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Investigation of heat capacity of Plutonium nitride

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ABSTRACT

An analytical equation for the heat capacity of Plutonium nitride (PuN) nuclear fuel has been performed by the use of the *n*-dimensional Debye function in the temperature range 298 - 3000 K. The results obtained for the heat capacity were compared with the literature data, and are found to be in good agreement. This agreement is shown that the present theoretical method would be useful to calculate the heat capacity of nuclear fuels such as Plutonium nitride.

1. Introduction

One of the important materials used in the field of nuclear energy is the plutonium isotope (Lai et al., 2020; Walters et al., 2002; Clark et al., 2008;). Compounds like nitrides and carbides are encouraging fuels for generation IV reactors due to their high melting points, high fuel density and high thermal conductivity, as well as being used as target materials for the transmutation of plutonium and minor actinides in fast reactor cores and accelerator driven systems. (Rodriguez, 1999; Sedmidubsky and Nova, 2005; Streit and Ingold, 2005; Clark et al., 2008; Kudinov et al., 2015; Rong Yang et al., 2018; Lai et al., 2020). Of these materials, PuN, which has simple rock-salt-type structure, can be essential for long-term storage. It is critical to specify the thermophysical properties of plutonium nitride (specific heat, thermal conductivity, entropy, etc.) to obtain information about nuclear fuel potential and fuel performance of plutonium nitride (Rong Yang et al., 2018). In this context, this material is of significant interest to both theorists (Fynn and Ray, 2007; Petit et al., 2009; Wen et al., 2013; Obodo and Chetty, 2013; Rong Yang et al., 2018) and experimentalists (Oetting, 1996; Tennery and Bomar, 1971; Hall et al., 1978; Havela et al., 2003). Hall et al. (1978) studied antiferromagnetic ordering of PuN within the framework of a maximum in magnetic susceptibility and specific heat. In theory, Rong Yang et al. (2018) investigated the structural, magnetic, electronic, dynamical and thermodynamic properties of PuN by using density functional theory, DFT+U and hybrid DFT. The substoichiometric behavior of PuN that is an important role in understanding its physical and chemical properties have been examined by Lai et al. (2020) using first-principles calculations, and they found that PuN is stable for only stoichiometric levels. The electronic, structural, and magnetic properties of PuN within the framework of the full-potential all-electron linearized augmented plane wave (FP-LAPW) plus local orbital method have been investigated by Fynn and Ray (2007). In this work, an analytical method of the heat capacity of nuclear fuels in framework of the n-dimensional Debye function have been introduced. To evaluate the obtained method, the heat capacity of PuN has calculated. The results we obtained in this study show that the method we used has reliable computational efficiency.

2. Computational Method

The heat capacity of materials at constant pressure is given by (Synoradzki et al., 2019; Eser and Koç, 2021; Eser et al., 2020; Bölükdemir et al., 2018).

$$C_P(T) = \gamma \mathrm{T} + 9\mathrm{nR}\left(\frac{1}{x_\mathrm{D}}\right)^3 \int_0^{x_\mathrm{D}} \frac{x^4 e^x}{(e^x-1)^2}\, dx \qquad (1)$$

In above equations, γ is the Sommerfeld coefficient, n is the number of atoms per formula unit, $x_\mathrm{D} = \theta/T$, where, θ is

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the Debye temperature, and γ is the gas constant. Considering the n-dimensional Debye function, equation (1) can be written as follows:

$$C_P(\beta, x_D) = \gamma T + 9R\Lambda_n(\beta, x_D) \quad (2)$$

$\Lambda_n(\beta, x_D)$ is the n-dimensional second kind Debye function, and can be expressed by the n-dimensional Debye function as:

$$\Lambda_n(\beta, x_D) = \frac{n}{n+1} x_D [D_n(\beta - 1, x_D) + D_n(\beta, x_D)] \quad (3)$$

Here, the n- dimensional Debye function $D_n(\beta, x_D)$ are defined by (Gökbulut et al., 2021)

$$D_n(\beta, x_D) = \frac{n}{x^n} \int_0^x \frac{t^n}{(e^t - 1)^\beta} dt. \quad (4)$$

As can be seen from the above equations, the solution of n-dimensional Debye functions is of prime importance. A solution of the Debye function defined in Mamedov et al. (2009) is given by

$$D_n(\beta, x_D) = \frac{n}{x_D^n} \lim_{M\to\infty} \sum_{m=0}^{M} n! \left\{1 - e^{-x_D w} \left(\sum_{k=0}^{n} \frac{(x_D w)^k}{k!}\right)\right\} w^{-(n+1)} \quad (5)$$

where $w = m + \beta$. $F_m(-\beta)$ are the binomial coefficients and defined as (Gradshteyn and Ryzhik, 1980):

$$f_m(n) = \frac{1}{m!} \prod_{i=0}^{m-1} (n - i). \quad (6)$$

3. Results and Discussion

In this study, we have proposed an analytical formulation for heat capacity of PuN nuclear fuel by the use of the n-dimensional Debye function. All calculations have been made with help of the Mathematica 8.0 program. The temperature dependence of the heat capacity for PuN is shown in Fig. 1, together with the literature data.

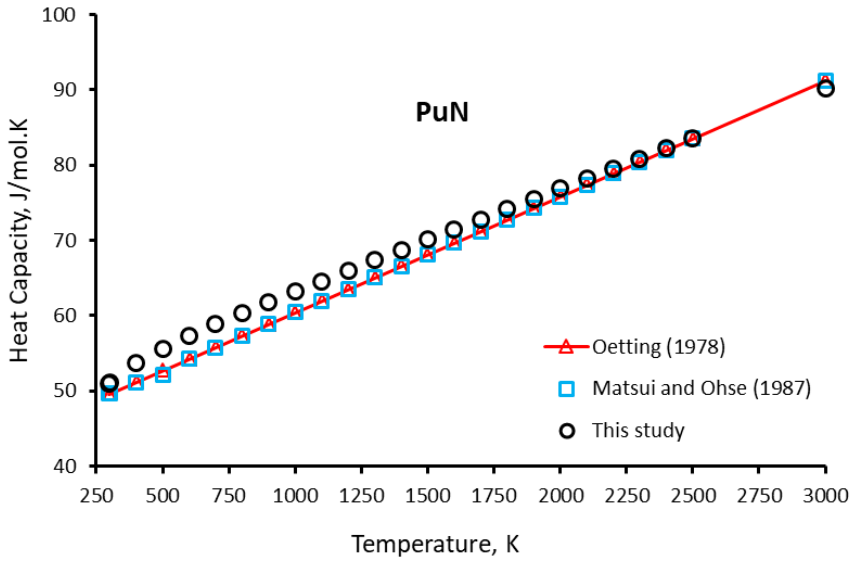

Figure 1. Temperature dependence of heat capacity of Plutonium nitride

Figure 1 is shown that the results of heat capacity obtained in this study are in good agreement with those of Oetting (1978) and Matsui et al. (1987). For instance, the derived value of PuN at 2300 K is 80.89 J/mol.K, which is in good agreement with the corresponding values of 80.46 J/mol.K (Oetting, 1978) and 80.47 J/mol.K (Matsui et al., 1987). As can be seen from Fig. 1, the best agreement with our values is found for temperatures higher than about 1000 K. The highest difference is seen in data of Matsui et al. (1987) at 500 K, about 6.62%.

4. Conclusion

In this study, the heat capacity, one of the most important thermophysical properties of PuN regarding the prediction of fuel temperature, has been calculated using a series of formulation obtained for the n-dimensional Debye function. It is seen that the obtained results on PuN were in agreement with the literature data. In a conclusion, it is suggested that the obtained method is general and can be used for calculation of thermodynamics properties of nuclear fuels over wide temperature range.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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