Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 131 (2017) 386–394

Energy Procedia

www.elsevier.com/locate/procedia

5th International Symposium on Innovative Nuclear Energy Systems, INES-5, 31 October – 2 November, 2016, Ookayama Campus, Tokyo Institute of Technology, JAPAN

Oxidation characteristics of lead-alloy coolants in air ingress accident

Masatoshi Kondo[a]*, Nariaki Okubo[b], Eriko Irisawa[b],

Atsushi Komatsu[b], Norito Ishikawa[b], Teruya Tanaka[c]

[a]*Laboratory for Advanced Nuclear Energy, Tokyo Institute of Technology*
2-12-1-N1-18, Ookayama, Meguro-ku, Tokyo, 152-8550 Japan
[b]*Nuclear Science and Engineering Center, Japan Atomic Energy Agency*
[c]*Fusion System Research Division, National Institute for Fusion Science*

Abstract

The chemical behaviors of lead (Pb) based coolants in the air ingress accident of fast reactors were investigated by means of the thermodynamic considerations and the static oxidation experiments for Pb alloys at various chemical compositions. The results of the static oxidation tests for lead-bismuth (Pb-Bi) alloys indicated that Pb was depleted from the alloy due to the preferential formation of PbO in air at 773K. Bi was not involved in this oxidation procedure. Pb-Bi oxide and Bi_2O_3 were formed after the enrichment of Bi in the alloys due to the Pb depletion. The thickness of the oxide layer formed on liquid LBE at static condition was approximately 1mm. The oxidation rates of the alloys were much larger than that of the steels, and became larger with higher Pb concentration in the alloys. The compatibility of Pb-Bi alloys with stainless steel was worse when the Pb concentration in the alloys became low, since the dissolution type corrosion was promoted by the Bi composition in the alloy. The Pb-Li alloys were oxidized as they formed Li_2PbO_3 and Li_2CO_3. Then, Li was depleted from the alloy. These oxides of the coolant can be generated in the low temperature region of the reactor system after the increase of oxygen concentration in the coolant system in the air ingress accident.

Peer-review under responsibility of the organizing committee of the 5th International Symposium on Innovative Nuclear Energy Systems.

Keywords: fast reactor; fusion reactor; accelerator driven system; liquid metal; air ingress accident

* Corresponding author. Tel.:+81-3-5734-3065; fax:+81-3-5734-2957.
E-mail address: kondo.masatoshi@lane.iir.titech.ac.jp

Peer-review under responsibility of the organizing committee of the 5th International Symposium on Innovative Nuclear Energy Systems.
10.1016/j.egypro.2017.09.473

1. Introduction

The conceptual designs of fast reactors cooled by liquid lead (Pb) or lead bismuth (Pb-Bi) eutectic (LBE, Pb44.5-Bi55.5) have been studied, and some of these designs have been discussed under the collaboration of the Generation IV International Forum (GIF) [1]. Liquid LBE is also the target material of accelerator driven system (ADS). The compatibility of the lead based coolant with some candidate structural materials is one of the important issues. However, authors found that the occurrence of the severe dissolution type corrosion and the corrosion-erosion [2] was suppressed by the formation of the protective oxide layer [3] on the steel surfaces by the control of the oxygen potential of the liquid alloy [4]. The protectiveness of the oxide layers was improved by the alloying elements of Al and Si in the steel [5]. The compatibility was improved by the anti-corrosion coating technology [6]. Another important issue is polonium (^{210}Po) production under a neutron flux. However, ^{210}Po was removed by the baking method [7]. Pb-17Li alloy is one of the candidate tritium breeders in the design of the fusion reactors, and the technical issues similar to the lead based fast reactors have been studied [8].

The chemical reactivity of Pb and LBE with water is negligibly small even at high temperature. This advantage enables to design the Pb-Bi-cooled direct contact boiling water type fast reactor [9]. The chemical reaction between these liquid metals and air is not violent, though these are oxidized by air. The air cooled type emergency core cooling system (ECCS) is adopted in some designs of the lead alloy cooled FRs and ADSs. The ECCS by the natural convection of atmospheric air is equipped in the reactor system of BREST-OD-300 [10]. The air cooled type reactor vessel auxiliary cooling system (RVACS), which is for passive decay heat removal of the reactor, is equipped in the design of the European Lead Fast Reactor (ELSY) [10] and the China lead-based research reactor (CLEAR-1) [11]. The ADSs of MYRRHA and XT-ADS also equips the RVACS in their design.

The air ingress accident is caused when these air-cooling systems become the route of air ingression into the liquid metal coolant. When a large quantity of oxides is formed by the air ingress situations, the pipe plugging [12] can be caused in the primary coolant system. In the same time, the degradation of the heat transfer efficiency due to the precipitation of oxides on the heat exchanger is also concerned. These situations cause the eventual temperature increase of the reactor core. Therefore, the air ingress situation causes a potential risk in some low probable situations of severe accidents in the reactors. The safety issues relating with air ingress accident have been studied for the design of high temperature gas-cooled reactor [13]. However, the chemical behaviors of the coolant in the air ingress situation for the lead alloy cooled reactor are not made clear so far.

The purpose of the present study is to make clear the chemical behaviors of Pb based coolant in air ingress accident and the oxidation characteristics of Pb alloys. The thermodynamic consideration on the air ingress accident in the LBE cooled reactor was performed. The oxidation characteristics of Pb alloy were investigated by means of the static oxidation tests and metallurgical analysis of the oxidized Pb alloys.

2. Thermodynamic consideration on air ingress accident in Pb alloy cooled reactor

Figure 1 shows the progression of physical and chemical events after air ingress accident in the LBE cooled reactor. Fig. 2 shows the relation between the oxygen potential in the LBE coolant and the Gibbs free energy for formation of some related oxides. The tube rupture in the air cooled type ECCS and RVACS is one of the possible causes for the air ingress accident. The chemical composition of air is $75.5N_2$-$23.1O_2$-1.29Ar-$0.05CO_2$ [wt%]. When the air ingress accident occurred in the low temperature region of the coolant system, the oxygen concentration in the coolant increases according to the dissolution of oxygen from the air bubbles ingressed into the coolant (Fig.2 (a)). When the total inventory of LBE coolant is roughly assumed as $1x10^4$ tones, the soluble oxygen in the coolant is estimated as 15kg based on the oxygen solubility at 673K [14]. Oxygen is promptly dissolved in the coolant due to the large affinity of the coolant with oxygen. The coolant system must have some oxygen control units based on the gas bubbling [2] and the mass exchanger [4]. However, the oxygen concentration in the fluid is controlled under equilibrium condition by these units. Therefore, the influence of the oxygen control by these units is limited under the air ingression situation, in which a great deal of air ingresses into the coolant system. When the air inflow rate is assumed as 100L/min in this accident, the oxygen concentration of the coolant reaches to the saturation condition within 10 hours. Nitrogen is not soluble in Pb and LBE [14]. The molecules of CO_2 and H_2O are chemically stable in the coolant (Fig.2). Therefore, the air bubbles are never collapsed in the coolant. The oxygen depleted bubbles