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Hypernetted chain calculations of molten uranium dioxide: Comparison of rigid ion potentials

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ABSTRACT

The first liquid state calculations within the hypernetted chain approximation for molten uranium dioxide using six different rigid ion model potentials are presented and discussed. Applicability of the existing rigid ion potentials to the molten phase of uranium dioxide is examined. Despite the minor differences, the radial distribution functions and static structure factors of all potentials are typical of a system which melted from a fast ion conducting phase. Only in one case, the breakdown in the local structure of oxygen ions in the solid state is reflected in the calculated coordination numbers. The behavior of static dielectric function is related to the onset of freezing and found to mimic that of the classical one component plasma at low values of plasma parameter. Finally, the enthalpy change is calculated and compared with molecular dynamic simulation results and experimental data.

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1. Introduction

Uranium dioxide is the main constituent material of corium and the most commonly used nuclear reactor fuel. It is also very interesting material with unusual high temperature structural feature in respect of a structural phase transition to superionic conducting phase below the melting point. Therefore, there have been a number of molecular dynamics simulation studies to investigate the time and temperature evolution of the structural, thermophysical and transport properties of uranium dioxide. In attempting such simulations description of interionic interactions in UO_2 with an effective rigid ion potential plays an important role. The rigid ion and the shell core model potentials developed by different authors have been compared by Govers et al. [1,2] in order to assess the range of applicability of the potentials in static and dynamic calculations. Devanathan et al. [3] have report the limitations of existing five different rigid ion model, which all have the same functional form, using in classical molecular dynamics simulations of the diffuse premelting transition, melting and defect production by 1 keV U recoils in UO_2. The rigid ion potentials have been placed into three groups based on the premelting transition from the lowest to the highest and oxygen mobility from the fastest to the slowest: Morelon et al. [4], Lewis and Catlow [5] and Arima et al. [6], and Basak et al. [7] and Yakub et al. [8] which are similar and have produced most significant results. Although, Govers et al. [1,2] have concluded that the choice of a potential is a difficult compromise between its range of applicability and its physical acceptability the partially charged rigid ion potentials generally predict the better agreement with experiment compared to the rigid ion potentials with completely charged ions.

In order to study the physical properties of a fluid system with a model potential one should employ either classical computer simulation techniques or adequate liquid state theories. In that respect, the integral equation theories (IET) [9] are relatively insubstantial method among the simulation techniques as IET are not exact and some approximations should be made. Nevertheless, IET are useful to test the hypothesis made on the properties of a real system such as the structural correlations and some thermophysical quantities proposed by the simulation and experiment. Two fundamental advantages of an approximate theory against simulation are those solution of IET does not require long computational time and is not limited to use of a finite system size. This paper is concerned with integral equation calculations of liquid uranium dioxide. We present the first theoretical calculations for radial distribution functions, static structure factors, longitudinal dielectric function and enthalpy change which were evaluated by solving the Ornstein–Zernike integral equation in conjunction with hypernetted chain (HNC) approximate closure. Six different rigid ion model potential existed in literature of which parameters have been fitted with respect to the solid state properties used in our calculations and tested for the applicability to model the liquid uranium dioxide.

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2. Interionic potentials

The type of the rigid ion model potential which has been widely accounted to model the interionic interaction for uranium dioxide is the Buckingham–Morse (BM) form [10]

$$\phi_{ij}(r) = \frac{Z_i Z_j}{r} + A_{ij} e^{\left(-\frac{r}{\rho_{ij}}\right)} - \frac{C_{ij}}{r^6} + D_{ij}\left(e^{-2\beta_{ij}\left(r-r_{ij}^0\right)} - 2e^{-\beta_{ij}\left(r-r_{ij}^0\right)}\right) \quad (1)$$

which was parameterized by different authors [4–8]. Recently, the Vashishta–Rahman (VR) form [11]

$$\phi_{ij}(r) = \frac{Z_i Z_j}{r} + \frac{H_{ij}}{r^{n_{ij}}} - \frac{P_{ij}}{r^4} - \frac{C_{ij}}{r^6} \quad (2)$$

has also been developed by the authors as an alternative model potential for UO_2 [12]. Parameters of BM and VR potentials have been taken from Devanathan et al. [3] as they present all potentials in common notation and units and Günay et al. [12], respectively, and are presented in Table 1.

The BM form produces unrealistic large attraction at very short O–O separations because of the dispersion term C_{ij}/r^6. This term has been ignored by Günay et al. [12] in their VR type potential. There are several ways in literature to overcome this problem [2,3,13] for which we have multiplied the dispersion term of O–O interaction with an exponential screening term $\exp(-\lambda/r)$ which hardened the potential at short distances but does not changes the behavior of the potential beyond the unphysical region. The criterion that we have chosen to determine the lowest value of the hardness parameter λ is the satisfaction of the accuracy condition in the HNC (see Section 4). The estimated values of λ for each potential are also given in Table 1. All potentials are presented in Fig. 1. Dash dotted lines in Fig. 1 show the O–O interaction without multiplying the dispersion term with $\exp(-\lambda/r)$.

3. HNC theory

For a two component homogeneous and isotropic liquid system such as molten UO_2 correlation functions are related in the form of Ornstein–Zernike (OZ) relation [9]

$$\gamma_{ij}(r) \equiv h_{ij}(r) - c_{ij}(r) = \sum_l n_l \int d\mathbf{r}' c_{il}(|\mathbf{r}-\mathbf{r}'|) h_{lj}(r') \quad (3)$$

where $h_{ij}(r)$ and $c_{ij}(r)$ are the total and the direct correlation functions, respectively for ionic components i and j. n_l is the number density of ions species l. The radial distribution functions $g_{ij}(r)$ are defined by $g_{ij}(r) \equiv h_{ij}(r) + 1$. The Fourier transform of the OZ equation is

$$\tilde{\gamma}_{ij}(k) = \frac{n\tilde{c}(k)^2}{1 - n\tilde{c}(k)} \quad (4)$$

The OZ equation yields several liquid state approximate closures for $g_{ij}(r)$. The hypernetted chain (HNC) approximation is one of the frequently used approximate integral equation scheme for ionic liquids. The HNC closure relation can be derived from the $c_{ij}(r)$ which is defined as [9]:

$$c_{ij}(r) = g_{ij}^{\text{total}}(r) - g_{ij}^{\text{indirect}}(r) \quad (5)$$

where $g_{ij}^{\text{total}}(r)$ is the radial distribution function $g_{ij}(r)$ which is related with the potential of mean force $w_{ij}(r)$

$$g_{ij}^{\text{total}}(r) = g_{ij}(r) = \exp\left[-\beta w_{ij}(r)\right] \quad (6)$$

where $\beta = (k_B T)^{-1}$. The indirect part of the $c_{ij}(r)$ is the radial distribution function without the direct interaction between ionic pair $\phi_{ij}(r)$

$$g_{ij}^{\text{indirect}}(r) = \exp\left\{-\beta\left[w_{ij}(r) - \phi_{ij}(r)\right]\right\}. \quad (7)$$

Exponential expansion of the indirect part leads to

$$c_{ij}(r) = g_{ij}^{\text{total}}(r) - \left\{1 + \beta\left[\phi_{ij}(r) - w_{ij}(r)\right] + \frac{1}{2!}\beta\left[\phi_{ij}(r) - w_{ij}(r)\right] + \ldots\right\}. \quad (8)$$

Considering only first two terms of the expansion and using the definition of the total part one can obtain the approximate direct correlation function in the form of

$$c_{ij}(r) = g_{ij}(r) - 1 + \beta\phi_{ij}(r) + \ln\left[g_{ij}(r)\right] \quad (9)$$

which called the HNC approximate closure and can be rewritten in commonly used form as

$$c_{ij}(r) = \exp\left\{-\beta\phi_{ij}(r) + \gamma_{ij}(r)\right\} - \gamma_{ij}(r) - 1. \quad (10)$$

Table 1
Parameters of the rigid ion potentials used in the present HNC calculations. The units of length are Å and the units of energy are eV.

Parameters	Arima	Basak	Lewis	Morelon	Yakub	Günay
Z_U	2.70	2.40	4.00	3.22724	2.2208	1.96
Z_O	−1.35	−1.20	−2.00	−1.61362	−1.1104	−0.98
A_{U-O}	55918.39	693.9297	1055.0	566.498	432.18	0.0
ρ_{U-O}	0.2020	0.327022	0.3949	0.42056	0.3422	0.0
C_{U-O}	0.0	0.0	0.0	0.0	0.0	8.294
D_{U-O}	0.0	0.57745	0.0	0.0	0.5055	0.0
β_{U-O}	0.0	1.65	0.0	0.0	1.864	0.0
r^0_{U-O}	0.0	2.369	0.0	0.0	2.378	0.0
H_{U-O}	0.0	0.0	0.0	0.0	0.0	243.63
P_{U-O}	0.0	0.0	0.0	0.0	0.0	4.032
A_{O-O}	979.17	1633.666	22764.3	11272.6	883.12	0.0
ρ_{O-O}	0.332	0.327022	0.149	0.1363	0.3422	0.0
C_{O-O}	17.3622	3.951	112.2	134.0	3.996	0.0
H_{O-O}	0.0	0.0	0.0	0.0	0.0	521.75
P_{O-O}	0.0	0.0	0.0	0.0	0.0	1.0
A_{U-U}	2.482×10^{13}	294.7593	0.0	0.0	187.03	0.0
ρ_{U-U}	0.072	0.327022	1.0	1.0	0.3422	0.0
H_{U-U}	0.0	0.0	0.0	0.0	0.0	1437.45
λ	1.35	0.98	2.97	4.47	1.07	–

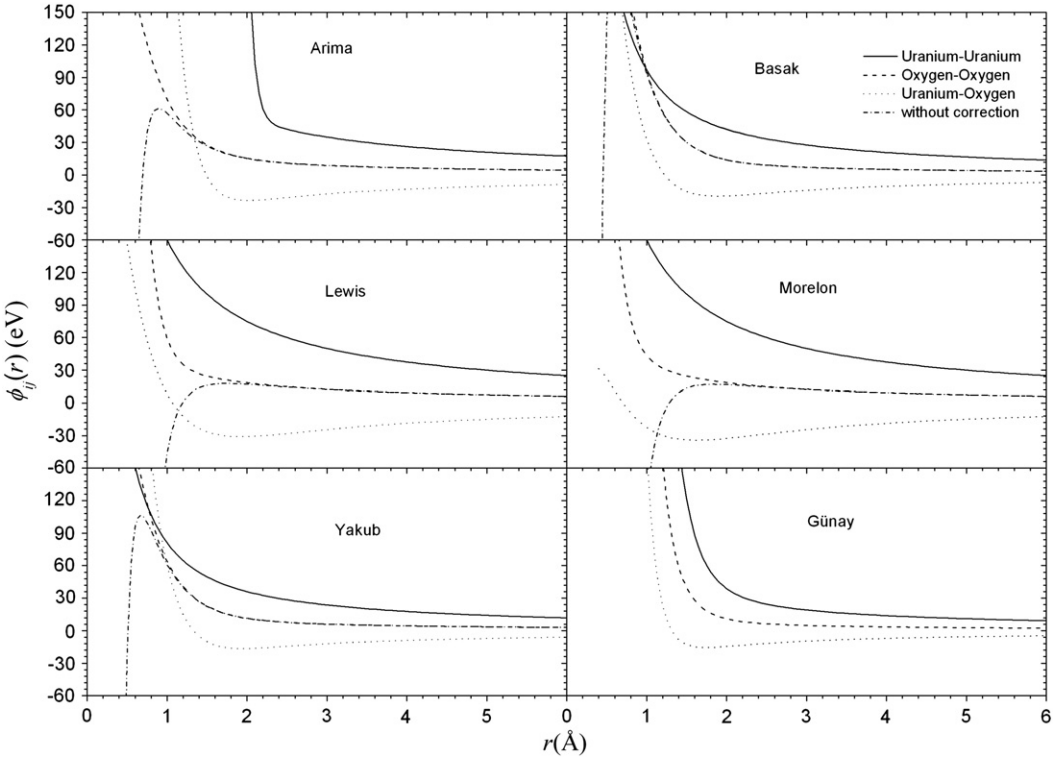


Fig. 1. Potential energies for UO_2.

4. Details of the HNC calculations

The OZ integral equation has been solved in conjunction with the HNC approximate closure using six different rigid ion potentials developed by Arima et al. [6], Basak et al. [7], Lewis and Catlow [5], Morelon et al. [4], Yakub et al. [8] and Günay et al. [12]. Numerical calculations have been carried out following the method originally proposed by Gillan [14] and Abernethy and Gillan [15].

Two traditional methods of solving such equations are called Picard and Newton–Raphson (NR) methods. However, both methods have their own disadvantages. The Picard method is generally very slow and a good initial estimation is needed for $\gamma_{ij}(r)$. On the other hand, the NR method requires a very difficult calculation and inversion of a matrix which has the dimension of 10^3. The method developed by Gillan [14] and Abernethy and Gillan [15] is basically combination of the Picard and NR methods. It has been shown that this method enables the calculations to reach an accurate solution of the OZ equation in about 20–30 iterations even with a poor initial estimation for $\gamma_{ij}(r)$. In this method $\gamma_{ij}(r)$ is decomposed into coarse part $\gamma_{ij}^{\text{coarse}}(r)$ and fine part $\Delta\gamma_{ij}(r)$. The coarse part is handled with the NR method while the Picard method is used for the fine part.

The elementary cycle of the method is as follows: make an initial estimation for $\gamma_{ij}^{k}(r)$; obtain $c_{ij}(r)$ from Eq. (10) (HNC) using $\gamma_{ij}^{k}(r)$ as input; obtain Fourier transform of $c_{ij}(r)$ and place it in Eq. (4) (Fourier transform of the OZ equation) to find the revised $\gamma_{ij}^{k+1}(k)$; finally the inverse Fourier transform of the $\gamma_{ij}^{k+1}(k)$ gives revised $\gamma_{ij}^{k+1}(r)$.

One then repeats the procedure given above using the new $\gamma_{ij}^{k+1}(r)$ as new input until satisfactory convergence has been obtained. The accuracy of the solution at each stage has been taken as $|\gamma_{ij}^{k+1}(r)-\gamma_{ij}^{k}(r)|<10^{-4}$. Initial estimations for $\gamma_{ij}(r)$ have been obtained from the solution of mean spherical approximation (MSA) [9] with charged hard-spheres to since UO_2 is an ionic system. $M=2048$ mesh points have been used at each run. The system has been gradually cooled down from an initial temperature T_i to the desired temperature T_f. The temperature T_f has been varied between 3000 K and 4500 K. The density of the liquid uranium dioxide for each T_f has been taken from Fink [16]. Mesh length in real and reciprocal spaces have been adjusted from the relation $\Delta r\cdot\Delta k=2\pi/M$. The units of length and energy used in HNC calculations are Å and $e^2/Å=14.399$ eV, respectively.

5. Results and discussions

The HNC calculations have been carried out for liquid UO_2 at various temperatures between 3000 K and 4500 K. In this section the structural properties are presented only at the freezing temperature 3120 K and number density 0.05928 ions/Å^3 [16] as an illustrative picture of all calculations. An important information on a quantitative measure of the spatial local order–disorder of the system structure can be obtained from the radial distribution function $g_{ij}(r)$. Calculated $g_{ij}(r)$ of molten UO_2 for all potentials displays the expected temperature dependence behaviors; as the temperature increases, peaks become lower and broader due to the larger thermal vibrations of the ions, and the average peak positions which correspond to the ionic separations do not change. Fig. 2 shows $g_{ij}(r)$ of molten UO_2 at the freezing temperature 3120 K and number density 0.05928 ions/Å^3. The $g_{OO}(r)$ has lower and broader principal peak with less pronounced oscillations beyond it compared to U–U pairs indicating the lower degree of correlations in the position on oxygen ions. Note that considering the $g_{OO}(r)$ and $g_{UU}(r)$ produced by Günay et al. [12] potential, the degree of correlation of uranium and oxygen ions are competing so it is hard to tell which one is more mobile. Another distinct feature is that oxygen ions display a charge penetration into the first coordination shell of unlike ions. Such type of behaviors are interpreted as the reflection of a system melted from a superionic or fast ion conducting phase in which oxygen ions are much more mobile and the sublattice constructed by the oxygen ions is liquid like. The charge penetration observed from Morelon et al. [4] and Günay et al. [12] potential is less significant. Bearing in mind that these do not validate the applicability of all model potentials studied here to UO_2 at molten phase, as all liquid state theories, the HNC has its limitations, i.e. one can not tell whether the system would melt at

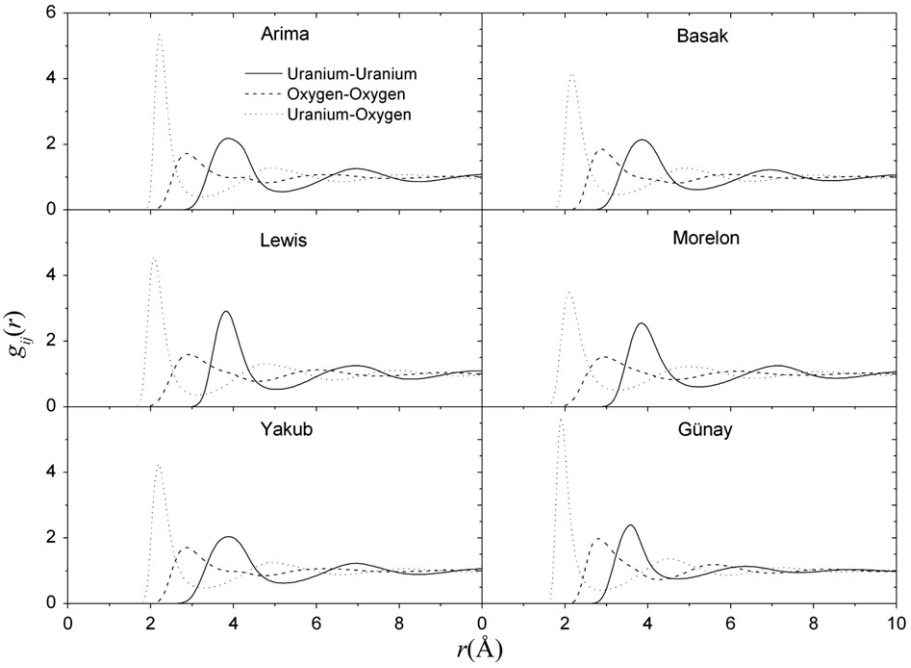


Fig. 2. Radial distribution functions $g_{ij}(r)$ of molten UO_2 at the freezing temperature 3120 K.

correct temperature and density just from the behaviors of the radial distribution functions.

The principal peak positions r_{ij}^m of the radial distribution functions which correspond to the interionic distances are given Table 2. The estimated approximate positions from our HNC calculations using BM type model potentials [4–8] are almost the same for each ionic pairs which are in accord with corresponding crystalline interionic distances $r_{UU}=3.86$ A, $r_{OO}=2.75$ A and $r_{UU}=2.36$A [17]. r_{ij}^m obtained by the VR type potential of Günay et al. [12] are lower than the interionic distances at solid state as they have also under estimate the lattice parameter in their molecular dynamics calculations.

The coordination number N_{ij} has been calculated from the area under the first peak of $g_{ij}(r)$ using the following relation:

$$N_{ij} = C_j n \int_{r_1}^{r_2} g_{ij}(r) d\mathbf{r} \tag{11}$$

where C_j is the concentration of ions of species j. Results are presented in Table 2. The CaF_2 type crystal arrangements of uranium and oxygen ions are $N_{UU}=12$, $N_{OO}=6$ and $N_{UO}=8$ [17]. The experimental percentage increase in volume $V_m - V_{300}/V_{300}$of UO_2 is about 15% [16]. Therefore significant changes in coordination numbers on melting, especially in O–O coordination, can be expected. While N_{ij} calculated from BM form of potentials alters about ± 2 the VR potential reveals more significant disruption of the ionic coordinations. For example, O–O coordination increases from 6 in solid phase to about 11 in liquid phase.

Table 2
The first peak positions r_{ij}^m (Å) of $g_{ij}(r)$, the coordination numbers N_{ij}, the periods of the AL partial structure factors Δk_{ij} (Å^{-1}) and the interionic distances $\Delta r_{ij}=2\pi/\Delta k_{ij}$ deduced from the HNC results at the freezing temperature of UO_2.

	Arima	Basak	Lewis	Morelon	Yakub	Günay
r_{UU}^m	3.85	3.85	3.80	3.85	3.90	3.60
r_{OO}^m	2.85	2.90	2.90	2.95	2.85	2.80
r_{UO}^m	2.20	2.15	2.10	2.10	2.20	1.90
N_{UU}	11	11	10	11	11	8
N_{OO}	7	8	6	6	7	11
N_{UO}	6	6	6	6	6	5
Δk_{UU}	1.30	1.60	1.80	1.66	1.60	1.72
Δk_{OO}	2.58	2.45	2.70	2.64	2.58	2.46
Δk_{UO}	2.70	2.76	2.95	1.35	2.70	3.13
Δr_{UU}	4.83	3.93	3.49	3.78	3.93	3.65
Δr_{OO}	2.43	2.56	2.33	2.38	2.43	2.55
Δr_{UO}	2.34	2.28	2.13	4.65	2.33	2.01

According to the HNC results using the potential by Günay et al. presented in Table 2 suggest that there is a breakdown in the local structure which has occurred either during the transition to the superionic phase and carried it through melting to the molten phase or during the melting, whereas other potential models signal that the local structure is almost retained while the system goes through the transitions to both the superionic phase and molten phase.

The Ashcroft–Langreth (AL) partial structure factors $S_{ij}(k)$ were determined as follows:

$$S_{ij}(k) = \delta_{ij} + 4\pi\left(n_i n_j\right)^{\frac{1}{2}} \int_0^{\infty} \left(g_{ij}(r)-1\right) \frac{\sin\ kr}{kr} r^2 dr \tag{12}$$

The AL partial structure factors of molten UO_2 at 3120 K for all potentials are presented in Fig. 3. The potential by Günay et al. produces lower principal peak for $S_{UU}(k)$ compared to that for $S_{OO}(k)$ which corresponds to lower degree of intermediate range ordering of uranium atoms in real space. This is not an expected case for the molten UO_2 which is supposed to be melted from the superionic phase in which the oxygen ions have the higher ionic conductivity.

We have also considered the periods of AL structure factors presented in Fig. 3 which correspond to the correlation distances $\Delta r_{ij}=2\pi/\Delta k_{ij}$ in radial distribution functions, in other words the distances between the ion pairs. The periods Δk_{ij} and corresponding distances Δr_{ij} are also listed in Table 2. Δr_{ij} values produced by Basak et al. [7], Lewis and Catlow [5], Yakub et al. [8] and Günay et al. [12] potentials are consistent with r_{ij}^m which has been determined from the position of the principal peaks of $g_{ij}(r)$. There are considerable differences between the distances estimated from $S_{ij}(r)$ and $g_{ij}(r)$, for uranium–uranium and uranium–oxygen using the potentials by Arima et al. [6] and Morelon et al. [4], respectively.

In this part of the article we aimed to study the static dielectric function, $\varepsilon(k)$, of molten uranium dioxide. The inverse of the longitudinal dielectric function, $\varepsilon^{-1}(k)$, of a charged particle system may be viewed as the response function of the system to either an external charge density

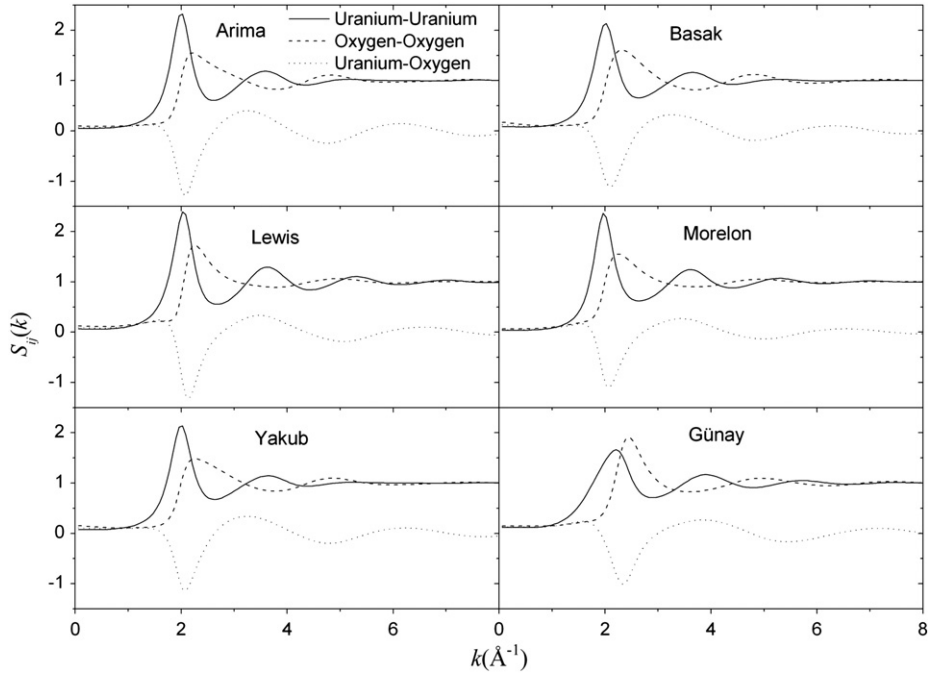


Fig. 3. Ashcroft–Langreth (AL) partial structure factors $S_{ij}(k)$ of molten UO_2 at the freezing temperature 3120 K.

or an electric field. Following the stability condition and the fluctuation–dissipation theorem, it has been shown that ionic fluids will be stable if $\varepsilon(k)<0$ for the values $k<(4\pi ne^2 S_{QQ}(k)/k_BT)^{1/2}$, $\varepsilon(k)>1$ for the values $k>(4\pi ne^2 S_{QQ}(k)/k_BT)^{1/2}$and $\varepsilon(k)=1$ as $k\to\infty$ [18,19]. For a rigid ions system obeying classical statistical mechanics, the $\varepsilon(k)$ is related to the charge–charge partial structure factor $S_{QQ}(k)$ as [12]

$$\varepsilon^{-1}(k) = 1 - \frac{4\pi ne^2}{k_B T k^2} S_{QQ}(k) \tag{13}$$

For such systems, in the region where $\varepsilon(k)$ is negative, it approaches, but does not attain, the value of zero around the position of the main peak of the $S_{QQ}(k)$ as the temperature decreases at molten phase. This behavior of $\varepsilon(k)$ signals the onset of freezing of an ionic melt. Studies showed that the $\varepsilon(k)$ of superionic systems, such as AgI, mimics the classical one component plasma (OCP) behavior better than the simple salts, such as NaCl, do [20–22]. The transition of the OCP to a Wigner crystal occurs at plasma parameter $\Gamma=(Ze)^2/k_BTa\cong 160$ when $\varepsilon(k)$ is closest to k-axis in the negative region, where $a=\left(3/4\pi n\right)^{1/3}$ is the mean ion radius.

We have obtained $\varepsilon(q)$, $q=ka$, for molten UO_2 using calculated values of $S_{QQ}(k)$ of HNC from the relation given by

$$S_{QQ}(k) = C_U S_{UU}(k) - 2(C_U C_O)^{1/2} S_{UO}(k) + C_O S_{OO}(k) \tag{14}$$

The temperature is varied between 3000 K and 4500 K. As the temperature reduces to the freezing value the maxima of $\varepsilon(q)$ approach to the q-axis but $\varepsilon(q)=0$ is never observed. This occurs only when the ions becomes localized on their lattice sites. The results at 3120 K are presented in Fig. 4. The $\varepsilon(q)$ of OCP for the plasma parameters $\Gamma=40$, 60 and 160 deduced from $S(k)$ of Roger et al. [23] is also included in Fig. 4. The $\varepsilon(q)$ of all model potential resembles the behavior of the OCP at the plasma parameter about 40 which is lower than the freezing value of OCP. The inset in Fig. 4 shows the approach of $\varepsilon(q)$ to q-axis in detail. The value of $\varepsilon(q)$ approaches to zero from negative region at $k=2.03$ Å^{-1}, 2.09 Å^{-1}, 2.15 Å^{-1}, 2.03 Å^{-1}, 2.33 Å^{-1} and 2.03 Å^{-1} and at the plasma parameter $\Gamma=123$, 97, 269, 175, 83 and 65 for Arima et al. [6], Basak et al. [7], Lewis and Catlow [5], Morelon et al. [4], Yakub et al. [8] and Günay et al. [12], respectively. It has been suggested that these behavior reflect the covalent nature of a superionic conductor (AgI) and the tendency of the ions in the molten phase to freeze in to the superionic conducting phase or, equivalently, the evidence of rigid uranium sublattice. It is also important to note that OCP and AgI are the charge symmetric systems whereas in the case of UO_2 the number of anions and cations are no longer the same and there is no charge symmetry. The inset in Fig. 4 reveals that the closest value of $\varepsilon(q)$ to zero in the negative region is varying between −0.045 and −0.04 in the case of BM type potentials and is about −0.005 for VR type potential.

The internal energy per particle of a two component liquid can be calculated from

$$\frac{E}{N} = \frac{3}{2}k_BT + \frac{n}{2}\sum_{i,j} C_i C_j \int g_{ij}(r)\phi_{ij}(r)d\mathbf{r}. \tag{15}$$

Enthalpy change of liquid UO_2 $\Delta H=H(T)-H(3120\ \text{K})$ has been calculated using the HNC results of $g_{ij}(r)$ and the model potentials $\phi_{ij}(r)$. Temperature dependence of ΔH per unit cell is presented in

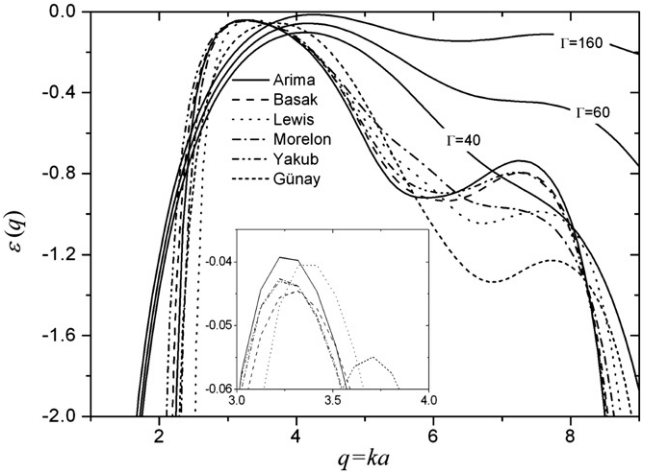


Fig. 4. Static dielectric functions $\varepsilon(k)$ of molten UO_2 at the freezing temperature 3120 K.

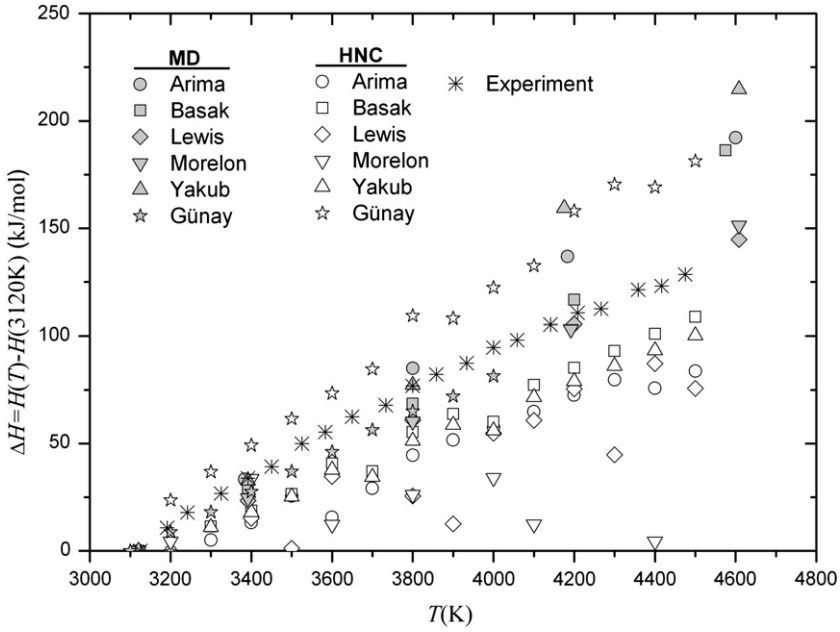


Fig. 5. Temperature dependence of enthalpy changes $\Delta H = H(T) - H(3120\ \mathrm{K})$ of molten UO_2.

Fig. 5 together with the enthalpy change of classical molecular dynamics simulations taken from Devanathan et al. [3] and Günay et al. [12], and of experimental data compiled by Fink [16]. It is known that in predicting the virial pressure, as any approximate theory, the HNC theory is internally inconsistent due to the approximations made on the bridge functions in exact closure. Nevertheless, comparing the results of HNC theory with the data of MD simulations and experiment can give important information on the capability of the model potentials in order to use to describe the uranium dioxide system at liquid phase. In general, the calculated liquid state enthalpy changes using the rigid ion potentials discussed in this article are in fair good agreement with the MD simulation results [3,12] and experimental data [16] except the results obtained using the potentials by Lewis and Catlow [5] and Morelon et al. [4]. Some of the ΔH of HNC by these two potentials are missing in Fig. 5 since unphysical enthalpy changes have been obtained at the corresponding temperatures. The $\Delta H$of Arima et al. [6], Basak et al. [7] and Yakub et al. [8] are very similar to each other and best agreed with the experimental data. Günay potential reasonable reproduces the experimental ΔH up to 4200 K beyond that discrepancies are increased.

6. Conclusions

Ornstein–Zernike integral equation together with hypernetted chain approximation of liquid state theories was employed to study liquid uranium dioxide. Existing six rigid ion model potentials which have been parameterized with respect to the solid state properties were used in our calculations and examined for the applicability to the molten phase. There are no significant differences between the potential models in considering the pair distribution functions and the static structure factors. All of them mimic the structural features of a typical system which melted from a superionic conducting phase. Apparent change of oxygen-oxygen coordination number calculated from the potential by Günay et al. reflects the breakdown during transitions in the substructure constructed by the oxygen ions. Interionic distances estimated from the static structures using the potentials of Arima et al. [6] and Morelon et al. [4] are not consistent. Longitudinal dielectric functions resemble that of classical one component plasma for the lower plasma parameter than the value at which one component plasma freezes. Therefore, the corresponding plasma parameters of Arima et al. [6], Lewis and Catlow [5], and Morelon et al. [4] were estimated too high to combine with the behavior of dielectric functions. The static dielectric function evaluated from Arima et al. potential is closest to zero. The closeness of dielectric function to *k*-axis is associated with the freezing. The enthalpy change calculated from the potentials of Lewis and Catlow [5] and Morelon et al. [4] provides unphysical values at some temperatures while the others reasonable reproduce the experimental data and MD simulation results. Overall, the hypernetted chain calculations, within the limitations, indicate that the potential of Yakub et al. looks the most suitable to study the liquid state properties of uranium dioxide.

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