abstract:

The lead titanate  $PbTiO_3$  was prepared by solid state reaction method . In this method ,the mixture of ( $PbO : TiO_2$ ) compounds in mole ratio [1:1] was calcined at  $900\text{ }^\circ\text{C}$  for 2 hr. The crystallisation and growth of tetragonal  $PbTiO_3$  phase (  $c/a = 1.039$ ) were investigated by using XRD technique. The dielectric properties of  $PbTiO_3$  were reported, in which the dielectric constant of  $PbTiO_3$  increases with increasing temperature in tetragonal phase to reach maximum value at curie temperature ( $486\text{ }^\circ\text{C}$ ), then decreases with increasing temperature in cubic phase. The loss factor increases with increasing temperature in different ratios The electric resistivity of  $PbTiO_3$  decreases with increasing temperature.

## تأثير درجة الحرارة على خواص العزل الكهربائي لمادة تيتانيت الرصاص $PbTiO_3$

د. جاسم محمد ياسين النعيمي

قسم العلوم

كلية التربية الأساسية / جامعة الموصل

### ملخص البحث:

تم تحضير مادة تيتانيت الرصاص  $PbTiO_3$  باستخدام طريقة تفاعل الحالة الصلبة. في هذه الطريقة تم حرق خليط المركبين ( $PbO : TiO_2$ ) وبنسبة مولية [1 : 1] بدرجة حرارة حرق  $900\text{ }^\circ\text{C}$  ولمدة 2 hr ؛ تم التحقق من تبلور ونمو الطور الرباعي لمادة  $PbTiO_3$  ( $c/a=1.039$ ) باستخدام تقنية حيود الأشعة السينية؛ تم قياس خواص العزل الكهربائي لمادة  $PbTiO_3$ ، حيث إن ثابت العزل لهذه المادة في الطور الرباعي يزداد مع زيادة درجة الحرارة ليصل أعلى قيمة له عند درجة حرارة كوري ( $486\text{ }^\circ\text{C}$ ) ومن ثم ينخفض مع زيادة درجة

الحرارة في الطور المكعبي؛ عامل الفقد لمادة  $\text{PbTiO}_3$  يزداد مع زيادة درجة الحرارة وبمعدلات مختلفة بينما تنخفض المقاومة الكهربائية مع زيادة درجة الحرارة.

## Introduction:

Lead titanate  $\text{PbTiO}_3$  is a perovskite ferroelectric having a high curie temperature of  $490^\circ\text{C}$ .  $\text{PbTiO}_3$  ceramics having a very small aging rate of dielectric constant, and have a small dielectric constant as compared with those of piezoelectric ceramics such as  $\text{BaTiO}_3$ . Therefore  $\text{PbTiO}_3$  ceramics would be of great promise as a stable piezoelectric material for high temperature and high frequency applications [1].  $\text{PbTiO}_3$  ceramics have many important ferroelectrical and piezoelectric technological applications in optoelectronics and , microelectronics, because of their polarization [2]. Various techniques have been applied to prepare lead titanate powders such as sol-gel method [3,4,5], hydrothermal method at  $110\text{-}200^\circ\text{C}$  [6] and solid- state reaction of mixed oxides [7,8,9], they reported that  $\text{PbTiO}_3$  is highly sensitive to the addition of certain cations, in which Niobium accelerate the densification process, increases the concentration of the lattice vacancies.

The objective of this paper is to prepare lead titanate  $\text{PbTiO}_3$  phase with good growth and to demonstrate that lead titanate can be produced by solid-state reaction with a good dielectric properties.

## Experimental section:

### 1-Synthesis:

To prepare  $\text{PbTiO}_3$  ceramic ,  $\text{PbO}$  ( purity 96 % - BDH ) and  $\text{TiO}_2$  (purity 98 % - BDH) were the chemicals chosen as starting raw materials and that were weighed according to the stoichiometric formula of the required composition in the phase diagram of  $\text{PbO-TiO}_2$  system [10]. By using solid- state reaction method, these chemicals were mixed with equal molar ratios with excess in  $\text{PbO}$  compound by ( 2.2 wt% ) because the loss in the weight of  $\text{PbO}$  compound in the calcination process [11]. The mixture was finely powdered using an agate mortar to obtain homogenous mixture and further grounded after adding ethanol. It was preheated at  $400^\circ\text{C}$  for 30 min and cooled to room temperature .Then ,the mixture was further grounded and calcined at  $900^\circ\text{C}$  for 2 hr .

The powder of calcined material was pressed into pellets in 20 mm diameter and 5 mm thickness. These pellets were sintered at  $1000^\circ\text{C}$  for 1 hr.

## 2- Characterization:

### 2.1- X-ray diffraction:

The calcined material was evaluated by using XRD analysis type PHILIPS PW1316/90 single open recorder / CuK<sub>α</sub> target .

The structural parameters (a) and (c) were calculated using following equation[12] :

$$d_{hkl} = \frac{a}{[h^2 + k^2 + l^2 (a^2/c^2)]^{1/2}} \dots\dots(1).$$

Where d<sub>hkl</sub> = d in Braggs law ( n λ=2dsinΘ ) , n=1 and λ= 1.54 .

### 2.2-Dielectrical properties:

The electrical resistance for a group of samples was tested using Ohm-meter with temperatures ( 32-250 °C ) after prepared by applying a silver electrode on surfaces of each samples . The electrical resistivity ( ρ ) was calculated using the equation (2). The capacity (C<sub>p</sub>) and the resistance of PbTiO<sub>3</sub> material in C-R circuits (R<sub>p</sub>) were measured using (Agilent 16451 Dielectric Test Fixture) with temperatures (30-490 °C) at 10 KHz. The dielectric constant (ε<sub>r</sub>) and the loss factor(ε<sub>r</sub>'') were calculated using the equations (3) and (4), respectively .

$$(\rho = R A / d ) \dots\dots\dots ( 2 )$$

$$\epsilon_r = C_p d / \epsilon_0 A \dots\dots\dots ( 3 ) .$$

$$\epsilon_r'' = d / \omega \epsilon_0 R_p A \dots\dots\dots ( 4 ) .$$

Where, A: surface area of sample , d : thickness

## Results and discussion:

Figure (1) shows the XRD pattern of the prepared PbTiO<sub>3</sub> phase. This figure shows a good growth in tetragonal PbTiO<sub>3</sub> phase (a=4.03,c=4.19) and ( c / a= 1.039), addition to appearance all main peaks that recognize the tetragonal PbTiO<sub>3</sub> phase such peaks (001), (100), (101), (110), (111), (200), (102) at 2 theta (22.6), (23.1), (31.3), (32.3), (39.1), (46.4), (49.8), respectively with high purity. Also this figure shows that there is a shift in 2 theta by (+ 0.2 degree) by compare it with the XRD patterns of PbTiO<sub>3</sub> that observed by Wang et al [3] and Nakagawa et al [13].

Figures 2,3,4 shows the dielectric constant , loss factor and electrical resistivity of  $\text{PbTiO}_3$  material as a function of temperature, respectively.

Figure (2) shows that the dielectric constant of  $\text{PbTiO}_3$  material increases slowly rate with increasing temperature in range (32-260 °C), then stay stable until (300 °C). After that, the dielectric constant increases in high ratios with increasing temperature until reaching maximum value (1183 ) at curie temperature (483 °C). In general , the dielectric constant increases with increasing temperature in the tetragonal  $\text{PbTiO}_3$  phase and then ,it decreases with increasing temperature in cubic  $\text{PbTiO}_3$  phase. This difference in behavior of the dielectric constant is because the change in the crystal structure of  $\text{PbTiO}_3$  from tetragonal to cubic phase, which leads to change in the dielectric properties of piezoelectric materials with temperature [1].

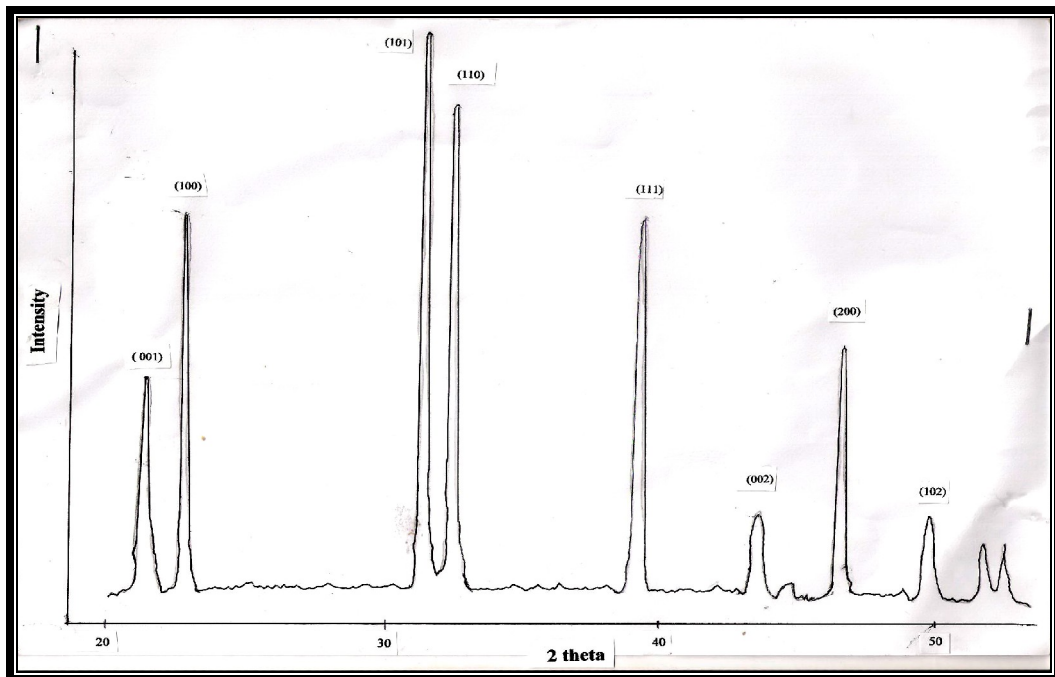


Fig.1: XRD pattern of prepared  $\text{PbTiO}_3$  phase at 900 °C for 2 hr.

The measured curie temperature was less than the theoretical value (490 °C ) by (- 4 °C), this shift because of the impurities in the starting materials. Figure (4) shows that the loss factor of  $\text{PbTiO}_3$  material increases with increasing temperature. To explain this behavior, the loss factor represents the loss in energy in the electric, dielectric materials, but in dielectric materials the loss in energy is variable and depending on the type of material, in which energy that is required to brake the resistance of viscous medium (interior friction) decreases with increasing temperature and that means, increasing in the alignment speed of dipoles and appearance the loss in the energy immediately [14].

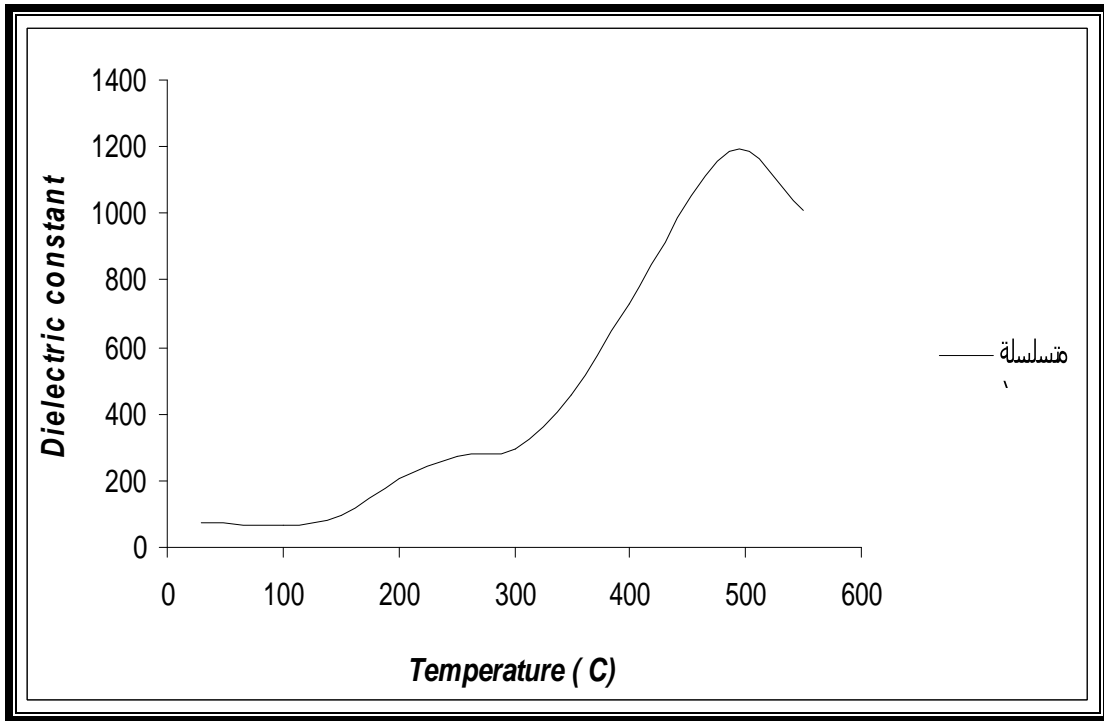


Fig.2: Dielectric constant of PbTiO<sub>3</sub> as a function of temperature

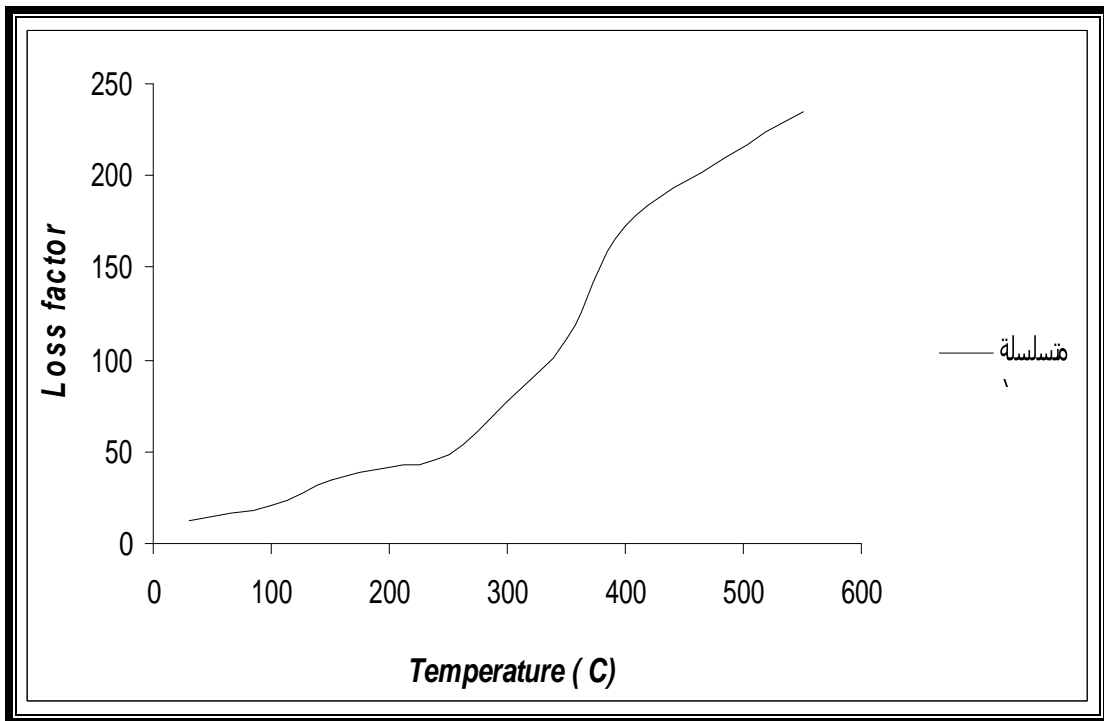


Fig.3 : Loss factor of PbTiO<sub>3</sub> as a function of temperature.

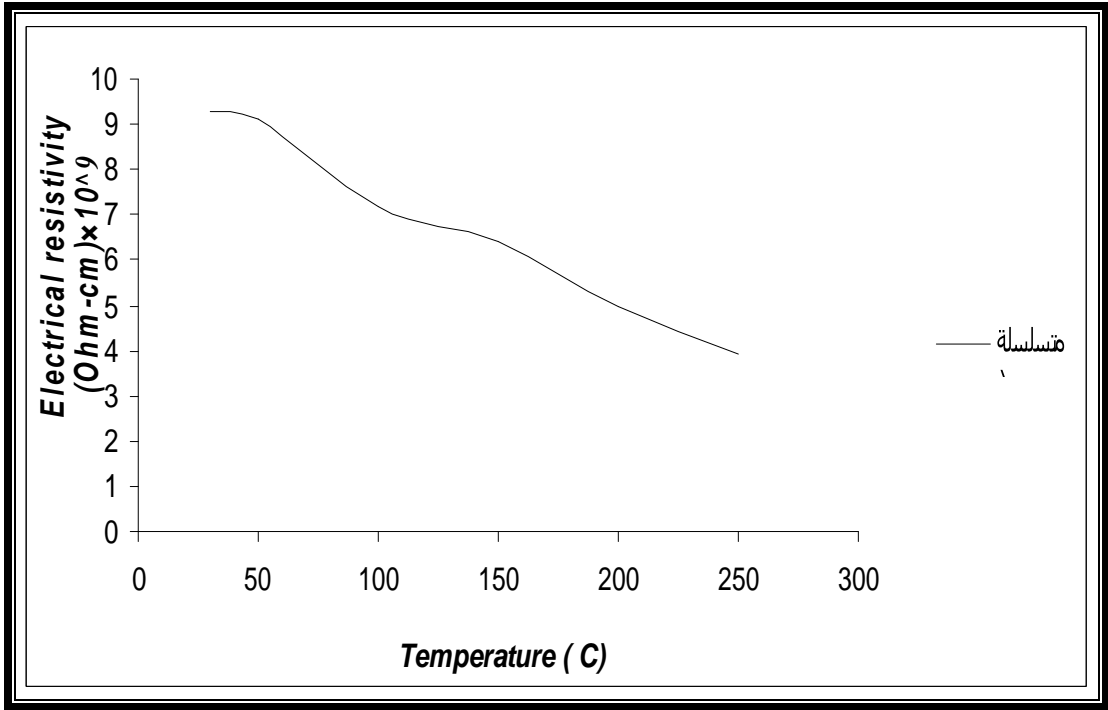


Fig.4: Electrical resistivity of PbTiO<sub>3</sub> as a function of temperature

Figure (4) shows that the electrical resistivity of PbTiO<sub>3</sub> material decrease with temperature increase ,in which a sharp decreasing over two temperature ranges (45- 103 C), up to 160 C and semi stable region in electrical resistivity over the temperature range (105-150 C).

From this figure, it can be seen that the electrical resistivity strongly depend on the temperature .This behavior can be explained as follows: the electrical conductivity of dielectrics may be either ionic or electronic or both. Ionic conductivity( $\sigma$ ) is simply due to the migration of positive or negative ions and it changes with temperature according to the equation

$$\sigma = \sigma_0 \exp (- E_a / K_B T) \dots\dots\dots (5)$$

Where  $E_a$  is the activation energy,  $K_B$  is Boltsmanns constant and T the absolute temperature. From equation (5) , it can be seen that the electrical conductivity increases. In other words, the electrical resistivity decreases with increasing temperature . Electronic conductivity in dielectrics arises from modification to the quantum mechanical band theory of solids, in which , at any finite temperature, electrons will be thermally excited into conduction levels, that lead to decreasing the electrical resistance [15].

## Conclusion:

The lead titanate  $\text{PbTiO}_3$  material prepared with good growth, purity in solid-state reaction method by calcination the mixture of ( $\text{PbO-TiO}_2$ ) as a raw materials at 900 C for 2 hr. The dielectric properties of  $\text{PbTiO}_3$  material strongly depend on the temperature, in which the dielectric constant increases with increasing temperature in tetragonal phase and it decreases with increasing temperature in cubic phase, the loss factor of  $\text{PbTiO}_3$  material increases with increasing temperature and the electrical resistivity decreases with increasing temperature obviously.

## References:

- 1- Rosen C.Z, Basavaj V, Newnham R; Piezoelectricity ; American Ins.phys.-New York, P175, 1992.
- 2- Chilibon, Marat J , Igrija R ; Ultrasound role in Sol-Gel processing of  $\text{PbTiO}_3$  ceramics; J. Optoelectronics and advanced mater, Vol.7, No.5 P 2727, 2005.
- 3- Wang Y, Xu G, Yang L ,Ren ; Preparation of single-crystal  $\text{PbTiO}_3$ , nanorods by phase transformation from  $\text{Pb}_2\text{TiO}_6$  nanorods; J. Alloys and compound, Vol.481, P127, 2009.
- 4- <http://www.j:ptio3sol5.htm>
- 5- Baodong Q ,Yuguo W, Peilin Z ; Size and temperature dependence of dielectric constant of ultra fine  $\text{PbTiO}_3$  particles ; Chin.phys.lett, Vol.11, No.8, P514, 1994.
- 6- Chien A T, Sachlaben J, Kim J H, Speck J S; Synthesis and characterization of  $\text{PbTiO}_3$  powders and heteroepitaxial thin films by hydrothermal synthesis; J.Mater.Res, Vol.14 ,No.8, 1999.
- 7- [http:// PbTiO<sub>3</sub>-PbZrO<sub>3</sub> - Pb\(Mn<sub>1/3</sub>-Nb<sub>2/3</sub>\)O<sub>3</sub>// 1310-0157,2007.](http://PbTiO3-PbZrO3-Pb(Mn1/3-Nb2/3)O3//1310-0157,2007)
- 8- Chandaratreya S S, Fulrat R M ; Reaction mechanisms in the formation of PZT solid solution ; J.Am.Cermic.Soc, Vol.64, No.7, P422, 1981.
- 9- [http:// resources .metapress .com/ pbf-preview.axd.-J.Mater.Sci.lett,](http://resources.metapress.com/pbf-preview.axd.-J.Mater.Sci.lett) Vol.11, 1992.
- 10- Barnard J, William R C; Piezoelectric ceramic; Academic press, P117 1971.
- 11- Othmer K; Encyclopedia of chemical technology; Vol.10, P13, Wiley-Inter, Pub, 1980.
- 12- Cullity B D; Elements of x-ray diffraction; Addison-Wesley Pub. Co.Inc, 1967.
- 13- Nakagawa T, Yanaguchi J, Okuyama M; Preparation of  $\text{PbTiO}_3$  ferroelectric thin films by chemical vapor deposition; Jap.J.Appl Phys, Vol.21, P655, 1982.
- 14- Harrop P J, ; Dielectrics; Butterworth-London 1972.
- 15- Odwyer J J ; The theory of electrical conduction and breakdown in solid dielectrics ; Clarendon Press. Oxford 1973.

This document was created with Win2PDF available at <http://www.daneprairie.com>.  
The unregistered version of Win2PDF is for evaluation or non-commercial use only.