Lead-Bismuth Eutectic Compatibility with Materials in the Concept of Spallation Target for ADS*

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Lead bismuth eutectic (LBE) is favored by spallation neutron sources and coolant in the sub-critical reactor at the accelerator driven nuclear transmutation system (ADS). Technical issues of ADS are material technology of how to compromise with flowing lead bismuth, high-energy proton accelerator technology and a sub-critical reactor system technology. This paper describes LBE technology developed at JAERI. First a scenario in order to realize the ADS is shown. The concept of spallation target test facility is introduced with a target design of thermo-fluid dynamics. Base data of flow rate and temperature of Pb-Bi during LBE circulation are described. The results of LBE loop operation under the flowing conditions of target design concept are reported. The stagnant corrosion tests were done to know the controlling parameters among the various steels. The tube-type oxygen sensor with having the solid electrolyte was studied. Cleaning techniques were developed to remove LBE from materials.

Key Words: Erosion, Corrosion, Electro-Magnetic Pump, Lead-Bismuth Eutectic, Spallation, Electro-Magnetic Flow Meter, Accelerator Driven System, Oxygen Sensor, Cleaning Techniques

1. Introduction

The ADS aims at reducing the amount of the minor actinide (MA) and the long-lived fission products (LLFP) in the high level radioactive waste. The Japan Atomic Energy Research Institute (JAERI) proposed the 800 MWt, nitride fueled, Pb-Bi eutectic cooled fast subcritical core driven by the spallation neutron source using Pb-Bi target and 1.5 GeV, 20 – 30 MW proton accelerator⁽¹⁾.

The scenario to realize the Accelerator Driven transmutation System (ADS) consists of four steps as follows:

Step-I, a development of base technology on a flow-

ing Pb-Bi, and establishment of base technology on controlling devices.

Step-II, a construction of the transmutation experimental facility equipped with Pb-Bi target and 200 kW proton beams and a zero-power critical assembly⁽²⁾.

Step-III, a construction of experimental ADS with 80 MWt, Pb-Bi target/coolant and partial MA nitride fuel region.

Step-IV, a construction of ADS transmutation plant with 800 MWt and Pb-Bi target/coolant, which is to transmute 250 kg MA annually.

Step-I program is under way and step-II program is planned under the joint project with the High Energy Accelerator Research Organization (KEK) on high intensity proton accelerator program called J-PARC (Japan Proton Accelerator Research Complex)⁽³⁾. A Transmutation Experimental Facilities (TEF) is planned for construction. The TEF consists of two concepts: ADS Target Test Facility and the Transmutation Physics Experimental Facility as shown in Fig. 1. The ADS target test facility aids in doing engineering tests for establishing a Pb-Bi spallation

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Fig. 1 Transmutation experimental facility

target technology⁽⁴⁾.

At TEF, materials irradiation examination will be carried out with research & development relevant to the Pb-Bi target and containment material performance. The target will be bombarded by protons, which are accelerated by the normal state conductivity linear accelerator to 400 MeV and by the superconductivity linear accelerator to 600 MeV. The incident proton cycle is 25 Hz with a long pulse of 0.5 ms.

In order to make design base for the ADS target test facility the research and development covers Pb-Bi technology, radiation effects of materials, target design with computer simulation and handling technology of spallation products. Pb-Bi technology consists of flowing loop test, stagnant corrosion test, the development of oxygen sensor and cleaning techniques. In the flowing loop test the experimental items are verification of feasibility of existing materials for the facility construction, the development of devices such as electro-magnetic pump (EMP), electro-magnetic flow meter (EMF), Pb-Bi level meter, filtering device and accumulation of experiences. Stagnant corrosion test mainly aims at corrosion test under controlled condition of oxygen potential. Oxygen sensors are being developed in order to achieve good performance at the low temperature around 450°C by using reference electrode such as Pt/air, In/In₂O₃ and Bi/Bi₂O₃.

2. Target Design for Test Facility

The incident proton power is 200 kW: 600 MeV and 0.333 mA at a frequency of 25 Hz with a pulse width of 0.5 ms. Technical issue is the target window performance. High-energy incident protons penetrate the target window and deposit heat in Pb-Bi and the target vessel. Materials were irradiated at SINQ target 3⁽⁵⁾ and tested at JAERI Hot Cell. Emphases must be put on the cooperative effect of the radiation and the flowing Pb-Bi on the materials in ADS.

Figure 2 shows a computer simulation (STAR-CD)⁽⁶⁾ of the velocity of Pb-Bi in the frame of the half model of the design concept of spallation target. Incident protons are injected from the left to the target. In the target

Fig. 2 The concept of Pb-Bi target

Fig. 3 Temperature distribution in Pb-Bi target (half model). Unit:°C

with co-axial double walls Pb-Bi flows to the direction indicated by arrows. Pb-Bi flows in the annular channel, returns at window and flows to the right side. There are plates in the center that would be samples for post irradiation examinations. This is an idea to know the cooperative effect by Pb-Bi and protons. Stripes in plotting showed the magnitude of velocity of Pb-Bi in arbitrary unit. Figure 3 shows the temperature distribution in the target near window. It appeared that the design base temperature and the flow velocity are about 450°C and 1 m/s at the target window, respectively.

3. Lead-Bismuth Loop

3.1 Description of test loop

A flow diagram of lead-bismuth circulating loop for materials corrosion was already reported⁽²⁾. The main circulating loop consists of an electro-magnetic pump (EMP), a heater, test specimen tubes at high and low temperatures, Pb-Bi filters, a surge tank, a cooler, an electro-magnetic flow meter (EMF), a surface-level meter, thermocouples, an observation window, and a drain tank. The Pb-Bi filters as well as the test specimen tube at low temperature were additional function. They were installed into the loop after first 3 000 hrs circulation test at the upstream of EMF, the downstream of EMP and the bypass line to the drain tank as shown in Fig. 4. EMP is a linear inductive type with an annular channel. EMF has two

Fig. 4 A flow diagram of Pb-Bi circulating loop

electrodes in contact with Pb-Bi that detect electromotive force in the magnetic fields. During operation, the maximum temperature, the temperature difference and the flow velocity of Pb-Bi at the specimen were 450°C, 50°C, and about 1 m/s, respectively. Oxygen was not actively controlled in the experiment. The argon gas of 99.95 mass% purity covered the top plenum void of the surge tank during operation. A portion of Pb-Bi was sampled from the adhered Pb-Bi on the specimen and melted in the retort after pre-treatment. Oxygen concentration in the sample was measured to be 1×10^{-3} mass% by infrared spectroscopic analysis. The saturation concentration of the oxygen in Pb-Bi at the operation temperature of 450°C is calculated as 3.2×10^{-4} mass%⁽⁷⁾.

3.2 Material preparation

Materials for tube and tanks that contact Pb-Bi in the loop were made of SUS316.

Testing specimen tube is a cold-drawn seamless type SUS316, which was produced as a tubing form with 13.8 mm outer diameter, 2 mm thickness and 40 cm length. The tube was solution-heat treated at 1 080°C for 1.5 mins. and then cooled rapidly. Acid washing was performed to remove the oxide layer after heat treatment in the furnace. Other tubes in the circulating loop, except for the testing tube, were cold-drawn products with 27.2 mm outer diameter and 2.5 mm thickness. This means that the maximum velocity of Pb-Bi will occur at the high-temperature specimen. The chemical composition of Pb-Bi is Bi 55/Pb 45 (wt%). The Pb-Bi filter consists of curling thin foils made of SUS430, which was packed in the cylindrical vessel as shown in Fig. 5.

3.3 Stability of circulating Pb-Bi parameters

In the first 3 000 hrs operation of the loop maximum temperature was kept to 450°C and the difference of temperature was 50°C. Figure 6 shows the stability of controlled temperature in the circulating loop during Jan.29 to Mar.25, in 2001, for example. However, flow control was not stable. Figure 7 shows instability of flow rate

Fig. 5 Filter element

Fig. 6 Stability of controlled temperature in the flowing loop

during operations. Scheduled flow rate was 5 liters per minutes: it corresponded to 1 m/s at the high temperature specimen and 0.2 m/s at the low temperature specimens, respectively. At a time of 1 300 and 2 500 hrs the Pb-Bi flow was interrupted by the calibration process of the electro-magnetic flow meter. The calibration was done by measuring a time that a particular volume of Pb-Bi in the surge tank drops into the dump tank under the gravitation force. Figure 8 shows the results of EMF calibration. In comparison with the initial condition of flow rate the susceptibility of EMF was enhanced after 1 300 hrs and kept stable. After the calibration of flow meter at 2 500 hrs the flow rate recovered abruptly. In these operations the input current in EMP was almost kept the same. This means the magnetic fields were almost constant and so the driving force to Pb-Bi was the same. Again the continuous decreasing of the flow rate was observed.

After operation specimen was removed from the loop and the cross section was examined by digital microscope. It appeared that the reason was attributed to an increase of flow resistance in the channel of EMP. Dissolved elements from stainless steel into Pb-Bi at high temperature precipitated in the flow channel accompanied with Pb-Bi in the

Fig. 7 Instability of EMF output in the first 3 000 hrs test

Fig. 8 Performance curve of electro-magnetic flow meter

upstream of EMP. However, the precipitation was not occurred in the downstream of EMP because the temperature was 400°C at the EMP inlet and presumably 420°C at the EMP outlet. As the precipitation site is limited in the lower temperature site, the narrowed channel was only the upstream area. These materials narrowed the flow channel of EMP and increased flow resistance. So the flow channel was double-widen before the second 3 000 hrs test. As results significant decreasing of flow rate was not observed as shown in Fig. 9 but still there was a scatter in the flow rate. The reason of small instability is under investigation.

Figure 10 shows the tip of EMF electrode. There are two electrodes in the flow meter at the opposite directions in the cross-section of the tube. Electromotive force will be provided between two electrode-tips, which contact with flowing Pb-Bi in the magnetic field. The tip shape was a circular cone with 0.5 mm height. But after 3 000 hrs in service the tip was eroded to 0.1 mm height. Further examination is needed to know the relation between the instability of flow rate measurement and erosion

Fig. 9 Instability of EMF output in the second 3 000 hrs test

Fig. 10 Eroded tip of EMF

Fig. 11 Specimens at high (top) and low (bottom) temperatures

of tips in the EMF.

3.4 Corrosion-erosion of SUS316

Main concern is compatibility of materials under flowing Pb-Bi. After the first 3 000 hrs test we examined high temperature specimen and tubes at low temperature. Then the low temperature specimen was added in the loop. After the second 3 000 hrs test we again examined high- and low-temperature specimens and observed the inner surfaces and the cross sections. Figure 11 shows the high-temperature and low temperature specimens. The low temperature specimen was already cut for the inspection in this figure. We used unpolished speci-

Fig. 12 Unpolished high-temperature specimen

Fig. 13 Polished high-temperature specimen

Fig. 14 Unpolished low-temperature specimen

mens at high and low temperatures in the first operation, and polished specimen at high temperature and unpolished specimen at low temperature in the second operation, respectively. Figure 12 shows the cross section of unpolished high-temperature specimen. The inner surface was locally corroded. Corrosion was not uniform in the test specimen: the corrosion depth was deeper in the downstream portion than the upstream in the test specimen. But it was unclear that this phenomenon was true or not in the overall circulating tubes at high temperature. The maximum depth was evaluated to 100 μm at the downstream and 30 μm at the upstream within the specimens, respectively. Figure 13 shows the cross section of polished high-temperature specimen after the second 3000 hrs test. The high temperature specimen was polished by sand paper of #800 before the test. It appeared that the corrosion depth was roughly 20 μm. This suggests polished surface enhances the endurance of material with Pb-Bi. Figure 14 shows the cross section of unpolished low-temperature specimen. The materials were not corroded. And precipitated materials were seen in the Pb-Bi adhered

Fig. 15 EDX analyses of low-temperature specimen

Fig. 16 A flake sample (right) cut from precipitated material (left) by means of focused ion beams

on the wall of SUS316 tube. Figure 15 shows the inner surface of the low-temperature specimens before and after the test. The surface of unused specimen was characterized by the creviced structure. This feature resulted from the acid washing in the manufacturing process as mentioned in the material preparation. The inner surfaces of high-temperature specimens are also seen like this morphology. There are precipitated materials on the tube. The precipitation materials consist of mainly Fe and Cr as a result of energy dispersive X-ray analyses and apparently look crystals. We analyzed one of the crystals by means of a focused-ion beam, X-rays and TEM. Figure 16 shows the process of making a flake sample cut from the crystal. Figure 17 shows TEM bright field image. As a result of the quantitative analysis of deposit crystal by X-rays, Fe and Cr were detected and the weight concentration ratio was roughly Fe: Cr=9:1. Nickel was not found in the polycrystals⁽⁸⁾. They were precipitations from Pb-Bi including impurities dissolved from SUS316 at the high temperature part. It is found that they are ferritic alloys. Consequently it is emphasized that the precipitation happened in the channel of EMP. And the flow resistance was increased as mentioned in the previous section. The precipitation materials are the same with mentioned here. The ferrite materials are easy to be trapped at the magnetic

Fig. 17 TEM bright field image: Grain boundary appeared in flake

Fig. 18 SEM image of the curling thin foils

field inside the pump. In addition we inspected the filter element after 3 000 hrs operation. The filter elements were also installed at lower temperature part as described in the section 3.1. Figure 18 shows SEM image of the curling thin foils, which consist of the filter element. The Fe-Cr polycrystals were buried in the Pb-Bi adhered on the foils.

4. Stagnant Corrosion Test

4.1 Corrosion test equipment

Figure 19 shows static corrosion equipment. The retort contacting liquid Pb-Bi is made of quartz⁽⁹⁾. Purging gas of a constant H_2/H_2O ratio is a good methodology to control oxygen concentration in liquid Pb-Bi. The test temperature will range between 350°C and 600°C. In the experiment reported here Ar gas containing several percentage of oxygen was used as cover gas over liquid Pb-Bi. PbO with about 1 mm thickness formed on the surface of the liquid Pb-Bi during the corrosion test. The specimens were pulled out from the liquid Pb-Bi when the experiment was completed and cooled in the cover gas. Although test specimens were washed in the silicon oil at about 170°C, adherent Pb-Bi could not be removed from

Fig. 19 Stagnant corrosion test equipment

Fig. 20 Corrosion depth in the stagnant tests

the specimen surface completely. The cross-section of the tested specimens was observed using optical microscope after cutting the corrosion specimens.

4.2 Materials compatibility in stagnant Pb-Bi

Tested materials in saturated oxygen content are pure Fe, 2.25Cr-Fe steel (Cr 2.18 mass%), F82H (Cr 7.72 mass%),⁽¹⁰⁾ Mod.9Cr-1Mo steel (Cr 8.41 mass%), SS410 (Cr 12 mass%), JPCA (Cr 14 mass%), and SS430 (Cr 16.24% mass%). Figure 20 shows the relationship between the corrosion depth and the Cr content in steels. Tested temperatures were 450 and 550°C in oxygen saturated Pb-Bi for 3 000 hrs, respectively. The corrosion depth of JPCA was 5 μm. In comparison with the results of the previous section, this corrosion depth is different from the results of flowing test. Anyhow it appeared that a corrosion rate was very affected by a flow speed of Pb-Bi. Other important results are the effect of Cr content on the corrosion thickness in Pb-Bi. JPCA showed minimum values of corrosion thickness among them. The corrosion thickness decreases with increasing Cr content.

Fig. 21 Oxygen sensor

5. Oxygen Sensor

The formation of the protective oxide layer is key issue to compromise with materials performance in Pb-Bi, especially at high temperature. The oxide layer is dependent on the oxygen content in Pb-Bi. The development of the on-line monitoring and controlling system of the oxygen content in the Pb-Bi has been studied.

First a tube-type oxygen sensor was made using the solid electrolyte of yttria-stabilized zirconia, YSZ (8-mole% Y_2O_3) as shown in Fig. 21. The basic characteristics have been investigated under the gas/gas and the gas/Pb-Bi melt environments; i.e. the reference gas is the He gas with 10% oxygen and the oxygen content of the measured gas is varied from 0.05% to 0.1%, and the air used as reference gas/Pb-Bi with saturated oxygen concentration. The temperature ranges from 350 to 600°C. The potentiometer for high impedance, KEITHLEY617, is employed. The dependency of the measured voltage on temperature was validated under the stable condition with sufficient holding time. The measured voltage under the stable condition was describable by the theoretical formulation in the gas/gas throughout the temperature range and in the air/Pb-Bi at the temperature higher than 450°C. The study is extended to other oxide references as In/ In_2O_3 and Bi/ Bi_2O_3 .

6. Cleaning Techniques

Pb-Bi cleaning technology is another key issue in order to reduce radiation exposure during maintenance service, to decontaminate exchanged components and to prepare specimens for the post irradiation examination.

Two types of cleaning methods were tested; those were silicon oil at 180°C and mixture of acetic acid, hydrogen peroxide and ethanol (1:1:1). Specimens were immersed in liquid Pb-Bi at 400°C during 250 hrs and pulled out from the retort. Then specimens were immersed during 90 mins. in the oil or in the mixed acid. Tested material was SUS316L at 30 mm length, 15 mm width and 2 mm thickness.

Figure 22 shows specimen before and after cleaning

Fig. 22 Macroscopic photographs and weight of specimen in the silicon oil

Fig. 23 Macroscopic photographs and weight of specimen in the mixed acid

in the silicon oil. Numerical values indicate weight of the specimen. It was found that blushing process was needed to remove Pb-Bi effectively for the specimens immersed in the silicon oil. Figure 23 shows that the mixed acid easily dissolved Pb-Bi and removed almost perfectly. But specimens themselves were also tinted as a result of chemical reaction.

7. Conclusions

In order to construct ADS target test facility in J-PARC project the research and development on the base technology for the lead-bismuth eutectic compatibility with materials in the concept of spallation target have been carried, which cover target design with computer simulation under the incident protons, flowing loop test, stagnant corrosion test, oxygen sensor and cleaning techniques. Obtained results are as follows:

- Corrosion rate of SUS316 under flowing Pb-Bi at 1 m/s at 450°C is 0.1 mm/3000 hrs.
- Fe and Cr were melted into lead bismuth from SUS316 in the high temperature part and deposited in the low-temperature part according to the difference of solubility. The polycrystals increased the flow resistance in the electro-magnetic pump.
- The corrosion thickness decreases with increasing

Cr content in the stagnant corrosion test at saturated oxygen concentration.

- Oxygen sensors are to be developed by using reference electrodes.
- In cleaning techniques, blushing process was needed to remove Pb-Bi effectively after immersion in the silicon oil. The mixed acid easily dissolved Pb-Bi and removed almost perfectly. But specimens themselves were also tinted.

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References

- (1) Tsujimoto, K., Sasa, T., Nishihara, K., Takizuka, T. and Takano, H., Study of Accelerator-Driven System for Transmutation of High-Level Waste from LWR, ICON-7, Tokyo, (1999).
- (2) Oigawa, H., Sasa, T., Ikeda, Y., Kikuchi, K., Maekawa, F., Meigo, S., Kasugai, Y., Tsujimoto, K., Nishihara, K., Konno, C., Kai, T., Takano, H. and Yamane, T., Conceptual Design of Transmutation Experimental Facility, Global 2001, Paris, (2001).
- (3) The Joint Project for High-Intensity Proton Accelerators, JAERI-Tech 99-056, August (1999).
- (4) Kikuchi, K., Sasa, T., Ishikura, S., Mukugi, K., Kai, T., Ouchi, N. and Ioka, I., Current Status of JAERI Spallation Target Material Program, J. Nucl. Mater., Vol.x, (2001), pp.34–42.
- (5) Dai, Y. and Bauer, G.S., Status of the First SINQ Irradiation Experiment STIP-I, J. Nucl. Mater., Vol.296 (2001), pp.43–53.
- (6) Star-CD User Guide, (2002) CD Adaco Japan Co., Ltd.
- (7) Martynov, P., Orlov, Yu., Efanov, A., Troinoy, V., Resanov, A. and Lavrova, O., Technology of Lead-Bismuth Coolants for Nuclear Reactors, Proc. ISTC-TITech Japan Workshop on Nuclear Reactor Technologies in Russia/CIS, (2001), pp.80–105.
- (8) Kikuchi, K., Kurata, Y., Saito, S., Futakawa, M., Sasa, T., Oigawa, H., Wakai, E. and Miura, K., Corrosion-Erosion Test of SS316 in Flowing Pb-Bi, J. Nucl. Mater., Vol.318 (2003), pp.348–354.
- (9) Kurata, Y., Futakawa, M., Kikuchi, K., Saito, S. and Osugi, T., Corrosion Studies in Liquid Pb-Bi Alloy at JAERI, J. Nucl. Mater., Vol.301 (2002), pp.28–34.
- (10) Tamura, M., Hayakawa, H., Tanimura, M., Hishinuma, A. and Kondo, T., J. Nucl. Mater., Vol.141–143 (1986), p.1067.