

Task 13

Developing a Sensing System for the Measurement of Oxygen Concentration in Liquid Pb-Bi Eutectic

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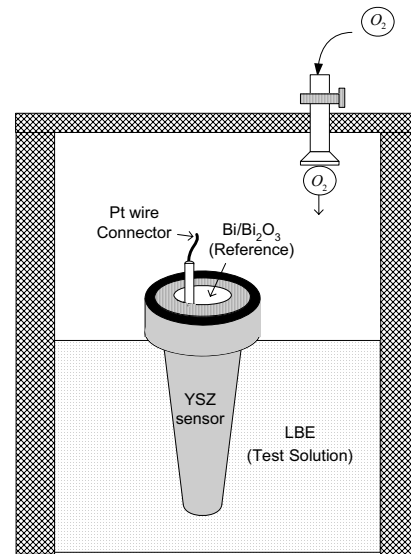
BACKGROUND

Lead-bismuth eutectic (LBE) is a candidate as a spallation target in sub-critical transmutation systems and as a coolant in nuclear programs. One of the primary concerns with LBE systems is the corrosion of stainless steel, the primary structural material used in nuclear systems. To mitigate this problem, trace levels of oxygen can be introduced into the system, causing the formation of a protective oxide layer at the interface between the LBE and steel. To protect the steel components, this oxide layer must be properly maintained. However, too much oxygen will produce unwanted oxide precipitation within the coolant and elsewhere in the system. With the current generation LBE systems, the stability of the oxide layer on the internal components is maintained through controlling the temperature of the system and the dissolved oxygen concentration in the coolant. Controlling these two operating parameters is the key to operating LBE systems and minimizing corrosion. While the temperature of the system is easy to measure, the concentration of dissolved oxygen in the LBE is more complicated.

Yttria-stabilized zirconia (YSZ) solid-electrolyte oxygen sensing systems are currently employed by Los Alamos National Laboratory (LANL) scientists to measure oxygen levels in the Delta Loop, an engineering-scale LBE experimental system. By measuring the voltage difference across the YSZ sensor, the oxygen concentration in test solutions can be determined relative to that in the reference solutions (the potentiometric method). The theoretical model for calculating oxygen concentration based on voltage measurements from YSZ sensors in static conditions is well understood. The real world performance of these systems, however, is less predictable.

One of the more significant challenges to measuring the dissolved oxygen concentration in LBE using the YSZ sensor is that the YSZ sensors are temperature-dependent. At high temperatures, the potential exists for electrons from bonding orbitals to become mobile and contribute to the electrical signal from the sensor. This component must be accounted for in electrical measurements, since it can be confused with the signal from the ionic conductivity.

Furthermore, device and material imperfections, such as irregular porous membrane and ohmic contributions, also contribute to deviations observed in the measured voltage response of the oxygen sensor system with respect to theoretical conditions. Therefore, there is a need to develop a complete set of calibration curves for YSZ sensor systems under various temperature and flow conditions in an LBE environment.



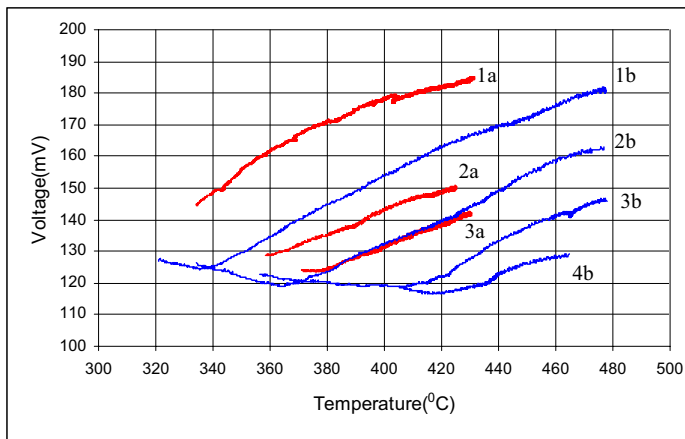
Schematic of YSZ Bi/Bi₂O₃ oxygen sensor in a heating chamber. The reference solution of Bi/Bi₂O₃ is filled and sealed inside the sensor core.

The primary goal of this research project is to examine the major factors impacting the performance of YSZ oxygen sensing systems. The research effort will serve to fill the gaps in the current sensor calibration/validation work and further the development of new sensors for oxygen concentration measurement in a nuclear environment. Ultimately, acquired data will lead to the full implementation of the instrumentation in the system. Through this work, the research group will also generate the calibration curves for the YSZ sensors over various temperature ranges.

RESEARCH OBJECTIVES AND METHODS

The research objectives of this project are as follows:

- To generate calibration curves of voltage versus oxygen concentration for the YSZ oxygen sensor system under various temperatures in liquid LBE.
- To determine the sensor characteristics of the YSZ sensor system.
- To determine oxygen dissolving rates in LBE under different temperatures in vitro.
- To study the effects of unwanted electrical conductivity, contributed by the mobility of the electrons at high temperatures, for more accurate oxygen measurement.
- To study alternative and promising oxygen measuring methods.



Sensor voltages at different oxygen concentrations (experimental results). Red lines are for sensor A and blue lines are for sensor B.

RESEARCH ACCOMPLISHMENTS

A set of calibration curves for output voltage versus temperature ranging from 300° C to 500° C under various oxygen concentrations in liquid LBE for the YSZ oxygen sensor have been produced. The current calibration strategy uses the direct injection of hydrogen and oxygen gases. Based on the experiments done so far, producing the correct level of oxygen in the system using the direct injection method does not appear to be adequate for producing the extremely low levels of oxygen concentration needed. To address this, the research team, along with their collaborators from LANL, switched to an alternative approach for controlling the oxygen level in the system. By varying the hydrogen to steam ratio in the system, it was possible to produce the low levels of dissolved oxygen needed. Experiments aimed at choosing a possible oxygen sensor to measure very low oxygen concentration in LBE using this technique are underway.

A new experimental apparatus for testing the oxygen sensors, based on an older version at LANL, is currently being developed at UNLV. The preliminary design of the system, referred to as the O₂ sensor pot, has been completed. The components for the system have been ordered, and the fabrication/assembly of the O₂ sensor pot is underway.

In parallel to this effort, the researchers have developed models of the proposed system. These models, designed to predict the dissolved oxygen concentration distributions in the experimental set-up, were used to assist in the design process and should serve as a vital tool to assist the research and sensor development. Using these models, the researchers hope to be able to examine the oxygen dissolving rates under various conditions including changing temperatures and inlet oxygen supply and to determine the diffusion coefficient of oxygen in liquid LBE under different temperatures through theoretical modeling and experimental measurement. The results obtained from the simulations also provide a means to cross-check and cross-validate with the experimental data.

A model of the oxygen-sensor and LBE experimental set-up was completed using FEMLAB. After several simulations, the investigators recognized that this program is not suitable for the simulation of the oxygen concentration and dissolving rate in LBE. A Lab View module for the acquisition and control of the apparatus has been designed and is undergoing testing and improvement.

FUTURE WORK

Calibrations that vary hydrogen to water steam ratio in the system will continue in order to control the dissolved oxygen concentrations at the required low levels. Once completed, these calibration curves will allow researchers to easily adjust the dissolved oxygen concentrations by simply varying the partial pressures of hydrogen and water steam. Additionally, calibrations for oxide dissociation limits will be conducted. Cross-calibration of different electrode sensors will also be undertaken.

YSZ sensor signals can also be processed using amperometric methods. In this method both anode and cathode are contained within the sensor assembly, and no electrical contact is made with the outside sample. The investigators hope to be able to design a new oxygen sensor system based on an amperometric method. This will allow investigators to compare the potentiometric and amperometric methods and determine which one is most conducive for the LBE system.

Techniques facilitating fully operational sensor systems will be obtained. In addition, these research efforts build a solid foundation for future development of new oxygen sensing systems.

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