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Density Measurement of Molten $NdCl_3$-NaCl and $NdCl_3$-KCl Systems by Gamma Ray Attenuation

Tadashi SATO*, Yoshihiro OKAMOTO and Toru OGAWA
Japan Atomic Energy Research Institute Tokai, Ibaraki 319-1195, Japan

Densities of molten chlorides mixture of $NdCl_3$-NaCl and $NdCl_3$-KCl systems have been measured using γ-ray attenuation method as a function of temperature. Gamma-ray spectra of Ho-166m radiation source were taken before and after passing through the molten chlorides in a quartz cell by a Ge-detector coupled with multi-channel pulse analyzer. The density was calculated with observed attenuation factor, cross section data of photons for relevant elements and length of the cell. The length of the cell was also determined with the attenuation factor observed on water, cross section data of photons for water and density of pure water at room temperature. The measured densities of molten $NdCl_3$-NaCl and $NdCl_3$-KCl systems were compared with literature data by dilatometric method. The excess molar volume isotherm at 1100 Kelvin for $NdCl_3$-NaCl and $NdCl_3$-KCl systems showed the maximum at $NdCl_3$ composition of around 0.2. A small negative excess molar volume was found at $NdCl_3$composition of 0.75 for $NdCl_3$-NaCl system.

KEYWORDS: molten salts, density, lanthanides, gamma-ray

I. Introduction

Data on densities of molten chlorides are required in the nuclear technology for plant design of nuclear incineration of trans-uranium elements by proton accelerator and high temperature chemical reprocessing of spent nuclear fuel. Reliable data on densities of alkaline chlorides are available, but data on densities of lanthanides chlorides and lanthanides chlorides-alkaline chlorides mixture are scarce.

II. Experimental

Materials:$NdCl_3$ was prepared from $NdCl_3 6H_2O$ by dehydrated at 200℃ in vacuum and sublimed in vacuum at 1000℃. Alkaline chlorides were dried at 500℃ in vacuum. Materials were handled in an argon-filled dry box which was controlled at a water content of <2ppm and oxygen content of <1ppm.

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

A schematic diagram of the experimental set-up for the γ-ray attenuation method is shown in **Fig.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 effect on the radiation source and 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 salt mixture of 3-6g with desired composition, it was sealed. Gamma-ray spectra were taken at various controlled temperature. The γ-ray spectra thus obtained were analyzed with a standard method. The attenuation at a given γ-ray energy is expressed by equation(1),

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

where I_i and I_{i0} are γ_i-beam intensities after and before passing through the materials. σ_i is total attenuation coefficient at the given γ-ray energy and for the given materials, ρ is density of the material and x is length of the specimen.

The total attenuation coefficient σ_i is expressed by equation(2),

$$\sigma_i = \sigma_{ie} + \sigma_{ic} + \sigma_{ip} + \sigma_{is} \qquad (2)$$

where σ_{ie} is the attenuation coefficient by photoelectric process, σ_{ic} is the attenuation coefficient by Compton scattering, σ_{ip} is the attenuation coefficient by pair-production and σ_{is} is the attenuation coefficient by coherent scattering. The attenuation coefficients were linearly fitted to the values in the literature[1]. By the three former process, the γ-ray with energy i gives its energy totally or partially, the γ-ray with energy i is thus attenuated. On the other hand, with the coherent scattering, the γ-ray with energy i does not change its energy and a strong anisotropic scattering occurs: some of the scattered γ-ray keeps its course and reaches the detector. The equation (2) must be replaced by equation (3),

$$\sigma_i = \sigma_{ie} + \sigma_{ic} + \sigma_{ip} + \sigma_{is} F \qquad (3)$$

where F is a probability of the scattered γ-ray coherently deviate from its original path.

From the attenuation factors, I_i/I_{i0} obtained with water and the total attenuation coefficients for water calculated with eq. (3) and the density of pure water at room temperature, the length of the cell was calculated by eq.(1). The density of the molten chloride was calculated in the same way from the attenuation factor with the total attenuation coefficient calculated for the constituent elements and the cell length obtained above. The factor F was chosen so as to minimize the standard deviation of the

*Corresponding Author, Tel +81-29-282-6083, Fax +81-29-6442
E-mail:sato@analchem.tokai.jaeri.go.jp

densities calculated from different energies of γ rays for a given sample for the whole densities measurement.

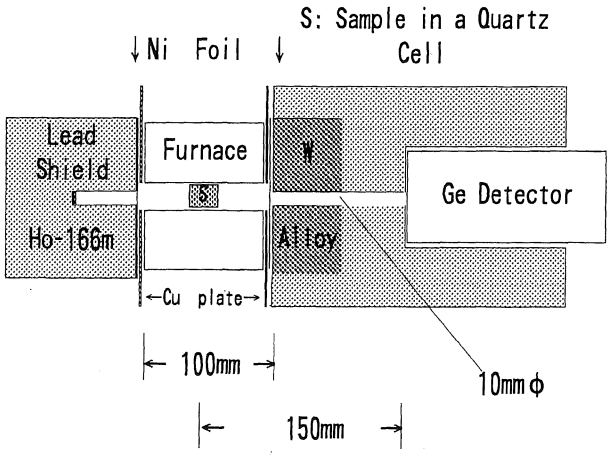

Fig. 1 Schematic diagram of the experimental set-up

III. Results and discussion

The results of density measurement by the γ -ray attenuation method for $NdCl_3$-KCl system are shown in **Fig.2** as a function of temperature. In this figure, the error bars represent one standard deviation.

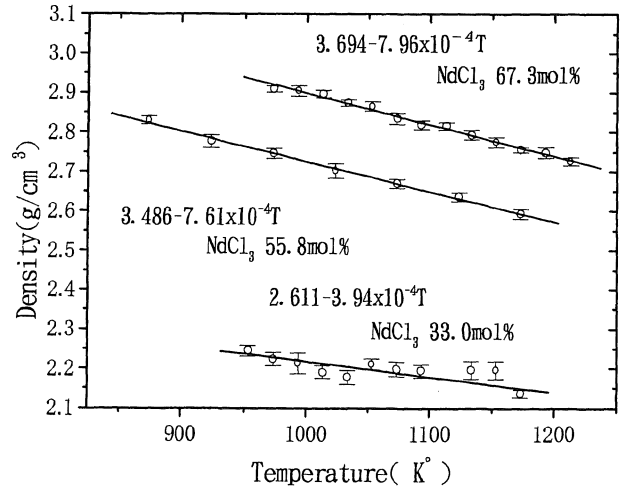

Fig. 2 Density of molten $NdCl_3$-KCl mixture as a function of temperature

The results of density measurement for $NdCl_3$-NaCl and $NdCl_3$-KCl systems are summarized in **table 1.**

The densities of the molten chlorides were expressed as a-bxT, where T is temperature in Kelvin.

The densities of $NdCl_3$-NaCl and $NdCl_3$-KCl systems at 1100 K are shown in **Fig.3** as a function of $NdCl_3$ composition together with literature values determined with dilatometric method reported by Mochinaga and Igarashi[2]. The values of densities of pure NaCl and KCl at 1100 K in this work were taken from Artsdalen and Yaffe[3].

Table1 Densities of $NdCl_3$-NaCl and $NdCl_3$-KCl system as a function of temperature.

System	$NdCl_3$	a	b
	mol%		$x10^{-4}$
$NdCl_3$-NaCl	24.6	2.646±0.065	4.10±0.60
	35.0	3.156±0.035	7.58±0.41
	49.7	3.528±0.017	8.15±0.18
	66.7	3.814±0.020	8.05±0.19
	100.0	3.971±0.063	6.72±0.54
$NdCl_3$-KCl	33.0	2.611±0.078	3.94±0.73
	55.8	3.486±0.022	7.61±0.22
	67.3	3.694±0.026	7.96±0.23

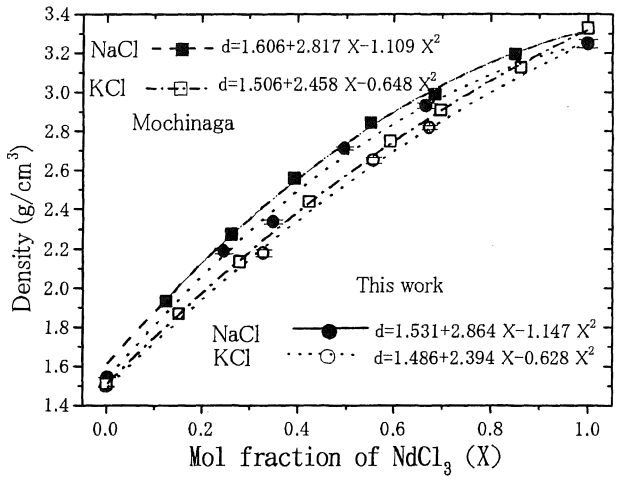

Fig.3 Densities of $NdCl_3$-NaCl and $NdCl_3$-KCl systems at 1100 K

The densities are expressed as secondary ordered polynomial equations. The results reported by Mochinaga and Igarashi are about 1 to 3 % larger than the densities obtained in the present work.

From the relationship between density of alkaline chlorides and neodymium chloride mixture and neodymium chloride composition, the binary excess molar volumes given as the deviation from additivity are obtained at 1100 K for of $NdCl_3$-NaCl and $NdCl_3$-KCl systems and depictured in **Fig.4**

Although, the excess molar volumes differ from each other, the tendencies are the same. The maximum occur at $NdCl_3$ composition around 0.2 and decreases toward higher $NdCl_3$composition. A small negative excess molar volume appears at $NdCl_3$composition of 0.75. These tendencies are also agreed with the results on lanthanum chloride alkaline chlorides mixture reported by Kim et.al[4].

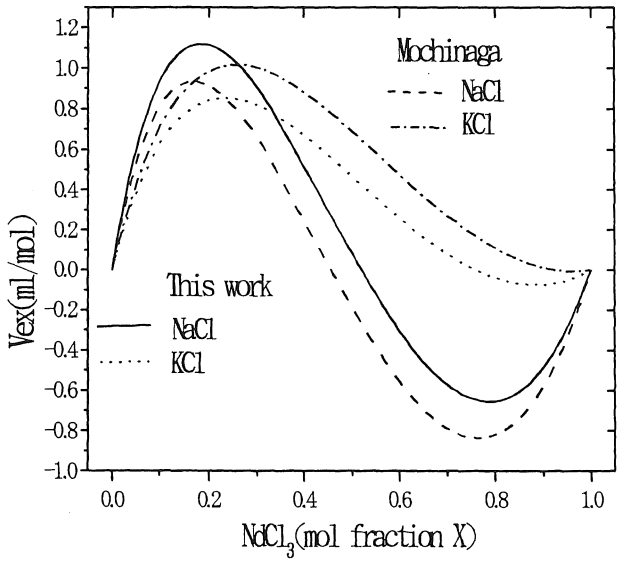

Fig.4 Excess molar volumes for $NdCl_3$-NaCl and $NdCl_3$-KCl systems at 1100 K

IV. Conclusion

The densities of molten $NdCl_3$-NaCl and $NdCl_3$-KCl systems were determined by γ-ray attenuation method. Isothermal equations at 1100 Kelvin for $NdCl_3$-NaCl and $NdCl_3$-KCl systems were $1.531+2.864X-1.147X^2$ and $1.486+2.394X-0.628X^2$, respectively. Where X is mol fraction of $NdCl_3$.

Reference

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