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Note

Dielectric properties of $PbSrWO_4$ and $PbBaWO_4$ compounds

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Abstract : $PbSrWO_4$ and $PbBaWO_4$ have been synthesised by the solid state reaction technique XRD patterns show them to be tetragonal. Dielectric constant (K') and Dielectric loss (K'') of $PbSrWO_4$ and $PbBaWO_4$ have been measured at 1 kHz in the temperature range of 300 to 1050 K. The log K' vs T as well as log K'' *vs* T plot of $PbSrWO_4$ and $PbBaWO_4$ shows rapid increase of dielectric constant above 590 K and 640 K, respectively.

Keywords : dielectric constant, dielectric loss, $PbSrWO_4$, $PbBaWO_4$

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Recently the optical and electrical properties of Scheelite structure have received much attention. $PbSrWO_4$ and $PbBaWO_4$ belong to Scheelite structure with tetragonal unit cell. The luminiscence properties, electrical conductivity and defect chemistry have been studied in the past years [1-4]. Dielectric studies are informative in the study of ferroelectricity and phase transition [5,6]. Besides electrical transport properties of 3d compounds are in general interesting from various points of view [7,8]. This paper reports dielectric constant and dielectric loss of $PbSrWO_4$ and $PbBaWO_4$.

The starting materials for the preparation of these compounds were PbO, SrO, BaO and WO_3 (of 99.99% purity, procured from Alfa Aesar, A Johnson Matthey Chemicals India Pvt. Ltd.). The stoichiometric amount of these oxides were mixed and heated in silica crucible for 50 h at a temperature of 1140 K followed by one intermediate grinding and final product was cooled down slowly. The prepared compounds undergo the following solid state reactions.

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$$PbO + BaO + WO_3 \xrightarrow[Air]{1140K} PbBaWO_4 + 0$$

The weight loss corresponding to oxygen on the right hand side of the reactions was observed in both cases.

To get confirmation regarding the complete formation of the compounds, X-ray diffraction pattern of the sample was taken at room temperature using CuKα line (λ = 0.15418 nm) as shown in Figure 1-2. From XRD pattern the values of d_{hkl} have been obtained using the relation

(1)

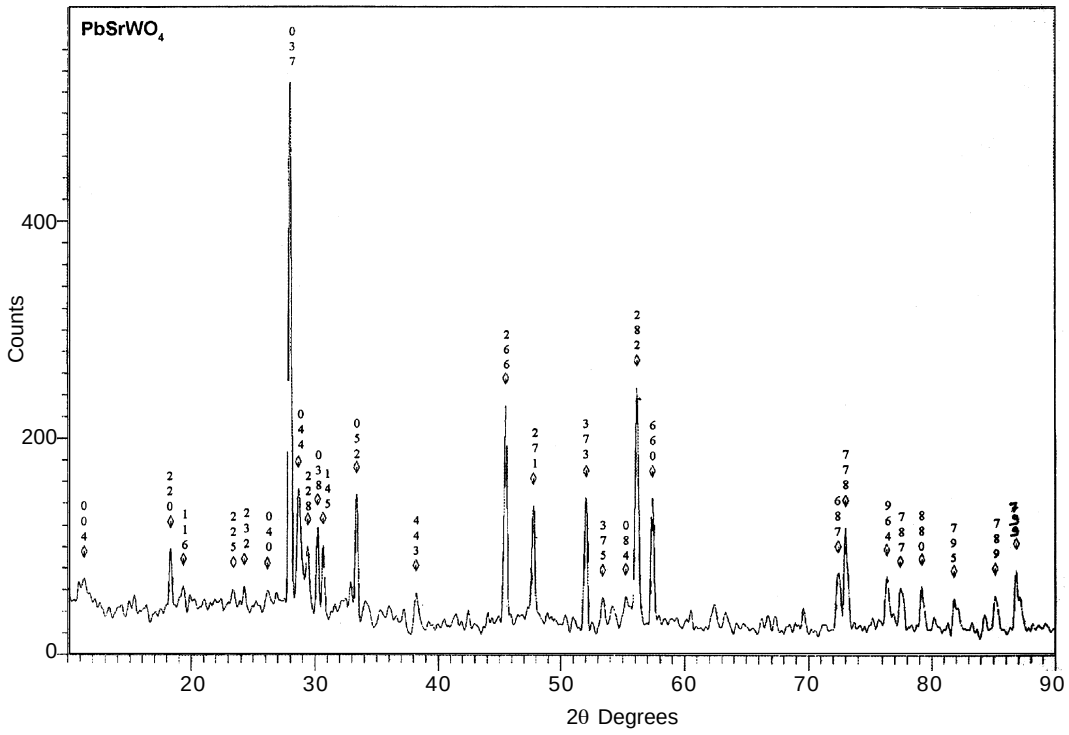

Figure 1. X-ray diffraction pattern of $PbSrWO_4$.

From d_{hkl} values, structure of the studied compounds were resolved following usual procedure. All the peaks have been assigned with proper *hkl* values. This confirms that the prepared compound has single phase and no unreacted part of the starting material was left. The unit cell parameters are given in Table 1.

Table 1. Structural parameters of tetragonal unit cell, calculated density (d_0), density of pressed pellets (d_p) and values of pore fraction (f).

Compounds	Unit cell	Lattice parameters		Density		Pore fraction
		a = b (nm)	c (nm)	d_0 ($kgm^{-3}\times10^{-3}$)	d_p ($kgm^{-3}\times10^{-3}$)	f
$PbSrWO_4$	Tetragonal	1.3630	3.1322	6.62	6.03	0.089
$PbBaWO_4$	Tetragonal	1.4681	2.3239	7.86	7.08	0.099

The prepared compounds were pressed at pressure of 8.22×10^{-8} Nm^{-2} to form pellets of circular cross section (Area ~ 0.9×10^{-4} m^2 and thickness ~ 0.3×10^{-2} m). The pellets were then sintered in air for 28 hrs at 1000K. The mounting of pellet was done between the rigid silver electrode of sample holder after coating with silver paint from both sides of the pellets.

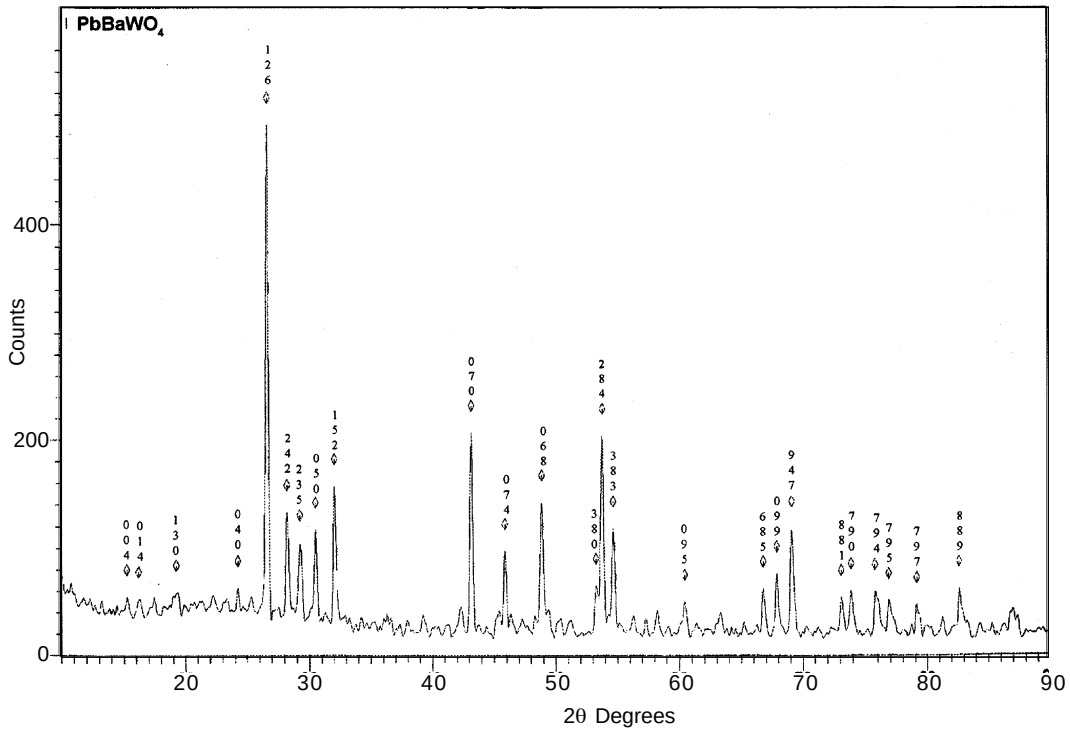

Figure 2. X-ray diffraction pattern of $PbBaWO_4$.

The dielectric constant and dielectric loss were obtained by measuring the capacitance (C) and quality factor (Q) of the sample at different temperatures and at a frequency of 1kHz, using LCR-Q meter model 928, Systronics India. The measurements have been done in both heating and cooling cycles, but no significance differences have been observed. The dielectric constant ($K^{/}$) and dielectric loss ($K^{//}$) of the material have been determined by using the following relations.

$$K' = \frac{Cb}{8.854 \times 10^{-12} A} \tag{2}$$

(3)

where b = the thickness, A = face area of the pellet, = frequency in Hz, R = resistance in and C_0 = the capacitance of the empty capacitor in F, respectively.

The variation of the dielectric constant (K') and dielectric loss (K'') with temperature at 1kHz is shown in Figure 3-4. It is seen from these plots that these compounds have high dielectric constant from 40 to 58 at 400 K. Since dielectric constant (K') seems to have almost no temperature dependence, these values may be taken as the room temperature values of the materials. The reported values of K' has been calculated using the capacitance value for the pressed pellets. The density of these pellets remains less than the theoretical density of these materials. This means pellets contain air pores. Therefore a correction

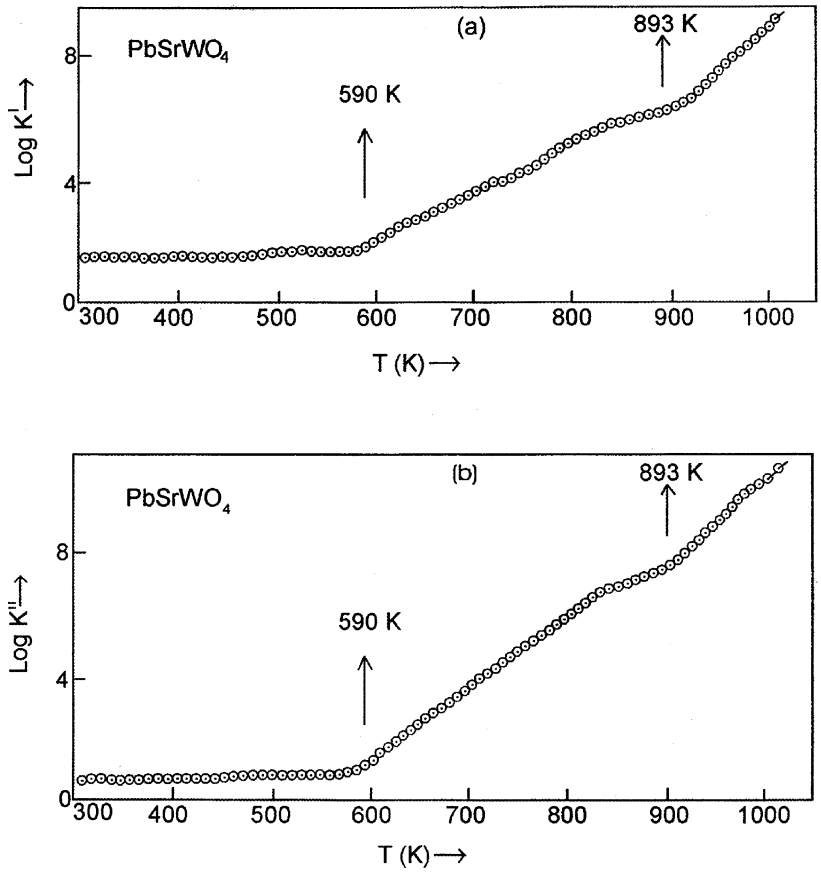

Figure 3. Plots of dielectric constant (log K′) and loss (log K″) against absolute temperature (T) for $PbSrWO_4$.

for pore fraction is essential to obtain the bulk value of the dielectric constant ( ) and is given by

. (4)

For low conducting solids K'_b, K/ and P are related by the relation [9]

. (5)

The evaluated values of are given in Table 2.

Table 2. The bulk value of dielectric constant ( ) of the studied compounds.

Compounds	at 400 K
$PbSrWO_4$	41
$PbBaWO_4$	60

$$f = \frac{d_0 - d_p}{d_0} = \left(\frac{K'_b - K'^3_b}{K'_b - P}\right)^3$$

$$K'_b = \frac{1 - P}{1 - P}$$

The values of $K^/$ of all these material become large as temperature is increased and validity of equation (5) becomes doubtful. Further this formula affects only the magnitude of but not the nature of temperature variation of $K^/$. Therefore we have not used this correction at higher temperatures.

The values of dielectric constant ($K^/$) of all the studied materials at temperature 400 K, 600 K, 800 K and 1000 K at frequency 1 kHz are given in Table 3.

Table 3. Dielectric constant ($K^/$) for studied tungstates at different temperature.

Compounds	Dielectric constant			
	400 K	600 K	800 K	1000 K
$PbSrWO_4$	40	120	1.90×10^5	9.12×10^8
$PbBaWO_4$	58	120	6.31×10^4	5.25×10^6

It is seen from the table that $PbBaWO_4$ has the highest dielectric constant at 400 K. The dielectric constant has a very slow increase with temperature upto 600 K. The systematic trend of $K^/$ variation reveals that polarization mechanism in all these tungstates are same. A relatively larger value of $K^/$ in case of $PbBaWO_4$ appears due to the presence of chemical impurities which forms some kind of donor centres and have larger polarizability. The dielectric constant has very slow increase at lower temperature. This shows that there is no chance for the existance of thermally generated charge carriers at lower side

of temperature. The lower value of $K^{//}$ below 600 K indicates that free charge carriers generated from impurities is also small. The slow increase of $K^{//}$ for all indicate that the thermally generated charge carriers below 600 K are small. This rules out the possibility of strong space charge polarizability. Well made electrode rules out the chance of interfacial polarization. Therefore this slow increase seems to be the combined effect of lattice and electronic polarizabilities of individual ions. The increase of these polarizabilities seems to compensate the slight decrease of polarizability due to decrease in the number of ions per unit volume following the lattice expansion with temperature. However, it must be noticed that the increase of $K^{/}$ with T is very slow in comparison to the variation one expects for ionic solids. This indicates that either thermal expansion of these materials is very small or they have some other kind of polarization mechanism.

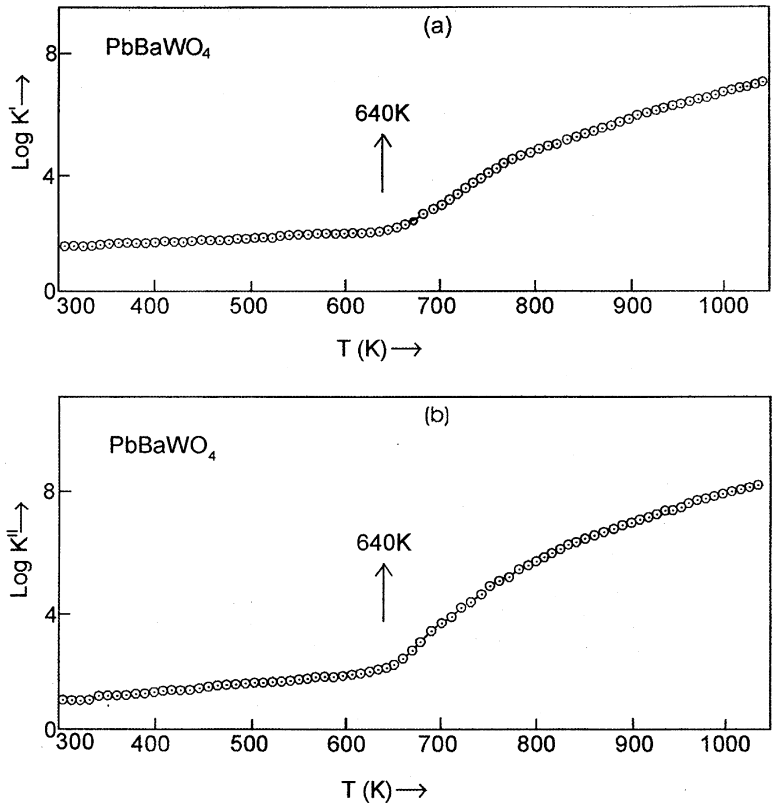

Figure 4. Plots of dielectric constant (log $K^{/}$) and loss (log $K^{//}$) against absolute temperature (T) for $PbSrWO_4$.

It is seen from Figures that dielectric constants ($K^{/}$) of these materials have much faster increase at higher temperature. The dielectric loss ($K^{//}$) also shows a rapid increase above certain critical temperature. It appears that higher increase in $K^{/}$ at higher temperature is due to space-charge polarization of thermally generated charge carriers. The break temperatures (T_k) observed in $K^{/}$ and $K^{//}$ are given in Table 4.

Table 4. Break temperature (T_k) obtained from dielectric constant and loss of studied tungstate.

Break temperature	Compounds	
	$PbSrWO_4$	$PbBaWO_4$
T_k	590	640

The dielectric constant ($K^{/}$) and loss ($K^{//}$) have very slow increase upto temperature T_k. However, above T_k, this increase becomes much faster. The value of T_k is different for different tungstates. The reason for faster increase of $K^{/}$ and $K^{//}$ at higher temperature is due to space charge effect of thermally generated charge carriers.

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