# Molecular Dynamics Simulation of Thermomechanical Properties of Mononitride Nuclear Fuel

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## 1. Introduction

Uranium-plutonium mixed nitride is a strong candidate fuel for advanced liquid metal cooled fast breeder reactors. It gives better neutron economy than oxide fuel and has high melting point, high fuel density and high thermal conductivity.

This kind of new fuel is under development-to-testing stage. Using molecular dynamics (MD) simulation to predict thermal and mechanical properties of the material will be very useful to understand the nitride fuel behavior.

The heat capacities of UN and PUN are estimated using MD simulation with temperature ranges from 300K to 2500K.

## 2. Methods and Results

### *2.1 Simulation Model*

Using the MOXDORTO molecular dynamics program [1], simulations of UN and PUN are performed with 4*4*4 cells (512 ions, U(PU) $^{3+}$: 256, N $^{3-}$: 256) in the same structure of NaCl type crystal. Table 1 shows the structure data used in the present simulation.

Table 1: Structure data of UN and PUN

UN

| Unit cell | 4.88 | 4.88 | 4.88 | 90 | 90 | 90 |
|---|---|---|---|---|---|---|
| | charge | x | y | z | | |
| U | 3+ | 0 | 0 | 0 | | |
| N | 3- | 0.5 | 0.5 | 0.5 | | |

PUN

| Unit cell | 4.905 | 4.905 | 4.905 | 90 | 90 | 90 |
|---|---|---|---|---|---|---|
| | charge | x | y | z | | |
| PU | 3+ | 0 | 0 | 0 | | |
| N | 3- | 0.5 | 0.5 | 0.5 | | |

Quantum correction is taken into account for Helmholtz free energy. Constant pressure-temperature (NPT) calculation and constant volume-temperature (NVT) calculation are performed. The lattice boundary is periodic and there is no edge or surface effect.

Morse model potential function including Morse type potential between cation-anion pair is used.

$$U_{ij}(r_{ij}) = \frac{Z_i Z_j e^2}{r_{ij}} + f_0(b_i + b_j)\exp\left(\frac{a_i + a_j - r_{ij}}{b_i + b_j}\right) + D_{ij}\{\exp[-2\beta_{ij}(r_{ij} - r^*_{ij})]\} - 2\exp[-\beta_{ij}(r_{ij} - r^*_{ij})], \quad (1)$$

where $Z_i$ and $Z_j$ are the effective partial electronic charges on the ith and jth ions, $r$ is the atom distance, $r_{ij}$* is the bond length of cation–anion pair in vacuum, and $a$ and $b$ are characteristic parameters which depend on the ion species. In this potential function, $D_{ij}$ and $\beta_{ij}$ describe the depth and shape of this potential, respectively. The parameters used are shown in Table2.

Table2: Potential parameters

UN

| | z | a | b | | $D_{ij}$ | $\beta_{ij}$ | $r^*_{ij}$ |
|---|---|---|---|---|---|---|---|
| $U^{3+}$ | -1.450 | 1.228 | 0.080 | $U^{3+}$ - $N^{3-}$ | 7 | 1.25 | 2.364 |
| $N^{3-}$ | 1.450 | 1.797 | 0.080 | | | | |

PUN

| | z | a | b | | $D_{ij}$ | $\beta_{ij}$ | $r^*_{ij}$ |
|---|---|---|---|---|---|---|---|
| $PU^{3+}$ | -1.450 | 1.196 | 0.080 | $PU^{3+}$ - $N^{3-}$ | 0.10 | 0.80 | 2.453 |
| $N^{3-}$ | 1.450 | 1.797 | 0.080 | | | | |

The equations of motion are integrated with the Verlet's algorithm [2]. The time step is $2.0\times10^{-15}$ s. The initial velocity is set randomly about 1.0 pm/fs.

Before simulation is started, an equilibrium running is performed at 300K for 50000 steps. The equilibrium is judged from the change of temperature ( ± 3%), pressure ( ± 10%), density ( ± 0.02%) and internal energy ( ± 0.3%). Then the temperature is increased to the desired temperature by simply scaling with 0.1K per step. $1\times10^4$ steps are taken in the simulation.

### *2.2 Results*

Fig. 1 and Fig.2 show the lattice parameters of UN and PUN obtained in the simulation. The results agree well with the experimental data.

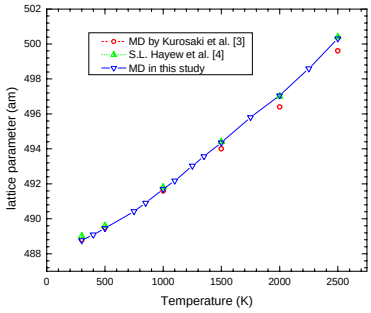


Fig. 1 The lattice parameter of UN with temperature

Lattice parameter is calculated from 300K to 2500K with pressure ranges from 0.1 MPa to 1.5GPa. The compressibility is estimated by following equation :

$$\beta = \frac{1}{a^3(P_0)}\left(\frac{\partial a^3(P)}{\partial P}\right)_T, \qquad (2)$$

where $a(P_0)$ is the lattice parameter at 0.1MPa.

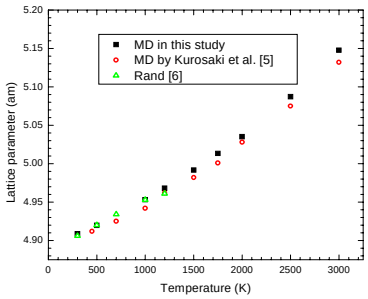


Fig. 2 The lattice parameter of PUN with temperature

The compressibility results of UN and PUN are shown in the Fig. 3 and Fig. 4. At low temperature, the difference between the present result and the experimental result comes from the varying sensitivity of the lattice parameters on temperature.

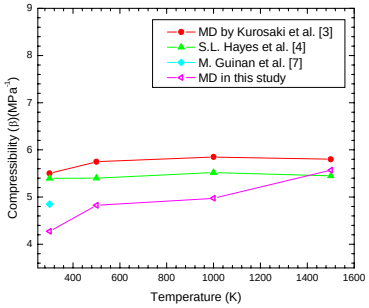


Fig. 3 The compressibility of UN with temperature

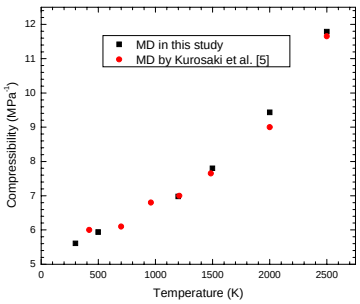


Fig. 4 The compressibility of PUN with temperature

The heat capacity at constant volume is evaluated from internal energy obtained by NVT MD simulation:

$$C_v = \left(\frac{\partial E}{\partial T}\right)_v. \qquad (3)$$

Fig. 5 and Fig. 6 give the heat capacity results of UN and PUN. The figures indicate that the result of UN in this study agrees well with the result reported by Kurosaki et al. But the result of PUN seems to agree with the Dulong-Petit law better than the result reported by Kurosaki et al.

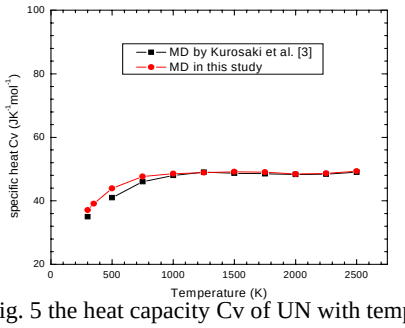


Fig. 5 the heat capacity Cv of UN with temperature

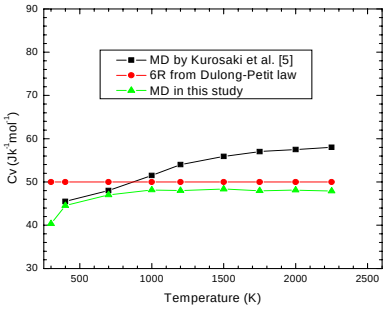


Fig. 6 The heat capacity Cv of PUN with temperature

## 3. Conclusions

MD NPT and NVT simulations are used in the present study to predict lattice parameter, compressibility and heat capacity at constant volume of the mononitride nuclear fuel. The results agree well with the reported data. The compressibility of UN is in good agreement with the experimental result at high temperature above 1400K. The heat capacity of PUN agrees with the Dulong-Petit law at temperature above 1000K. This indicates that the Morse-type potential function describes the crystal parameters well and the MD simulation can be used to evaluate the heat capacity at constant volume of nitride nuclear fuel.

## ACKNOWLEDGEMENT

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