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Preparation and ultrasonic study of sodium potassium tantalate ($Na_{1-x}K_xTaO_3$) mixed system

Manish Uniyal*, S C Bhatt & Sidharth Kashyap
Department of Physics, H N B Garhwal University, Srinagar (Garhwal) 246 174, India

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Ultrasonic velocity of the piezoelectric ceramic pellets of ferroelectric material $Na_{1-x}K_xTaO_3$ (X=0, 0.2, 0.3, 0.4, 0.5, 0.6 and 1.0) have been investigated at temperature 32 ºC and frequency 5 MHz with the help of ultrasonic c-scan system developed at NPL, New Delhi. The samples have been prepared by the conventional solid-state reaction method and sintering process.

Keywords: Ultrasonic velocity, Ceramic pellets, Ferroelectric materials, Solid state reaction method

1 Introduction

Sodium potassium tantalate ($Na_{1-x}K_xTaO_3$) piezoelectric ceramics and single crystals, piezoelectric/epoxy composites have been used as the active layer in ultrasonic transducers, particularly in high frequency probes for medical imaging. These composites offer several advantages in comparison to monolithic piezoelectric ceramics or polymers. High coupling coefficient, low acoustic impedance, better acoustic matching to the human body, adjustable dielectric constant and mechanical flexibility are some of the benefits of SPT piezoelectric composites[1]. Ultrasonic testing of materials is one of the widely used methods of nondestructive testing, in which beams of high frequency sound waves generally of 0.5 MHz to 25 MHz are used to detect the cracks, laminations, shrinkages, cavities and other discontinuities. Inclusion and other inhomogeneties in the material can also be detected using partial reflection or scattering of ultrasonic waves. It has high sensitivity for detecting flaws, can penetrate extremely thick sections and need access to only one surface of the testing material.

The propagation of ultrasonic wave in any substance has become a fundamental test to investigate its properties. The velocity and attenuation coefficients are the basic propagation constants, which are related to the microscopic structure of the materials. Sodium potassium tantalate (SPT) with perovskite structures are widely used for transducer applications with broad ranges of technologically important dielectric, piezoelectric, ferroelectric and electro-optic properties. Cross[2] has predicted, theoretically, the phase diagram of $KTaO_3$-$NaTaO_3$ mixture from phenomenological arguments. Due to the immense importance of these materials, ultrasonic investigation has been carried out by several researchers[3-9]. Also ultrasonic velocity and thermodynamic parameters are widely used to study of molecular interactions[10-12] in pure liquid, aqueous solutions and liquid mixtures.

Ultrasonic propagation velocity in $KTaO_3$ has been measured by Barrett[13]. In the study the ultrasonic propagation velocity was measured using a coherent pulse -C. W. technique, as described by Klerk[14], with a measurement frequency of about 200 MHz. The ultrasound was generated by a CdS transducer evaporated onto a [100] cleavage face of the sample. In our study we have measured the ultrasonic velocity for the composition of $NaTaO_3$, $Na_{0.8}K_{0.2}TaO_3$, $Na_{0.7}K_{0.3}TaO_3$, $Na_{0.6}K_{0.4}TaO_3$, $Na_{0.5}K_{0.5}TaO_3$ and $Na_{0.4}K_{0.6}TaO_3$ mixed ceramics at 32 ºC and frequency 5 MHz at the transverse section of the prepared samples.

2 Ultrasonic Transducers

Ultrasonic transducers are composed of three main components; a piezoelectric material, a backing material and one or multiple matching layers, sodium potassium tantalate (SPT) piezoelectric materials are the core component of ultrasonic transducers for generating the ultrasound beam as well as receiving the echo signal. The acoustic performance of transducers is prominently influenced by electromechanical and dielectric properties of the piezoelectric layer. For medical imaging applications, broadband transducers are required. The bandwidth (BW(%)) of the transducers is defined as[15]:

$$BW=f_H\text{-}f_L/f_c \times 100$$

where, f_c, is center frequency, and f_L and f_H are low and high frequencies at -6dB of the frequency response spectrum.

*Corresponding author (E-mail: dr.manishuniyal@yahoo.com)

Broadband transducers can be operated at multiple transmit/receive frequencies. Transmit at lower frequencies followed by receiving the echo signal at higher frequencies enhances the sensitivity and resolution during a medical imaging event[16]. Usually piezoelectrics with high dielectric constant suggests higher acoustic output pressure. This is particularly important for matrix arrays with small element size, with high permittivity materials for a better electrical impedance matching. On the other hand, lower dielectric permittivity is required in high frequency single element transducer to improve the resolution and sensitivity of the transducers.

3 Preparation Techniques

Here we have used the conventional solid state reaction method and sintering process followed by the procedure employed by Tennary and Hang[17].

The following compositions of sodium potassium tantalate ceramic were prepared by conventional solid state reaction and sintering process. The prepared samples, with varying compositions, of the systems have been prepared as given below:

(a) $Na_{1-x}K_xTaO_3$

1 $NaTaO_3$
2 $Na_{.9}K_{.1}TaO_3$
3 $Na_{.8}K_{.2}TaO_3$
4 $Na_{.7}K_{.3}TaO_3$
5 $Na_{.6}K_{.4}TaO_3$
6 $Na_{.5}K_{.5}TaO_3$
7 $Na_{.2}K_{.8}TaO_3$
8 $KTaO_3$

Sodium tantalate has an orthorhombic symmetry at room temperature and it becomes cubic as potassium is mixed in $NaTaO_3$. At 50/50 composition, anomalous behaviour was reported. $Na_{.5}K_{.5}TaO_3$ composition is very important from this point of view and remarkable other properties.

The starting materials used for our samples were, Na_2CO_3, K_2CO_3, and Ta_2O_5 having purity 99%. The solid-state reaction involved for preparation of these compositions is as under:

$$2Na_2CO_3(1-x)+ 2K_2CO_3(x)+ 2Ta_2O_5= 4Na_{1-x}K_xTaO_3+4CO_2+O_2,$$

where, x can vary from 0 to 1, as (0, 0.2, 0.3, 0.5, 0.6, 0.8 and 1).

The required quantity of each reagent is calculated using the following procedure:

To prepare a sample or pellet of sodium potassium tantalate of 3 g, one requires:

Amount of Na_2CO_3 required= 3× Molecular weight of Na_2CO_3/Mol. wt. of system $(NaKTaO_3)\times2$

Amount of K_2CO_3 required= 3× Molecular weight of K_2CO_3/Mol. wt. of system $(NaKTaO_3)\times2$

Amount of Ta_2O_5 required= 3× Molecular weight of Ta_2O_5/Mol. wt. of system $(NaKTaO_3)\times2$

All the samples were prepared by conventional sintering method (solid state reaction method). The quantities of the reagents required for each composition were calculated from the above-mentioned formula.

The starting material Na_2CO_3, K_2CO_3 and Ta_2O_5 were dried at 200 ºC for one hour to remove absorbed moisture. K_2CO_3 has to be handled very carefully in order to prevent it from absorbing moisture during the material formation process.

Different compositions of $Na_{1-x}K_xTaO_3$ were prepared by weighing sodium carbonate, potassium carbonate, and tantalate in proper proportions. Each composition was manually dried mixed for 60 min and then wet mixed using reagent methyl alcohol and mullet mortar and pestle, for 60 min. The mixture was calcined in a platinum crucible, in air, at 950 ºC for one hour for carbonate removal. After cooling in dry air, the calcined mixtures were weighed to ensure complete carbonate removal.

The pre-sintered mixture was ground and pressed into pellets of 10 mm diameter applying pressure of 2.5 tons. All the pellets were placed on a platinum crucible and sintered, in air, at 1200 ºC for 24 h. The sintered pellets were electroded using air-drying silver paste for dielectric measurement. The sintering conditions for all the compositions are given in Table 1.

Some of the prepared ceramic pellets of mixed $Na_{1-x}K_xTaO_3$ system shown in Fig. 1. Colour found on

Table 1 — Sintering conditions for $Na_{1-x}K_xTaO_3$ system of all the compositions.

Compositions	Mixing time (h)	Cacining temperature (ºC)	Cacining time (h)	Final milling time (h)	Sintering temperature (ºC)	Sintering time (h)
$Na_{1-x}K_xTaO_3$	2	950	2	1	1200	24

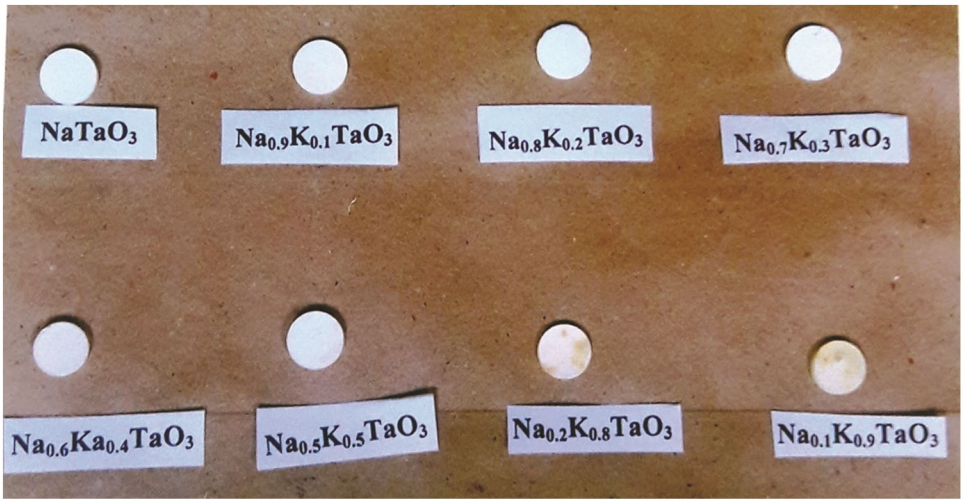

Fig. 1 — Some of the prepared ceramic pellets of $Na_{1-x}K_xTaO_3$ system.

the pellets is due to the binder (poly (vinyl alcohol)) which is used during the preparation of these ceramic pellets. Characterization, dielectric measurements, electrical conductivity and theoretical and experimental properties have been investigated in our previous result[18-27].

4 Characterizations

To characterize the material in the present study, X-ray diffraction pattern of the samples at room temperature were obtained on a SEIFERT X-ray diffractometer made by Bruker, using Cu-K filter radiation of 1.540598 Å wavelength. The instrument is well calibrated with the silicon standard sample. Peak indexing was done by using joint committee on power diffraction standards (JCPDS) data cards. From the observed diffraction pattern, lattice spacing d was determined, which was used to determining the perovskite lattice parameters. The unit cell parameters were determined using the Auto-X computer software, which includes CRYSFIRE and FULLPROF software.

The lattice parameters for $NaTaO_3$ and $KTaO_3$ were obtained from the X-ray diffraction patterns and given in the Table 2.

Table 2 — Lattice parameters of $NaTaO_3$ and $KTaO_3$.

	$NaTaO_3$		$KTaO_3$	
	Present study data	NBS and data from literature[28]	Present study data	NBS and data from literature[29]
a	5.525	5.4941	3.985	3.9885
b	7.419	7.7508	3.983	3.9885
c	5.49	5.5130	3.981	3.9885

5 Speed and Attenuation of Ultrasonic Waves in Solids

In isotropic solids, both transverse and longitudinal waves can be propagated. The velocity of shear wave in an extensive medium is $c_S = \sqrt{(G\rho)} = \sqrt{\{E/2\rho(1 + \sigma)\}}$, E being Young's modulus, G the rigidity modulus and σ is Poisson's ratio, and this is also the velocity of torsional waves in thin cylindrical bars. The speed of longitudinal or irrotational waves in an extensive medium is given by $c_L = \sqrt{\{(K + \frac{4}{3} G)/\rho\}} = \sqrt{\{E(1 - \sigma)/\rho(1 - 2\sigma)(1 + \sigma)\}}$, K being the bulk modulus. In straight uniform bars and in tubes thin compared with a wavelength, the speed of longitudinal waves is $c_E = \sqrt{(E/\rho)}$. Surface waves propagating along the surface of an extensive solid are generally known as Rayleigh waves and propagate with speed $c_{SR} = ac_S$, where a is the least positive root of the equation.

$$\frac{a^6}{8(1 - a^2)} + a^2 = \frac{1}{1 - \sigma}$$

In anisotropic solids, which may have as many as 21 independent elastic constants, there may exist, for a given direction of the wave normal, three distinct displacement vectors each associated with a distinct plane wave velocity. Out of these three waves, one is analogous to the longitudinal and the others to transverse waves in the isotropic case. The directions of the respective displacement vectors are mutually

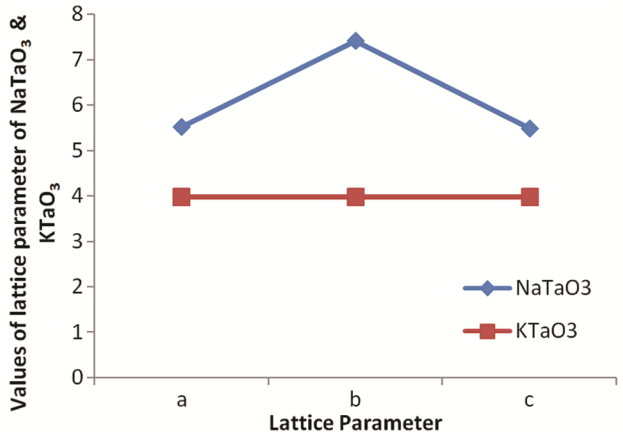

Fig. 2 — Values of lattice parameters for the composition of $NaTaO_3$ and $KTaO_3$

Table 3 — Variation of ultrasonic velocity by mixing potassiun in sodium tantalite.

Sample ($Na_{1-x}K_xTaO_3$) (x=0, 0.2, 0.3, 0.4, 0.5, 0.6 and 1)	Ultrasonic velocity$\times 10^4$ m/s
$NaTaO_3$	0.3026
$Na_{0.8}K_{0.2}TaO_3$	0.2679
$Na_{0.7}K_{0.3}TaO_3$	0.2701
$Na_{0.6}K_{0.4}TaO_3$	0.2626
$Na_{0.5}K_{0.5}TaO_3$	0.2526
$Na_{0.4}K_{0.6}TaO_3$	0.2448
$KTaO_3$	0.1838

orthogonal and are in general oblique to the wave normal.

6 Measurement

In the present study we have investigated the ultrasonic velocity in pure and mixed system of $Na_{1-x}K_xTaO_3$ where (x=0, 0.2, 0.3, 0.4, 0.5, 0.6 and 1). The velocity of ultrasonic wave has been measured at the transverse section of the prepared samples at frequency 5 MHz and at temperature 32 ºC with the help of ultrasonic c-scan system developed at NPL, New Delhi. Variation of ultrasonic velocity by mixing K on $NaTaO_3$ is tabulated in Table 3 and shown in Fig. 2.

7 Results and Discussion

The ultrasonic study of the prepared samples gives an understanding of the acoustical velocity and mechanism that could lead to the device application avenues of the present SPT system. From Fig. 3 and Table 3, it is observed that as soon as potassium is mixed with sodium tantalate at temperature 32 ºC and frequency 5 MHz, *i.e.*, K on $Na_{1-x}K_xTaO_3$, the

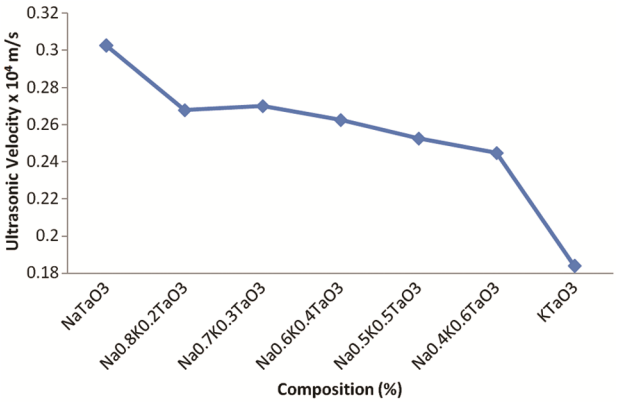

Fig. 3 — Variation of ultrasonic velocity by mixing potassium in sodium tantalate

ultrasonic velocity decreases continuously. From Fig. 1 it is observed that with increasing amount of K (Potassium) in place of sodium in $Na_{1-x}K_xTaO_3$, the attenuation coefficient for propagation of ultrasonic's in the mixed system is calculated to be 0.0577 Neper.

8 Conclusions

No abrupt changes in velocity occurred, even after the mixing of K in $NaTaO_3$. The detectable change in velocity varied from 0.3026×10^4m/s for $NaTaO_3$, 0.2679×10^4m/s for $Na_{0.8}K_{0.2}TaO_3$, 0.2448×10^4 m/s for $Na_{0.4}K_{0.6}TaO_3$ and 0.1838×10^4 m/s for $KTaO_3$ depending on the mixing, atomic arrangement, attenuation and hence on the temperature. From X-ray patterns, it was found that the structure of $NaTaO_3$ showing orthorhombic in nature while $KTaO_3$ is cubic, which is in agreement with the previously reported results[27].

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References

1 Wang D Y, Li K & Chan H L W, *Sens Actuators Phys*, 114 (2004) 1.
2 Cross L E, *Philos Mag*, 1 (1956) 56.
3 Bhatt S C & Semwal B S, *Indian J Pure Appl Phys*, 25 (1980) 174.
4 Bhatt S C & Semwal B S, *Solid State Ionics*, 23 (1987) 77.
5 Bhatt S C, Baluni G N, Naithani U C & Semwal B S, *Indian J Pure Appl Phys*, 26 (1988) 356.

6 Garland C W & Donald N, *Phys Rev*, 177 (1969) 971.
7 Havlin S, Litov E & Sompolinsky H, *Phys Rev B*, 13 (1976) 4999.
8 Litov E & Garland C W, *Phys Rev B*, 8 (1970) 971.
9 Barret H H, *Phys Rev*, 2 (1969) 178.
10 Jahagirdar D V, Arad B R, Patil C S & Shankarwar A G, *Indian J Pure Appl Phys*, 38 (2000) 645.
11 Sekar D M & Ramkumar D, *J Pure Appl Ultrason*, 24 (2002) 63.
12 Riyazuddeen & Islam N, *J Pure Appl Ultrason*, 19 (1997) 16.
13 Berret H, *Phys Lett* A, 26 (1968) 217.
14 Klerk *J*, *Rev Sci Inst*, 36 (1965) 1540.
15 Hagh N M, Jadidian B, Ashbahian E & Safari A*, IEEE Trans Ultrason Ferroelectr Freq Control*, 55 (2008) 214.
16 Chen J & Panda R, *IEEE Ultrason Symp*, 1 (2005) 235.
17 Tennery V J & Hang K W, *J Appl Phys*, 39 (1968) 4749.
18 Singh K, Uniyal M, Bhatt S C & Semwal B S, *Srilankan J Phys*, 1 (2001) 13.
19 Uniyal M, Singh K, Bhatt S, Bhatt S C, Pant R P, Suri D K & Semwal B S, *Indian J Pure Appl Phys*, 41 (2003) 305.
20 Uniyal M, Bhatt S C, Bhandari R, Ramchandra R M S, Pant R P & Semwal B S, *Indian J Pure Appl Phys*, 42 (2004) 341.
21 Bhatt S C, Singh K, Uniyal M & Semwal B S, *Indian J Pure Appl Phys*, 45 (2007) 613.
22 Uniyal M & Bhatt S C, *Indian J Pure Appl Phys*, 49 (2011) 350.
23 Uniyal M & Bhatt S C, *Indian J Pure Appl Phys,* 50 (2012) 248.
24 Bhatt S C & Uniyal M, *Int J Mater Chem*, 2 (2012) 47.
25 Uniyal M, Bhatt S C & Gairola R P, *Indian J Pure Appl Phys*, 50 (2012) 915.
26 Uniyal M & Bhatt S C *Indian J Pure Appl Phys*, 52 (2014) 391.
27 Vousden, *Acta Crystallogy*, 4 (1951) 373.
28 Wemple S H*, Phys Rev*, 137A (1965) 1575.
29 Shirane G, Newnham R & Pepinsky R, *Phys Rev*, 96 (1954) 581.