Iron Corrosion-Resistance in Lead-Bismuth Eutectic Coolant by Molecular Dynamics Method

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Abstract. In this present work, we report numerical study of iron corrosion in interaction with lead-bismuth eutectic. The goal of this work is to study how the oxygen can be used to reduce the corrosion rate of iron in lead-bismuth eutectic. The molecular dynamics method was applied to simulate corrosion process. By evaluating the diffusion coefficients, RDF functions, MSD curves of iron and also observed the crystal structure of iron before and after oxygen injection we concluded that a significant and efficient reduction can be achieved by injecting about 2% of oxygen into lead-bismuth eutectic.

Keywords: lead-bismuth eutectic coolant, corrosion, oxygen content, molecular dynamics method.

1. Introduction

Lead-bismuth eutectic (LBE) has been a potential candidate as a coolant material in advanced nuclear reactors design. It is also well known the steel cladding was severely corroded when they are exposed to LBE directly at high temperatures [1-2]. This crucial issue has presented a critical challenge in the use of LBE and then a full knowledge of corrosion characteristics and how to reduce corrosion are essential for safety of heat transfer systems in reactors. In our previous work we have used MD method to calculate the diffusion coefficient of iron in liquid lead/LBE [3]. In the present work we report the effect of oxygen injection into LBE to reduce the corrosion rate of iron in LBE by using MD method. We predicted the most effective oxygen injection for efficient iron corrosion reduction.

2. Fundamental theory

Molecular dynamics is a simulation technique in which the interacting atomic system is allowed to evolve for a specified period of time. In our work we used the Lennard-Jones (LJ):

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

where σ and ε are the LJ potential parameters. To maintain the validation of LJ potential in our simulation, we used the potential parameters as reported by Shu and Davies for Fe-Fe and Pb-Pb interaction [4]; using the potential parameter of O-O interaction as reported by Lemmon and

Jacobsen [5]; and using the Bi-Bi interaction from the fitting of EAM pair potential [6]. Meanwhile for cross-interaction: Fe-Pb, Fe-Bi, Fe-O, Pb-Bi, Pb-O, and Bi-O we approached using the popular Lorentz-Berthelot formula:

$$\sigma_{AB} = \frac{(\sigma_{AA}+\sigma_{BB})}{2} \quad \text{and} \quad \varepsilon_{AB} = (\varepsilon_{AA}.\varepsilon_{BB})^{1/2} \tag{2}$$

Table 1 summarizes the parameters of LJ potential that we used in our work. All simulations in our work was carried out by using the **MOLDY** molecular dynamics code [7].

Table 1. *The Lennard-Jones parameters that used in this research*

Pair Interaction	σ (Å)	ε (eV)	Pair Interaction	σ (Å)	ε (eV)
Fe-Fe	0.4007	2.3193	Fe-Pb	0.2766	2.7540
Pb-Pb	0.1910	3.1888	Fe-Bi	0.1538	2.6846
Bi-Bi	0.0590	3.0500	Fe-O	0.0639	2.7836
O-O	0.0102	3.4280	Bi-O	0.0245	3.1490
Pb-Bi	0.1061	3.1194	Pb-O	0.0441	3.2184

2.1 Diffusion Coefficient Calculation

Corrosion may be understood as a degradation of structural materials into its constituent atoms due to the chemical reactions with environment**.** The LBE coolant creates high temperature environment for reactor cladding that in turn causes corrosion. The degradation occurs due to high solubility of certain elements of the steel (Ni, Cr and Fe, mainly) into LBE [8]. Then liquid-metal corrosion for the most part simply depends on the solubility of the structural material/solid metal in the stagnant liquid metal.. In our work the corrosion phenomena was credible as a diffusion process where a fraction of Fe atoms leave their crystals bulk toward a high temperature coolant of liquid metal. We calculated the diffusion coefficients using three below successive equations:

$$MSD = \langle |\vec{r}(t) - \vec{r}(0)|^2 \rangle \tag{3}$$

$$D = \lim_{t\to\infty} \frac{\langle |\vec{r}(t) - \vec{r}(0)|^2 \rangle}{6t} \tag{4}$$

where *MSD* is mean-square displacement, *D* is diffusion coefficient, *r* is position of an atom and *t* is time. The temperature dependence of diffusion coefficient *D*(*T*), is evaluated by using the Arrhenius formula,

$$D(T) = D_0 \exp(-A/RT) \tag{5}$$

where *T* is a temperature, *R* is the universal gas constant, and *A* is an activation energy of diffusion.

3. Simulation System

We modeled the corrosion by placing the iron (1729 atoms in bcc crystal structure, with lattice constant a = 2.8286 Å) in the center of LBE as in *figure1*. The gray color is Pb atoms, cyan color is Bi atoms and the indigo color is Fe atoms.

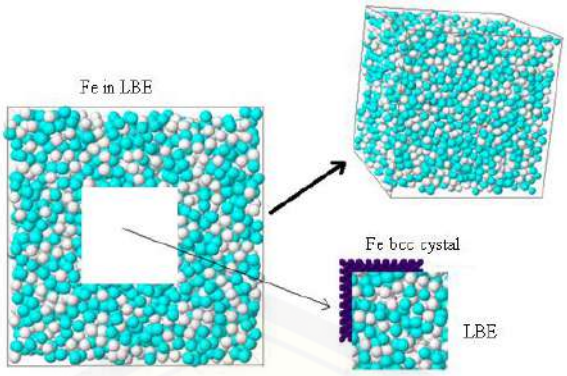

Figure 1. *Initial atomic configuration for FeLBE system (with Jmol at http://www.jmol.org).*

The liquid LBE was made from 2540 of Pb atoms (45.5%) and 3037 of Bi atoms (55.5%), with LBE density of 0.0274 atoms/Å^3. Dimension of whole system (FeLBE) is (63.2×63.2×63.2) Å^3.
To study the effect of oxygen injection then we spread the oxygen atoms evenly into LBE coolant with certain percentages (compared to the total number 5577 atomsof PbBi coolant): 28 (0.5%), 42(0.75%), 58(1.0%), 68(1.5%), 114(2.0%), 140(2.5%), 170(3.0%), and 192(3.5%) into coolant. The simulation was carried out at 750 ^{o}C. Then we calculate the diffusion coefficients to study the corrosion characteristic of iron.

4. Results and Discussion

4.1 Iron Diffusion in LBE without Oxygen Injection

Figure 2 is the plot of (1/*T*) vs. log *D*(*T*) from simulation data. The graph has linear equation:

$$logD(T) = -\frac{1020}{T} - 7.28 \qquad (6)$$

with correlation coefficient R^2 = 0.984. We can derive from this equation that the temperature dependent of iron diffusion coefficient in LBE is :

$$D(T)_{0\%} = 5.28 \times 10^{-8} \exp(-2340.18/T) \qquad [m^2/s] \qquad (7)$$

Using this equation we can compute the diffusion coefficient at 750 ^{o}C, is:

$$D(T = 750°C)_{0\%} = 2.33 \times 10^{-9} \qquad [m^2/s] \qquad (8)$$

S.Banerjee has reported that the iron diffusion in LBE is $D_{Fe \rightarrow LBE}$ = 2.27 ± 0.11 x 10^{-9} m^2s^{-1} = (2.16 – 2. 38) x 10^{-9} m^2 s^{-1} at temperature 750 $^\circ$C [1]. Our calculation Eq.8 then is still in the range of Banerjee's result.