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ABSTRACT. $Pb_{1-x}Ni_xTiO_3$ (PNT) with x varying between 0 and 1 is synthesised by sol-gel technique and then processed in microwave at 1000ºC. The X-ray diffraction confirms the cubic structure in PNT. The electrical conduction and frequency response is investigated on the basis of impedance spectroscopy. PNT exhibited a negative temperature coefficient of resistance and non-Debye type of relaxation for all the recorded temperature.

Introduction. Ferroelectric ceramics are widely used in spintronic, optoelectronics as sensors and transducers. Ferroelectric with perovskite structure can exhibit a wide range of application in semiconductor devices. The fundamental interest of ferroelectric and ferromagnetic coexistence is to cause an electric polarisation by applying magnetic field or magnetisation in applied electric fields. In non-volatile memory devices writing data bit with an electric field by creating magnetic field used to read the data will be possible [1-2]. Researchers are interested in microwave sintering technique for ceramic due to its advantages over conventional sintering.Microwaves when passing through a specimen the molecular dipoles oscillate with the applied microwave frequency (2.45GHz in the present case) and produce alarge amount of latent heat which is again absorbed the material results in sintering. The process is quite fast compared to conventional heating with added benefits of low energy consumption, cost, sintering temperature and uniform densification [3-4].The present work is focused on thesol-gel synthesis of $Pb_{1-x}Ni_xTiO_3$ (PNT) sintered using microwaves. The sol-gel technique is adopted in lieu of its ability to produce high purity, ultrafine nanoparticles at low processing costs [5-7].

Experimental Procedure.

Lead acetate Pb $(CH_3COOH)_2$ $3H_2O$, Nickel nitrate Ni $(NO_3)_2$ $6H_2O$ and Titanium Butoxide Ti $(C_4H_9O)_4$ were taken as starting materials to prepare PNT. To prepare PNT, stoichiometric ratios of starting materials are taken. Lead acetateand nickel nitrateare dissolved in pure glacial acetic acid. The prepared solution is dehydrated at 100°C for 10min and cooled down to room temperature. After cooling down, Titanium Butoxide added drop by drop to that solution then continuous stirring for 30 min at room temperature. Then the mixture of deionized water and ethanol are addedat a slow rateso as to initiate hydrolysis and prevent fast gelation of the solution. Thus prepared gel is heated at 100°C for 2 hours in theoven to obtain the PNT powder. For the calcination step, microwave furnace is used to at 730°C for 45min at a rate of 30°C per min.The green powder samples aregrindedat 8 hours using agate mortar. Thegrinded powders are sintered at 1000°C using microwave furnace. The prepared samples are used in different characterization. For making a pellet 1 cm diameter die is used with PVA as abinder.

Results and Discussion. Structural analysis

X-ray diffraction:

The X-ray diffraction pattern of PNT powder is calcined at 730°C at 45min at the rate of30°C/min using microwave furnace. Fig.1 shows the X-ray diffraction of PNT, x=0.0 and x=1.0 isfor lead titanate (PT) and nickel titanate (NT). It confirms the tetragonal structure for P4mmspace group for x=0.0 (PT) and rhombohedral structure for R3 space group for x=1.0 (NT). The transformation from tetrahedral to rhombohedral occurred gradually with increase in x. Pseudo cubic structure can be noticed for x = 0.4 to 0.6 in agreement with earlier reports [8-9]. The co-existence of tetragonal and rhombohedral (probably-cubic) phase [1].

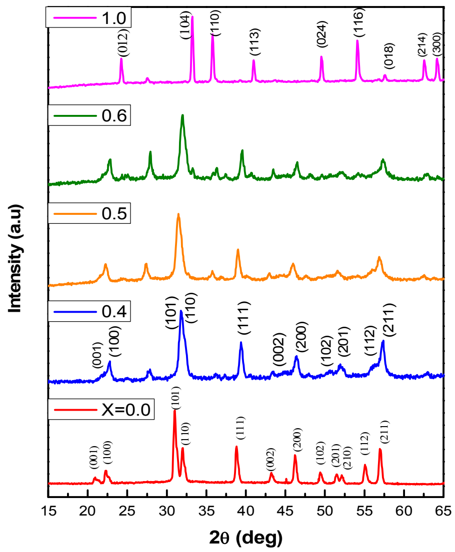

Fig. 1. XRD pattern of PNT.

Electrical Conductivity Measurements

Complex impedance analysis

Complex impedance spectroscopy technique is used to investigate the electrical properties of materials. Ferroelectric ceramics are in general electrically heterogeneous. The variation of real with theimaginary part of the impedance is known as Nyquist plots. In general Nyquist plot is asemicircular arc for understanding the grains and grain boundary effect on the electrical property of the materials. Complex impedance spectroscopy is a powerful non-destructive testing tool to examine the effect of microstructure on electrical behaviour. Complex impedance analysis is the representation of impedance *Z*, electrical modulus *M* and admittance *Y* as complex parameters. A Debye-type relaxation in the ceramics results in a semicircular Nyquist plot with its centre lying on the abscissa. However, the centre is expected to be lying below the abscissa for non-Debye type relaxations. The complex impedance is generally represented as below [8]:

$$Z^* = Z' + iZ" = \frac{R}{1+\left(\frac{\omega}{\omega_o}\right)^{\alpha}} \quad (1)$$

where α is the measure of electrical deviation from theideal response and can be estimated from the centre of the Argand semicircle.

The deviation α tends to zero for mono-dispersive Debye type relaxations and it is non-zero and positive for non-Debye and poly-dispersive type of relaxations. The Nyquist plots of PNT at selected temperatures are shown in Fig. 2. The contribution to electrical response in polycrystalline solids is by grains, grain boundaries and space charge. A semicircle at alow frequency would indicate space charge relaxation, at theintermediatefrequency, it would be grain boundaries and at high frequency, it is due to grains. The Cole-cole plots show single semicircle indicating the dominance of grains over grain boundaries and space charge effect. Furthermore, all the relaxations are found to be thenon-Debye type with the centre of the semicircular arcs lying below the abscissa. The electrical responses from impedance spectroscopy can always be represented as anequivalent electrical circuit as series of parallel RC elements. In the present case, single parallel RC ($R_b \| C_b$) circuit represents the relaxation of grains to the applied field. The values of Rb, Cb and relaxation time τ are estimated using relaxation Eqn. (2) and are presented in Table 1.

Table 1. Resistance, Capacitance, Relaxation time and Conductivity value at different temperature.

Ratio	Temperature (°C)	R_b (ohm)	C_b ($\times 10^{-10}$ farad)	τ ($\times 10^{-7}$sec)	σ_{dc} (S/cm)
X=0.0	440	764.90	2.97	2.27	6.13×10^{-3}
	460	451.20	4.41	1.99	0.010
	480	303.12	7.50	2.27	0.015
	490	259.37	8.77	2.27	0.018
	500	244.47	8.14	1.99	0.019
	520	233.60	6.81	1.59	0.020
	540	169.05	1.04	1.76	0.027
X=0.4	440	22656.25	0.35	7.96	1.51×10^{-4}
	460	8233.17	0.64	5.30	4.17×10^{-4}
	480	3635.81	1.09	3.98	9.46×10^{-4}
	500	1985.57	2.00	3.98	1.73×10^{-3}
	520	1075.12	3.70	3.98	3.19×10^{-3}
	540	640.02	4.97	3.18	5.37×10^{-3}
X=0.5	440	5161.29	0.617	3.18	9.03×10^{-4}
	460	1941.10	1.17	2.27	2.40×10^{-3}
	480	1168.75	1.36	1.59	3.99×10^{-3}
	500	933.77	0.852	0.79	4.99×10^{-3}
	520	640.10	0.829	0.53	7.28×10^{-3}
	540	609.27	0.522	0.31	7.65×10^{-3}
X=0.6	440	35009.61	0.758	26.5	1.23×10^{-4}
	460	6269.47	1.26	7.96	6.87×10^{-4}
	480	2392.78	1.66	3.98	1.80×10^{-3}
	500	1382.52	0.575	0.79	3.11×10^{-3}
	520	1057.78	0.752	0.79	4.07×10^{-3}
	540	854.08	0.621	0.53	5.04×10^{-3}
X=1.0	440	7250480.77	0.0732	530.7	3.77×10^{-7}
	460	2207932.69	0.0721	159.0	1.24×10^{-6}
	480	1385576.92	0.111	159.0	1.97×10^{-6}
	500	938701.92	0.0565	53.0	2.91×10^{-6}
	520	533653.84	0.0596	31.8	5.13×10^{-6}
	540	475096.15	0.0335	15.9	5.76×10^{-6}

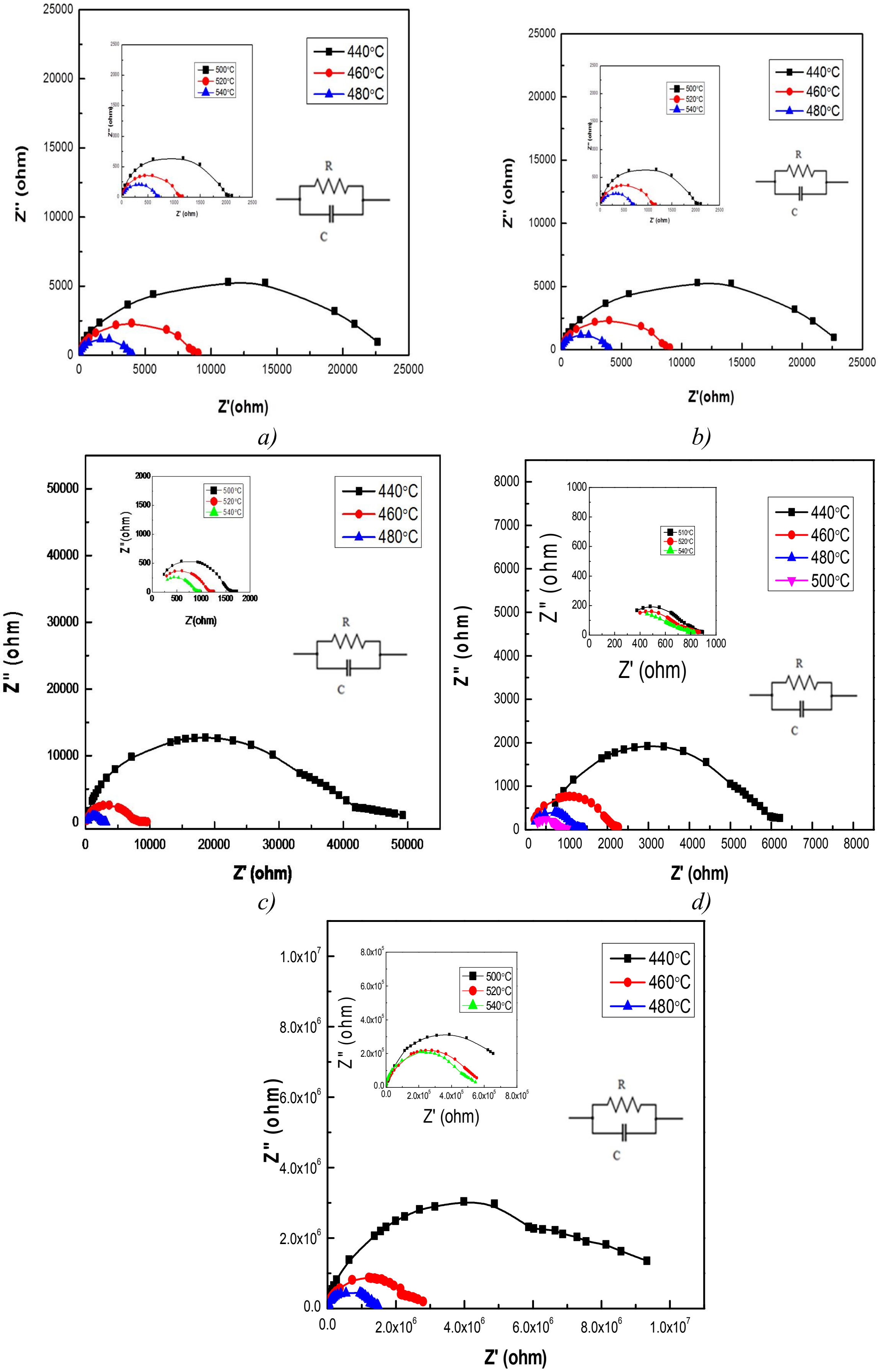

Fig. 2. (a) x=0.0 (b) x=0.4 (c) x=0.5 (d) x=0.6 (e) x=1.0 shows the cole- cole plot at different temperatures.

$$\tau = \frac{1}{\omega} = R_b C_b \tag{2}$$

where $\omega=2\pi\nu_{max}$, ν_{max} denotes applied frequency corresponding to the arc maximum, R_b and C_b are bulk resistance and bulk capacitance respectively.

The conductance of the material is calculated using the relation (3):

$$\sigma_{dc} = \frac{l}{AR_b} \tag{3}$$

where l and A are the thickness and cross-sectional area of the pelletized sample.

These values are also tabulated in Table 1. It is also noticed that bulk resistance decreases with increase in temperature indicating negative temperature coefficient of resistance.

Summary. The lead nickel titanate ($Pb_{1-x}Ni_xTiO_3$, PNT) is synthesised by sol-gel route followed by microwave sintering. X-ray diffraction confirms the cubic structure for $x = 0.4$ to 0.6. The impedance analysis of PNT is studied for grain and grain boundary effects. PNT exhibited thenon-Debye type of relaxations at all temperatures recorded. An equivalent RC circuit is proposed in each case along with estimated resistance, capacitance and relaxation time that is observed to be very low of the order of 10^{-7}sec. DC electrical conductivity is also estimated at selected temperatures. It is noticed that PNT is a negative temperature coefficient material.

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