γ-Ray Attenuation Studies on NaI

A.S. Madhusudhan Rao[1], K. Narender[2]

Abstract—Temperature dependent γ- ray attenuation studies have been carried out by a γ- ray densitometer fabricated in our laboratory. The linear attenuation coefficients (μ_l) of NaI as a function of temperature in the range 300K-900K have been determined using different energies of γ-beam viz., Am (0.0595MeV), CO(1.173MeV & 1.332MeV). The coefficient of temperature dependence of density has been reported. The variation of density and thermal expansion of NaI in the temperature range have been studied and compared with the results available in the literature. The temperature dependence of linear attenuation coefficient, density and thermal expansion has been represented by second degree polynomials. Volumetric thermal expansion coefficient as a function of temperature has been reported by γ-ray attenuation studies for the first time.

Index Terms—Linear attenuation coefficient, density, thermal expansion,

1 INTRODUCTION

THE study of temperature dependence of fundamental thermophysical properties such as density and thermal expansion of solids is very important for understanding temperature variation of other properties like elastic constants, refractive indices, dielectric constants, thermal conductivity, diffusion coefficients and other heat transfer dimensionless numbers. Thermal expansion of solids is of technical importance as it determines the thermal stability and thermal shock resistance of the material. In general the thermal expansion characteristics decide the choice of material in high temperature applications in science and technology. Number of methods have evolved for the determination of density and thermal expansion of solids at high temperature Thermal expansion studies on alkali halides have been reported by several workers using X-ray diffraction [1-3], dilatometry [4, 5], Fabrey - Perot interference method [6] and by other theoretical models [7-14]. Using γ-ray attenuation technique W.D. Drotning [15] measured thermal expansion of isotropic solid materials at high temperatures. He studied thermal expansion of Aluminum at high temperatures and such studies have been extended by him to study the thermal expansion of other materials such as metals and glasses in the condensed state [16]. Authors employed γ-radiation attenuation technique for the determination of thermo physical properties of alkali halides by using γ-beam of energy Cs (0.66MeV) [17]. The γ- radiation attenuation technique for the determination of thermo physical properties in the condensed state offers several advantages over other methods at high temperatures. This is possible because the γ-ray is not in any kind of physical or thermal contact with the material and hence the thermal losses are also reduced and in addition eliminates sample and probe compatibility problem.

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Present work focuses the studies on temperature dependence of γ-ray attenuation, density and thermal expansion of Na in the temperature range 300K-900K using different energies of γ- beam viz., Am (0.0595MeV), CO (1.173MeV & 1.332MeV) for the first time. We also compare with the data available in literature in the temperature range 300K-900K. The present study has been carried out using a γ-ray densitometer fabricated in our laboratory which includes a programmable temperature controlled furnace (PTC).

2 THEORY

The technique of γ- ray attenuation method is based on the fundamental equation

$$I = I_0 \exp [- \mu \rho l] \quad (1)$$

where I_0, the intensity of γ- ray before passing through the sample, I, the intensity of γ- ray after passing through the sample, μ, the mass attenuation coefficient of the sample, ρ, the density of the sample and l, the thickness of the sample. It is clear from Equation (1) that any change in the temperature of the solid is accompanied by change in it's density causing a change in the measured intensity. The density and thermal expansion of the materials studied in the present work have been determined following the method suggested by Drotning [15]. The relation between coefficient of volumetric thermal expansion (α_ρ) and coefficient of linear thermal expansion (α_l) is given by

$$\alpha_\rho \equiv -3\, \alpha_l (1 - 2\, \alpha_l \Delta T). \quad (2)$$

where α_ρ and α_l are mean values over a temperature interval. $\Delta T = T_2 - T_1$ such that $\alpha_l = (l_2 - l_1) / (\Delta T) l_1$ and

$$\alpha_\rho = (\rho_2 - \rho_1) / (\Delta T)\, \rho_1$$

where $\rho_1 = \rho(T_1)$, $l_1 = l(T_1)$, etc.

Rewriting Equation (2) as

$$(\Delta T)^2 \alpha_l\, \alpha_\rho = z - (\Delta T)\, \alpha_l - (\Delta T)\, \alpha_\rho, \quad (3)$$

where z is defined by

$$z = \ln [I (T_1) I_0 (T_2) / I(T_2) I_0(T_1)] / (\mu \rho_1 l_1) = (\rho_2 l_2 / \rho_1 l_1) - 1 \quad (4)$$

Substituting for α_ρ from Equation (2) gives

$$-3 (\Delta T)^2 \alpha_l^2 (1 - 2\, \alpha_l\, \Delta T) = z - (\Delta T)\, \alpha_l + 3(\Delta T)\, \alpha_l (1 - 2\alpha_l \Delta T),$$

(5)

which can be rewritten as

$$6x^3 + 3x^2 - 2x - z = 0 \quad (6)$$

where $x \equiv (\Delta T)\,\alpha_l$ (7)

The intensities of γ-radiation with sample I and without sample I_0 are recorded at every temperature. At room temperature T_1, thickness of the sample l_1 is measured and using Equation (1) μ is determined. Further measurements of I and I_0 at different temperatures enable the determination of z by Equation (4) and hence x can be found from the solution of Equation (6). From the value of x, mean linear thermal expansion (α_l) can be determined as a function of temperature.

3 EXPERIMENTAL

NaI has NaCl structure. The pellet of NaI is prepared by compressing the NaI powder at a pressure of 2000 psi. The weight of the powder is 20.0 gm and the thickness of pellet is 1.40 cm. The pellet is sintered at 600 K. and then firmly mounted on the round sample holder made of flat stainless steel strip inserted into an air tight quartz tube. The precise sample temperature was measured using a thermocouple sensor. The thermocouple sensor tip was mounted on the sample holder ensuring a perfect physical contact with the sample for recording precise sample temperature. The sample holder along with the sample and thermocouple was slid through a cork into an air tight quartz tube and was fixed firmly. A diffusion pump was then connected to the sample holder tube for evacuation. For inert atmosphere, argon gas was introduced into the quartz tube through the sample holder tube. Then the quartz tube assembly along with the sample was slid into the programmable temperature controlled (PTC) furnace and fixed at appropriate position ensuring a perfect alignment of sample with collimation on either sides. A programmable temperature controlled furnace with sample inside the air tight quartz tube is introduced in the γ- radiation path allowing the beam to pass through the sample and to the detector without any interruption. The temperature of the sample is varied to study the attenuation at various temperatures. The PTC furnace was programmed in such a way that the furnace temperature is increased by 25K in every step starting from room temperature, and stabilizes there for 5 minutes. Temperature equilibrium is achieved within 5 minutes and temperature accuracy is ±1% of set point temperature after stabilization. Heating rate between each step of 25K is 5K/min. At each temperature the γ- ray counts of Am (0.0595MeV), CO (1.173MeV & 1.332MeV) with sample (I) and without sample (I_o) were detected and recorded using a multichannel analyzer. The recording of γ - ray counts was done for a period of 20 minutes at each programmed temperature. After a time of 20 minutes of isothermal holding, the difference of programmed and sample temperature (error) was ±1K of set point. Measurement of γ- ray attenuation counts at every step of temperature was repeated a minimum of nine times before and after the sample was introduced and the average value was considered in all our calculations. The cooling rate varied between 10K/min from 1300K-800K, 6K/min from 800K-500K, 4K/min from 500K-400K and 2K/min thereafter up to 300K. The γ- ray counts were recorded while heating and cooling the sample. This procedure was repeated until the desired temperature range was covered in each case. The gamma radiation detector used in our study is a sodium iodide – thallium activated detector. The 0.0762 m diameter and 0.0762 m thick crystal is integrally coupled to a 0.0762 m diameter photo multiplier tube (PMT). The PMT has a 14 pin base and can be mounted on two types of PMT preamplifier units. The one used in our study is a coaxial in-line pre-amplifier. The detector has a resolution of 8.5% for 0.662MeV of ^{137}Cs.

4 RESULTS AND DISCUSSION

The results obtained for the temperature dependence of the linear attenuation coefficient (μl) of NaI at different energies of γ- beam are presented in Table 1. The density (ρ) and coefficient of linear thermal expansion (α) of NaI are presented along with data available in literature in Table 1. The measurements have been carried out in solid phase only. The experimental data obtained in the present work for the density; also the linear attenuation coefficient and coefficients of thermal expansion have been fit to a least-squares quadratic polynomial of the form

$$\rho(T) = a + bT + cT^2 \quad (8)$$

Since the measurements have been made in the limited temperature range the coefficient of volumetric thermal expansion (CVTE) was calculated using the equation

$$\beta = \frac{1}{\rho}\left(\frac{d\rho}{dT}\right) \quad (9)$$

where dρ/dT is the first derivative of density with respect to the absolute temperature which is determined from Equation (8). The mass attenuation coefficients (μ) of NaI for γ-energies of Am (0.0595MeV), CO (1.173MeV & 1.332MeV) are determined to be 658.90×10^{-3} m^2kg^{-1}, 5.34×10^{-3} m^2kg^{-1} and 4.99×10^{-3} m^2kg^{-1} respectively. The experimental values reported in the present work are in good agreement with the values calculated from National Institute of Standards and Technology (NIST-X-COM).

The density of NaI decreases from a value of 3670 kgm^{-3} at 300 K to a value of 3178kgm^{-3} at 900 K giving rise to a decrease of about 13.40% in this temperature range. The decrease in density in NaI with temperature is due to the increase in the equilibrium concentration of thermally generated Schottky defects. The values of density seem to have not been affected much with irradiation of γ-rays. The values of coefficient of linear thermal expansion at different temperatures obtained in the present work agree well with data reported in the literature. The values of coefficient of linear thermal expansion obtained in the present work are given in Table 1 along with the values reported in the literature. Rapp and Merchant [18] reported the values of thermal expansion of NaI in the temperature range 300-600 K using a high precision differential dilatometer technique. The agreement between the values of present work and the values reported by them is good. Sunil and Sharma [19] calculated thermoelastic proper-

ties of NaI at high temperature using Born model for ionic solids in the temperature range 300-900 K. The agreement between the calculated and measured results is good, although, the calculated values are slightly greater than the measured values.

However, the results on variation of density and linear attenuation coefficient of NaI with temperature are not available from other methods for comparison. The temperature dependence of μ_l for γ-energies of Am (0.0595MeV), CO (1.173MeV & 1.332MeV) have been represented by following equations respectively,

$$\mu_l(T) = (2500.05) + (-4.03\times10^{-1})T + (-5.22\times10^{-5})T^2 \quad (10)$$

$$\mu_l(T) = (20.32) + (-3.27\times10^{-3})T + (-4.23\times10^{-7})T^2 \quad (11)$$

$$\mu_1(T) = (18.99) + (-3.05\times10^{-3})T + (-3.96\times10^{-7})T^2 \quad (12)$$

The temperature dependence of ρ and $\Delta l / l$ have been represented by following equations,

$$\rho(T) = (3805.97) + (-6.12\times10^{-1})T + (-7.91\times10^{-5})T^2 \quad (13)$$

$$\Delta l / l(T) = (8.25\times10^{-3}) + (-9.54\times10^{-6})T + (9.49\times10^{-8})T^2 \quad (14)$$

The data obtained for α and β have been fit to a second degree polynomial in T and are given by equations,

$$\alpha(T) = (36.67\times10^{-6}) + (1.39\times10^{-8})T + (44.70\times10^{-12})T^2 \quad (15)$$

$$\beta(T) = (1.63\times10^{-4}) + (5.94\times10^{-8})T + (2.46\times10^{-11})T^2 \quad (16)$$

The values of coefficient of linear thermal expansion (α) obtained in the present work for NaI have been shown in Fig.1 along with the results reported in literature by Rapp and Merchant [18], Sunil and Sharma [19] for comparison. The values of coefficient of volumetric thermal expansion (β) obtained in the present work for NaI have been shown in Fig.2.

The uncertainty in the measured physical parameters depends on uncertainty in the furnace temperature and measurement of the mass attenuation coefficient, which has been estimated from errors in intensities I_0, I and thickness (l) using the following relation (17) [20]

$$\Delta(\mu_m) = \frac{1}{\rho l}\left[\left(\frac{\Delta I_0}{I}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \left(\ln\frac{I_0}{I}\right)^2 + \left(\frac{\Delta l}{l}\right)^2\right]^{1/2} \quad (17)$$

Where ΔI_0, ΔI and Δl are the errors in the intensities I_0, I and thickness (l) respectively. In this experiment, the intensities I_0 and I have been recorded for the same time and under the same experimental conditions. Estimated error in these measurements does not exceed 2%.

5 CONCLUSION

The results on the variation of linear attenuation coefficient for different energies of gamma beam, density and thermal expansion with temperature of NaI have been reported and these variations have been represented by second degree polynomials. The steep fall observed in the values of density in the temperature range 300-375 K is due to evaporation of water in the sample in this temperature range. The values obtained for coefficient of thermal expansion using γ-ray attenuation technique agreement is good with the data from other methods. This renders reliability on the data obtained for linear attenuation coefficient and density at different temperatures using γ-ray attenuation technique.

5 FIGURES

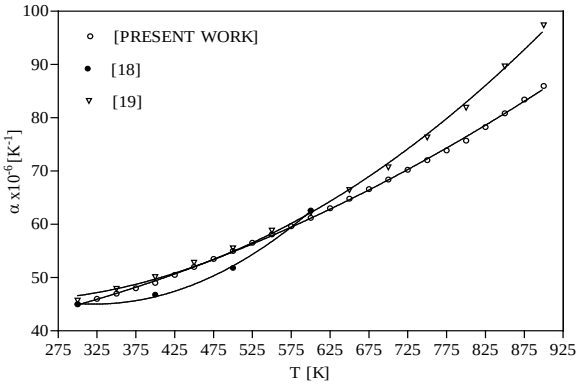

Fig.1. Comparison of coefficient of linear thermal expansion of NaI with available data in literature [18-19] and present work

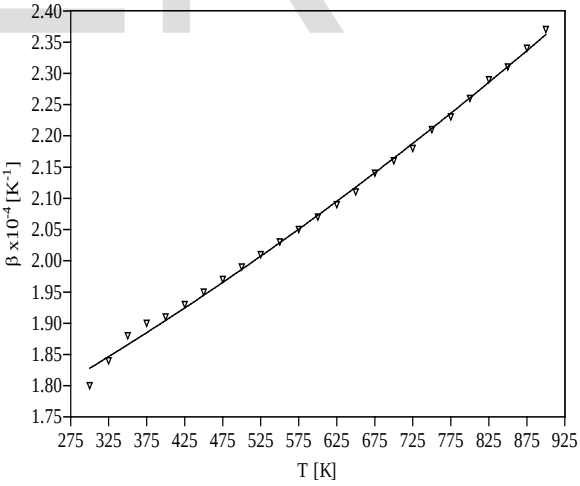

Fig.2. Variation of coefficient of volumetric thermal expansion of NaI with temperature

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Table 1

Variation of linear attenuation coefficient for different gamma energies, density and coefficient of linear and volume thermal expansion of NaI with temperature.

T [K]	μ_l [m^{-1}]			ϱ [kgm^{-3}]	-d ϱ/dT	(Δl/l) 10^{-3}	α [$10^{-6}K^{-1}$]			β [$10^{-4}K^{-1}$]
	Am (0.0595) MeV	CO					[Present work]	[18]	[19]	
		1.173 MeV	1.332 MeV							
300	2418.5	19.60	18.31	3670	0.659	13.50	45.00	45.00	45.67	1.80
325	2379.6	19.28	18.02	3611	0.663	14.97	46.00			1.84
350	2340.8	18.97	17.72	3552	0.667	16.45	47.00		47.88	1.88
375	2333.5	18.91	17.67	3541	0.671	18.02	47.99			1.90
400	2326.9	18.86	17.62	3531	0.675	19.60	48.99	46.80	50.10	1.91
425	2318.4	18.79	17.55	3518	0.679	21.50	50.49			1.93
450	2309.8	18.72	17.49	3505	0.683	23.40	51.99		52.80	1.95
475	2300.6	18.64	17.42	3491	0.687	25.45	53.49			1.97
500	2291.3	18.57	17.35	3477	0.691	27.50	54.99	51.80	55.50	1.99
525	2281.5	18.49	17.28	3462	0.695	29.72	56.54			2.01
550	2271.6	18.41	17.20	3447	0.699	31.95	58.10		58.83	2.03
575	2261.7	18.33	17.13	3432	0.703	34.34	59.65			2.05
600	2251.1	18.24	17.05	3416	0.707	36.72	61.20	62.60	62.17	2.07
625	2239.9	18.15	16.96	3399	0.711	39.42	63.00			2.09
650	2228.7	18.06	16.88	3382	0.715	42.12	64.80		66.43	2.11
675	2216.9	17.96	16.79	3364	0.719	45.01	66.60			2.14
700	2205.0	17.87	16.70	3346	0.723	47.88	68.40		70.70	2.16
725	2193.2	17.77	16.61	3328	0.727	50.96	70.23			2.18
750	2180.6	17.67	16.51	3309	0.731	54.04	72.05		76.30	2.21
775	2168.1	17.57	16.42	3290	0.735	57.30	73.88			2.23
800	2155.6	17.47	16.32	3271	0.739	60.56	75.70		81.90	2.26
825	2140.4	17.34	16.21	3248	0.743	64.64	78.27			2.29
850	2125.3	17.22	16.09	3225	0.746	68.72	80.85		89.63	2.31
875	2109.5	17.09	15.97	3201	0.75	73.05	83.42			2.34

900	2094.3	16.97	15.86	3178	0.754	77.39	85.99	97.37	2.37

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