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Interaction Between Liquid Lead and FeNi Material Using Molecular Dynamics Simulation

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Abstract. The liquid lead corrosion is often described as a damage of the metal surface due to the high solubility of atoms of the metal that flowing into the liquid metal medium through the diffusion process. This research was conducted to examine the performance of FeNi alloys in liquid lead at various compositions, especially the metal structure conditions due to interactions between metal atoms at high temperatures. To see the performance of this FeNi alloy steel, the parameter that you want to know is the diffusion coefficient of its constituent elements. The potential used is the Lennard-Jones potential. This research uses the LAMMPS molecular dynamics simulation software. From the simulation works can be concluded that the lowest diffusion coefficient of Fe in liquid lead which produces the strongest structure is at the composition (concentration) 65% Fe and 35% Ni with related diffusion coefficient of 5.8582 x 10-12 m2/s, where at this value the corrosion of FeNi in Liquid lead can already be reduced till 77.32%.

Keywords: Liquid lead corrosion, Molecular dynamics, FeNi Alloy, Composition, LAMMPS

Introduction

In general, the definition of corrosion is the damage that occurs to a material due to a chemical reaction. The presence of a substance that acts as an electrolyte that functions as a conductor of electricity causes the corrosion process to occur [1]. Corrosion of liquid metal is a special kind of corrosion called hot corrosion where the phenomenon is that there is no electron transfer (chemical reaction) due to the solubility of solid metals, so sometimes it can only be done by looking at the physical properties of the material such as the diffusion coefficient [2]. The term corrosion for it is often described as damage to the metal surface due to the high solubility of steel/iron atoms into the liquid metal medium through the diffusion process.

Generally, the basic daily energy needs of a country can be met by using oil, coal and natural gas. However, the availability of conventional energy sources is very limited and over time, the amount will be depleted due to the increasing consumption rate according to the times. The use of nuclear fuel in the process of generating electricity in nuclear reactors is an effective way to produce large amounts of energy that is deemed capable of meeting future energy needs. Reactor design that utilizes the concept of nuclear fission reactions is the basis for the development of nuclear reactors that are operated at this time and for many years to come [3]. One of the design of the Gen-IV nuclear reactor is a liquid metal cooled fast nuclear reactor (exp. lead) which has

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many advantages, one of which is being able to transfer large amounts of heat from the reactor core out to the electric turbine system [4].

However, in addition to the advantages and disadvantages, the fast reactor cooled by liquid lead metal still has drawbacks that must be resolved. One of them is the presence of corrosion in the steel metal material covering the uranium fuel in the nuclear reactor system. At the operating temperature of the reactor (high temperature) this steel material which interacts with the liquid metal coolant (functions to transfer heat from the nuclear reactor fission reaction to the electric turbine) according to experimental data, experiences very high corrosion. The diffusion of atoms of the wrapping steel material into the liquid metal which results in corrosion is caused by the interaction with the liquid metals [5].

One type of future reactor (Gen-IV) that uses liquid heavy metals as a coolant and does not use water as a coolant is the Fast Breeder Reactor (FBR). From a chemical and physical point of view, this liquid lead (Pb) refrigerant is a very favorable coolant candidate for the design of fast nuclear reactors. The lead (Pb) liquid metal coolant does not react actively with water or air so it does not trigger an explosion due to the chemical reaction process. This liquid metal also has high thermal conductivity and heat capacity, making it an efficient heat transfer medium [6].

In general, there are several types of reactor concepts that have been developed, one of which is LFR (Lead cooled Fast Reactor) which is cooled by liquid metal such as lead (Pb), lead with bismuth (Pb-Bi), and also LBE (Lead Bismuth Eutectic). One way to maximize the utilization of energy generated from a reactor is to choose the type of material so that it does not experience unmanageable corrosion. The interaction between liquid lead (Pb) coolant with structural materials and fuel cladding at high temperatures which can cause easy corrosion is a common problem that occurs in this type of reactor [3].

Nuclear reactor systems can also be designed more optimally in terms of economy and safety by taking into account the selection of cooling materials used. Things that must be considered are low melting point values but have high boiling points, heat resistance, corrosion system formation, low swelling ability, high heat transfer and so on [3].

Theoretical Background

Diffusion is a slow process in the process of moving atoms in the system, while the phenomenon that occurs due to a potential gradient in the material system is called diffusion transport. The value of the diffusion coefficient increases substantially with increasing temperature, as in chemical reactions. The overall rate of the process is limited when diffusion slows down. There are two things that are distinguished in predicting the diffusion process, namely self-diffusion which is the average displacement of molecules and transport diffusion which is a collection of molecules due to driving forces. To describe the interactions between particles, atoms or molecules using a force field. Newton's second law of motion equations in numerical form are used in molecular dynamics simulations. Newton's second law states that the acceleration of a particle is proportional to the net force on the particle and inversely proportional to its mass,

$$a_i = \frac{F_i}{m_i} = \frac{d^2 r_i}{dt^2} \quad (1)$$

where a_i is the acceleration of particle i, F_i is the net force acting on the particle, m_i is the mass of particle i, r_i is the position of particle i and t is time [7].

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Material system input in the form of material structures either in crystal form or in other forms is versi necessary in dynamic molecular simulations [8]. Crystalline structure are very simple solids formed in certain elements, where the atoms occupy the positions of the lattice [9].

Computational methods that can be used to predict the static and dynamic properties of a system, which are derived directly from interactions at the atomic or molecular level, are the understanding of molecular dynamics [10]. Molecular dynamics is used to predict the physical quantities that you want to know based on the designed model and based on the given input. Using the potential function according to the atomic trajectory in solving or finding Newton's equations of motion in this method [2].

The Lennard-Jones potential to describe metals interaction was used in this study. The experimental data fittings were obtained from the Lennard-Jones potential parameters consisting of (σ) and (ε) [11]. In general, the Lennard-Jones potential equation is as follows:

$$U(r) = k\varepsilon\left[\left(\frac{\sigma}{r}\right)^{n} - \left(\frac{\sigma}{r}\right)^{m}\right] \quad (2)$$

The following Table 1 lists the Lennard-Jones parameters for some of the materials used in this study:

Table 1 Lennard-Jones Potential Parameters
(Data source from [2])

Pair interaction	ε (eV)	σ (Å)
Fe – Fe	0.4007	2.3193
Pb – Pb	0.1910	3.1888
Ni – Ni	0.6	1.9

For simulations, we use the LAMMPS molecular dynamics program that contains a set of classical molecular dynamics codes, which can be used to simulate the behavior of up to billions of particles in their solid, liquid and gaseous states. LAMMPS integrates Newton's equations of motion for collections of atoms, molecules and macroscopic particles that interact with initial state or boundary condition forces [12].

To visualize the material system, we use the Ovito program. The use of OVITO is to visualize 2D and 3D models. OVITO is an open-source software that is used as a support in analyzing data from molecular dynamics simulations, such as LAMMPS [13].

Materials and Methods

Materials

a) Iron

Iron is a metal material needed by the world to build life [14]. Pure ferrous metal is soft and malleable, silver in color and looks shiny. Iron has an FCC (Face Centered Cubic) structure at high temperatures (> 910°C) and changes back to a BCC (Body Centered Cubic) structure at a temperature of 1390°C. Iron has an atomic radius of 126 pm and a melting point of 1535°C [15].

b) Lead

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Lead is the heaviest metal of all carbon metals, has a blue-gray color with a shiny surface and a soft texture. This lead can usually be found in soil. Lead has characteristics, among others, cannot conduct electricity, sound, vibration well and has a melting point at a temperature of 327°C [16].

c) Nickel
Nickel has a melting point of 1455°C with an atomic radius of 124 pm. Nickel is resistant to atmospheric corrosion in the passive state at normal temperatures [15].

Methods
This research begins with creating a simulation system. The simulation system is modeled in the form of a cube measuring 66.20Å x 66.20Å x 66.20Å. The system model used is a simulation system consisting of metal alloys, namely iron (Fe) and nickel (Ni) placed in liquid lead. The simulation cube contains 5698 atoms of liquid lead with an empty center that will be used as a metal alloy for FeNi. The metal alloy FeNi contains 1214 atoms.

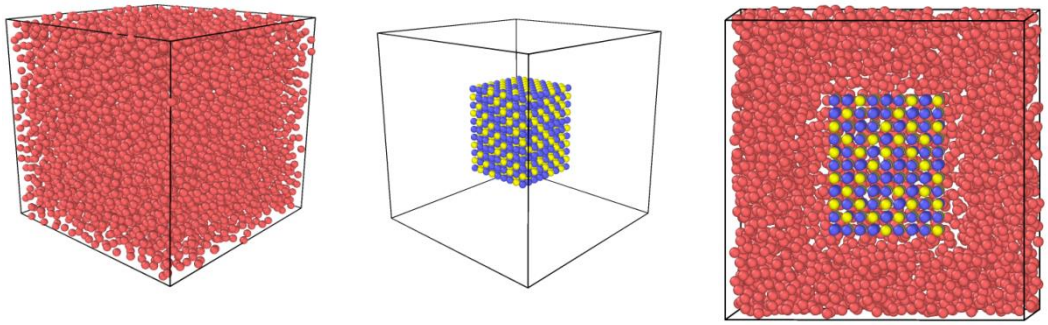

Figure 1. Visualization (a) Liquid Lead (b) FeNi Alloy Metal (c) Modeling of FeNi Alloys in Liquid Lead

Figure 1 shows a visualization of the simulation system model in this study, where liquid lead is shown in red, iron atoms are shown in dark blue, and the alloying element (Ni) is shown in yellow. Figure 1(a) is liquid lead with an empty middle that will be used as a metal alloy for FeNi, figure 1(b) is an alloy of iron and nickel, figure 1(c) is a slice of the combined system model between liquid lead and FeNi alloy.

Then create an input script for LAMMPS. The simulation input used in this study is an input file that has been prepared and contains several quantities in the form of variables that affect the state of the system such as metal crystal shape, crystal constant, atomic number, atomic weight, potential shape and parameters, number of simulation steps, temperature simulation, simulated pressure, calculated physical quantities.

The calculation of the diffusion coefficient (D) is done by calculating the slope value obtained from the gnu plot results. The results of these calculations will be represented in the form of a diffusion coefficient curve that shows the relationship between the concentration (%) of each variation in the composition of the FeNi metal alloy and the diffusion coefficient.

Then the analysis of the results and the visualization of the simulation obtained is in the form of an analysis of the structure of ferrous metal with the visualization of the simulation results in the

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form of a crystal structure analysis/CNA (Common Neighbor Analysis) used to visualize the data analyzed using the OVITO program. The results of running the input file on the LAMMPS generate a data dump file in the form of the XYZ position and then enter it in OVITO to observe the shape and number of crystal structures that are still left from the results of running simulations through color coding in CNA analysis so that based on the visualization, conclusions can be drawn for simulation research that has been carried out done.

Results and Discussion

This research is computational research that uses molecular dynamics simulation to predict and study the characteristics of FeNi alloys in liquid lead with various compositions. Simulations were carried out on several variations in the number of nickel atoms combined with iron atoms. Simulation data were analyzed by calculating MSD, diffusion coefficient and visualization of metal crystal structures.

To describe the value of the rate of diffusion indirectly can use the diffusion coefficient, which is directly proportional to the rate of corrosion of iron. In general, the higher the diffusion coefficient, the higher the corrosion rate. Conversely, the lower the diffusion coefficient, the lower the corrosion that occurs. The following is Table 2 correlation of the concentration of FeNi alloy composition with the value of the diffusion coefficient from the analysis of the MSD plot.

Table 2. Diffusion Coefficient Values based on Variations in Concentration of FeNi Alloys in Liquid Lead

No	Composition Concentration (%)	D (m^2/s)
1	0.5	6.3093×10^{-11}
2	5	1.0938×10^{-11}
3	10	5.2904×10^{-11}
4	15	5.1675×10^{-11}
5	20	8.5228×10^{-11}
6	25	7.4160×10^{-11}
7	35	5.8582×10^{-12}
8	40	3.5164×10^{-11}

Table 2 shows the data for the value of the diffusion coefficient that has been obtained from the MSD plot results after performing the simulation. Diffusion coefficient data can be used to analyze the corrosion of iron that occurs in liquid lead metal. These data are ferrous metal mixed with nickel (FeNi in Pb). The analysis of the results of the diffusion coefficient will clearly be displayed in the form of a curve as shown in Figure 2.

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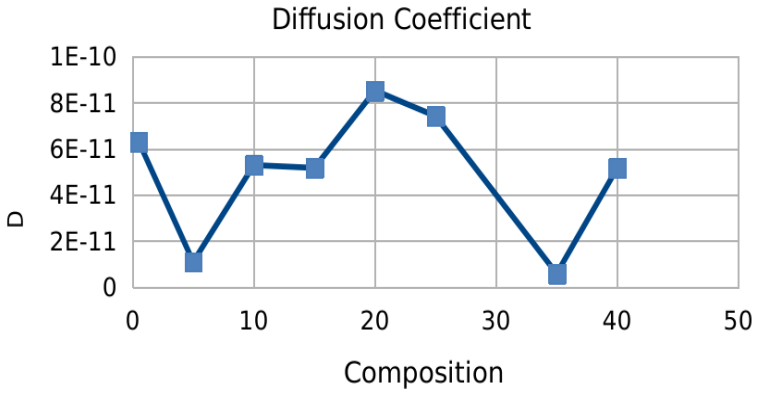

Figure 2. Diffusion Coefficient of Iron in Liquid Lead

Figure 2 shows the diffusion coefficient value curve. Based on the simulation, variations in FeNi concentration at a certain percentage will decrease the rate of iron diffusion which indicates a low corrosion rate. The value of the lowest diffusion coefficient is in the range of 5.8582 x 10^{-12} m^2/s – 1.0938 x 10^{-11} m^2/s. FeNi alloys in liquid lead which have small diffusion coefficient values are at concentrations of 5% and 35%. Concentration of 95% Fe + 5% Ni showed a diffusion coefficient value of 1.0938 x 10^{-11} m^2/s, and concentration of 65% Fe + 35% Ni showed a diffusion coefficient of 5.8582 x 10^{-12} m^2/s.

Based on the diffusion coefficient, the calculation of the corrosion rate using MSD slope analysis shows that the right composition between Fe and Ni which produces the most stable material structure (small damage rate) when interacting with metal is at a concentration of 65% Fe + 35% Ni which is strengthened by analysis of the amount of structure crystals using CNA on the OVITO program which showed that the crystals were also better at a concentration of 65% Fe + 35% Ni. This is in accordance with Ernik's research (2017), which states that Fe-18%Ni steel is able to reduce corrosion in liquid lead by lowering the diffusion value of iron and Fe-20%Ni is able to maintain FeNi steel structure by reducing the amount of damage to the atomic structure. Based on the results obtained, it shows that the differences between Ernik's research (2017) and experiments are quite different, due to the difference in the number of atomic interactions and the program used, in previous studies using 3 atoms (Fe, Ni, and Cr) and using the MOLDY program, while in This research only uses 2 atoms (Fe and Ni) and uses the LAMPS program. So it can be said that in this study the best percentage of composition to produce superior material in the form of corrosion-resistant steel is the composition of FeNi with a concentration of 65% Fe + 35% Ni. The smaller the value of diffusion, the smaller the level of damage that occurs and produces the most stable material structure when interacting with liquid metal.

Conclusions

Based on research on the effect of liquid lead on the structure of FeNi alloys in various compositions using molecular dynamics methods, it can be concluded that the right composition between Fe and Ni which can produce the most stable material structure (small damage rate) when interacting with liquid metal is the composition of FeNi with a concentration of 65% Fe + 35% Ni. The lowest diffusion was produced at 65% Fe + 35% Ni which was able to reduce corrosion up to 77.32%.

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