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Temperature dependence diffusion coefficients of iron, boron and iron-boron calculated by molecular dynamics method

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Abstract. It has been calculated the diffusion coefficient of iron, boron and iron-boron system as temperature function using molecular dynamics simulation method. The diffusion coefficient is very important for knowing the physical processes. However, the diffusion coefficient data are not always available from experimental measurements, as so many applications using this data as an input of calculation. The computational molecular dynamics method shows a powerful tool for predicting the needed properties of material under consideration. In this work we predict the diffusion coefficient based on the Lennard-Jones potential under scheme of Lorentz-Berthelot mixing formula as the atomic interaction of material for molecular dynamics simulation. From this work we have determined the temperature dependence diffusion coefficient: $D_{Fe}(T)=5.20\times10^{-7}.\exp(-393.82/T)$, $D_B(T)=1.74\times10^{-6}.\exp(-297.62/T$, $D_{B\rightarrow Fe-B}(T) =2.50\times10^{-6}.\exp(-411.29/T)$ and $D_{Fe\rightarrow Fe-B}(T)=5.274\times10^{-9}.\exp(-930/T)$ in the unit of m^2/s.

1. Introduction

The diffusion coefficients are very important to study the mechanism and processes of physical phenomena in order to know the possible potential applications. In this research we study the diffusion proccesses of iron-boron system. The steels or other structural materials in a nuclear power plant have usually iron element as a major component. The coolant in the heat transfer system of reactor can be made from many possible materials and some of specific materials can be injected into the coolant for specific purpose as for corrosion inhibition. In our previous computational (molecular dynamics) work [1-6] we injected a small certain concentration of oxygen into high temperature molten liquid lead coolant (and also lead bismuth eutectic) in order to reduce the corrosion attack. We also applied the nitrogen as other inhibitor to know its effect to lower the corrosion rate of iron [7]. Experimentally, the use of oxygen for reducing corrosion rate of steels in liquid metal have also been investigated intensively by other researchers [8,9].

In current days Boron element shows an intensive application as a neutron absorber in nuclear reactor systems. The use of boron is interesting due to its low cost of production and fabrication,

besides of its popular high thermal absorption cross section. The mixing phenomena of coolant in a nuclear reactor power plant is an important mechanism for intrinsic safety against dilution of boron. In a PWR reactor, the boric acid is added into the coolant in such a way as to compensate the excess reactivity of the fuel [10]. Non-uniformity in the mixing of boron into coolant is also one of the most important safety topics of LWR (Light Water Reactors). The mixing phenomena is not only because of importance in nuclear safety, but also for structural integrity of nuclear materials [10]. So knowing the thermodynamics properties of boron are also important due to the integraty has correlation with atomic diffusion of materials.

The diffusion data of materials are also very important for studying synthesis and processes in materials. Many phenomena in nuclear power plant are studied by many methods using the diffusion data. Moving Particle Semi-implicit (MPS) method can study and predict the properties of the eutectic reaction during nuclear reactor accident, where solid–liquid system of eutectic interaction was developed [11]. The eutectic interaction is an important phenomenon which possible affects in the failure of reactor fuels, integrity of core structures and pressure vessel (RPV). The theoretical formulation in the MPS method includes the diffusion data of materials under investigation [12]. The diffusion data are often needed in the mode of temperature dependence. In the MPS method applies several fundamental models such as heat conduction, melting, solidification, surface tension, etc., which supports a more fundamental approach to analyse the physical mechanism. It is reported that eutectic formation rate is maintained by diffusion process [12, 13].

In this current work we study the self-diffusion and inter-diffusion coefficient of iron and boron. The iron is the major component of steels that widely used in nuclear reactor. Boron is material that often used in nuclear reactor applications, as control rods and mixed materials of coolants. The chemistry of the reactor coolant is maintained by the chemical and volume control system, which is designed to control water's chemical composition. The advantage of this system is to control the primary-water boron content as a function of the nuclear reactor's power level [14]. The degradation of steels in nuclear power plant (may) has relation with composition of coolant materials [1-7]. In our work we predict the temperature dependence of diffusion coefficient of iron and boron. In molecular dynamics simulation we used the Lennard-Jones (LJ) potential, eventhough this may not so accurate for metal systems. However, because this is very easy to be applied for many systems so we can apply it as a preliminary study. We also concern about diffusion at very high temperature where there was a high diffusion of iron in liquid lead coolant. The diffusion coefficients from molecular dynamics calculation then can be used as an important input for many calculations as in the MPS simulation [14,15].

2. Theory

The many models are suggested to describe the interactions among atoms or molecules in the substances [16]. One of the most frequently used is a simple fluid model, Lennard-Jones (LJ) potential:

$$u(r) = 4\varepsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^{6} \right] \tag{1}$$

where ε is the depth of potential energy well and σ is the separation distance at which the potential is zero, and for different components we used the Lorentz-Berthelot for simulations:

$$\sigma_{AB} = \frac{(\sigma_{AA} + \sigma_{BB})}{2} \qquad \text{(Lorentz rule)} \tag{2}$$

$$\varepsilon_{AB} = \left(\varepsilon_{AA} \times \varepsilon_{BB} \right)^{1/2} \qquad \text{(Berthelot rule)} \tag{3}$$

The diffusion coefficient that describing the diffusion of elements may be calculated by following equations as below:

$$MSD = \left\langle \left| \vec{R}(t) - \vec{R}(0) \right|^2 \right\rangle \qquad \text{(Mean Square Displacement)} \tag{4}$$

$$D = \lim_{t \to \infty} MSD/6t \qquad \text{(Einstein relation)} \qquad (5)$$

$$D(T) = D_0 \exp(-A/\Re T) \qquad \text{(Arrhennius formla)} \qquad (6)$$

where t is time, T temperature, A activation energy for diffusion to be happen and $\Re$ is a gaseous constant. When we use the MOLDY molecular dynamics code, the MSD calculation has been facilitated by this software [18,19].

3. Procedure of Simulation

Procedure to determine the diffusion coefficient by Moldy code can be described as below steps:

1. Preparing two input files: (1) specification file and (2) control file
    The specification file stores about: element, mass, charge, potential energy (see Table 1), number of atom, dimension of box simulation, coordinates of atoms or molecules. The control file (See Table 2) stores: pressure, temperature, number of simulation steps, etc.
2. Installation. We can install moldy in serial or parallel mode.
3. Simulation: iron material prepared inside boron material and simulation is done for some several high temperatures (500°C, 600°C, 700°C, 800°C, 900°C). By using Eq. (4), (5) and logarithmic of Eq. (6) then we can compute the $D(T)$. Figure 1 is detail view of the iron-boron system.
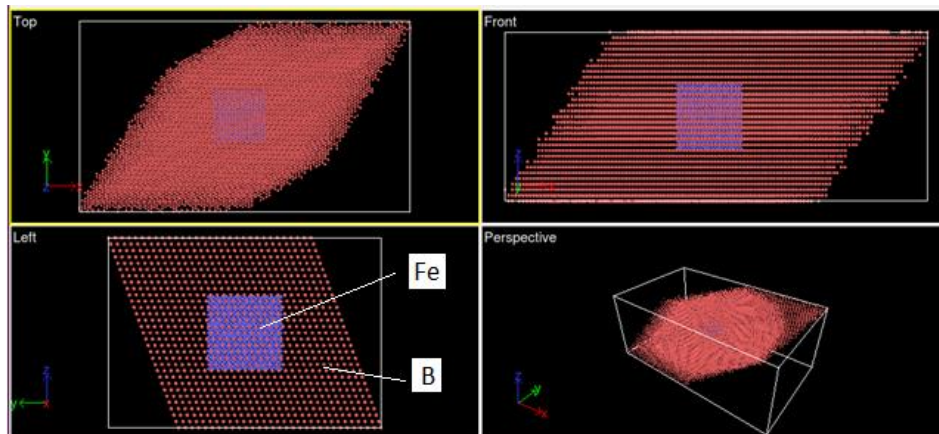

Figure 1. Vizualisation of Iron-Boron system before simulation with Ovito [20]

Table 1. Input data in specification file

Input	iron	Boron	iron-boron
mass	55.847	10.811	
crystal unit	bcc	Rhombohedral ($\alpha=\beta=\gamma=58.06°$)	Fe (bcc) in B (rhombohedral)
lattice constant	2.8665	5.06	
number of atoms	8192	26996	Fe = 8192 B = 25577
σ (Å)	2.3193 [21]	3.5430 [22]	2.9311
ε (eV)	0.4007 [21]	0.0041 [22]	0.0406

Table 2. Input data in control file

| Input of control	Values
begin-dump	20000
begin-average	20000
temperatures	*** (in kelvin)
lattice-start	1
const-temp	1
scale-options	2
const-pressure	4
pressure	*** (in Mpa)
dump-level	3
nsteps	60000
Step	0.0001
Subcell	2
cutoff	8.5125

4. Results and Discussions

After simulations, we can extract and compute the MSD. Then using the Eqs.(4 – 6) we can plot the pictures as shown in Figure 2, 3, 4 and 5.

4.1 Simulation of iron
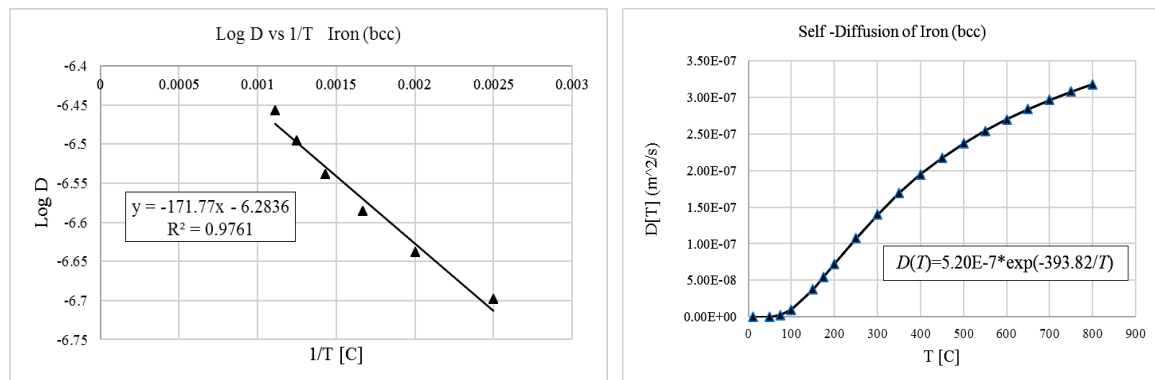
Figure 2. Plot log D vs $1/T$ and the $D(T)$ diffusion coefficient calculation of Iron (Fe)

4.2 Simulation of Boron
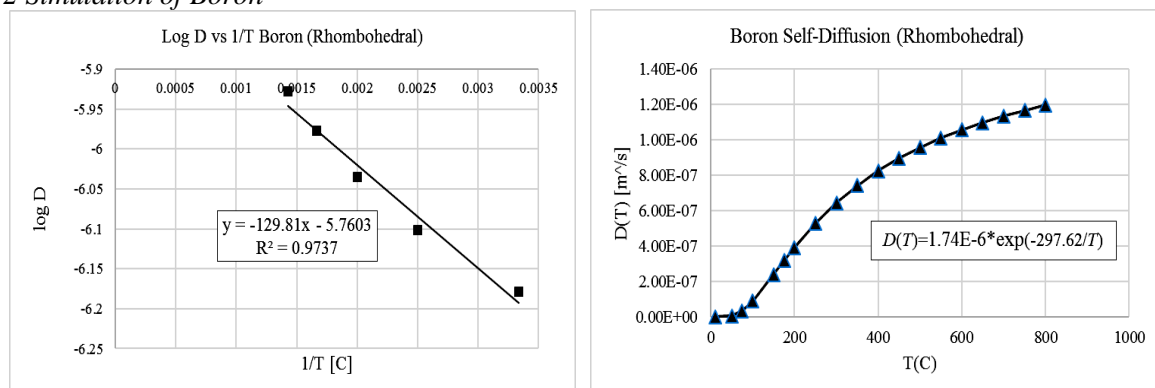
Figure 3. Plot log D vs $1/T$ and the $D(T)$ diffusion coefficient calculation of Boron (B)
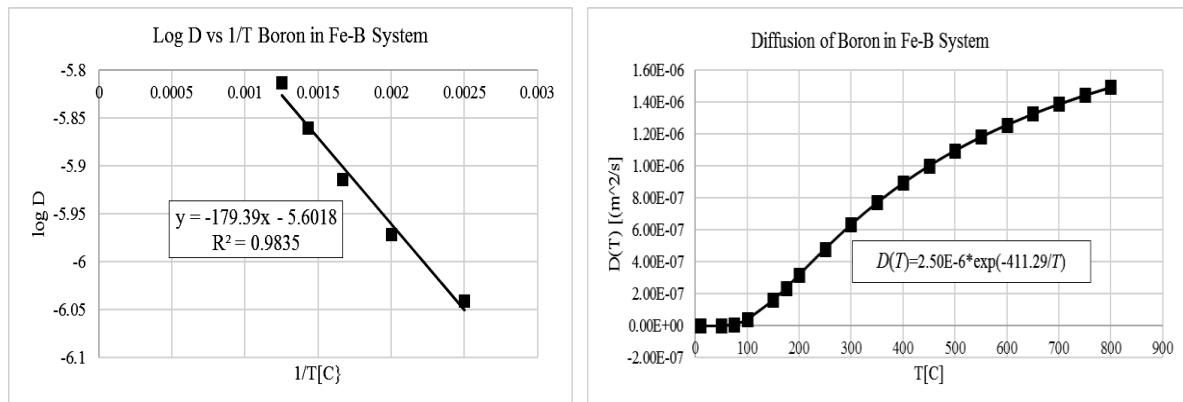
4.3 Simulation of Boron (B) in Fe-B system

Figure 4. Plot log *D* vs 1/*T* and the *D*(*T*) diffusion coefficient calculation of Boron in Fe-B

4.4 Simulation of Iron (Fe) in Fe-B system

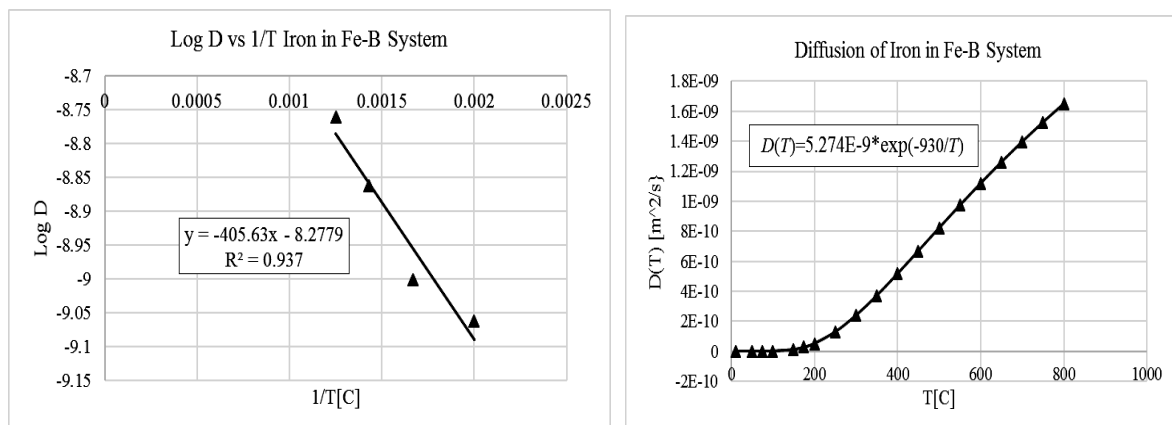**Figure 5**. Plot log *D* vs 1/*T* and the *D*(*T*) diffusion coefficient calculation of iron in Fe-B

Lets we collect all *D*(*T*) from figures Fig.2 to Fig.5:

$$D_{Fe \rightarrow Fe}(T) = 5.20 \times 10^{-7}.\exp(-393.82/T) \qquad [m^2/s] \qquad (7)$$

$$D_{B \rightarrow B}(T) = 1.74 \times 10^{-6}.\exp(-297.62/T \qquad [m^2/s] \qquad (8)$$

$$D_{B \rightarrow Fe-B}(T) = 2.50 \times 10^{-6}.\exp(-411.29/T) \qquad [m^2/s] \qquad (9)$$

$$D_{Fe \rightarrow Fe-B}(T) = 5.274 \times 10^{-9}.\exp(-930/T) \qquad [m^2/s] \qquad (10)$$

By MOLDY molecular dynamics simulation we have derived the diffusion coefficients of materials. Further, these results need to be checked and verified with available experimental data. However, we can underline from those results that:

1. Those results can be used as a first prediction in applications as far as the experimental data are not available. For example, those diffusion data can be used as an input in MPS simulation.

2. We can always make a correction for more accurate results by choosing a better potential energy function in our molecular dynamics simulation, or changing the geometry/configuration of simulated materials. In our work, we put the iron material in the center of borom material.

5. Conclusion

From our work we have shown and derived the diffusion coefficient of materials. The temperature dependence diffusion coefficients $D(T)$ have been derived based on the Einstein relation and Arrhenius formula. In this work the temperature dependence diffusion coefficient of B, Fe, B in FeB, Fe in Fe-B have been calculated as in Eq.(7) to Eq.(10). The molecular dynamics method can also describe other thermodynamics properties of materials and processes in which the experimenal data may not available or there are difficulties in experimental measurements as processes in the reactor core during reactor operation.

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