Comparative study of thermophysical properties of Nickel in liquid phase

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Abstract. The results of molecular dynamics simulations of the self-diffusion coefficient (SDC) and viscosity coefficient (VC) of liquid nickel (Ni) in a temperature range of 1720 – 2500K are presented. The temperature dependence of the SDC is determined numerically by molecular simulations with the Embedded Atom Method Finnis-Sinclair (EAMFS) potential as implemented in the Virtual Nano Lab 2017.2. The temperature dependence agrees with the Arrhenius law. Results obtained for SDC are found to overestimate available experimental results, while results for VC underestimated experimental values. Also, the validity of Stokes-Einstein relation was tested in liquid Ni and was found to be valid at all temperatures.

Keywords: Liquid Ni, molecular dynamics, thermophysical properties.

1 Introduction

Thermophysical properties such as self-diffusion and viscosity coefficients are fundamental properties that are related to features of dynamics as well as the structure of particles of liquid materials (Morioka et al. 2002, Echendu et al. 2011). To enable successful material design from molten state and the modeling of solidification of alloys in the metallurgical industry, one requires an adequate understanding of these thermophysical properties, and their dependence on temperature, pressure and composition. Also, the knowledge of these properties helps to distinguish the liquid phase of a material from the solid phase, since atoms in solid phase only self-diffuse when defects exist, while in liquid phase the atoms diffuse from one place to the other when defects are either present or absent (Mei and Davenport 1990).

However, only a few experimental data are available for transition metals (Iida and Guthrie 1988), and the available results for diffusion coefficients in metallic melt usually show deviations up to one hundred percent (Banish and Lyle 1999). In line with this, Meyer (2010) reiterated that SDCs are normally
overestimated in a range of ten to one hundred percent and the temperature dependence deviates from the exact value. This motivated both experimental and theoretical investigations over the years.

Some of the earlier studies in this field are as follows. Qian et al. (1990) used the first principle liquid theory to compute the activation energy for diffusion in liquid sodium and found that the temperature dependence of SDC follows the Arrhenius behavior. Mei and Davenport (1990) performed a computer-based experiment to study the self-diffusion of liquid Ag, Au, Cu, Ni, Pd, and Pt. They employed the Embedded Atom Model (EAM) potential in their simulation at temperature 1720K and obtained a SDC for Ni as 3.894 X 10^-9 m²/s. Alemany et al. (1998) in their work did an MD simulation using the Voter and Chen version of the embedded Atom Model (VC-EAM) to study the dynamic properties of liquid Ni, and calculated diffusion constants and shear viscosity for liquid Ni at T= 1775K and T= 1875K. Also, Alemany et al. (1999) carried out an EAM molecular dynamics study of self-diffusion coefficient of Ni impurity in liquid Al and obtained results that are consistent with experimental values at T= 1091K and T= 1250K, respectively. Meyer et al. (2008), in their experimental study, determined the SDCs for liquid Ni by using the Electromagnetic Levitation with Quasielastic Neutron Scattering (ELQNS) technique. They obtained results for Ni at different temperatures, and observed Arrhenius behavior of temperature dependence self-diffusion coefficients. Lu et al. (2012) also carried out a MD simulation on the relationship between self-diffusion and viscosity in liquid metals. They employed EAM to study Ni and Cu and Modified-EAM (MEAM) to study Ni-Si alloys within a temperature range of 1500K–2200K. In their study, they showed that the self-diffusion and viscosity coefficients obey the Arrhenius law down the under-cooled state. Results from their study overestimated available experimental (Meyer et al. 2008) and theoretical values (Alemany et al. 1998). Meyer (2015) did an experimental measurement of self-diffusion coefficients for liquid metals (Ge, Ti, Ni, Cu, and Al) using the Quasielastic Incoherent Neutron Scattering (QINS) technique. The self-diffusion coefficient results for these liquid materials were found to be described by Arrhenius law temperature dependence within their melting points. For the nickel, results are slightly different from the experimental results obtained by Meyer et al. (2008) at T= 1750, 1810 and 1870K, respectively. Rozas et al. (2016) as well calculated thermophysical properties for liquid Ni at the melting point and obtained results that overestimate experimental data for both viscosity and self-diffusion.

From the aforementioned, it is clear that discrepancies exist between experimental and theoretical results for self-diffusion and viscosity coefficients of the transition metal (Ni). Therefore, there is a need for further studies to get correct and consistent results for these thermophysical properties that have useful applications. Hence, in this study, we employed the EAMFS potential to study the self-diffusion and viscosity coefficients for Ni
melt in a range of temperature, and compared our results with both experimental and theoretical results. We discovered that the temperature dependence follows the Arrhenius behavior. Also, the validity of Sutherland-Einstein relation is verified for liquid nickel and found to be valid.

2 Computational methods

We prepared a system with FCC crystal structure of nickel in space group 225 Fm-3m (Kings, 2002), using periodic boundary conditions on a supercell size of \(10a_0 \times 10a_0 \times 10a_0\) (where the lattice constant \(a_0 = 3.5249 \, \text{Å}\)) with 4,000 atoms. The simulations were performed using ATK-force field code (AtomistixToolKit 2017; Griebel et al. 2007) as implemented in the Virtual Nano Lab 2017.2 (AtomistixToolKit, 2017). The Embedded Atom Method Finnis-Sinclair potential (Jess and Brian 2000; Sheng et al. 2011; Daw and Baskes, 1984) is used to treat the interactions between atoms of liquid Ni. In the EAMFS (Mendelev et al. 2012), the energy of the system is given by

\[
E_{\text{tot}} = \frac{1}{2} \sum_{i<j} V(r_{ij}) + \sum_i F(\rho_i)
\]

(1)

\[
\rho_i = \sum_j \phi(r_{ij})
\]

(2)

where \(E_{\text{tot}}\) is the total energy, \(V(r_{ij})\) is the potential function, \(\phi(r_{ij})\) is the electron density contribution from atom \(j\) to atom \(i\), \(F(\rho_i)\) is the energy required to embed an atom into the electron density at site \(i\), and \(\rho_i\) is the total electron density at an atomic position \(i\) calculated through the linear superposition of electron density contributions from neighboring atoms. The supercell of crystalline nickel was first heated from 1500K to its melting point (1726K) for 200,000 steps at intervals of 2000 within a time integration step of one femto second (fs) at zero pressure using the NPT Martyna Tobias barostat. This enables us to increase the temperature gradually to its melting point. This implies that the ordered solid Ni has finally become a liquid that only exhibits a short-range order. Then, the liquid Ni is allowed to undergo annealing to bring the system to equilibrium. This is achieved by equilibrating the supercell structure of Ni melt at a constant temperature of 1726K, using the same NPT for 200,000 steps with Maxwell-Boltzmann velocity type. This eliminates any memory effect of the first solid nickel on the physical properties of liquid Ni obtained from the first supercell structure. Finally, the equilibrated Ni melt is annealed for another 200,000 steps using the same velocities from the second step of MD simulation as initial velocities. In this step, we used the supercell of the equilibrated Ni melt at a constant temperature of 1726K to collect enough statistical data for calculating the self-diffusion coefficients of liquid Ni. The entire process is repeated for a
range of temperatures below and above the melting point to obtain the thermophysical properties. Then, the Stokes-Einstein relation \( \eta = k_B T/2 \pi \sigma D \) is used to calculate the viscosity coefficients at different temperatures.

3 Results and Discussion

The self-diffusion coefficient \( D \) is calculated from the mean-square displacement (MSD) of the atomic particles in MD simulations (Quantumwise 2017) with

\[
D = \frac{1}{2} \left\langle X^2(t) \right\rangle,
\]

(3)

\[
\left\langle X^2(t) \right\rangle \approx \frac{1}{T_{MD} - t} \int_{t'}^{t_{MD} - t} \frac{1}{N} \sum_{j=1}^{N} \left[ r_j(t' + t) - r_j(t') \right] dt',
\]

(4)

where \( \left\langle X^2(t) \right\rangle \) is mean-square displacement of the metallic atoms in liquid phase calculated at the observation time \( t \), \( N \) is the total number of Ni atoms in the supercell, \( r_j(t + t') \) and \( r_j(t') \) are atomic position coordinates at time \( t + t' \) and \( t' \), with \( j=1, 2, \ldots, N \), \( t' \) is the initial time and \( t \) is the time interval of observation adopted to extract the atomic diffusion coefficient from the time dependent mean-square displacement. It is important to note that the observation time; \( t \) is always less than the total simulation time \( T_{MD} \) in the molecular dynamics calculation of liquid. As \( t \) goes close to \( T_{MD} \), the mean-square displacement becomes noisy because it is no longer linearly dependent on \( t \). Therefore, for exact calculation of the mean-square displacement only simulations with values of \( t \) much lesser than the simulation time are considered.

Figure 1(a) reports the radial distribution function \( g(r) \) as a function of \( r \) (Å) at 15 different temperatures. It clearly shows that \( g(r) \) converged very well at all simulation temperatures. At \( T=1720 \)K, the radial distribution function has the largest peak at the position 2.425 Å, while at \( T=1726 \)K – 2500K the peak of \( g(r) \) decreases with increasing temperature. The atomic self-diffusion coefficients are averaged over all Ni atoms using Eq (3). As the ordered solid Ni transforms into Ni melt, a short-range order is observed to be preserved on melting. It shows a well-defined peak around 2.425 Å and 2.475 Å, suggesting that this is the dominant nearest neighbor distance between atoms in nickel melt at the respective temperatures. The \( g(r) \) is observed to be temperature dependent. It decreases with increasing temperature. This means that as the temperature keeps increasing atoms in the Ni melt gradually becomes disorderly.
Fig. 1. Radial distribution function $g(r)$ for liquid Ni at different temperatures (a, left), and mean square displacement for liquid Ni at temperature ranging from 1720K to 2500K (b, right).

Figure 1(b) reports simulation time profile of the MSD at several temperatures from 1720K to 2500K. It is observed that in this range of temperatures, the MSD changes linearly with time and the slope of the mean-square displacement increases remarkably with temperature. This implies that metallic Ni has completely melted, and the system has undergone a transition from solid phase to liquid phase. The self-diffusion coefficient is obtained by taking the slope of mean-square displacement at the respective temperatures. It is observed to increase with increasing temperature as reported in Figure 2. This is so because the Ni atoms move more radically at higher temperatures. The results obtained are compared with the SDCs reported from other theoretical studies and experimental results as shown in Table 1. At simulation temperatures 1720, 1726, 1750, 1810, 1870 and 1920K, our SDC results overestimated the experimental results obtained by ELQNS and QINS. Though, the two experimental results also report different values at 1750, 1810 and 1870K. The calculated self-diffusion coefficients from this study are seen to be closer to experimental results obtained by the QINS technique (Meyer, 2015).

At $T=1720$K, theoretical results obtained using the EAM potential (Mei and Davenport 1990) overestimated the experimental results from QINS by 34.27%, while our results from the EAMFS overestimated the experimental result by 15.68%. Results from our calculations at $T=1720$K appears to be closer to experimental results than the EAM result. At $T=1775$K and 1875K, results from this study are higher than results obtained from the VC-EAM (Alemany et al. 1998) by 0.275 and 0.066, respectively.
Table 1. Simulated values of self-diffusion coefficient $D$ (in 10^{-9} m^2/s), viscosity coefficient $\eta$ (in 10^{-3} Pa.s), radial distribution $g(\AA)$ and position $r(\AA)$ for liquid Ni compared with both experimental and other theoretical results at different temperatures (T).

| $T$(K) | $D$  | $\eta$ | $g(\AA)$ | $r(\AA)$ | Present study | Other theoretical results | Experiment |
|--------|------|--------|----------|----------|---------------|--------------------------|------------|
| 1720   | 3.355 | 4.639  | 2.6808   | 2.425    | 3.894^c       | -                        | 2.90^c     |
| 1726   | 3.453 | 4.523  | 2.6712   | 2.475    | 3.639^e       | 3.217^c                  | 2.96^c     |
| 1750   | 3.791 | 4.178  | 2.6567   | 2.475    | 3.81^c        | 3.144^c                  | 3.54^b     |
|        |       |        |          |          | 3.10^c        |                          |            |
| 1775   | 3.875 | 4.149  | 2.6572   | 2.425    | 3.60^d        | 4.96^d                   | -          |
|        | 3.984 | 3.071^c| 4.24^c   | 2.976^c  | 3.75^b        | 3.46^c                   |            |
| 1810   | 4.029 | 4.065  | 2.6195   | 2.475    | 4.685^e       | 2.826^e                  | 4.07^b     |
|        |       |        |          |          | 3.80^c        |                          |            |
| 1850   | 4.351 | 3.848  | 2.6010   | 2.425    | 4.38^d        | 4.32^d                   | -          |
|        | 4.72^e| 2.815^e|          |          |              |                          | -          |
| 1920   | 4.635 | 3.748  | 2.5655   | 2.425    | 5.07^e        | 2.715^e                  | 4.2^c      |
| 1940   | 4.834 | 3.631  | 2.5479   | 2.475    | 5.23^c        | 2.673^e                  | 4.77^b     |
| 1950   | 5.010 | 3.522  | 2.5556   | 2.425    | -             | -                        | 3.77^f     |
| 2000   | 5.568 | 3.251  | 2.5369   | 2.425    | 5.71^c        | 2.556^c                  | -          |
| 2113   | 6.282 | 3.044  | 2.4830   | 2.425    | 6.64^c        | 2.365^c                  | -          |
| 2307   | 7.703 | 2.710  | 2.3831   | 2.425    | -             | -                        | -          |
| 2500   | 9.585 | 2.360  | 2.3195   | 2.425    | -             | -                        | -          |

^a Mei and Davenport (1990), ^b Meyer et al. (2008), ^c Meyer 2015, ^d Alemany et al. 1998, ^e Lu et al. (2012), ^f Assael et al. (2012).

There are no experimental results at these temperatures to compare our results with. At $T$=1720K to 2113K, we also compare our results to those obtained by Lu et al. (2012) using modified-MEAM. Their results overestimated experimental results (QINS) at $T$=1726, 1750, 1810, 1870 and 1920K by 22.95%, 22.9%, 22.54%, 23.29% and 20.71% respectively. Results from the current study are observed to be closer to experimental values because our results overestimated experimental results at these temperatures only by 15.68%, 16.65%, 22.29%, 16.44% and 17.34% respectively. The difference between our theoretical results and other theoretical values is expected because self-diffusion coefficients obtained from different potentials varies. While the disparity between the calculated self-diffusion coefficients and experimental results may be due to lack of uniformity of atomic ordering in the liquid phase of nickel. The temperature dependence of self-diffusion coefficients calculated with the EAMFS potential is also compared with results obtained from both experiments and theory as reported in Figure 2(a) and Table 1. It increases with increasing temperature. This is in line with the well-known Arrhenius behavior. We can see how our calculations...
overestimated the experimental results in Figure 2(a). The temperature dependence of the calculated viscosity coefficients compared with both theoretical and experimental results are reported in Table 1 and Figure 2(b). The shear viscosity of liquid Ni decreases with increasing temperature. When compared with experimental results, we found that our calculation underestimated the oscillating cup-experimental results by 16.6% at 1750K, 12% at 1850K, 6.58% at 1950K and 8.58% at 2000K. Also, when compared with other theoretical calculations, our results appear higher than results obtained with MEAM as shown in Figure 2(b). It is clear that results from this study are closer to experimental data than results obtained from MEAM and VC-EAM (Assael et al. 2012).

Following the work of Lu et al. (2012), we tested the validity of Sutherland-Einstein (S-E) relation (Sutherland, 1905; Einstein, 1905) in liquid Ni. Since this equation gives the relationship between the viscosity and self-diffusion coefficients: \( D = k_B T / 4 \pi r \eta \), we calculated the product of \( D \eta \) which linearly depends on temperature (\( T \)).

\[
D\eta = \left( \frac{k_B}{4\pi r} \right) T
\]

\[\text{(5)}\]
Table 2. Calculated values of $D\eta$ for liquid Ni compared with other theoretical results at different temperatures.

| $T$(K) | Calculation | Other theoretical results |
|--------|-------------|--------------------------|
|        | $D\eta$ ($10^{-11}$ J/m) | $D\eta$ ($10^{-11}$ J/m) |
| 1720   | 1.558       | -                        |
| 1726   | 1.563       | 1.171$^{(c)}$            |
| 1750   | 1.585       | 1.198$^{(c)}$            |
| 1775   | 1.608       | 1.223$^{(c)}$, 1.786$^{(d)}$ |
| 1810   | 1.639       | 1.262$^{(c)}$            |
| 1850   | 1.675       | -                        |
| 1870   | 1.694       | 1.324$^{(c)}$            |
| 1875   | 1.698       | 1.329$^{(c)}$, 1.892$^{(d)}$ |
| 1920   | 1.739       | 1.377$^{(c)}$            |
| 1940   | 1.757       | 1.398$^{(c)}$            |
| 1950   | 1.766       | -                        |
| 2000   | 1.812       | 1.459$^{(c)}$            |
| 2113   | 1.913       | 1.570$^{(c)}$            |
| 2307   | 2.089       | -                        |
| 2500   | 2.264       | -                        |

$^{(d)}$(Alemany et al. 1998), $^{(e)}$(Lu et al. 2012)

As reported in Table 2 and Figure 3, the product $D\eta$ appears to be linearly increasing with increase in temperature. Suppose the Sutherland-Einstein equation holds for liquid Ni, we obtain the atomic radius of Ni to be $r$=1.213 Å by fitting the data with Eq. (5). This is approximately equal to our simulation atomic radius ($r$=1.2125Å) being half the position of the first peak of the radial distribution function (Figure 1(a)). This indicates that the
Sutherland-Einstein relation is obeyed in liquid Ni. Also, since the self-diffusion and viscosity coefficients of the Ni melt obey the Arrhenius law, the product \((D\eta)\) can be written as

\[
D\eta = D_\eta \exp\left(\frac{-E_\eta}{RT}\right) \eta_\eta \exp\left(\frac{E_\eta}{RT}\right) = A_\eta \exp\left(\frac{\Delta E}{RT}\right)
\]

where \(\Delta E = E_\eta - E_\eta\), \(A_\eta = D_\eta \eta_\eta\), \(R\) is gas constant. Figure 3 displays the product \(D\eta\) as a function temperature. The gradual change of the slopes confirms the linear dependence on temperature.

4 Conclusions

The self-diffusion and viscosity properties of liquid Ni in a temperature range of 1720–2500K were calculated using MD simulations with EAMFS potentials. The results obtained are compared with available experimental and theoretical results. The self-diffusion coefficient results obtained in this study are found to overestimate the experimental values, while results for viscosity coefficient underestimated experimental results. The temperature dependence of these properties agrees with the Arrhenius law. The Sutherland-Einstein relation was found to be valid in liquid nickel.

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