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Thermophysical Properties of NaCl, NaBr and NaF by γ-Ray Attenuation Technique

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ABSTRACT

The *γ*-ray densitometer has been designed and fabricated in our laboratory and carried out studies on temperature dependent *γ*-ray attenuation and thermo physical properties of NaCl, NaBr and NaF. The linear attenuation coefficients (μl) for the pellets of NaCl, NaBr and NaF as a function of temperature have been determined. The coefficients of temperature dependence of density have been reported. The variation of density and thermal expansion of NaCl, NaBr and NaF in the temperature range of 300 K - 1000 K have been studied and compared with results available in the literature. The temperature dependence of density and thermal expansion has been represented by linear equations. Volume thermal expansion coefficients have been reported.

Keywords: Linear Attenuation Coefficient; Density; Thermal Expansion; *γ*-Ray Densitometer

1. Introduction

Density and Thermal expansion are fundamental thermo physical properties of solids. The study of temperature dependence of these properties is very important in understanding the 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 for the construction of metrological instruments and in the choice of container material in nuclear fuel technology. Number of methods have evolved for the determination of density and thermal expansion of solids at high temperature like Archimedean method, pycnometry, dilatometry, electromagnetic levitation, Method of maximal pressure in gas bubble, method of sessile drop, hydrostatic weighing, high temperature electrostatic levitation and gamma ray densitometry. 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 and type 303 stainless steel at high temperatures and such studies have been extended by him to study the thermal expansion of metals and glasses in the condensed state [16]. 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.

As NaCl, NaBr and NaF are isotropic solids; we extended, for the first time, the *γ*-ray attenuation technique, to carry out the studies on temperature variation of *γ*-ray attenuation and thermo physical properties of NaCl, NaBr and NaF. In this communication, we report the temperature dependence of linear attenuation coefficient of *γ*-radiation, density and thermal expansion of NaCl, NaBr and NaF in the temperature range 300 K - 1000 K. In order to carry out this work, we have designed and fabricated a *γ*-ray densitometer and a programmable temperature controlled furnace (PTC) which can reach up to a temperature of 1300 K in our laboratory. The data obtained in the present work for coefficient of linear thermal expansion of NaCl, NaBr and NaF as a function of temperature have been compared with experimental and theoretical data available in literature.

2. Theory

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

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

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_1) is given by

$$\alpha_\rho - 3\alpha_1\left(1 - 2\alpha_l \Delta T\right) \quad (2)$$

where α_ρ and α_l are mean values over a temperature interval. $\Delta T = T_2 - T_1$ such that

$$\alpha_l = \left(l_2 - l_1\right)/\left(\Delta T\right) l_1 \text{ and } \alpha_\rho = \left(\rho_2 - \rho_1\right)/\left(\Delta T\right)\rho_1$$

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

Rewriting Equation (2) as

$$\left(\Delta T\right)^2 \alpha_1 \alpha_\rho = z - \left(\Delta T\right)\alpha_1 - \left(\Delta T\right)\alpha_\rho, \quad (3)$$

where z is defined by

$$\begin{aligned} Z &= \ln\left[I\left(T_1\right)I_0\left(T_2\right)/I\left(T_2\right)I_0\left(T_1\right)\right]/\left(\mu\rho_1 l_1\right) \\ &= \left(\rho_2 l_2 \rho_1 l_1\right) - 1 \end{aligned} \quad (4)$$

Substituting for α_ρ from Equation (2) gives

$$\begin{aligned} &-3\left(\Delta T\right)^2 \alpha_l^2 \left(1 - 2\alpha_l \Delta T\right) \\ &= z - \left(\Delta T\right)\alpha_l + 3\left(\Delta T\right)\alpha_l\left(1 - 2\alpha_l \Delta T\right) \end{aligned} \quad (5)$$

which can be rewritten as

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

where

$$x \equiv \left(\Delta T\right)\alpha_l \quad (7)$$

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

3.1. γ-Ray Densitometer

The experimental setup used for determination of density of materials utilizing γ-ray attenuation technique is called a γ-ray densitometer. 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 block diagram of γ-ray densitometer is shown in **Figure 1**.

3.1.1. Stage-1

Consists of source housing, programmable temperature control (PTC) furnace with sample holder inside the air tight quartz tube, lead and stainless steel collimators, detector housing. The γ-radiation detector used in our study is a sodium iodide-thallium activated detector. The 3 inch diameter and 3 inch thick crystal is integrally coupled to a 3 inch 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 ^{137}Cs.

3.1.2. Stage-2

Consists of amplifier which is fed to multi-channel analyzer where γ-ray spectrum is analyzed.

3.1.3. Stage-3

Consists of PC with Nets win Software and printer to calculate and to store the data. PTC furnace is an electric muffle furnace consisting temperature sensors along with programmable logic controllers (PLC) used for monitoring and recording the temperature. Type K thermocouple is used in the furnace for wide operating temperatures. The operation of furnace is monitored with the help of electronic panel. The cross sectional view of γ-ray densitometer is shown in **Figure 2**.

The samples studied in the present work were in the form of pellets. The pellets were prepared with fine powder of NaCl, NaBr and NaF with a diameter of 20 mm with varying thicknesses with a die set by applying hydraulic press pressure. NaCl, NaBr and NaF pellets were sintered at a temperature of 800 K for densification. The pellet was 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

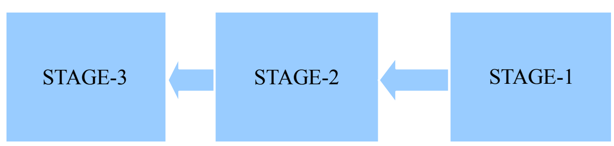

Figure 1. Block Diagram of γ-ray densitometer.

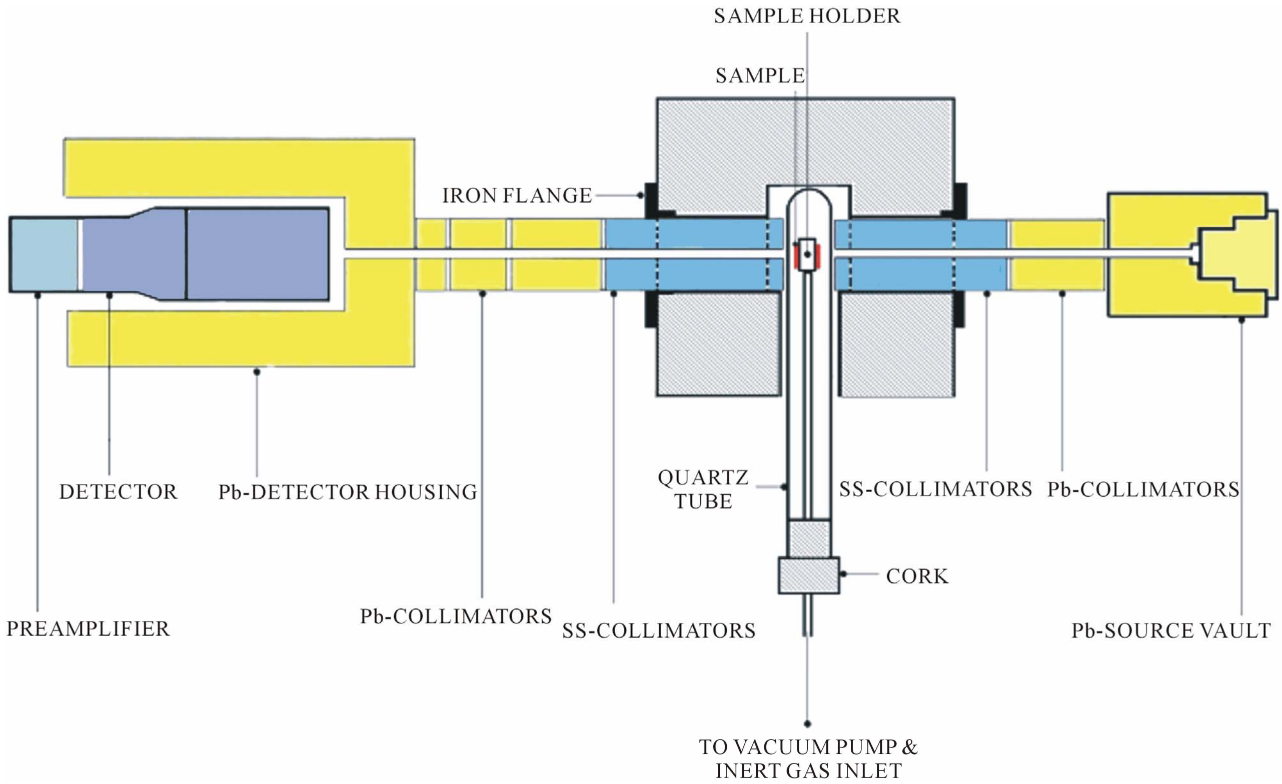

Figure 2. Cross-section view of *γ*-ray densitometer.

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. The PTC furnace was programmed in such a way that the furnace temperature is increased by 25 K in every step starting from room temperature, and stabilizes there for a certain length of time. At each temperature the γ-ray counts of ^{137}Cs with sample (I) and without sample (I_o) were detected and recorded using a multichannel analyzer. Measurement of γ-ray attenuation counts at every step of temperature was repeated a minimum of nine times. 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.

4. Results and Discussion

The results obtained for the temperature dependence of the linear attenuation coefficient (μ_l), density (ρ) and the coefficient of linear thermal expansion (α) of NaCl, NaBr and NaF are summarized in **Table 1**. The measurements have been carried out in solid phase only. The experimental data obtained in the present work for the density have been fit to a linear equation of the form

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

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

$$\beta = (1/\rho)(\mathrm{d}\rho/\mathrm{d}T) \quad (9)$$

where $(\mathrm{d}\rho/\mathrm{d}T)$ is the first derivative of density with respect to the absolute temperature which is determined from Equation (8). The variation of linear attenuation coefficient and variation of density with temperature of NaCl, NaBr and NaF have been shown in **Figures 3** and **4** respectively. The mass attenuation coefficients (μ) of of NaCl, NaBr and NaF are found to be 7.428×10^{-3} m^2·kg^{-1}, 7.164×10^{-3} m^2·kg^{-1} and 7.289×10^{-3} m^2·kg^{-1} respectively these results agree well with the calculated values 7.434×10^{-3} m^2·kg^{-1}, 7.187×10^{-3} m^2·kg^{-1} and 7.367×10^{-3} m^2·kg^{-1} respectively from National Institute of Standards and Technology (NIST-X-COM).

4.1. NaCl

The density of NaCl decreases from a value of 2165 kg·m^{-3} at 300 K to a value of 1904 kg·m^{-3} at 1000 K a

Table 1. Variation of linear attenuation coefficient, density and coefficient of linear thermal expansion of NaCl, NaBr and NaF with temperature.

	NaCl			NaBr			NaF		
TK	μ_l m^{-1}	ρ kg·m^{-3}	$\alpha(10^{-6})$ K^{-1}	μ_l m^{-1}	ρ kg·m^{-3}	$\alpha(10^{-6})$ K^{-1}	μ_l m^{-1}	ρ kg·m^{-3}	$\alpha(10^{-6})$ K^{-1}
300	16.08	2165	39.80	23.00	3210	41.75	18.65	2558	33.70
325	15.86	2135	40.23	22.66	3163	42.35	18.42	2528	34.17
350	15.63	2105	40.67	22.32	3116	42.94	18.20	2497	34.63
375	15.59	2099	41.78	22.26	3108	43.59	18.16	2492	35.27
400	15.54	2093	42.90	22.21	3100	44.23	18.12	2486	35.90
425	15.50	2087	43.65	22.14	3090	45.18	18.08	2480	36.51
450	15.46	2081	44.40	22.07	3081	46.12	18.04	2475	37.12
475	15.42	2075	45.10	22.00	3072	47.02	18.00	2469	37.71
500	15.37	2069	45.80	21.94	3062	47.91	17.95	2463	38.30
525	15.32	2062	46.97	21.86	3051	49.07	17.91	2456	39.03
550	15.27	2055	48.13	21.78	3040	50.23	17.86	2450	39.76
575	15.22	2050	48.52	21.67	3025	52.53	17.81	2444	40.33
600	15.18	2044	48.90	21.56	3010	54.82	17.77	2437	40.90
625	15.14	2039	49.05	21.47	2998	56.05	17.71	2430	41.70
650	15.11	2034	49.20	21.39	2985	57.29	17.66	2423	42.50
675	15.04	2025	50.50	21.28	2971	58.96	17.62	2417	42.85
700	14.98	2017	51.80	21.18	2956	60.63	17.57	2411	43.20
725	14.93	2010	52.57	21.07	2941	62.41	17.51	2403	44.15
750	14.88	2003	53.33	20.96	2925	64.20	17.45	2394	45.10
775	14.82	1995	54.52	20.83	2907	66.44	17.40	2388	45.50
800	14.75	1986	55.70	20.70	2890	68.68	17.36	2381	45.90
825	14.69	1977	56.95	20.57	2871	70.91	17.30	2373	46.77
850	14.62	1968	58.20	20.44	2853	73.13	17.23	2364	47.63
875	14.55	1959	59.55	20.28	2831	76.09	17.18	2356	48.37
900	14.48	1949	60.90	20.12	2809	79.04	17.12	2348	49.10
925	14.39	1937	63.19	-	-	-	17.06	2340	49.77
950	14.29	1924	65.47	-	-	-	17.00	2332	50.43
975	14.22	1914	66.84	-	-	-	16.93	2323	51.42
1000	14.14	1904	68.20	-	-	-	16.86	2313	52.40

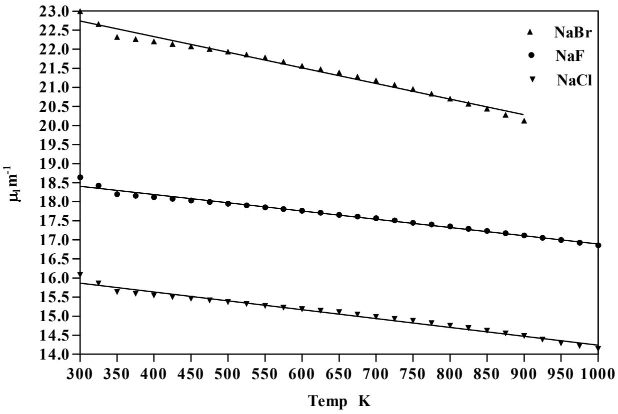

Figure 3. Variation of linear attenuation coefficient of NaCl, NaBr and NaF with temperature.

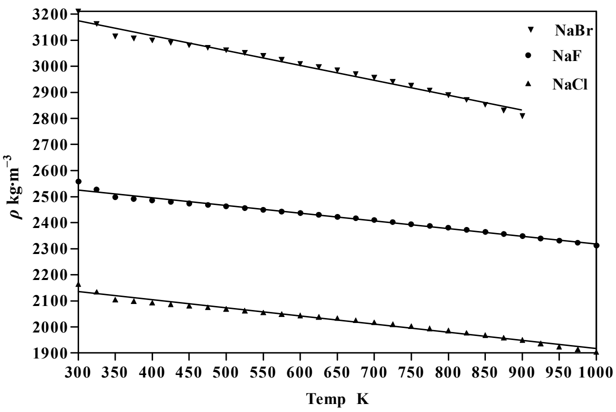

Figure 4. Variation of density of NaCl, NaBr and NaF with temperature.

decrease of about 12.06%. The temperature dependence of density is a negative linear function of temperature. For NaCl the temperature dependence of density is represented by linear equation

$$\rho(T) = (2230 \pm 5.822) + \left(-3.130\times10^{-1} \pm 8.527\times10^{-3}\right)T \quad (10)$$

The coefficient of temperature dependence of density is −0.3130 kg·m^{-3}·K^{-1} and the coefficient of volume thermal expansion is 1.446 × 10^{-4} K^{-1} in the temperature range 300 K - 1000 K. The thermal expansion increases linearly with temperature and the results on thermal expansion in the temperature range from 300 K to 1000 K have been analyzed by least squares method and is represented by the linear equation

$$\Delta l/l = (-15.140 \pm 1.596) + \left(7.734\times10^{-2} \pm 2.337\times10^{-3}\right)\Delta T \quad (11)$$

4.2. NaBr

The density of NaBr decreases from a value of 3210 kg·m^{-3} at 300 K to a value of 2809 kg·m^{-3} at 900 K a decrease of about 12.49%. The temperature dependence of density is a negative linear function of temperature. For NaBr the temperature dependence of density is represented by linear equation

$$\rho(T) = (3345 \pm 10.15) + \left(-5.715\times10^{-1} \pm 1.620\times10^{-2}\right)T \quad (12)$$

The coefficient of temperature dependence of density is −0.5715 kg·m^{-3}·K^{-1} and the coefficient of volume thermal expansion is 1.78 × 10^{-4} K^{-1}. The thermal expansion increases linearly with temperature and the results on thermal expansion in the temperature range from 300 K to 900 K have been analyzed by least squares method and is represented by the linear equation

$$\Delta l/l = (-21.20 \pm 2.2) + \left(9.475\times10^{-2} \pm 3.511\times10^{-3}\right)\Delta T \quad (13)$$

4.3. NaF

The density of NaF decreases from a value of 2558 kg·m^{-3} at 300 K to a value of 2313 kg·m^{-3} at 1000 K a decrease of about 9.58%. The temperature dependence of density is a negative linear function of temperature. For NaF the temperature dependence of density is represented by linear equation

$$\rho(T) = (2614 \pm 4.964) + \left(-2.962\times10^{-1} \pm 7.270\times10^{-3}\right)T \quad (14)$$

The coefficient of temperature dependence of density is −0.2962 kg·m^{-3}·K^{-1} and the coefficient of volume thermal expansion is 1.16 × 10^{-4} K^{-1} in the temperature range 300 K - 1000 K. The thermal expansion increases linearly with temperature and the results on thermal expansion in the temperature range from 300 K to 1000 K have been analyzed by least squares method and is represented by the linear equation

$$\Delta l/l = (-10.10 \pm 0.7465) + \left(5.975\times10^{-2} \pm 1.093\times10^{-3}\right)\Delta T \quad (15)$$

The results obtained for the coefficient of thermal expansion in the present work agree well with the values reported from X-ray studies, dilatometry and theoretical models [17-22] as seen from **Figures 5-7** of NaCl, NaBr and NaF respectively. However, the results on variation of density and linear attenuation coefficient of NaCl, NaBr and NaF with temperature are not available from other methods for comparison.

5. Conclusions

The pellets were prepared with fine powder of NaCl,

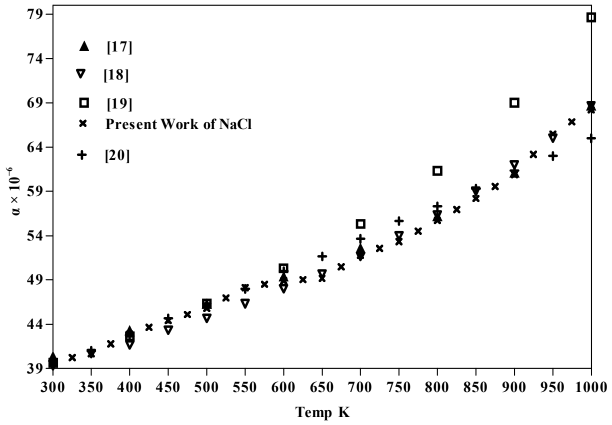

Figure 5. Comparison of coefficient of linear thermal expansion of NaCl with available data.

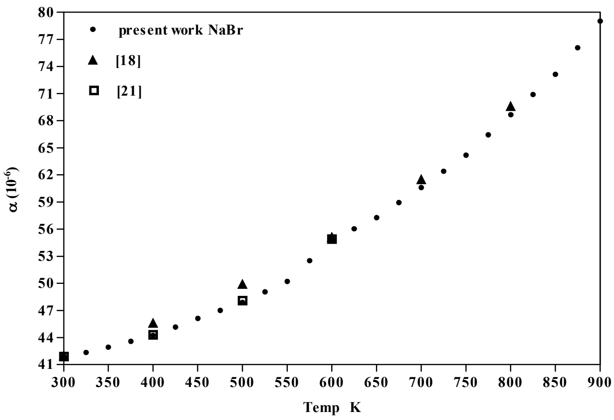

Figure 6. Comparison of coefficient of linear thermal expansion of NaBr with data available.

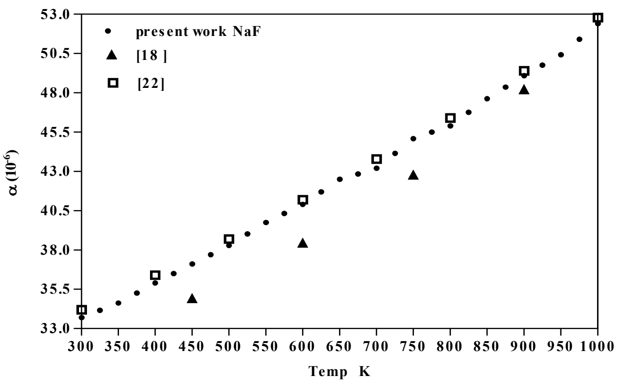

Figure 7. Comparison of coefficient of linear thermal expansion of NaF with data available.

NaBr and NaF with a diameter of 20 mm with varying thicknesses with a die set by applying hydraulic press pressure. The γ-ray attenuation measurements have been made using a γ-ray densitometer designed and fabricated in our laboratory. The results on the variation of linear attenuation coefficient, density and linear thermal expansion with temperature of these pellets have been reported and these variations have been represented by linear equations. The results on these pellets by using γ-ray attenuation technique are being reported for the first time.

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