Density of Molten Terbium Chloride

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The density of molten terbium chloride at 993 to 1213 K, as measured by the γ-ray attenuation method, is found to be $d = [3.937 \pm 0.028 - (4.76 \pm 0.25) \cdot 10^{-4} T]$ g/cm^3. The characteristic state parameters of molten $TbCl_3$ are found to be 1129 K and 78.0 ml for T^* and v^*, respectively.

Key words: Density; Terbium Chloride; Holmium-166m; Gamma-ray Attenuation; Molten Salt.

1. Introduction

Data on the densities of molten chlorides are required in the nuclear industry. As for the lanthanide chlorides, such data exist for $LaCl_3$ [1, 2], $PrCl_3$, $NdCl_3$, $GdCl_3$, and $DyCl_3$ [3], and $NdCl_3$ [4]. The present paper reports the density of molten $TbCl_3$, measured by the γ-ray attenuation method.

2. Experimental

Anhydrous (99.99%) $TbCl_3$ was obtained from Aldrich (Milwaukee, WI, USA) and used without further purifications.

It was handled in an argon-filled dry box with a water content < 2 ppm and an oxygen content < 1 ppm.

A ^{166}Ho radiation source (2.27 MBq) was used because of its desirable decay characteristics ($t_{1/2} =$ 1200y, up to 8 photons per disintegration, more than 10% branching ratio). A multi-channel pulse height analyzer, coupled with a Ge-detector (1.7 keV FWHM at 1332.5 keV, 23% relative efficiency) was used to obtain the γ-ray spectrum of the ^{166}Ho.

A schematic diagram of the experimental set-up is shown in Figure 1.

The radiation source was placed in a lead shield. A tungsten alloy and lead were used as collimator. The sample in a quartz cell was set in a furnace. Nickel foils and cupper plates were used to prevent thermal effects on the radiation source and the Ge-detector.

Gamma-ray spectra were first taken with an empty quartz cell and the cell filled with water. After the cell was charged with anhydrous Terbium chloride, it was sealed. The γ-ray spectra were taken at various controlled temperatures and analyzed with a standard method.

The attenuation of γ-ray is expressed by

$$I_i = I_{0i} \exp(-\sigma_i \rho x), \tag{1}$$

where I_i and I_{0i} are the γ_i beam intensity after and before passing through the material, ρ is the density of the material, and x the length of the specimen. The total attenuation coefficient σ_i is given by

$$\sigma_i = \sigma_{ie} + \rho_{ip} + \rho_{ic} + \rho_{is}, \tag{2}$$

where σ_{ie}, ρ_{ip}, ρ_{ic} and ρ_{is}, are the attenuation coefficient by the photoelectric process pair-production, Compton scattering, and coherent scattering, respectively. The attenuation coefficients were linearly fitted to the values given in [5].

A γ-ray loses its energy totally by the photoelectric process or by 1.01 MeV through pair-production. In the Compton scattering process, the energy i' of a γ-ray scattered from a γ-ray with the energy i is given by

$$i' = i/(1 + (i/mc^2)(1 - \cos\theta)), \tag{3}$$

where θ is scattering angle, m the rest mass of the electron, and c the velocity of light. For $\theta \to 0$ the difference between i' and i becomes too small to be resolved with the γ-ray spectrometer coupled with the Ge-detector. The contribution of the scattered γ-ray with energy i' to the γ-ray with energy i is constant for a given detection system at a given γ-ray energy, as can be seen from (3). As water is used to obtain the cell

Parameters for the calculation of the cell length and the density of molten terbium chloride at 1033 K.

γ-ray from ^{166m}Ho		Calculation of the cell length with water Density of water at 25°C = 0,9956						Calculation of the density of molten $TbCl_3$ at 1033 K				
Energy	Blanch	I_0	Attenuation Coefficient for water (cm^2/g)			I	Cell	Attenuation Coefficient for $TbCl_3$ (cm^2/g)			I	Density of $TbCl_3$
MeV	%	Peak area (counts)	Coherent (σ_{is})	$\sigma_{ie}+\sigma_{ic}$ $+\sigma_{ip}$	Total (corrected)	Peak area (counts)	Length (cm)	Coherent (σ_{is})	$\sigma_{ie}+\sigma_{ic}$ $+\sigma_{ip}$	Total (corrected)	Peak area (counts)	g/cm^3
0,1844	73,9	8904943	0,001617	0,13873	0,13948	6961624	1,773	0,03216	0,43764	0,46569	507290	3,459
0,2805	30,1	3151251	0,000715	0,11990	0,12023	2544085	1,788	0,01476	0,19793	0,21081	866443	3,444
0,4109	11,7	954535	0,000332	0,10365	0,10381	793725	1,785	0,00715	0,11939	0,12563	443321	3,432
0,5298	10,3	677814	0,000200	0,09382	0,09392	573883	1,780	0,00437	0,09369	0,09751	373464	3,437
0,7117	59,3	3211229	0,000111	0,08249	0,08254	2775452	1,775	0,00246	0,07480	0,07694	2005902	3,438
0,7523	13,2	689414	0,000099	0,08042	0,08046	597464	1,787	0,00221	0,07206	0,07399	437646	3,453
0,8103	63,3	3113580	0,000086	0,07768	0,07772	2715648	1,767	0,00191	0,06863	0,07029	2023719	3,446
0,8306	10,7	520903	0,000082	0,07681	0,07685	454767	1,775	0,00182	0,06753	0,06911	340731	3,453
			$F = 0,46$			Average	1,779	$F = 0,872$			Average	3,445
						σ	0,008				σ	0,009

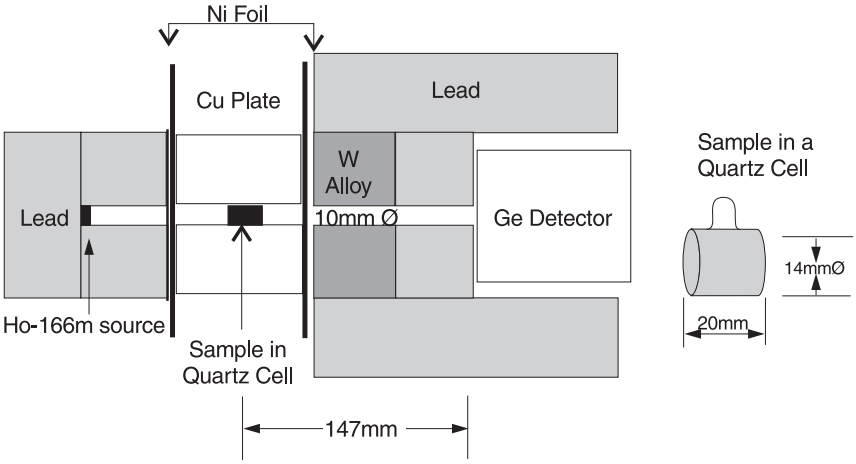

Fig. 1. Experimental set-up for the γ-ray attenuation method.

Table 2. Characteristic state parameter T^* and v^*.

Compounds	$T^*(k)$	$v^*(ml)$	ref.
$LaCl_3$	1133,0	77,12	[1]
$PrCl_3$	1138,8	78,05	[3]
$NdCl_3$	1186,0	78,90	[4]
$GdCl_3$	1132,9	77,87	[3]
$TbCl_3$	1127,8	78,02	This work
$DyCl_3$	1559,2	85,25	[3]

Calculated from density data.

length, there is no contribution of scattered γ-rays to the density measurements. On the other hand, with the coherent scattering the γ-ray energy i does not change and a strong anisotropic scattering occurs: some of the scattered γ-rays keeps their course and reach the detector. Equation (2) must therefore be replaced by

$$\sigma_i = \sigma_{ie} + \rho_{ip} + \rho_{ic} + \rho_{is}F, \tag{4}$$

where F is the probability that the scattered γ-ray deviates coherently from its original path. From the attenuation factors, $I_i = I_{0i}$ obtained with water and the total attenuation coefficients for water calculated with (4) and the density of pure water at room temperature, the length of the cell was calculated by (1). As the contribution of ρ_{is} to σ_i decreases rapidly with increasing energy of the γ-ray, the factor F was chosen so as to minimize the standard deviation of the length of the cell calculated from γ-rays with different energies. For the calculation of the density of the melt, the factor F was chosen so as to minimize the standard deviation of the density of molten terbium chloride calculated from γ-rays with different energies.

3. Results and Discussions

The parameters used for the calculation of the cell length and the density of molten $TbCl_3$ at 1033 K are

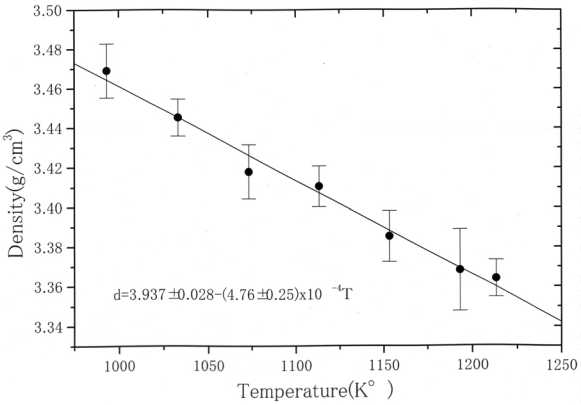

Fig. 2. The densities of molton tertium chloride as a function of temperature.

summarized in Table 1.The length of the cell was obtained with a standard deviation of about 0.5%. The density d of molten $TbCl_3$ as a function of temperature is shown in Figure 2. It resulted in the empirical equation $d = [3.973 \pm 0.028 - (4.76 \pm 0.25) \cdot 10^{-4} T]$ g/cm^3, where T is temperature in Kelvin.

According to corresponding-state correlations [6, 7], the molar volume v and viscosity η of many monovalent and divalent molten salts can be predicted using the characteristic state parameters T^*, v^* and η^*:

$$\tilde{V} = 0.7452 + 0.1049\tilde{T} + 0.1502\tilde{T}^2, \quad (5)$$

$$\ln\tilde{\eta} = -1.998 + 2.065\tilde{T} - 0.0573\tilde{T}^8, \quad (6)$$

where $\tilde{T}$ is $\frac{T}{T^*}$, $\tilde{V}$ is $\frac{v}{v^*}$, $\tilde{\eta} = \frac{\eta}{\eta^*}$.

v^* and η^* are the molar volume and viscosity at T^*. T^* is best determined [7] from density measurements over a sufficient temperature range to find the thermal expansivity. Assuming that the corresponding-state correlations can be applied to trivalent molten salts, T^* is calculated to be 1127.8 K for molten terbium chloride.

The characteristic state parameters T^* and v^* of the lighter lanthanide chlorides from $LaCl_3$ to $GdCl_3$ differ considerably from that of the heavier $DyCl_3$, as can be seen in Table 2. The characteristic state parameters T^* and v^* of $TbCl_3$ are similar to those of the lighter lanthanide chlorides.

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