Overview of Recent Studies Related to Lead-Alloy-Cooled Fast Reactors

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Abstract. The recent progress of the studies related to LFRs are summarized. The compatibility of materials with lead alloys has been clarified under steady and transient temperature conditions. Higher Cr content, Si and Al addition and Al-Fe alloy-coating improved corrosion resistance. The Al-Fe alloy-coated steel was not corroded even high temperature transient conditions. The ceramics of SiC and and Si_3N_4 are expected to be used as cladding material for high temperature LFR. For the consideration of mass transport, the diffusion coefficient of Ni could be measured using Capillary methods. A new bubble visualization method in LBE with gamma-ray radiography was developed. The thermal interaction of LBE and lead droplets with sub-cooled water, and the fragmentation of droplets were investigated, and the visualization of volatile liquid in high temperature liquid were achieved.

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1. INTRODUCTION

Lead alloys are attractive candidate coolants and target materials for lead alloy-cooled fast reactors (LFR) and the accelerator driven system (ADS), respectively, where the lead alloy includes both of pure lead and lead-bismuth eutectic (LBE or 45%Pb-55%Bi). Feasibility studies of LFRs have been performed so far for the **L**BE-Cooled **L**ong-**L**ife **S**afe **S**imple **S**mall **P**ortable **P**roliferation-**R**esistant **R**eactor (LSPR) (53 MWe/150MWt) [1-3], the Pb-Bi-cooled Direct Contact Boiling Water Small Fast Reactor (PBWFR) (150MWe/450MWt) [4], etc. Various studies have been also performed for the development of LFRs. The present paper overviews our recent studies for the following items:

(1) Compatibility of structural and cladding materials with lead alloys
- Corrosion under steady condition
- Corrosion under transient condition
- Employment of ceramics for cladding materials
- Properties of lead alloys related to corrosion

(2) Safety issues
- Water injection into reactor for decay heat removal
- Accident in direct contact of water/steam-lead alloy

2. COMPATIBILITY OF STRUCTURAL AND CLADDING MATERIALS WITH LEAD ALLOYS

2.1 Corrosion under Steady Condition

2.1.1 Background

Lead alloys are corrosive to steels in general, and the corrosion rate of steels increases in about one order of magnitude with an increase in temperature by 100°C. Therefore, structural and cladding materials and operating temperature must be chosen based on compatibility data of the materials with lead alloy at high temperature.

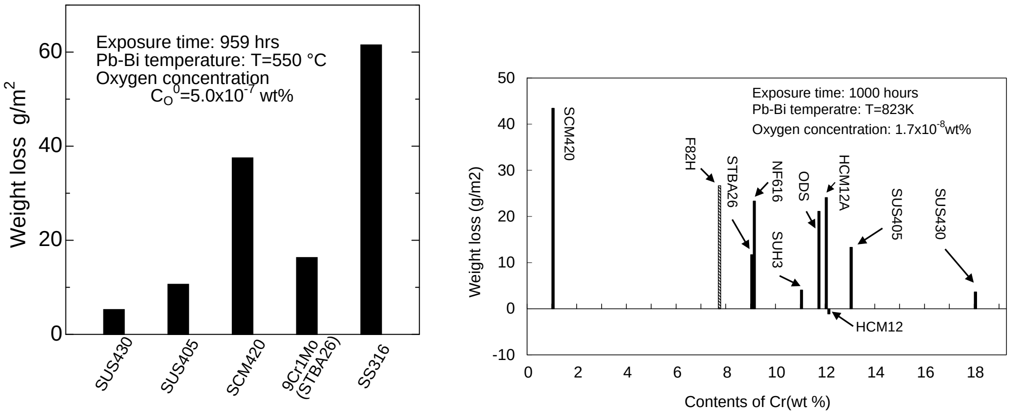
The compatibility of the materials should be investigated taking account of the factors below.

(1) The primary coolant temperature ranges in 350-400°C for core inlet, and 450-550°C for core outlet in most of the LFRs. Therefore, the structural materials in the cold leg will be exposed to lead alloy at the temperature less than 400°C, where corrosion rate is low. Stress due to pumping and coolant static pressures acts on reactor vessel. The pressure in reactor vessel is as high as 7MPa in PBWFR.
(2) On the other hand, the structural materials in the hot leg will be exposed to lead alloy at the temperature higher than 450°C, where corrosion rate is high. Steam generator (SG) tubes in the hot leg are exposed to the high temperature lead alloy and inner steam pressure.
(3) In these cases, cladding tubes at hot spot may reach 650°C and have inner pressure of fission product (FP) gases. In addition, in UTOP, ULOF and ULOHS events, the temperature of cladding materials at hot spot reaches as high as 700-800°C even for a short time.
(4) The materials such as cladding and SG tubes will be fabricated by welding. The effects of the welding on corrosion are also important.
(5) Mass transport of dissolved metals in lead alloy has a large influence on liquid metal corrosion, or dissolution type of corrosion. The transport is dominated by diffusion of metals in a flow circuit. The diffusion coefficients of metals in lead alloy are important factors for the estimate of each rate.

Therefore, the compatibility studies were performed taking account of the factors mentioned above.

2.1.2 Commercialized Steels

Corrosion experiment in flowing LBE was conducted for structural materials such as commercialized steels under the hot leg condition of 550°C [5]. Oxygen potential was adequately chosen so that oxide protection layers were formed on steel surfaces. LBE velocity was 1 and 2 m/s. It was found that single or multiple oxide layers formed on the steel surfaces protected the material from corrosion. Ferritic-martensitic steels with higher Cr content were more corrosion-resistant. Austenitic stainless steel, SS316, was corroded by selective dissolution of Ni into LBE. The result of weight loss of 25 g/m^2 in HCM12 and HCM12A corresponds to corrosion depth of 0.9 mm for thirty year-operation, and that of 60 g/m^2 in SS316 corresponds to that of 2 mm for the same operation period. Even if SG tubes are made of HCM12 or HCM12A, they must be replaced in shorter operation period than reactor life time.



(a) 959 h, 2m/s (b) 1,000 h, 1 m/s

FIGURE 1. Result of corrosion experiment in flowing LBE at 550°C.

In the same type of LBE corrosion experiment, serious erosion occurred when the oxygen concentration was lower than the Fe oxide formation conditions, i. e., the oxygen concentration of 2x10^{-9} wt% [6]. This suggests the necessity of adequate control of oxygen potential in lead alloy.

2.1.3 Al- and Si-added Steels

The addition of Si and Al improves the corrosion resistance drastically [7]. That is because very thin and stable Si- or Al-oxide layers in the thickness of approximately 1 μm is formed and self-healed. Their oxide formation potentials are very low, the corrosion is inhibited even under very low oxygen concentration in LBE. Si- and Al-rich steels, SUH3 (10Cr-1Mo-2Si), NTK04L (18Cr-3Al) and Recloy10 (18Cr-1Al-1Si), were immersed in a flowing LBE for 500 to 2,000 h under the oxygen concentration of 1.7x10^{-8} - 1x10^{-6}wt%. It was found that the addition of Si and Al is good for improvement of corrosion resistance for cladding steels.

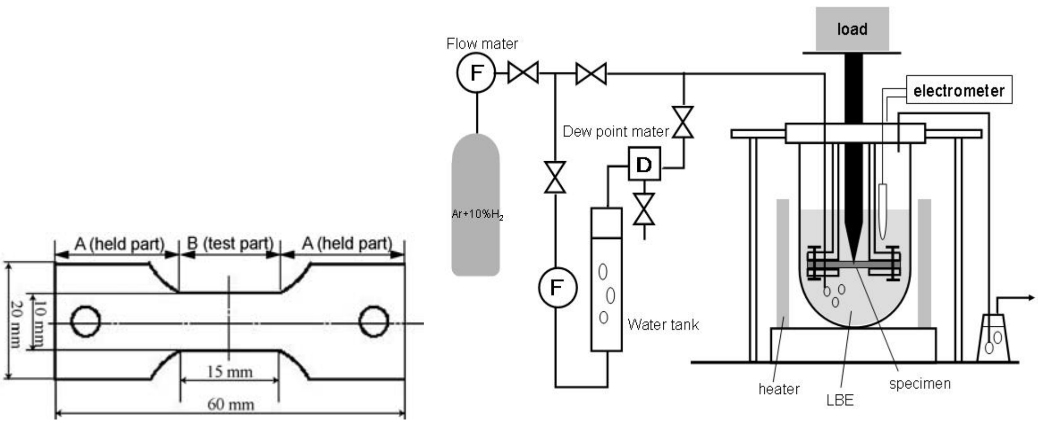
2.1.4 Surface-coated Steels with and without Bending Load

It is possible to improve the corrosion resistance only by changing the surface of material with Al-Fe alloy coating. Thus, the test steels, T91, E911 and ODS, were surface-treated and Al-alloyed by pulsed electron beam (GESA—GepulsteElektronenStrahlAnlage). Then, a corrosion test was performed for the specimens in a flowing LBE at 550ºC under the oxygen content of 10^{-6} wt% for 2000 h [8].

As the other surface Al-alloying technique, the unbalanced magnetron sputtering (UBMS) method with Al and SS304 targets was adopted [9, 10]. Surface-coated steels were tested by means of stirred-type LBE corrosion test at 700 ºC for 1000 h. Oxygen concentration was $4.5x10^{-7}$ - $5x10^{-6}$wt%. The results showed that a very thin oxide layer was formed on the coating layer and protected the material from corrosion attack.

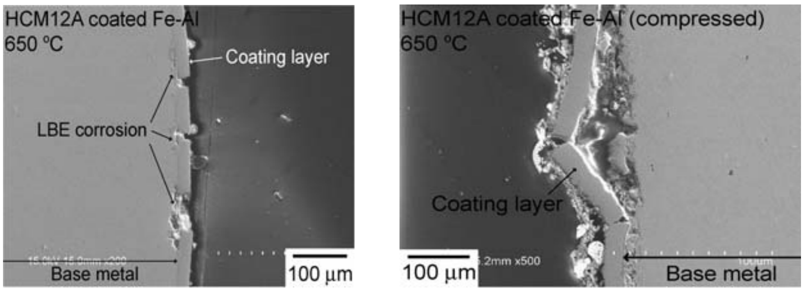
External stress acts on structural materials, and influence the corrosion process in certain cases [11]. A SS316 tube filled with LBE ruptured due to cyclic thermal stress. Liquid metal embrittlement took place in the tube wall.

The stress has the influence of damage on the Fe-Al alloy coating layer [12]. It was investigated under bending stress condition (**FIGURE 2**). LBE pool was at the low oxygen concentration (up to 5.2×10^{-8} wt%) and 550 and 650°C. Under a tensile stress condition, Fe-Al alloy-coated HCM12A exhibited the penetration of LBE into the base metal and dissolution of base metal to LBE around cracks of the coating layer (**FIGURE 3** (a)). Under a compression stress condition, the coating layers detached from the base metal (**FIGURE 3** (b)). LBE corrosion was not observed appreciably at 550°C. At 650 °C, Fe-Al coating layer was not corroded by LBE, and the penetration of LBE to base metal and dissolution of base metal into LBE occurred.



(a) Specimen (b) Experimental set-up for bending and corrosion test

FIGURE 2. Corrosion test under loading.



(a) Tensile stress (b) Compression stress

FIGURE 3. Cross sections of HCM12A specimen with Fe-Al alloy coating after corrosion test at 650 °C under loading for 240 h.

Heat treatment to the FeAl coating layer for strengthening had no appreciable effect on corrosion of material under the bending stress [13]. Again, the coating layer was effective to reduce the surface of the matrix to be corroded by LBE.

2.1.5 Welded Steels

Compatibility of welded structure with LBE at high temperature should be taken into account for the design of LFRs. Thus, the effect of welding on steel corrosion was investigated at 600ºC for 500 h [14]. Several welding methods, TIG, YAG laser and electron beam welding, were applied to the steel: HCM12A. Oxygen concentration was ～4.7x10^{-6}wt%. Grain structure around fusion zone was coarse, and grain structure in base metal was typical ferritic-martensitic. Cr-rich spinel oxide layer was much thicker in the fusion zone: 18-30μm, than in the base metal: 10-15μm. The same type of experiment was conducted in LBE at 650ºC under oxygen concentration of 7x10^{-7} wt% for 500 h [15]. Oxide layer was larger in the weld zones than base metal.

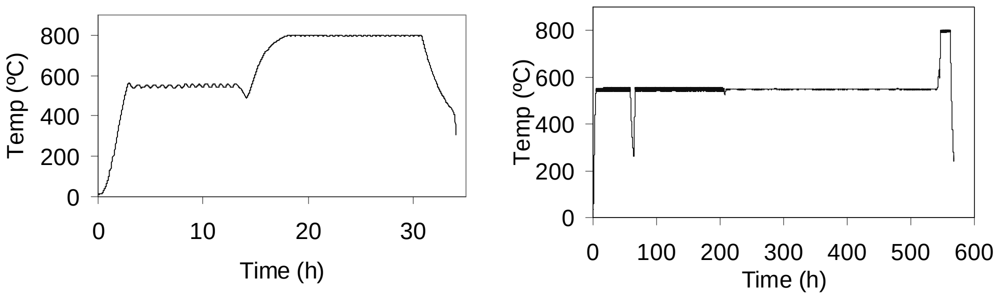
2.2 Corrosion under Transient Condition

2.2.1 Background

Fuel and cladding temperatures at hot spot will reach 700-800 ºC at the UTOP and ULOF events in LSPR [16]. The cladding temperature at hot spot reaches 792 ºC in ULOHS (Unprotected Loss of Heat Sink)/ULOF (Unprotected Loss of Flow) event in PBWFR [17]. It is concerned that lead alloy may become much corrosive at the transient temperature higher than 700ºC.

2.2.2 Corrosion of Steels in Lead-Bismuth Eutectic under Transient Temperature Conditions

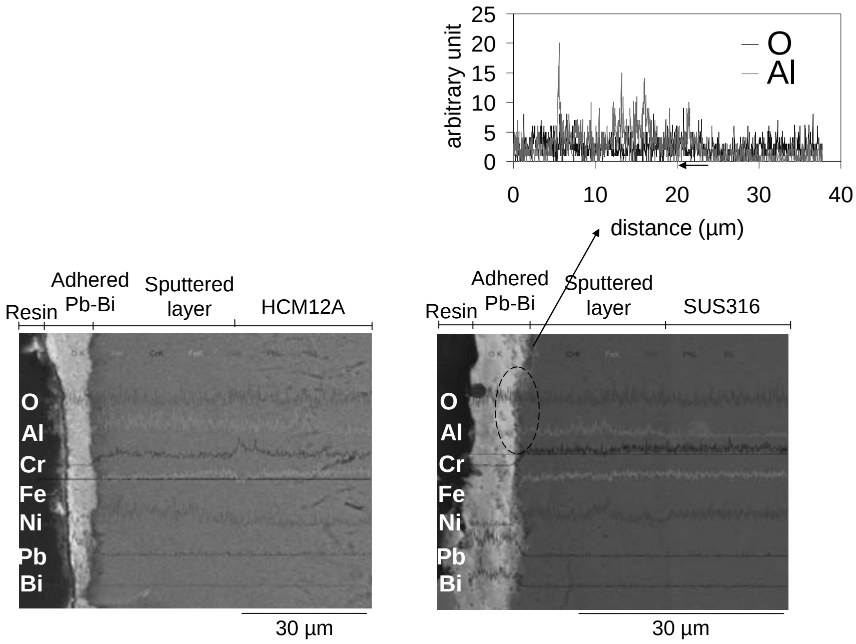
The corrosion characteristics of candidate materials was investigated under transient temperature conditions **[18]**. The test temperature conditions are shown in **FIGURE 4**. The concentration of oxygen was 10^{-6} (550 ºC) - 2 x 10^{-5} (800 ºC) for the transient case 1 and 10^{-6} wt% (550 ºC-800 ºC) for the transient case 2.



(a) Case 1 (b) Case 2

FIGURE 4. Transient temperature conditions.

FIGURE 5 (a) shows that the Al-Fe-coating layer remains intacted on the base metal. The coating layer with ~20μm in thickness was the same as before the immersion test. There is no penetration of LBE into the matrix of material and dissolution of constituent metals from the matrix into LBE. **FIGURE 5** (b) shows that the coating layer remains intacted on the base surface with ~10 μm in thickness, which was the same as that before the test. There was no penetration of Pb-Bi into the matrix of material and dissolution of constituent metals to Pb-Bi.



(a) Case 1 (b) Case 2

FIGURE 5. Al-Fe-sputtering-coated steels after transients.

2.3 Employment of Ceramics for Cladding Materials

2.3.1 Background

Ceramics materials, silicon carbide (SiC) and its composites, may be promising cladding and structural materials for high temperature LFR, since they are expected to be compatible with high temperature lead-alloys [19]. They have the superior high-temperature properties, thermo-chemical stability, irradiation tolerance, inherent low activation and low-after heat properties. The neutronic performance of the core and corrosion resistance of ceramics with lead alloys were investigated.

2.3.2 Neutronic Performance of Core with Ceramics Cladding

Neutronics of a reactor core with SiC cladding and structure was compared with that with steel cladding and structure analytically for small lead-cooled fast reactors [19]. The analytical result indicated that neutron energy spectrum was slightly softer in the core with the SiC cladding and structure than that with steel cladding and structure. In other words, the SiC type core can be designed smaller than the steel type core.

2.3.3 Compatibility of Ceramics with Lead Alloys

Corrosion characteristics of ceramic materials SiC and Si_3N_4 in a flowing lead-bismuth (Pb-Bi) for 2,000 h were investigated compared with those of high chromium steels [20]. The surfaces of the specimens were smooth without corrosion after the exposure of the specimens to the flowing Pb-Bi. No appreciable oxide layer was formed, and no chemical segregation layer was observed near the surfaces. The weight losses were negligibly small, and were much lower than those of 9Cr steels. Corrosion test of SiC and Ti_3SiC_2 in a pot was carried out in stagnant lead alloys at 700ºC [10]. The ceramics exhibited high corrosion resistance.

2.4 Properties of Lead Alloys Related to Corrosion

2.4.1 Background

One of main factors which cause steel corrosion in LBE is the dissolution of metallic elements in steels, such as Fe, Cr and Ni, into the liquid LBE. The diffusion characteristics of these dissolved elements in the liquid LBE are important factors for prediction of dissolution type of corrosion and the transport of the elements in a LBE circuit. Especially, Ni has high solubility into LBE and the preferential dissolution of Ni causes the dissolution corrosion of austenitic stainless steels.

2.4.2 Diffusion of Metal Elements in Lead-Bismuth Eutectic

The diffusion coefficient of Ni in LBE was determined in the temperature range from 550 to 650 ˚C by means of Capillary method [21]. The inner diameter of the capillaries was 2 mm, and the length was 50 mm. The Ni concentration distribution in LBE was determined by means of the inductively coupled plasma mass spectroscopy (ICP-MS) method. The diffusion coefficients of Ni were determined from the measure profiles.

3. SAFETY ISSUES

3.1 Water Injection into Reactor for Decay Heat Removal

3.1.1 Background

The conceptual designs of LFRs are equipped with decay heat removal systems: PRACS and RVACS which work with natural circulation of primary coolant. However, if the coolant level down terminates the natural circulation, the function of decay heat removal is lost. In order to avoid the core disruptive accident (CDA) in this event, a water can be injected into the core like the emergency core cooling systems (ECCS) of LWRs. However, there is the possibility of re-criticality in the water injection.

3.1.2 Criticality of Core in Case of Water Injection

A scenario for prevention of the CDA with the ECCS was examined for PBWFR. The possibility of the re-criticality was evaluated analytically [22]. It is found that the reactor becomes super-critical when the core is filled with water (**TABLE 1**). It is necessary to prepare the ECC system with boric acid and additional backup rods for sub-critical core in water injection.

TABLE 2. Result of effective multiplication factor in PBWFR.

Condition	k_{eff}
Full Pb-Bi without CR insertion	0.996
Pb-Bi with full CR insertion	0.881
Water without CR insertion	1.077
Water with full CR insertion	1.060

3.2 Accident in Direct Contact of Water/Steam-Lead Alloy

3.2.1 Background

One of the advantage of LFR is the elimination of the secondary lead alloy loop and installation of SG inside the reactor vessel. However, there is additional concern of SG pipe break. Rapid vaporization of the discharged water into LBE will bring sloshing motion of the heavy primary coolant and impact pressure to the reactor structures. It is necessary to avoid the mechanical damage to the structures. Although there is no SG pipe break in PBWFR, the feed water injected into LBE coolant may induce violent boiling. It is necessary to make the interaction of LBE with water mild in the chimney.

3.2.2 Visualization of Bubbles in Lead-Bismuth Eutectic

For the experimental study of LBE-water interaction, a new bubble visualization method was developed by means of gamma-ray radiography [23]. ^{60}Co with 11 TBq was used as a γ –ray source, and a multi-crystal Gd_2O_2S (Tb) scintillator was employed as a γ –ray detector. The bubble behavior was visualized successfully. Void fraction distributions were obtained by processing the visualized images.

3.2.3 Thermal Interaction of Lead Alloy Droplet with Subcooled Water

The lead alloy droplet and water micro interaction was experimentally clarified [24]. Characteristics of thermal-hydraulic interaction of LBE and lead droplets with subcooled water in water pool were investigated. Two kinds of interaction zones (deformation and fragmentation) and three kinds of interaction zones (solidification, deformation and fragmentation) were observed. The fragmentation zone (FZ) could be bounded by two border lines: spontaneous nucleation temperature and minimum film boiling temperature.

Fragmentation behavior of molten lead alloys droplet in water was investigated experimentally by releasing liquid LBE and lead droplets into a pool of subcooled water [25]**.** The fragmentation occurred when the temperature of the interface was higher than the spontaneous nucleation temperature of water and lower than the minimum film boiling temperature.

Preliminary experiments at low temperature jet boiling were performed by using ethanol as jet, and transparent fluorinert (FC-3283) as hot liquid, to understand the fundamental physical phenomena of the jet boiling of a water jet into LBE [26]. Jet boiling occurred at bottom part of jet first and developed to the upper part of jet within very narrow area around jet.

4. CONCLUSIONS

Fundamental studies have been performed for development of LFRs. The recent progress of the studies are summarized below.

The compatibility of candidate structural and cladding materials has been clarified under steady and transient temperature conditions. Higher Cr steels, Si- and Al-added steels and Al-Fe alloy-coated steels exhibited good performance of corrosion resistance as structural materials, cladding materials and special high temperature materials, respectively. The Al-Fe alloy-coated steel was not corroded even high temperature transient conditions. The ceramics of SiC and and Si_3N_4 are expected to be used as cladding material for the core of high temperature LFR, since they were highly corrosion resistant both in a flowing condition and in high temperature condition. For the consideration of mass transport in corrosion process, the diffusion coefficient of metallic impurity, Ni, could be measured using Capillary and ICP-MS methods. A new bubble visualization method in LBE using the gamma-ray radiography was developed. The thermal interaction of LBE and lead droplets with sub-cooled water, and the fragmentation of droplets in the same system were investigated, and the visualization of injection of volatile liquid into high temperature transparent liquid were achieved.

REFERENCES

1. S. Zaki, H. Sekimoto, "A Concept of Long-Life Small Safe Reactor, Potential of Small Nuclear Reactors for Future Clean and Safe Energy Sources," Elsevier, Amsterdam, pp. 225-234 (1992).
2. H. Sekimoto, S. Zaki, "Design Study of Lead- and Lead-Bismuth-Cooled Small Long-Life Nuclear Power Reactors Using Metallic and Nitride Fuel," *Nucl. Tech.*, **109**, pp.307-313 (1995).
3. M. Takahashi, S. Uchida, K. Hata, T. Matsuzawa, H. Osada, Y. Kasahara, N. Sawa, Y. Okubo, T. Obara, E. Yusibani, "Pb-Bi-Cooled Direct Contact Boiling Water Small Reactor," *Prog. in Nucl. Ener.*, **47**/1-4, pp. 190-201 (2005).
4. M. Takahashi, S. Uchida, Y. Kasahara, "Design Study on Reactor Structure of Pb-Bi-Cooled Direct Contact Boiling Water Fast Reactor (PBWFR)," *Prog. in Nucl. Ener.*, **50**, pp.197-205 (2008).
5. M. Kondo, M. Takahashi, N. Sawada and K. Hata, "Corrosion of Steels in Lead-Bismuth Flow," *J. of Nucl. Sci. Tech.*, Vol.**43**, No.2, pp.107-116 (2006).
6. M. Kondo, M. Takahashi, T. Suzuki, K. Ishikawa, K. Hata, S. Qiu, H. Sekimoto, "Metallurgical Study on Erosion and Corrosion Behaviors of Steels Exposed to Liquid Lead-Bismuth Flow, " *J. of Nucl. Mater.*, **343**, pp.349-359 (2005).
7. M. Kondo, M. Takahashi, "Corrosion Resistance of Si- and Al-rich Steels in Flowing Lead-Bismuth," *J. of Nucl. Mater.*, **356**, pp. 203-212 (2006).
8. A. Heinzel, M. Kondo, M. Takahashi, "Corrosion of Steels with Surface Treatment and Al-Alloying by GESA Exposed in Lead-Bismuth", *J. of Nucl. Mater.*, **350**, pp. 264-270 (2006).
9. A. K. Rivai, M. Takahashi, "Compatibility of Surface-Coated Steels, Refractory Metals and Ceramics to High Temperature Lead-Bismuth Eutectic," *Prog. in Nucl. Energy*, Vol. **50** (2008) pp.560-566.
10. A. K. Rivai, M. Takahashi, "Corrosion Investigations of Al-Fe-coated Steels, High Cr Steels, Refractory Metals and Ceramics in Lead Alloys at 700ºC," *J. of Nucl. Mater.*, **398** (2010) pp.146–152.
11. M. Kondo, M. Takahashi, "Metallurgical Analysis of a Tube Ruptured in the Lead Bismuth Corrosion Test Facility", *J. Nucl. Sci. and Tech.*, **43**, No.2, pp.174-178 (2006).
12. E. Yamaki, M. Takahashi, "Corrosion Resistance of Fe-Al Alloy-Coated Ferritic/martensitic Steel under Bending Stress in High Temperature Lead-Bismuth Eutectic," *J. of Nucl. Sci. and Tech.* **48**, No.5, (2011) pp. 797-804.
13. E. Y.-Irisawa, S. Numata, M. Takahashi, "Corrosion Behavior of Heat-treated Fe-Al Coated Steel in Lead-bismuth Eutectic under Loading," *Prog. in Nucl. Energy,* **53** (7), pp.1066-1072 (2011) (in print).

14. A. Pramutadi A. M., M. Takahashi, "Corrosion Properties of Welded Ferritic-Martensitic Steels in Liquid Lead-Bismuth at 600ºC", *J. of Pow. and Ener. Sys.*, **5**, No.1, (2011) pp.69-76.
15. A. Pramutadi A. M., M. Takahashi, "Study on Corrosion of Welded Steel for LBE-Cooled Fast Reactors," *Prog. in Nucl. Ener.,* **53** (7), pp.1073-1077 (2011) (in print).
16. S. Zaki, H. Sekimoto, "Safety Aspect of Long-Life Small Safe Power Reactors for Various Core Configuration," *Ann. Nucl. Ener.*, **22**, No.11, pp.711-722, (1995).
17. M. Takahashi, S. Uchida, Y. Yamada, K. Koyama, "Safety Design of Pb-Bi-Cooled Direct Contact Boiling Water Fast Reactor (PBWFR)," *Prog. in Nucl. Ener.*, **50**, pp.269-275 (2008).
18. A. K. Rivai, M. Takahashi, "Corrosion Characteristics of Materials in Pb–Bi under Transient Temperature Conditions," *J. of Nucl. Mater.*, Vol. **398** (2010) pp.139–145.
19. A. K. Rivai, M. Takahashi, "Design Study of Small Lead-Cooled Fast Reactor Cores Using SiC Cladding and Structure," *J. of Pow. and Ener. Sys.*, **1**, No.2, pp.134-142 (2007).
20. M. Takahashi, M. Kondo, "Corrosion Resistance of Ceramics SiC and Si3N4 in Flowing Lead-bismuth Eutectic," *Prog. in Nucl. Ener.,* **53** (7), pp.1061-1065 (2011) (in print).
21. E. Yamaki-Irisawa, S. Numata, M. Takahashi, W. Wu, "Experimental Study on Diffusion of Metals in Lead-Bismuth Eutectic in A Thin Tube," *19th Int. Conf. on Nucl. Eng., (ICONE19)*, October 24-25, 2011, Osaka, 2011, ICONE19-43499.
22. M. Takahashi, A. R. Khalid, V. Dostal, Novitrian, Y. Yamada, "Safety Design/Analysis and Scenario for Prevention of CDA with ECCS in Lead-Bismuth-Cooled Fast Reactor," *2007 International Cong. on Adv. in Nucl. Pow. Plants (ICAPP 2007)*, May 13-18, 2007, Nice, France, Paper 7280.
23. Minoru Takahashi, Takanori Yumura, Isao Yoda, Rongyuan Sa, "Visualization of Bubbles Behavior in Lead-bismuth Eutectic by Gamma-ray," *18th Int. Conf. on Nucl. Eng. (ICONE18)*, May 17-21, 2010, Xi`an, China, ICONE18-29522.
24. R. Sa, M. Takahashi, "Experimental Study on Lead Alloy Droplet / Subcooled Water Interaction," *J. of Ener. and Pow. Eng.* **5**, No.7, (2011) pp.579-589.
25. R. Sa, M. Takahashi, K. Moriyama, "Study on Thermal Interactions of Lead Alloy and Water," *Prog. in Nucl. Energy*, **53** (7), pp.895-901 (2011).
26. R. Sa, M. Takahashi, "Experimental Study on Thermal Interaction of Ethanol Jets in High Temperature Fluorinert," *19th Int. Conf. on Nucl. Eng., (ICONE19)* Oct. 24-25, 2011, Osaka, Japan, ICONE19-43422.