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## Short Communication

# A Physical Reason Why Molten Lead-bismuth Alloy has Advantage of Being Used as a Potential Coolant for Fast Nuclear Reactors Design

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**Abstract.** The development of new energy sources from nuclear energy is increasingly becoming the choice of many countries. One type of nuclear reactor that is promising to be developed in the future is a liquid lead-bismuth cooled fast nuclear reactor. The important use of this liquid bismuth eutectic (LBE) for coolant of fast nuclear reactor has long been a potential choice. The LBE with composition of Pb and Bi commonly used and about Pb 44.5wt% and Bi 55.5%wt. What is so special about this composition? This study seeks to see the physical reasons why the fast reactor nuclear design uses LBE with a composition of about Pb 44.5wt% and Bi 55.5%wt. From the result of the molecular dynamics simulation of iron in the LBE at a temperature of 1023K, it can be observed that the use of LBE liquid metal coolant with that composition caused the least damage effect (corrosion) to iron compared to using other compositions. It can be concluded from the calculation of the CNA values (Common Neighbour Analysis) of the iron structure in the LBE which shows that the crystal structure of iron in the LBE is the most stable for that composition. This simulation result is extraordinary because it explains the popular choice of the PbBi composition for the coolant candidate of fast nuclear reactors.

**Keywords:** molecular dynamics, liquid metal corrosion, CNA, LBE, Pb - Bi coolant

Nuclear power plants have provided much electricity energy for the world. The IAEA reports that there are about 437 nuclear power reactors in operation and 69 nuclear power reactors under construction in 31 countries around the world in the year 2016 (Kanti and Sanathkumar, 2016).

The development and construction of thermal reactors have been a research and development program by several member countries of the International Atomic Energy Agency on fast reactor design, cooled by liquid, metal, gas or others (IAEA, 2012). For the development of nuclear energy concepts to meet consumer needs, it was known that the properties of lead-bismuth alloy coolants in lead cooled fast reactor (LFR) systems offer many potential advantages for reactor designs and applications (Allen and Crawford, 2007).

Molten liquid Pb-Bi coolants (often called LBE = lead-bismuth eutectic) offer a number of interesting properties: chemically inert with air and water (unlike sodium coolant), low vapour pressure over the relevant temperature interval range, high boiling temperature (compared with sodium coolant) and others (Loewen and Tokuhiro, 2003). However, it was also already known for a long time that Pb - Bi coolants at the temperatures of interest can be very corrosive to structural materials.

The study reported by Wang *et al*. (2021) about liquid metal causing high corrosion phenomena to the structural materials (EP823, T91, ODS and authentic steels) in the reactors.

The LBE is one kind of the heavy liquid metals family (HLM). The HLM compatibility with structural materials including corrosion and liquid metal embrittlement is among the main challenges preventing widespread applications such as fast nuclear reactors. Researchers have designed a rotating disc experiment to study flow accelerated corrosion of 316L steel in LBE at a temperature of 600 °C and upto 3.14 m/s linear velocity for 150 h. They found that above 2 m/s LBE affected zone is less sensitive to flow velocity and that erosion starts at 3 m/s in Osman (Anderoglu *et al*., 2021). This means that the liquid metal corrosion and its flow velocity are very important to be considered in applications. Another corrosion experiment of steels in

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a flowing Pb - Bi environment has also been studied. Specimens were exposed to the melt at 300 °C and 470 °C for upto 3116 h. The flow velocity was also close to 2 m/s and the oxygen concentration in Pb - Bi was maintained at 1-2 x 10-6 wt% for ensuring stable protection of steels against liquid metal corrosion at least for that period (Barbier and Rusanov, 2001). The research studies that observed the relation between corrosion rates and corrosion mechanisms we also done where selected steel was exposed to high velocity (above 2m/s) in flowing LBE at a temperature of 500 °C (Woloshun and Rubio, 2013).

The investigation of LBE coolant for nuclear reactor designs is carried out not only experimentally but also theoretically and computationally. Authors have studied LBE material computationally using the molecular dynamics simulation method for many years (Arkundato *et al*., 2019; Arkundato *et al*., 2015; Arkundato *et al*., 2014; Arkundato *et al*., 2013a; Arkundato *et al*., 2013b; Arkundato *et al*., 2012). From these works they have shown the effect of LBE on the degradation of material (iron) and its corrosion inhibition using oxygen injection (for appropriate limited very small concentration in LBE), mostly using the Lennard-Jones potential for simulation. The behaviour of iron, lead and oxygen using the molecular dynamics methods investigated by Soontrapa (2009) and the EAM potential for some specific accurate calculated. They show the oxidation of iron during the corrosion process.

For above simulation, (Arkundato *et al*., 2019; 2015; 2014; 2013a; 2013b; 2012) directly used the LBE coolant with composition of about 44.50 wt% Pb and 55.5 wt% Bi. They used the composition that is usually applied in many kinds of literature. It is in our current work we want to investigate the reason why many researchers use LBE with composition 44.5 wt% Pb and 55.5 wt% Bi. We will simulate the effect of LBE with various compositions on the structural stability of iron crystals for checking different composition.

**Molecular dynamic simulation.** The examined material of our simulation was the Pb-Bi (in several eutectic compositions) that prepared and placed covering the iron BCC crystals in the center. The simulation run is based on Newton's equation of motion $Fi= -\nabla U(r_{ij})$ for studying the characteristics of simple fluids, where U is the interatomic potential. The simulation was carried out using moldy molecular dynamic software. Moldy is one of many molecular dynamic programs for studying the statistical mechanics of small molecules, atoms or ions (Refson, 2000). This code has been developed to analyze many structures of molecules and crystals, like modeling molecular liquids, solids, crystals, polymers and so forth. By simulation, we can reach the thermodynamics structure of many forms of material and learn many physical phenomena of it. We can analyze the structure of materials before and after simulation at certain temperatures and pressures.

**Lennard-Jones potential.** Our work uses Lennard – Jones potential which is one of some potential types which is supported by moldy code. This potential is widely used for intermolecular potential in the classical simulation of molecular dynamics. The equation of this potential is:

$$U(r) =4e[(S/r)^{12}-(S/r)^{6}] \quad (1)$$

where;

e and S is the parameter of Lennard – Jones Potential (Wang *et al.*, 2019) and $r=|r_1-r_2|$, $r_1$ and $r_2$ denoted the vector position of two particles. The physical description of the Lennard-Jones potential can be visualized in Fig. 1.

Every pair interaction that we used in this work as shown in Table1.

In equation (1), S is the distance at which the atom-atom potential energy U is zero and it is often known to the "size of the atom", whereas, e is the depth of potential well that is also known to as "the dispersion energy".

**LBE hot corrosion.** Normally, the corrosion degree of metal can be known as the degradations of materials structural as a result of chemical reactions with the environment. For LBE corrosion phenomena at high temperature this is often known as "the hot corrosion

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

| Pair interaction | e(eV) | S(Å) |
|---|---|---|
| Fe-Fe | 0.4007 | 2.3193 |
| Pb-Pb | 0.1910 | 3.1888 |
| Bi-Bi | 0.0590 | 3.0500 |
| Pb-Bi | 0.1061 | 3.1194 |
| Fe-Pb | 0.2766 | 2.7540 |
| Fe-Bi | 0.1538 | 2.6846 |

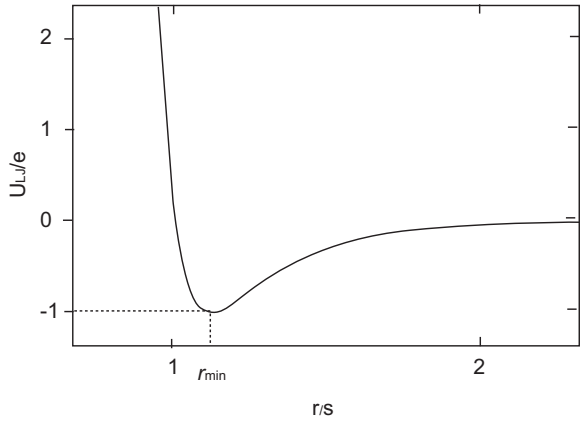


**Fig. 1.** The Lennard-Jones potential of two particles as a function of distance r (Shekaari and Jafari, 2021).

type" where there is no transfer of electrons during the reaction. So, we can describe LBE corrosion as a diffusion process. Many atoms from a metal surface experience high solubility flowing to the LBE medium (Zhang and Li, 2008).

The degradation can occur due to the high solubility of certain elements of the steel into the LBE as a coolant (Moosa, 2008). In this work the corrosion phenomena were considered as a diffusion process of Fe atoms leaving their crystals bulk toward into high temperature LBE coolant. In this research, we used the CNA (common neighbour analysis) method to determine the iron corrosion level of Fe BCC metal in LBE coolant by identifying the iron structures after corrosion simulation (Stukowski, 2010). In the current work, this corrosion phenomenon will be simulated at temperature of 1023K. As stated earlier, the purpose of this research is to find out what is the best composition of Pb and Bi in LBE which has the potential to be used as a coolant. To find out the best composition we will do many Pb-Bi simulations with different compositions of Pb and Bi. The best composition choice is the composition which shows the lowest level of iron damage. This can be known by choosing the highest CNA value of iron which describes the most stable BCC structure of iron after undergoing diffusion due to corrosion.

**Common neighbour analysis.** The CNA value of the iron structure can be calculated by using the common neighbour analysis (CNA) menu on OVITO an open visualization tool (Stukowski, 2010). This CNA method has its origin in paper of Honeycutt and Andersen (1987). They encode *n*-body clusters using their neighbourhood relationship. They generalized this method to a larger set of clusters to characterize the structure of a material. OVITO (Open Visualization Tool) is open-source program for the visualization of scientific data for molecules and other particle bases. This tool can be used to calculate a number of crystals using the CNA analysis menu (Stukowski, 2010).

**Details of simulation.** Our work is simulating the BCC iron crystal placed in the center of LBE for different compositions of Pb and Bi concentration. The simulation was carried out using Moldy code at temperature 1023K, using NPT ensemble, at vacuum pressure. Illustration of our simulation procedure can be seen in the following steps:

**Pre-processing step.** The simulated material system is prepared in the specification file, containing information: Iron BCC crystal in the center of LBE Fig. 2, lattice constant of Fe is 2.8286 Å, number of atom Fe = 10745, Pb-Bi = 45006, potential parameters as in Table 1 and LBE compositions were prepared for 11 cases of simulations in the form of liquid state as shown in Table 2.

**Processing step.** To run the simulations (Fe in Pb-Bi), the needed information is stored in the control input as: Nose Hoover Thermostat scheme, temperature = 1023K; pressure = 0 (Andersen constant pressure mechanism); n steps = 30000 (number of integration) and step = 0.0001 (time step of integration).

**Post-processing step.** The calculation of physical property (CNA value) can be done in the following ways: extract the dump file of Moldy simulation using

**Table 2.** Percentage of Fe-BCC structure that is still available after corrosion simulation

| Cases | Pb (wt%) | Bi (wt%) | Fe BCC (%) |
|---|---|---|---|
| Case 1 | 49.78% | 50.22% | 52.70% |
| Case 2 | 39.80% | 60.20% | 52.40% |
| Case 3 | 29.82% | 70.18% | 53.10% |
| Case 4 | 19.86% | 80.14% | 51.70% |
| Case 5 | 9.92% | 90.08% | 53.80% |
| Case 6 | 89.92% | 10.08% | 51.90% |
| Case 7 | 79.86% | 20.14% | 51.50% |
| Case 8 | 69.82% | 30.18% | 51.50% |
| Case 9 | 59.79% | 40.21% | 51.70% |
| Case 10 | 44.51% | 55.49% | 54.00% |
| Case 11 | 55.06% | 44.94% | 53.20% |

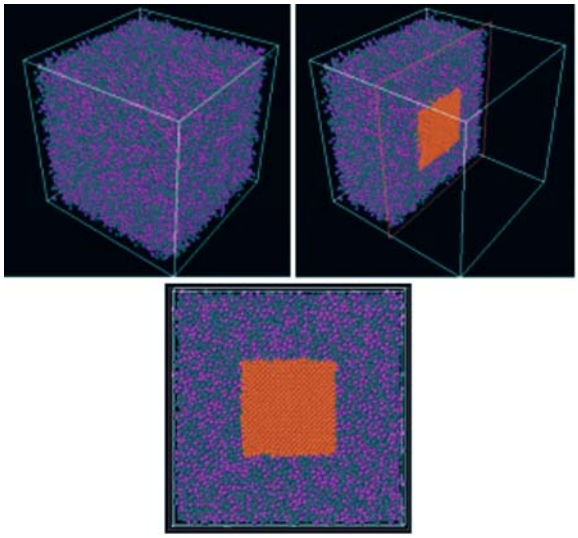

**Fig. 2.** Fe in LBE (Pb-Bi) after simulation.

MDAVPOS utility as in the manual Moldy (Refson, 2000), open the extracted file (in XYZ format) using Ovito code, calculate CNA value as in manipulation menu of the Ovito code for /iron structure only and compare every CNA values of iron for all composition of Pb-Bi.

Purple and gray particles are Pb-Bi and the orange particles in the center of the Pb-Bi system and iron. The number of Fe atom is 10745, prepared in the form of a BCC crystal structure. is 45006 in the form of a liquid state. The distance between Pb-Bi and Fe atom is equal to **S** parameter.

The LBE affects the structural change of iron after experiencing corrosion Fig. 2. The quality of corrosion depended on the compositions of Pb-Bi of LBE. Several compositions of Pb-Bi coolant have been simulated at a temperature 1023K. This range is still relevant to (Montanari *et al*., 2019) results between 125 °C to 720 °C. The CNA values of Fe material after corrosion simulation for certain different LBE compositions can be seen in Table 2. In our simulation work we ran 11 simulations of different cases (compositions).

It is from many experiments, is known that the liquid bismuth eutectic (LBE or Pb-Bi) causes high corrosion to structural materials such as iron or steels. After some time of interaction, many atoms of Fe on the surface of BCC iron crystal will be dissolved into LBE. Iron undergoes high corrosion, so the number of BCC structures will be reduced by no more than 100%. The number of these BCC structures can be known from the CNA value of iron materials after experiencing corrosion. Column 4$^{th}$ of Table 2 gives information on how much % of the BCC of the structure after it has undergone corrosion. In this case the different compositions of Pb and Bi will relate to the % number of the BCC structure of the iron remaining during corrosion process. From Table 2 it is clear that mixing of Pb and Bi in the ratio of Pb = 44.51 wt% and Bi = 55.49 wt% gives a better effect on the stability of the iron material against this liquid Pb-Bi attack. Lead and Bismith composition seems to cause minimum corrosion on the iron compared to use other compositions. Therefore, this simulation work has shown that the use of Pb-Bi as a coolant for a fast nuclear reactor will provide more safety if using a composition of about Pb = 44.5wt% and Bi = 55.5 wt%. In case 5, with a comparison of Pb = 9.92 wt% and Bi = 90.08 wt%, the results are close to case 10 which is 53.80%. Why is that? The explanation for this seems to require in-depth follow-up research and is not reviewed in this study. However, if we look at the selling price of these two materials, the price of bismuth is very much more expensive than the price of lead. Therefore, it is certainly not recommended to use such LBE with Bi = 90.08 wt%.

## Conclusion

Some compositions of Pb-Bi liquid have been chosen and simulated to prove which composition of Pb-Bi is the best as a coolant candidate for the LFR energy system. This composition produces the smallest structural damage of Fe BCC crystal. By using Ovito CNA analysis, it has been known that 54% of Fe BCC crystals did not affect by Pb-Bi coolant. Then, the promising Fe-Pb composition of LBE coolant should be used for the concentration of Pb 44.5 wt% and Bi 55.9wt% for the potential application of LFR system.

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**Conflict of Interest.** The authors declare they have no conflict of interest.

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