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

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Task 13

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BACKGROUND

Although liquid lead-bismuth eutectic (LBE) is a good candidate for the coolant that can be employed in a subcritical transmutation blanket, it is known to be very corrosive to stainless steel, the material used in the containment structure. 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. The proper formation of this oxide layer largely depends on the accurate measurement and subsequent control of the oxygen concentration in liquid LBE.

Yttria Stabilized Zirconia (YSZ) oxygen sensors, using molten bismuth saturated with oxygen as the reference, have been utilized to accurately measure the concentration of oxygen dissolved in LBE. 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.

For the past three years, calibration of YSZ oxygen sensors has been performed using both the experimental setup located in LANL and one apparatus designed by TRP Task 13 team members. A set of calibration curves (voltage vs. temperature, ranging from 300 to 500 ^{o}C, under various oxygen concentrations in liquid LBE) has been obtained. Different sensor references and signal wires have also been tested. Numerical simulation has been conducted to study the oxygen transfer in the calibration task.

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.

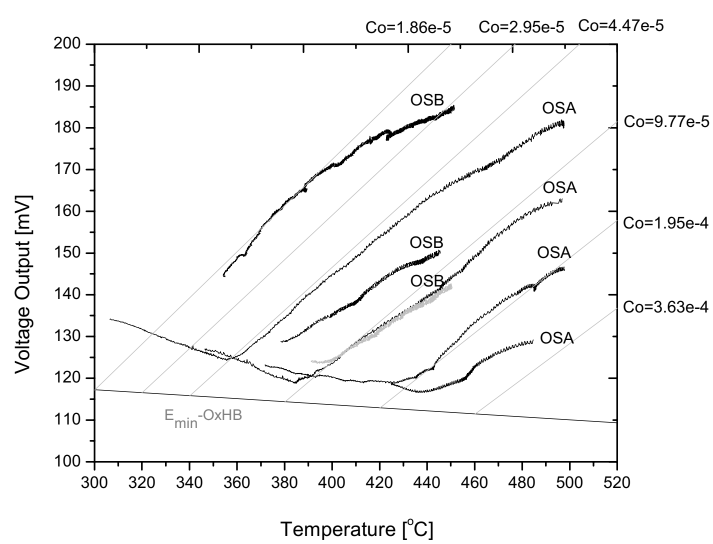
RESEARCH ACCOMPLISHMENTS

The first experimental setup consisted of a temperature controlled U-shape container, gas supplies and exhaust, a residual gas analyzer (RGA), a high-impedance electrometer, and a PC for data acquisition. The container is tightly sealed from outside atmosphere using a conflat flange except for gas inlet and outlet, and openings for insertion of thermocouple or RGA signal wire. Flexible heating tapes around the container can heat the liquid metal to the required temperature under the control of a temperature controller. The thermally insulated container was placed on a rocker to provide fluid motion that promotes mixing and homogenization of oxygen concentration in LBE.

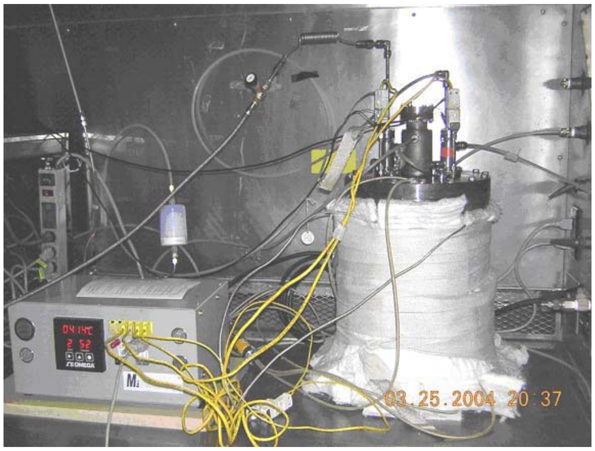
Although some calibration results were obtained using this setup, it had several shortcomings. For instance, it was impossible to heat the system to reach the desired high temperature (up to 750 ^{o}C). In addition, direct injection of the O_2/H_2 method adopted in this system is unlikely to produce a required extremely low oxygen level (ppm to tens of ppb) in liquid LBE. These problems in part were solved in another apparatus later developed by TRP Task 13 team members.

Calibration curves obtained from Oxygen Sensors A and B are shown in the figure below. It can be seen that after some initial transients, voltage-temperature curves first follow constant oxygen concentration lines according to the Nernst equation, and then turn to concentration saturation line.

- The slopes of experimental and theoretical curves are almost identical, ranging from 0.33 to 0.5. This indicates that the YSZ sensors are of high sensing quality;
- Being almost consistent with the theoretical results (a little above the theoretical calculation), the turning points clearly shown on the experimental curves for the sensor b indicate the regions at which the oxygen saturation occurs;
- Overlapping of the calibration curves for sensors A and B indicates the consistency in the sensors of the same design.



Sensor calibration results from two different sensors A & B.



Experimental apparatus developed at UNLV for the testing of the YSZ oxygen sensors.

FUTURE WORK

A number of practical problems have been encountered in this project including the contamination of the sensor chamber due to the leakage of liquid metal, and failure of the sensor due to the oxidization of the signal wire. These problems can become more serious when the sensor is used in a high temperature environment (up to 750 ^oC).

It has further been realized that for high temperature measurements, the main sources of oxygen measurement error result from the electronic contribution to the electrical conductivity of the electrolyte as well as the chemical reactivity between the electrolyte and electrode systems. As such, the reduction of the sensor's measurement errors can be achieved by proper selection of the solid electrolyte and the reference system.

Furthermore, a coating technology may be needed for better sealing of the sensors to operate in high temperature for prolonged hours.

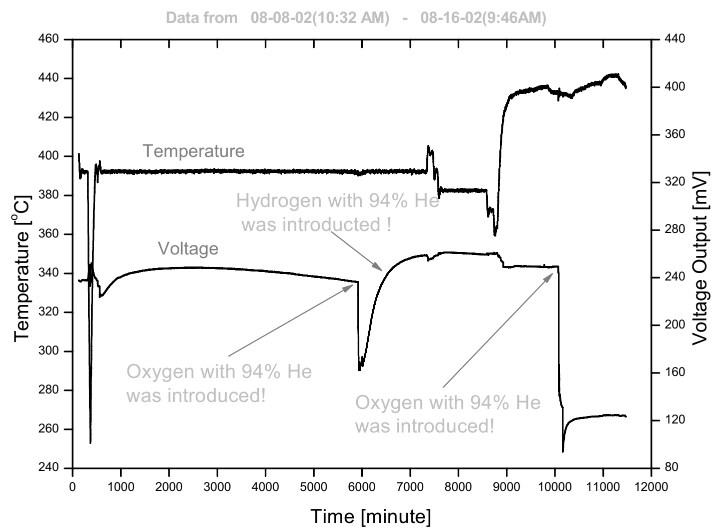
All of the above mentioned issues have to be considered to improve the YSZ sensors to operate in high temperatures. In addition, the improved high temperature sensors must be calibrated by intensive experimental work.

The future research work for this task shall include following components:

1) Try to mitigate the fluid that may leak into the sensor chamber.
2) Test different sensor signal wire materials and determine the best signal wire which can stand high temperature without oxidization.
3) Determine the protection of electrolyte of sensors at high temperatures.
4) Solve the assembly and sealing problems of sensors at high temperatures.
5) Fully test the new oxygen calibration apparatus and calibrate the new and improved sensors spanning the whole operational temperature range (350 ^oC to 750 ^oC), particularly, in temperature higher than 500 ^oC.
6) Develop silicon nitride and anodized alumina coating technologies for high temperature sensors.

ACADEMIC YEAR HIGHLIGHTS

- A manuscript entitled "Enhancement of Oxygen Transfer in Liquid Lead and Lead-Bismuth Eutectic by Natural Convection" was accepted for publication by *International Journal of Heat and Mass Transfer*.
- A paper entitled "Instrumentation of YSZ Oxygen Sensor Calibration in Liquid Lead-Bismuth Eutectic" was accepted by IEEE International Symposium of Circuits and Systems for the track Chemical Sensors, May 2005.



Response time to oxygen or hydrogen introduction using Sensor A.

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