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Study on control of oxygen concentration in lead–bismuth flow using lead oxide particles

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Abstract

Performance of mass exchanger type oxygen control system for the control of oxygen concentration in a flowing lead–bismuth (Pb–Bi) was investigated in a low temperature region of a Pb–Bi circulation loop. Oxygen dissolved and diffused from lead oxide (PbO) particles into the melt or oxide precipitated in the mass exchanger. The electromotive force (EMF) of an oxygen sensor installed in a high temperature region of the loop indicated the changes of oxygen concentration in the loop with the temperature changes of the PbO particles reasonably. The measured EMF agreed well with theoretical result obtained using $\Delta G^0_{\text{Pb–Bi–O equation}}$ in the Nernst equation. The expression of oxygen solubility in the melt in the mass exchanger, C_s, was derived from the data as $\log C_s = A + B/T$, where the constant A ranged from −4000 to −4600, and the constant B ranged from 1 to 3.5 depending on the temperature of the melt.

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1. Introduction

Conceptual designs of lead–bismuth (45Pb–55Bi) cooled fast reactors (LFRs) and Pb–Bi target type accelerator driven system (ADS) have been proposed by a number of researchers [1–5]. The compatibility of cladding, structural and window materials in the Pb–Bi flow at high temperature is one of the critical issues.

A self-healed oxide layer formed on steel surface inhibits a liquid metal corrosion [6], and reduces the corrosion rate in the flowing Pb–Bi [7]. In order to keep a stable condition of the self-healed layer, an oxygen content in the Pb–Bi should be controlled adequately by an oxygen control system.

One of the control methods of oxygen concentration in liquid lead alloy is a chemical reaction of a mixture gas of H_2, steam and Ar with the melt. The controllability of this method was reported in Refs. [6,8]. The reaction rate increases at higher temperature, and the equilibrium of oxygen potentials in the gas and in the melt can be attained faster by increasing the gas-melt contact area. However,

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an exchange rate of oxygen molecules between the gas and the melt depends on oxygen content in the gas.

A faster response of oxygen concentration with oxygen control may be obtained by immersing solid oxide in the melt in a mass exchanger vessel because of its higher oxygen content [9,10]. The temperature of solid lead oxide (PbO) and the melt is controlled to have desired oxygen solubility. Oxygen concentration becomes equilibrium through dissolution of oxygen into the melt or oxide precipitates from the melt on the solid lead oxide and inner surface of the mass exchanger vessel.

This method allows us not only to control the oxygen concentration but also to measure oxygen solubility in the melt. The experimental data of the oxygen solubility is needed to determine oxygen concentration from measured electromotive force (EMF) of oxygen concentration using a solid-electrolyte type oxygen sensor [11,12]. The oxygen concentration in the melt has been estimated using the correlations of oxygen solubility obtained in Russia [13] and in Germany [14] from measured EMF of the sensor. However, there is no experimental data of the oxygen solubility at low temperature.

The purposes of the present study are to investigate the controllability of oxygen concentration in a flowing Pb–Bi for the mass exchanger type oxygen control system and to determine the oxygen solubility in the liquid Pb–Bi.

2. Experimental

2.1. Pb–Bi forced convection loop

Fig. 1 shows a schematic of a Pb–Bi forced convection test loop used in the present study. The volume of the liquid Pb–Bi in the loop is 22 l. The detail of the loop was described in Ref. [15]. The loop consists of a high temperature region made of STBA26 steel (9Cr–1Mo) and a low temperature region made of SS-316 (18Cr–12Ni–2Mo). The corrosion test section and the oxygen sensor are in the high temperature region, and the expansion tank, the electromagnetic pump and the electromagnetic flow meter [16] are in the low temperature region. A by-pass line that has the electromagnetic flow meter, the bellow valves and the PbO reaction vessel was added in the low temperature region. The temperature of solid PbO particles in the PbO reaction vessel was controlled to have a desired oxygen concentration in the Pb–Bi loop.

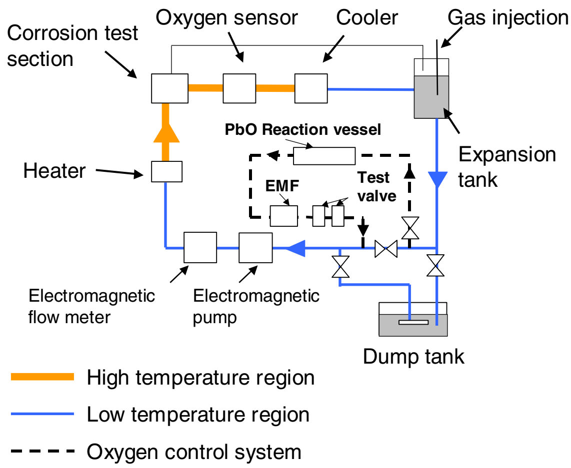

Fig. 1. Pb–Bi corrosion test loop.

Liquid Pb–Bi was circulated by the electromagnetic pump, and the flow rates were measured by the main and by-pass electromagnetic flow meters. The Pb–Bi temperatures at several locations of the loop were monitored using sheathed thermocouples inserted into the flow.

2.2. Oxygen sensor

The oxygen concentration in the melt at the outlet of the corrosion test section was measured by the solid-electrolyte type oxygen sensor. Fig. 2 shows a schematic of the sensor. The sensor cell was made of a solid-electrolyte conductor: yttria stabilized zirconia (Y_2O_3–ZrO_2). In/In_2O_3 and air were tested as reference electrodes in the previous studies [7,8]. In the present experiment, an oxygen-saturated Bi melt

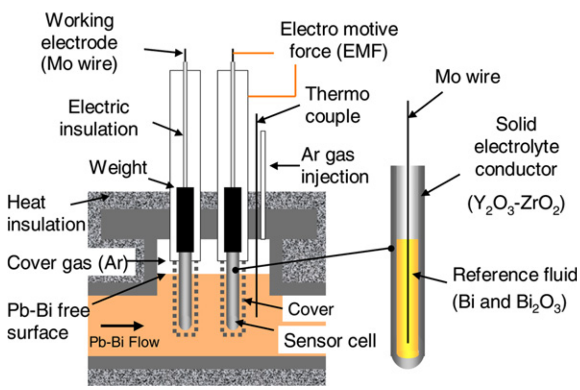

Fig. 2. Oxygen sensor.