

Material Challenges for Corrosive Environments and High Temperatures in Lead-Cooled Fast Reactor

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Abstract

Research on one of the generation IV reactors, the Lead-Cooled Fast Reactor (LFR), began in the 1950s. The development of this reactor continues until now. However, there are material challenges in the development of LFR. The use of liquid Pb or Pb-Bi as an LFR coolant poses a significant challenge due to severe corrosion. Researchers have tested various materials such as steel, ceramics, composites, and refractory alloys in liquid Pb or Pb-Bi environments to assess their corrosion resistance. These materials have shown improved radiation performance at high temperatures and have been developed (such as ODS, FeCrAl, SS316, AISI 316 EP823, AISI 304, and HCM12A). However, these materials are not yet sufficiently compatible with corrosion performance. The results indicate that no metal or ceramic material currently proves to be completely resistant to corrosion and radiation over the long term. The LFR system is intriguing but has limited applicability until suitable construction material designs are further identified. Therefore, further research in this field is crucial to address material challenges and unlock the potential applications of LFR in the future. Future research should focus on developing materials with high corrosion resistance to ensure the longevity and safety of the reactor. Additionally, materials must be able to withstand high radiation conditions to maintain structural integrity and reactor performance. It is also necessary to develop new materials such as stainless steel, ceramics, and refractory alloys that can meet all the requirements for LFR.

Keywords: LFR, liquid Pb, corrosive, material challenges

INTRODUCTION^{1*}

Research and development of LFR began in the 1950s (IAEA 2012). A.I. Leipunsky, an Academician at the Institute of Physics and Power Engineering (IPPE) of the Soviet Union, initiated research and design of LBE (Lead-Bismuth Eutectic) as a reactor coolant in the early 1950s. The main goal of IPPE in the Soviet Union was to develop nuclear submarines. The United States also began research on lead-based submarines around the same time as the Soviet Union (Macdonald and Buongiorno 2002). The LFR research agenda was resumed in the 1990s (Alessandro Alember et al. 2020). Then, in 1997, Russia proposed the concept of the Lead-cooled BREST (Orlov et al. 2005) as a medium and small power reactor (Zrodnikov et al. 2006). This was followed by the European Union, which developed the conceptual designs for ALFRED (Advanced Lead

Fast Reactor Demonstrator) and ELFR (European Lead Fast Reactor) as part of the EU FP7 LEADER (Lead-cooled European Advanced Demonstration Reactor) project (Grasso et al. 2014). Japan has also developed several innovative LFR concepts, such as the PBWFR (Pb-Bi Cooled Direct Contact Boiling Water Fast Reactor) (Pramono et al. 2005), SLPLFR (Steam Lift Pump Type LFR) (Nakazima et al. 2006) and CANDLER (Constant Axial shape of Neutron flux, nuclide number densities and power shape During Life of Energy producing reactor) (Sekimoto 2010). China has also developed a lead-based reactor called the China Lead-based Reactor (CLEAR) for the ADS China project. CLEAR is a pool-type research reactor cooled by LBE with a capacity of 10 MWth, named CLEAR-I (Zhang et al. 2020) (DOE 2002) (Cinotti et al. 2010). Overall, Russia and European are considered to be the most advanced in LFR research and development with significant

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contributions in reactor design and technology. The Lead-Cooled Fast Reactor is one of six advanced and promising Generation IV reactor types (Achuthan et al. 2021). An illustration of the shape of the Lead-cooled Fast Reactor is depicted in Figure 1.

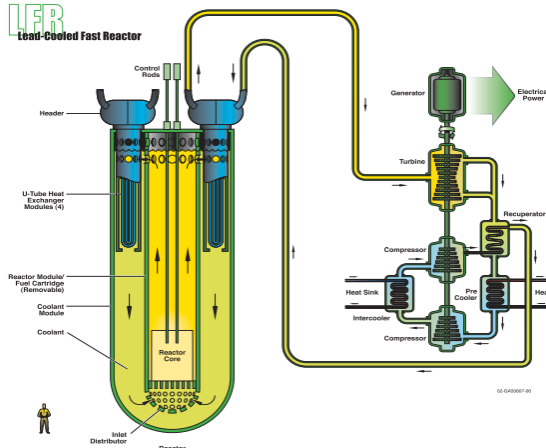


Figure 1. Illustration of the Lead-cooled Fast Reactor (DOE 2002)

The LFR is a reactor cooled by liquid lead (Pb) or lead-bismuth (Pb-Bi) and operates on a fast neutron spectrum at high temperatures (up to 1743°C). The LFR system ranks highly in terms of sustainability because it uses a closed fuel cycle and offers resistance to proliferation and physical protection due to its long-lasting core. This system is considered excellent in terms of safety and economics. Safety is enhanced by the choice of inert coolant. The LFR is designed for power generation and hydrogen production (Cinotti et al. 2010) (IAEA 2019) (Allen and Crawford 2007a) (IAEA 2019). One of the coolants for LFR is Lead-Bismuth Eutectic (LBE), which has a lower melting point compared to pure lead. The use of LBE as a coolant also helps prevent the freezing of the primary coolant. One of the countries using LBE is Russia. Russia wants to switch from LBE to pure lead (Pb) due to high costs (Allen and Crawford 2007a). Additionally, the temperature for lead (Pb) coolant in LFR is typically around 500°C, and the fuel temperature is 1432 K (Sofu 2019). The effect of radiation is also important for LFR because it can avoid swelling (Allen and Crawford 2007a). Based on the existing advantages, the phase diagram of the lead system is illustrated in Figure 2. Behind its advantages LFR also has many drawbacks that need to be addressed to ensure its success in applications such as Lead (Pb) or Lead-Bismuth (Pb-Bi) coolant can cause severe corrosion of reactor construction materials. This requires the development of materials that are highly resistant to corrosion. The materials used in LFRs must be able to withstand very high operating temperatures and under high radiation conditions. Many existing materials do not fully meet these

requirements. The use of lead or lead-bismuth as a coolant can be very expensive, mainly due to the need for specialized corrosion and radiation resistant materials. Complex LFR design require advanced technology and extensive development, which can take significant time and cost.

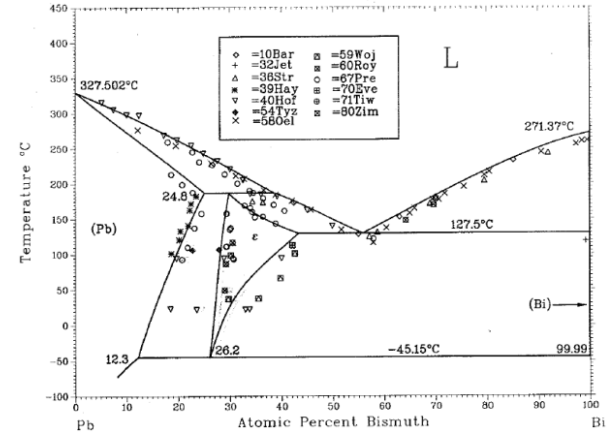


Figure 2. Phase diagram of the lead system (Allen and Crawford 2007a)

Therefore, based on the advantages and disadvantages of LFR mentioned in previous paragraph, materials for LFR must meet three criteria: adequate mechanical properties (strength, flexibility, and toughness) within the range of operating temperatures, stress, and dose; adequate dimensional stability (resistance to swelling and thermal, as well as irradiation) within the range of operating temperatures, stress, and dose; and finally, resistance to corrosion, stress corrosion, and embrittlement due to liquid metals under all operating conditions. Examples of low-temperature LFR materials include the BREST or INEEL burner designs, which are most likely metals, particularly austenitic stainless steel or ferritic/martensitic steel (Allen and Crawford 2007a). HT9 is a ferritic or martensitic steel developed for power generation in the 1960s and introduced into the fast reactor and fusion material programs in the United States in the 1970s. Table 1 summarizes the evolution of five generations of high-temperature materials over the past 60 years, starting from 21/4Cr-1Mo (T22) material (Hejzlar et al. 2004).

Table 1. Evolution of ferritic/martensitic materials for the power generation industry (Allen and Crawford 2007b)

Generation	Year	Steel modification	10^5 hours rupture strength (MPa)	steel	Maximum temperature (°C)
0	1940-60		40	T22, T9	520-538

1	1960-70	Addition of Mo, Nb, and V to simple Cr-Mo steels	60	EM12, HCM9M, HT9, HT91	565
2	1970-85	Optimisation of C, Nb, and V	100	HCM12, T91, HCM2S	593
3	1985-95	Partial substitution of W for Mo and addition of Cu	140	NF616, E911, HCM12A	620
4	Future	Increasing W and adding Co	180	NF12, SAVE12	650

Based on the previous explanation, HT9 is a first-generation steel with limited temperature capability. Table 1 shows that EM12, a duplex ferritic/martensitic steel, has been researched in France for implementation in fast reactors in Germany (W. Nr. 1.4914) and the UK (FV448). However, Generation IV steel contains cobalt, making it unsuitable for nuclear use (Allen and Crawford 2007a).

The purpose of this review is to understand the development of LFR and the main challenges for the LFR design developed to date. One of the main challenges in the development of LFR is the need for materials that can withstand extreme operating conditions, including corrosive environments, high radiation exposure, and high pressure and temperature. Therefore, materials that are corrosion-resistant, radiation-resistant, and mechanically compatible are crucial for the Lead-cooled Fast Reactor (LFR) to support the reactor's sustainability and efficiency. Without materials that meet these three aspects, there will be an increase in operational

costs, potential system failures, and safety risks. Thus, investment in research and innovation in materials is a key factor in the development of next-generation reactor technology, particularly LFR, making it one of the GEN IV reactors as a safe, efficient, and sustainable energy solution for the future.

EXPERIMENTAL METHOD

This methodology is designed to create a review of the Lead-cooled Fast Reactor (LFR), a Generation IV nuclear reactor design that promises high efficiency and the ability to effectively manage nuclear waste. The systematic steps in preparing this review are as follows.

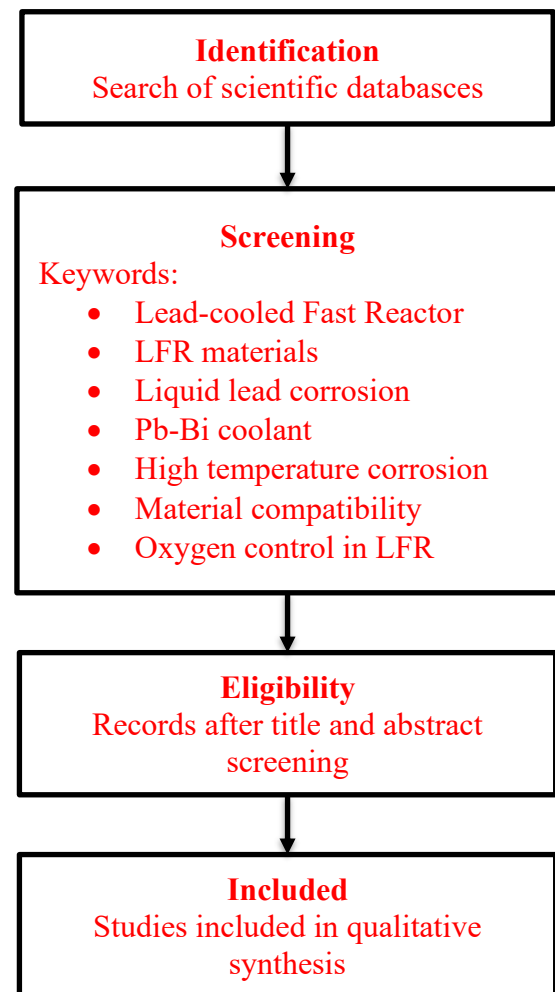


Figure 3. Visual Flowchart

1. **Identification** Collect relevant literature from various sources such as scientific journals (Scopus, Springer, Science Direct, etc.), books, and international conferences. The selected literature includes a brief description and advantages of LFR, the main

challenges in LFR design, and future challenges.

2. **Screening** After collecting the literature, the next step is to screen it based on relevance. This screening focuses on topics relevant to the review. Additionally, it focuses on sources with strong scientific methods and validated results.
3. **Eligibility** Screened articles are thoroughly read and evaluated on whether they contain experimental or simulation data relevant to the material challenges of LFR, and whether the methodology is valid and reliable.
4. **Included in Review** The 35 sources were selected as the primary basis for this literature review. These articles were analyzed and synthesized to produce the results and discussion section.

RESULTS AND DISCUSSION

Challenges of Liquid Pb/Pb-Bi Environmental Materials

a. Corrosion

In Pb or Pb-Bi cooled reactors, corrosion is a problem that is still being researched today. Corrosion of LFR structural materials is caused by chemical interaction between the molten metal (Pb or Pb-Bi) and the metal surface at high temperatures (400-600°C). This process is exacerbated by the high diffusivity of ferrous metals and chromium into the liquid coolant. In addition, Pb-Bi has the potential to form intermetallic compounds such as PbCrO_4 , leading to structural degradation. Significant temperature differences also cause atoms from structural materials and cladding to move from high-temperature regions to low-temperature regions. The accumulation of these atoms in cold regions forms deposits that trigger corrosion reactions. The rate of corrosion in materials immersed in liquid Pb is affected by how quickly the atoms of the material can mix and dissolve into the Pb or Pb-Bi metallic liquid. The presence of barriers such as oxides, nitrides, and other compounds on the surface of the material, as well as the presence of impurities or impurities in lead liquids and structural materials, can inhibit the movement of atoms during the dissolution process. This will reduce the dissolution rate and ultimately affect the level of corrosion damage.

One way to improve the resistance of containers to liquid lead is to regulate the amount of

oxygen that reacts with the surface of the container. This process will produce an oxide layer that serves as a protective shield, thereby reducing the rate of dissolution and corrosion. (Wu et al., 2002).

A summary by Zhang and Li (2004) provides an overview of Pb-Bi properties, corrosion mechanisms, corrosion mitigation including oxygen control, and corrosion models. Zhang and Li summarized the knowledge of Pb-Bi corrosion as follows:

1. At very low oxygen, both austenitic and ferritic-martensitic steels can undergo dissolution, even at low temperatures.

2. From 300 to 470°C, with sufficient oxygen (>10-4 ppm), a protective oxide film can be formed on austenitic and ferritic-martensitic steels. The cross section type can be seen in Figure 3.

3. For temperatures above 550°C, austenitic steels undergo rice dissolution and ferritic-martensitic steels form a very thick layer and potentially unstable oxides appear. These thick oxides are susceptible to erosion at high flow rates. At temperatures between 470°C and 550°C, the corrosion behavior of structural steels appears to transition from oxidation to dissolution and is acceptable using oxygen control.

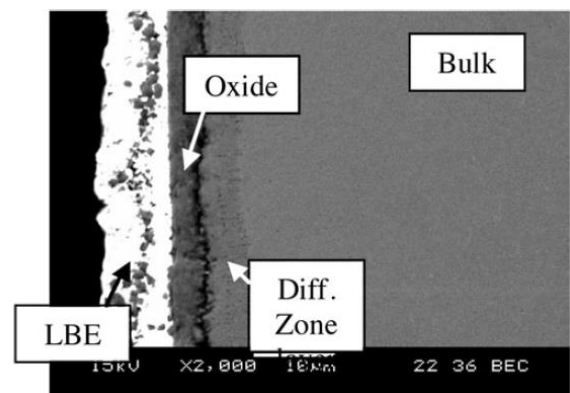


Figure 4. Cross-Section of HCM12A alloy with SEM exposed in lead-bismuth eutectic at 530°C for 600 hours shows oxidation and diffusion zones. (Machut, 2007)

Several studies were conducted for ceramic and metal refractory corrosion tests. (Runge *et al.*, 2003) tested the corrosion response of V-4Cr-4Ti alloys exposed to Pb at 650 to 800°C for 1000 hours. These significant changes to the alloy open up new opportunities in the development of materials capable of withstanding extreme conditions in fusion reactors. SiC-SiC Nicalon composite materials were also tested in Pb 800°C for 1000 hours (Kurata and Futakwa, 2004). Pb enters the composite surface and the structure forms a reaction phase associated with silica residues inside the Nickel fibers. Fazio et al. (2003) studied the corrosion of AISI Type 316L, T91,

W, and Mo in an oxygen environment with very low concentrations (3.1×10^{-10} to 7.3×10^{-8} wt%) for an exposure duration of up to 1500 hours. At this low oxygen potential, W and Mo have corrosion rates twice as large as steel. Based on previous testing, protective oxides are not expected at higher oxygen potential for refractory alloys. Then the corrosion mechanism of SS316 immersed in liquid Pb under Ar at 650°C and 750°C is shown in Figure 5. (Wang et al., 2024).

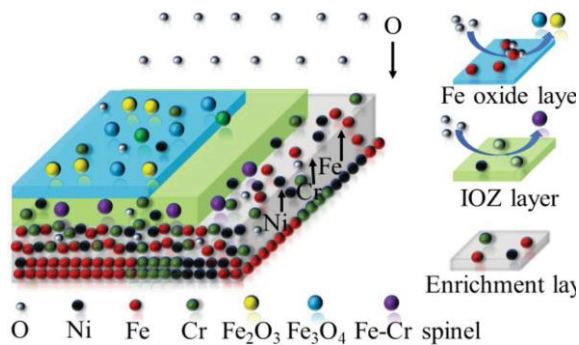


Figure 5. Corrosion mechanism of SS316 immersed in liquid Pb under Ar at high temperature (Wang et al., 2024).

b. Effect of Temperature and Oxygen Concentration

A precisely controlled oxygen concentration can form a passive layer on the metal surface, usually in the form of Cr_2O_3 . However, if the concentration is too low, solution corrosion will dominate, while too high a concentration can cause over-oxidation. The influence of parameters such as temperature and oxygen concentration on material compatibility with Pb or Pb-Bi is also being studied. The higher the temperature, the higher the kinetic energy of the particles in the system. Where this can cause an increase in the rate of chemical reactions, including corrosion reactions (Alzam et al., 2021). On the other hand, high temperatures can change the properties of Pb or Pb-Bi to become more brittle, so this can accelerate the rate of corrosion due to the surface being easily damaged or scratched. In addition, the increase in temperature in general can increase the solubility of oxygen in molten metals. Dissolved oxygen is active with metals forming oxides and accelerating the corrosion process. The increasing concentration of oxygen results in a great opportunity to react with metals. This will accelerate the formation of oxides and increase the rate of corrosion (Adhi et al., 2018).

Experiments to overcome rust problems in austenitic and ferritic steels involve setting the right amount of oxygen at 550° C. This method has proven to be effective because there is no dissolution

of the steel constituent materials into the surrounding environment. To control corrosion, it is necessary to control oxygen levels. This aims to reduce the rate of dissolution of the materials that make up the structure. Russian researchers have successfully developed an oxygen level control technique for systems using Pb alloys (references). The KALLA laboratory in Germany has conducted further research and confirmed that by regulating the balance between oxygen, hydrogen, and water, we can control the rate of corrosion in the material. (Loewen and Tokuhito, 2003).

Approaches to Addressing Material Challenges

a. New Material Development

Russia developed EP823 steel specifically for use in Pb-Bi (Barbier et al., 2001). EP823 is a 12 Cr ferritic or martensitic steel with a high silicon content (1-1.3 wt%). No comparable American steel was produced at the time. The development of steel for LFR may require the development of similar alloys. Materials such as T91, HT9, and 316L stainless steel have been tested, but have not shown long-term resistance to LFR environments. Recent research explores the use of ODS steels.

In order to obtain better high-temperature creep-strength of ferritic steels, dispersion-strengthened (ODS) oxide steels have been produced and evaluated. This steel has high temperature strength with good Y-Ti-O particle distribution. This is contrary to the precipitation of carbides in conventional high-temperature steels. Effective reinforcement by carbide is a typical approach with applications from many steels, not possible above $\approx 650^\circ\text{C}$, because carbides are unstable. Reinforced with different oxide dispersions, these steels have been developed in different alloys (Klueh et al., 2001). The Japan Atomic Energy Agency has developed advanced ODS steels with increased high-temperature strength (Yamashita et al., 2002) and tested as a fuel cladding in irradiated test assemblies in Russia. In addition, this steel is produced with difficult and costly mechanical alloy metallurgical techniques. Therefore, the joining of steel is a challenge because the separation of oxide particles to the surface from the welding zone cannot be adequately handled. Therefore, although ODS steel promises to be a steel that can operate at a temperature of 800°C , it is still in the development stage to date.

Austenitic stainless steels are considered for LFR components with lower temperatures and lower radiation doses. The austenitic stainless steels AISI 304 and AISI 316 have been used extensively in the LWR industry as well as fast reactor development

programs. The performance of the 316 is superior and will be a prime candidate for the austenitic component in LFR (Wang, 2023).

Research on the corrosion characteristics of materials in Pb-Bi under transient temperature conditions has been carried out in the range of 550-800°C. There were two cases in which transient temperature conditions were performed, for the first case, the specimen was soaked in Pb-Bi at a temperature of 550°C for 12 hours, the temperature was raised to 800°C and stored for 12 hours. As for the second case, the specimen was immersed in Pb-Bi at 550°C for about 500 hours, the temperature was raised to 800°C and stored for 15 hours. The materials tested are Al-SUS304 surface coated steel with sputtering HCM12A and SUS316, HCM12A high chromium steel, Recloy10 and SUS430, Mo refractory metal, SiC and Ti3SiC2 ceramics. The results showed that there were no traces of Pb-Bi corrosion on Al-SUS304 sputtering coated steel, refractory metals, and ceramics for all cases of transient temperature conditions. On the other hand, Pb-Bi corrosion attacks were found for high chromium steel specimens in all cases (Rivai and Takahashi, 2010).

Another material under consideration is FeCrAl alloy. Testing of five FeCrAl-RE alloys was carried out and exposed to liquid Pb for up to 10,000 hours at 550 °C. The test matrix consisted of three 10 wt.% Cr alloys, with Al content ranging from 4 to 8 wt.% (10Cr-4Al, 10Cr-6Al and 10Cr-8Al), one alloy without the addition of reactive elements (RE) (10Cr-6Al), and one reference alloy with 21 wt.% Cr and 5 wt.% Al (21Cr-5Al). No alloy is severely corroded, however Pb penetrates to a relatively large extent into the porous oxides of the low-performance alloys. An oxide scale as thick as 100 nm, partially composed of alumina (Al₂O₃), was observed for a high-performance 10Cr-6Al alloy. Fe10CrAl-RE alloy as a whole shows excellent corrosion resistance so it can be proposed as a promising new alloy for applications in liquid Pb environments (Ejentsam et al., 2013).

In another study, Fe-10Cr-4Al was tested by exposure to liquid Pb at a temperature of 750°C for up to 1970 hours. Two exposures with different test conditions were performed: one with the addition of oxygen (as H₂O) to liquid Pb and the other without oxygen. This experimental alloy shows general oxidation properties and good self-healing properties. The test results show that this particular steel category has the potential to operate in liquid Pb at very high temperatures with very little oxidation (Dömstedt, 2019).

Recently, (Wang et al., 2024) conducted a study to see the corrosion mechanism and distribution of SS316 material tested in Pb liquid at temperatures of

650°C and 750°C for 100 hours. The test specimen is a piece of SSS316 pipe in the form of an arch. Oxidation corrosion first predominates at 650°C. The outer surface layer of corroded SS316 is a loose layer of iron oxide and the inner layer is spinel solid Fe-Cr. However, the selective dissolution of the dominant Ni and Cr initially at 750°C and O diffuses to the interface increases over time, then an inner oxide layer is formed with Ni, Cr, and Fe in the inner layer of the matrix. The size of the pores formed by dissolution gradually increases with the increasing corrosion time and the partially dissolved area turns whole. Selective dissolution corrosion on corroded SS316, dominant at 750°C over time. The results show that SS316 has good corrosion resistance in liquid Pb.

b. Controlled Oxygen Usage

The principle of controlled oxygen flow utilization is based on the properties of liquid Pb-Bi which is relatively more inert compared to metal components in steel. So that if there is a flow of oxygen, iron oxide and chromium oxide (Cr) will be formed on the surface of the steel. The formation of an oxide layer on the surface of the steel will result in the solubility of the steel in the liquid to be smaller, so that the wear resistance becomes longer in a period of 2-3 years. Schematically, the process of forming the oxide layer is shown in Figure 6.

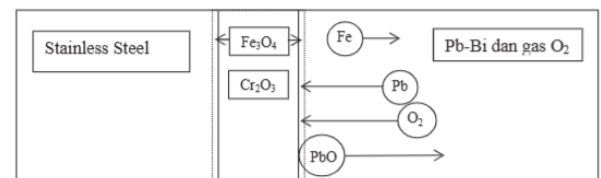


Figure 6. Metal surface coating process to inhibit corrosion (Maulana, 2023)

Several studies have been conducted on the corrosion of structural materials in liquid Pb-Bi such as using a material test loop (MTL) facility that can circulate the metal liquid with a hydraulic thermal system and an oxygen control system. To analyze the chemical reactions and fluid flows that occur, STAR-CD and CHEMKIN programs are used with volume control methods and chemical kinetics. The equations used in STAR-CD for fluid analysis and mass transfer are: continuity equation, momentum equation, transport equation and energy equation. The analysis is done macroscopic with the software.

Material Simulation for Corrosion Research

Research for the atomic level has been carried out using molecular dynamics methods on liquid Pb-Bi

materials in liquid sodium (Qi and Takahashi, 2003). In this method, the system is modeled to consist of a number of atoms interacting with each other with certain potential energies. The speed and position of the atoms can be traced against time. These atoms are assumed to be classical particles and their germs satisfy Newton's laws. The results of this simulation concluded that there was a discrepancy with the results of the experiment. A possible cause is the inaccurate use of Born-Mayer's potential energy in this case. So it is necessary to look for more accurate potential models.

Then researchers from Tokyo Tech (Qi and Takahashi, 2003) conducted a study on steel corrosion in liquid pb-Bi using the Ab-initio method. This simulation was carried out against SS316 and Fe crystals. The result obtained is that Fe crystals cannot diffuse with liquid Pb-Bi, but it is technically difficult to obtain such crystals so there are usually defects in Fe such as voids. This defect allows Pb or Bi atoms to penetrate the material. So there is a slight solubility of Pb in Fe caused by the defect (Qi and Takahashi, 2003). Meanwhile, the simulation of SS316 steel containing nickel and chromium elements is carried out only by calculating the potential energy of the interaction that occurs between these atoms with Pb and Bi. So in this study, only qualitative data was obtained for SS316 steel, not to the calculation of the diffusion coefficient of the particle distribution area at any given time.

Based on previous research and the explanation that has been explained starting from the introduction chapter, the summary of material performance in the LFR environment is as follows.

Material	Main Composition	Corrosion Resistance	Max Temp (°C)	Advantages	Disadvantages
T91	Fe-9Cr-1Mo	Medium	~550	Widely used and available	Degraded without oxide layer
SS316	Fe-Cr-Ni-Mo	Low	~500	Radiation resisters, easily available	High solution corrosion
HT9	Fe-12Cr-1Mo	Medium-High	~600	Structurally stable	Need strict

					oxygen control
ODS steel	Fe-Cr-Al+oxide	High	>650	High temperature and irradiation resistance	Complex manufacturing process
Aluminacoated	Steel+Al ₂ O ₃	Very high	~650 - 700	High corrosion resistant	Coating stability not yet proven

CONCLUSION

The emerging global energy market is motivating the consideration of innovative nuclear energy technologies. The refrigerant properties of liquid Pb and Pb-Bi allow for the design of lead-cooled fast reactor (LFR) systems with good innovation. However, the realization of such a system requires further technological development to overcome problems with materials. Steel alloys with improved high-temperature radiation performance have been developed (such as ODS, FeCrAl, SS316, AISI 316 EP823, AISI 304, and HCM12A), but these materials are still not suitable enough to withstand corrosion performance and thus will hinder application in LFR systems for at least a decade. No metal or ceramic material has been definitively proven to resist corrosion and radiation in LFR conditions. The LFR system is attractive, but has limited applicability until a suitable construction material design is found to be further identified. Many research projects are still limited to the laboratory scale, and long-term durability tests under reactor conditions are needed. The development of an accurate oxygen monitoring system is also key to success of LFR applications. It is estimated that in the next 10-15 years, a combination of new materials and surface protection strategies could lead to viable solutions for industrial use. Therefore, the continuation of research is essential to support a safe and efficient nuclear energy future.

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