Task 21

Theoretical Modeling of Protective Oxide Layer Growth in Non-isothermal Lead Alloy Coolant Systems

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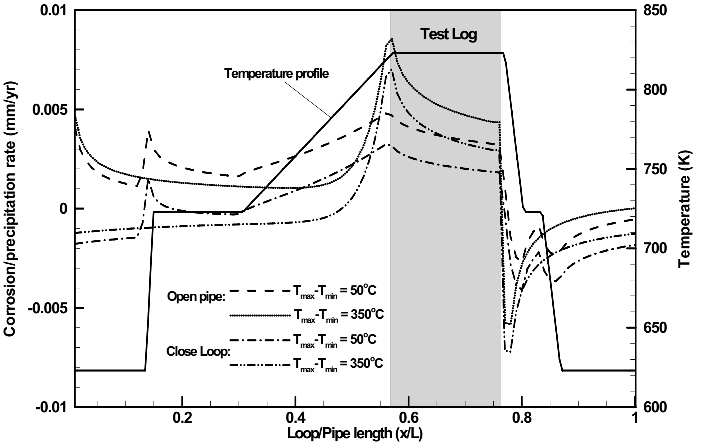
BACKGROUND

In advanced nuclear energy systems, lead-alloys (e.g., lead, lead-bismuth eutectic) emerge as strong candidates for transmutation and advanced reactor systems as nuclear coolants and high-power spallation neutron targets. However, it is widely recognized that corrosion of materials caused by lead-alloys presents a critical barrier to their industrial use. A few experimental research and development projects have been set up by different groups such as LANL to study the corrosion phenomena in their test facilities and to develop mitigation techniques and materials.

One of the central or main techniques in lead-alloys coolant technology under development is to use active control of oxygen thermodynamic activity (OTA) to provide protective oxide layers. Setting OTA in flowing lead-alloys makes corrosion highly dependent upon the oxygen concentration and the oxidation processes at materials surfaces. The active oxygen control technique exploits the fact that lead and bismuth are chemically less active than the major components of steels, such as Fe, Ni, and Cr. By carefully controlling the oxygen concentration in LBE, it is possible to maintain an iron and chrome based oxide film on the surfaces of structural steels, while keeping lead and bismuth from excessive oxidization that can lead to precipitation contamination. Thermal analysis has given an ideal oxygen level range in a non-isothermal lead-alloys coolant system. However, in a practical coolant loop, the proper oxygen level depends not only on thermal factors but also on hydraulic factors (system operating temperature, temperature profile, flow velocity, etc.). In addition, the oxygen distribution in a non-isothermal lead-alloys coolant system is still unclear. The optimal oxygen levels still need to be investigated.

The goal of the proposed research project is to provide basic understanding of the protective oxide layer behaviors and to develop oxide layer growth models of steels in non-isothermal lead-alloys (lead or lead-bismuth eutectic) coolant systems. Precise studies and simulations of all hydrodynamics with thermal conditions encountered in practical coolant loop systems by use of different flowing conditions in the laboratory are difficult and expensive, if not impossible. Therefore it is important and necessary to develop theoretical models to predict the protective oxide layer behaviors at the design stage of a practical lead-alloy coolant system, to properly interpret and apply experimental results from test loops, and to provide guidance for optimization in lead-alloys nuclear coolant systems. The research project, therefore, is aimed at filling the gaps of protective oxide layer growth and the oxygen concentration level before lead-alloys nuclear coolant is ready for programmatic implementations and industrial applications.



Corrosion rate under different axial temperature distributions.

RESEARCH OBJECTIVES AND METHODS

The research objectives are:

- To elucidate the mechanism of the protective oxide layer growth of steels in static, non-isothermal flowing lead-alloys coolant systems with oxygen concentration level control.
- To elucidate the mechanism of mass transport of oxygen, corrosion products in the multi-phase system.
- To develop oxidation growth models of steels in lead-alloy coolant systems.
- To clarify the dependence of oxidation process on the hydraulic factors (system operating temperature, temperature profile, flow velocity, etc) and the oxygen concentration distribution and level.
- To clarify the optimal oxygen concentration levels in practical coolant system scales.
- To interpret the experimental results from test loops and to apply them to the design of practical nuclear coolant systems.

The research goals are:

- To understand the difference in oxidation behaviors between different types of structure materials.
- To incorporate the present oxide layer growth model to our previous kinetic corrosion model.
- To develop a general numerical code that can predict the oxygen concentration level, the oxidation growth rate and the corrosion rate in practical lead-alloys coolant systems.

Kinetic Model

An improved kinetic model was developed to predict the mass transfer controlled corrosion/precipitation in non-isothermal LBE pipe/loop systems. Two sets of mass transfer equations were solved separately both in the turbulent core region and sub-layer region. The improved model was applied to the DELTA loop at Los Alamos National Laboratory. The DELTA loop is a non-isothermal closed loop and is used to study the corrosion of various materials in the flowing LBE system. The temperature profile is shown in the figure on the opposite page. This figure also shows the predicated corrosion/precipitation rate by the present model in the pipe/loop flow in the DELTA Loop. From this figure, one can find that there also exists a precipitation region in the open pipe case and this precipitation region occurs beside the highest temperature region because of the large axial temperature difference.

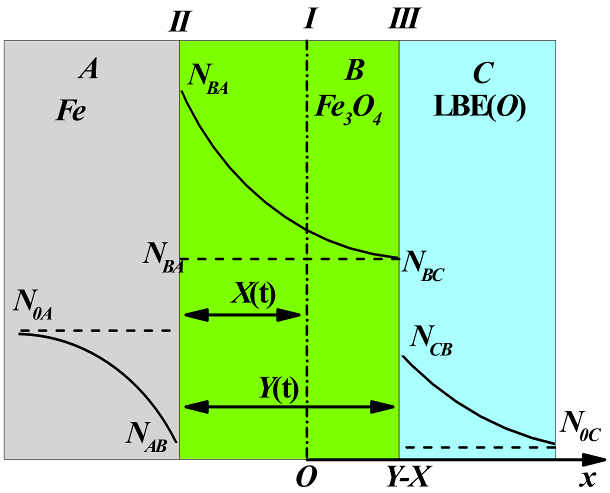
Numerical Analysis

Numerical analysis of the coupled natural convection and corrosion product transfer in a two-dimensional circular loop was made to study the corrosion product under the active oxygen controlled model.

Kinetic Oxide Growth Model

A kinetic oxide growth model in Liquid LBE was developed for the pure iron exposed liquid LBE with oxygen controlled. A schematic plot is shown in the figure below. The oxide Fe_3O_4 layer grows toward both in internal side (II surface) and external side (III surface). The following assumptions were made:

- The interfaces are local equilibrium and the processes do not affect the kinetics of oxidation.
- The growth of the oxide Fe_3O_4 is limited by the diffusion of iron.
- The diffusion of oxygen anion is neglected.
- The oxide layer growth and the consumption of metal obey the Wagner's parabolic law.
- The interfaces are flat plat.



Schematic plot of the structure of pure iron exposed to LBE with oxygen controlled. A region: Metal Fe; B region: Metal oxide Fe3O4 ; C region: Liquid LBE with oxygen; I: Original metal surface; II: Metal-oxide Interface; III: Oxide-LBE interface. X : Depth of metal consumption; Y: Width of Oxidation Layer.

ACADEMIC YEAR HIGHLIGHTS

- ♦ "Corrosion behaviors of U.S. steels in flowing lead–bismuth eutectic (LBE)" was published in *Journal of Nuclear Materials*, 2005,336, 1–10.
- ♦ "Oxidation Mechanism of Steels in Liquid–Lead Alloys," was published in *Oxidation of Metals*, 2005, 63, 353-381.
- ♦ "Modeling corrosion and precipitation in non-isothermal LBE pipe/loop systems," was submitted to *Journal of Nuclear Science and Technology*, 2005, in press.
- ♦ "Dynamics of high-temperature oxidation accompanied by 3 scale removal and implications for technological applications," was submitted to *Journal of Nuclear Materials*, 2005, in press.
- ♦ "A improved Kinetic Corrosion Model in Non-isothermal loop/pipe Systems," was presented at the ANS Student Conference, Columbus, OH, April 14-16, 2005.

FUTURE WORK

The next phase of the project involves accomplishing the following tasks:

- Illustration of the oxidation process mechanisms in oxygen control lead-alloys systems.
- Identification of the protective oxide layer growth rate and the dependence on thethermal and hydrodynamic factors of the entire coolant loop.
- Optimal operation conditions for oxygen control lead-alloys systems.
- Analytical models for various limiting process regimes.
- Development correlations and tools for calculations of the oxidation rate, oxygen concentration level and distribution, and oxygen consumption.

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