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IOP Conf. Series: Journal of Physics: Conf. Series **1170** (2019) 012010 doi:10.1088/1742-6596/1170/1/012010

Performance of the Fe-Ni-Cr steel alloy in high temperature molten liquid lead

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Abstract. Degradation of steels used in the fast nuclear reactor that using liquid metals as a coolant material is still a major problem. Whereas, the design of the fast nuclear reactors as an IVth generation reactor has many advantages. Many investigations have been done with the purpose of finding the best steel alloy material and finding the best mechanism of corrosion reduction of material. Theoretically, to predict a needed novel material and also knowing the mechanism of corrosion inhibition, we can use computational methods as (MD) molecular dynamics method. Using molecular dynamics, we can explore new potential materials and new promising corrosion inhibition mechanism based on the calculated properties of materials. In the current MD work, we study Fe-Ni-Cr performance if used in a high-temperature molten lead liquid (coolant) and observing how this material has the possibility in fast nuclear reactor applications. We also studied the mechanism of corrosion inhibition based on the injection of oxygen gas into the coolant. From MD simulation we have got an important conclusion about the FeNiCr performance that may be useful for nuclear reactor design.

1. Introduction

In the current days, the need for energy supply is very crucial to support all our daily activities. Unfortunately, the popular energy sources such as a hydrocarbon or geothermal are very limited for a long time of purposes. It is reasonable to expect and use nuclear energy as a possible and promising solution for our need of much energy. The fast nuclear reactor is a type of nuclear reactors where the primary coolant may a liquid metal, as a lead liquid. The extraordinary thermophysical properties of liquid lead as a coolant giving many advantages for applications, particularly related to its heat transfer properties as conductivity [1]. The lead (alloys) coolant have a much higher density than the traditional water coolant, and they can remove heat more rapidly and also allow much higher power

density. This makes the lead coolant is attractive in a situation where size and weight are important, like on ships and submarines.

However, though there are many advantages to using liquid metals, there are disadvantages. The difficulty related to corrosion phenomena of using the lead or lead-bismuth metals coolant is an example [2,3]. This is a current major issue and problem in fast nuclear reactor design. There is a need a comprehensive data for reactor applications related to the liquid metals corrosion. The costs are also an important thing for using a liquid metal as coolant material [1]. To overcome this problem, we need to find a mechanism of corrosion inhibition or using new corrosion resistant materials. Operating the lead cooled reactor at the temperature of 550 ^{o}C is readily achievable but 800^{o}C is envisaged with the support of advanced novel materials to provide lead corrosion resistance at high temperature where it would enable thermochemical hydrogen production. For real application in nuclear reactors design, there is the need for having a comprehensive data (experimentally or computationally) about liquid metal (lead) corrosions. In this current work, we want to study computationally the corrosion problem of steel material due to the liquid lead coolant application. We want to study how to inhibit the corrosion and also how the used steel material performance under high-temperature molten lead. We use the molecular dynamics simulation method that is one of the powerful computational tools. For inhibition, we will use popular oxygen agent, and for steel material, we use nickel-chromium-iron for preliminary study. As we know nickel-base alloys are used in many applications where they are subjected to high-temperature environments. Nickel-chromium alloys or alloys that contain more than about 15% Cr are used to provide resistance at temperatures exceeding 760^{o}C as inside of reactor core. Nickel-chromium-iron alloys have been developed to provide high strength and excellent corrosion resistance. This group of alloys led the way to higher strength and resistance to elevated temperatures. For using steels in liquid metals, Fe-alloys materials with Cr will be a choice [4]. Considering a very high-temperature environment of Fe-alloys in liquid metal, then in this study we simulate a material with low thermal expansivity and a modulus of elasticity unaffected by change with temperature. The material as the elinvar type with chromium content 5% and nickel 36% will be studied in this [5]. For simplicity, we study the FeNiCr alloy with composition 60%, 35%, 5%.

2. Methods

2.1 Potential Energy

The system of materials is composed of atoms. Among atoms, there are interaction forces that mathematically can be described by potential energy function. For different form, type or phase of materials there may also be a different kind of interaction (potential energy). There are many kinds of interaction potential: the van der walls, hard-sphere, Lennard-Jones (LJ), ionic, Sutton-Chen, Gupta and the embedded atomic method (EAM), etc. The LJ potential is the most popular one that may be applied for many materials or systems. The EAM potential is generally suggested for metal systems [6]. The LJ potential is in a very simple form then is very easy for implementation in the simulation. However, for metal systems, the LJ potential is usually not recommended and one should use another type of potential like the EAM potential for more accurate calculation. However, creating the EAM potential is not easy because we need quantum mechanical calculation and time-consuming calculation. So the LJ potential is still used in a simulation, especially for preliminary calculation or just for easy prediction of phenomena. In our current work then we still use the LJ potential for predicting and calculating the physical properties of corrosion liquid lead phenomena [7-9].

The Lennard-Jones potential can be stated as [10],

$$u(r)=k\varepsilon\left[(\sigma/r)^{n}-(\sigma/r)^{n}\right] \quad (1)$$

Where ε and σ are the potential parameters and k is a proportional constant. Especially for n = 12 and m = 6 and k = 4, then we have a popular (12-6) LJ potential function,

$$u(r) = 4\varepsilon\left[(\sigma/r)^{12} - (\sigma/r)^{6}\right] \quad (2)$$

Eq.(2) will support the interactions between pairs of atoms or molecules. The values of ε and σ for a type of atom can be estimated based on fitting of experimental data. Having two different types of atoms then we need to have cross-interaction potential parameters. Following the popular **Lorentz-Berthelot** mixing formula then the cross interaction parameters are [7-9]:

$$\sigma_{AB} = (\sigma_{AA} + \sigma_{BB})/2 \quad (3)$$

$$\varepsilon_{AB} = \sqrt{\varepsilon_{AA} \times \varepsilon_{BB}} \quad (4)$$

Table 1 shows potential parameters for a pair of atoms: Fe-Fe, Pb-Pb, Ni-Ni, Cr-Cr, Fe-Ni, Fe-Cr, Pb-Ni, Pb-Cr, Ni-Cr.

Table1. Values of potential parameters ε and σ

Pair Interaction	ε (eV)	σ (Å)	Reference
Fe-Fe	0.4007	2.3193	[7-9]
Pb-Pb	0.1910	3.1888	[7-9]
O-O	0.0102	3.2480	[7-9]
Ni-Ni	0.3729	2.2808	[10]
Cr-Cr	0.413	2.336	[10]
Fe-Pb	0.2766	2.7541	Eq.(3) & (4)
Fe-Cr	0.4068	2.3275	Eq.(3) & (4)
Fe-Ni	0.3865	2.3000	Eq.(3) & (4)
Pb-Cr	0.2808	2.7623	Eq.(3) & (4)
Pb-Ni	0.2668	2.7348	Eq.(3) & (4)
Ni-Cr	0.3924	2.3083	Eq.(3) & (4)
Fe-O	0.2766	2.7451	Eq.(3) & (4)
Pb-O	0.0441	3.2184	Eq.(3) & (4)
Ni-O	0.0617	2.8544	Eq.(3) & (4)
Cr-O	0.0648	2.8819	Eq.(3) & (4

2.2 Diffusion Coefficient

In the classical molecular dynamics (MD), generally we solve the Newton equation of motion $\boldsymbol{F} = \mathrm{d}\boldsymbol{p}/\mathrm{d}t$ based on the potential function $U(r)$ via equation $F = -\mathrm{d}U/\mathrm{d}r$. We need to solve the equation of motion numerically based on a specific algorithm of the integration method. There are many schemes of numerical integration, one of them is the Beeman algorithm, that is also adopted in our current work. Creating a good MD program (code) is another difficult work and not is our current focus. Rather than developing a new own MD code, we use a powerful available code as the Moldy code that was developed by Keith Refson [11].

The result of the Newton equation is the trajectories of the atoms and using the statistical mechanic's formulation we can compute many physical properties. In our work, we will focus on the calculation of diffusion coefficient of materials, especially for diffusion of iron in lead liquid because the corrosion of materials (in the reactor) actually can be seen from the degradation of material by dissolution of elements of material (iron or steel) that flowing into a liquid lead. The diffusion calculation can be computed by using the equations as below [12]:

$$MSD = \left\langle \left| \vec{R}(t) - \vec{R}(0) \right|^2 \right\rangle \quad \text{(mean square displacement)} \qquad (5)$$

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

Basically, in our work, we want to find the lowest diffusion coefficient iron in the liquid lead ($D_{Fe \to Pb}(T)$)of material FeNiCr alloys that showing lowest corrosion whereas we also injected a certain oxygen concentration into the liquid lead coolant.

3. Result and Discussion

3.1. Simulation Method

Before running a simulation using the MOLDY code, we need to determine the geometry and structure of simulated materials in a certain coordinate of atoms. This means we should determine all coordinates of atoms in FeNiCrPb material before corrosion inhibition and coordinate of atoms of FeNiCrOPb for corrosion inhibition. The Fig.1 shows the material structure of FeNiCr in liquid Pb (blue) (visualized by Ovito code) [13]).

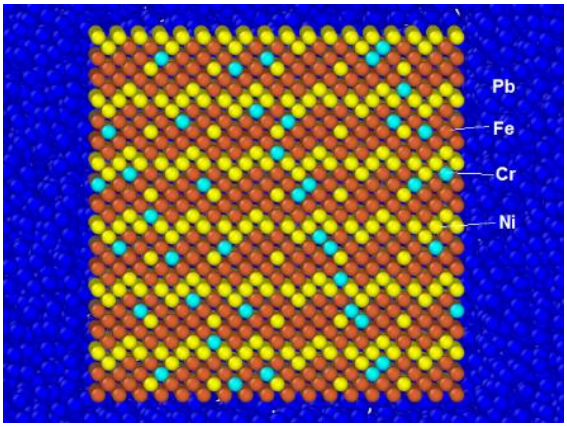

Figure 1. FeNiCr system before the simulation

The simulation procedure of MD simulation that can be described as follow:

1. Simulations run using Moldy code [11] with control input:
   NPT ensemble, Anderson mode, zero pressure, Nose-Hoover thermostat and 0.0001 ps of time size, modified Beeman integration algorithm, LJ potential energy, n steps = 60000, begin-average = 20000. The number of atoms and coordinates of atoms is based on the Fig.1 configuration.
2. Simulating a certain temperature (we focus on temperature T = 1023 K).
3. Calculate MSD and $D(T)$ of iron after complete simulations.
4. Compare the calculated coefficient diffusion of iron with available experimental data.
5. Do conclusions.

3.2. Simulation and Calculation Results

The output of simulation can be extracted (by using msd and mdavpos Moldy utilities)to find MSD and coordinates of atoms. The msd command line will produce msd file of diffusing material, and mdavpos command line will produce the xyz coordinate of atoms of diffusing material. Using Eq. (5) And (6) then we can calculate the diffusion coefficient of iron in liquid lead metal. We compute the iron diffusion $D_{Fe\rightarrow Pb}$ (T = 1023K) with oxygen injection and without oxygen injection. The setup of simulated material is Fe(60%)Ni(35%)Cr(5%) placed in the center of liquid lead (Pb). The oxygen used for corrosion inhibition in this simulation was taken about 0.75% of total lead coolant atoms. In detail number, the system was comprised from 6448 Fe atoms, 3581 Ni atoms, 716 Cr atoms, 45006 Pb atoms, and 340 O atoms. The mass of Fe is 55.845 amu, Pb is 207.19 amu, O is 15.998 amu, Cr is 51.9668 amu, and Ni is 58.693 amu [14]. By this configuration, the outer dimension of the simulation box is 123 x 124 x 125.1 $Å^3$.

The temperature was set 1023K with a reason there is experimental data at this temperature [1]. Before oxygen injection (pure Fe in liquid Pb) in the liquid lead then the computed diffusion of iron is $D_{Fe\rightarrow Pb}$ = 1.42 $\times 10^{-9}$ m^2/s. Adding the Ni and Cr atoms into iron (so we have FeNiCr) still without oxygen then we have $D_{Fe\rightarrow Pb}$ = 1.34 $\times 10^{-9}$ m^2/s. Injecting oxygen into liquid lead randomly and immersed pure iron into them (Fe in PbO) then we have $D_{Fe\rightarrow Pb}$ = 7.38 $\times 10^{-12}$ m^2/s and then immersed FeNiCr into the liquid lead with oxygen (FeNiCr in PbO) we have $D_{Fe\rightarrow Pb}$ = 4.00 $\times 10^{-10}$ m^2/s.

The clear results of the simulation can be explained below. The diffusion coefficient $D(T$=1023K) of iron in lead liquid can be determined by Eq. (6). This equation can be calculated by computing the MSD values, and this value can be generated by msd command line as explained in Moldy manual. It is from the simulation the addition of Ni and Cr at certain percentage will decrease the diffusion rate of iron from $D_{Fe\rightarrow Pb}$ = 1.42 $\times 10^{-9}$ m^2/s to $D_{Fe\rightarrow Pb}$ = 1.34 $\times 10^{-9}$ m^2/s. The very important thing is adding the oxygen into the liquid lead at a certain small percentage (0.75%). In this study, we use this percentage with a reason as a result in our previous study for lowest diffusion [8]. We see it put FeNiCr into the liquid lead with oxygen (FeNiCr in PbO) then we have $D_{Fe\rightarrow Pb}$ = 4.00 $\times 10^{-10}$ m^2/s. There is a significant reduction using oxygen and not using oxygen. So the importance of this study has shown that the MD simulation can be used to predict the materials that will be used for nuclear applications with promising physical properties. For example the property of the material with lowest diffusion coefficient under high-temperature liquid lead environment as in nuclear reactor. Figure 2 is the performance of FeNiCr alloy with several compositions Fe-Ni-Cr.

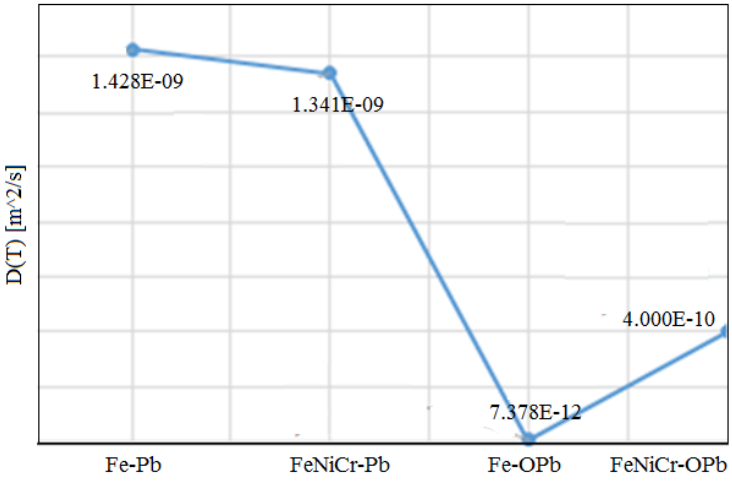

Figure 2. The diffusion of Pure Fe and Fe-alloys in liquid lead

4. Conclusions

The simulation work has shown important results as following:

1. The use of Cr, Ni to create FeNiCr steel alloy has strengthened the material from an attack of high temperature liquid molten lead, rather than it is in the mode of pure iron. This can be seen from reduction of diffusion from $D_{Fe\rightarrow Pb} = 1.42 \times 10^{-9}$ m^2/s to $D_{Fe\rightarrow Pb} = 1.34 \times 10^{-9}$ m^2/s.
2. Injecting the certain small concentration of oxygen into a liquid molten lead, it has also reduced the corrosion significantly. Injecting oxygen 0.75% (of number of lead atoms) has reduced the diffusion value from $D_{Fe\rightarrow Pb} = 1.34 \times 10^{-9}$ m^2/s to $D_{Fe\rightarrow Pb} = 4.00 \times 10^{-10}$ m^2/s for FeNiCr alloy.
3. The important view of this work is the MD simulation can be used as a powerful tool to predict the new materials with expected valuable properties for future applications.
4. The suggestions for future works, we need more detail of simulation for many different oxygen concentrations for getting comprehensive results. Also, we need to use more accurate potential energy rather than LJ Lorent-Bertholet Potential, to get better results.

Acknowledgment

The authors thank the DRPM DIKTI Republic of Indonesia for financial support of this project under the scheme of Hibah Fundamental (PD-UPT) 2017-2018 Contract Number: SP DIPA-042.06.1.401516/2018: 5 December 2017".

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