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

Yitung Chen

University of Nevada, Las Vegas, yitung.chen@unlv.edu

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Task 21: Theoretical Modeling of Protective Oxide Layer Growth in Non-isothermal Lead-Alloys Coolant Systems

Quarterly Progress Report 10/01/04- 12/31/04

UNLV-AAA University Participation Program

Principle Investigator: Yitung Chen

Co-Principle Investigators: Huajun Chen, Jinsuo Zhang, and Jichun Li

Purpose and Problem Statement

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. It is widely recognized that the corrosiveness of the lead-alloys is a critical obstacle and challenge for which it can be safely used or applied in the nuclear coolant systems. Active oxygen control technique can promote the formation of the “self-healing” oxide films on the structural material surface, drastically reducing steel corrosion and coolant contamination. Many experiments of steels exposed to flowing lead-alloys have been carried out to study the protective oxide layer behaviors. However, the experimental data are still very incomplete at present and can not provide the dependence of the oxide behaviors on the system operating temperature, temperature profiles along the lead-alloys loop, oxygen concentration, flow velocity, etc. In addition, oxygen distribution in a non-isothermal lead-alloys coolant system is not well understood. 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-alloys 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.

Personnel

Principle Investigator:

- Dr. Yitung Chen (Mechanical Engineering)

Co-Principle Investigators:

- Dr. Huajun Chen (Mechanical Engineering)
- Dr. Jinsuo Zhang (LANL)
- Dr. Jichun Li (Mathematics)

Graduate Students:

- Mr. Taide Tan, Ph.D Graduate Student, (Mechanical Engineering)

National Laboratory Collaborators:

- Dr. Ning Li, LBE Team Leader, LANL

Management Progress

Budget Issues:

- N/A

Student Issues:

- N/A

Management Problems

No management problem issues at this time.

Technical Progress

For studying the oxygen distribution in non-isothermal LBE system, experimental results from Delta loop are analyzed. Analyses on sensor output show that the fully turbulent flow leads to a uniform oxygen concentration all over the loop and there is no significant delay of sensor response to a change of the operating condition. Taking into account the chemical reaction at the liquid/oxide interface, the possible oxide behaviors are analyzed, providing means to achieve the optimized oxide layer thickness. At corrosion zones, the iron is removed by mass transfer corrosion. Three typical behaviors of the oxide layer at the corrosion zone can be observed: the thickness is constant if the amount of removal of corrosion products by mass transfer corrosion equals to that of diffusion through the oxide layer; the thickness decreases due to dissociation if the amount of removal by mass transfer is larger than that by diffusion through the oxide; the thickness increases due to new oxide formation if the amount of removal by mass transfer corrosion is less than that by diffusion. Taking into account the mass transfer rate is proportional to the surface corrosion product (mainly iron) concentration in the liquid which is determined by the oxygen concentration at equilibrium state, it is possible to obtain an optimized oxide thickness by controlling the oxygen level, avoiding the oxidation corrosion due to the formation of very thick oxide layers and directly dissolution due to the formation of very thin oxide layers.

At precipitation zones, the iron comes from two sources: precipitation from the liquid and diffusion from the steel. Because the amount of iron by diffusion through the oxide layer decreases with the oxide thickness increasing, the oxide formation at liquid/oxide interface will be dominated by iron precipitation finally. Therefore, controlling the precipitation rate through changing the liquid flow velocity and the oxygen concentration can prevent very thick oxide formation which may result in oxide spallation or flow blockage at precipitation zones.

Corrosion of steel in flowing LBE system can be affected by hydrodynamic parameters. Available models to predict the corrosion rate in LBE loop mainly focuses on the influence of the local conditions, in particular the velocity of the liquid LBE and the local temperature [1]. And the axial temperature profile, or the axial profile of the boundary concentration of the corrosion product, is usually neglected in the analysis of corrosion kinetics.

A new model to predict the behaviors of corrosion/precipitation in non-isothermal and multi-modular LBE loop system has been developed [2, 3]. However, according to their assumption, this model only consider the Schmidt number is very large and the variation of the corrosion product concentration is confined in a thin incompressible boundary layer, which is in the linear boundary layer of the LBE turbulent flow. And also this model doesn't take the effect of turbulent diffusion on the corrosion into account. As a fact, the Schmidt number for the corrosion product in LBE liquid is 150, and the concentration boundary layer should exist between the viscous sublayer and buffer region the corrosion transport is by both molecular conduction and eddy diffusion. So this model needs to be corrected.

The effect of turbulence diffusion on the corrosion in the LBE Loop system has been checked. Based on the surface renewal concept, we first examined the diffusion of corrosion production in the thin wall-layer by considering that it is periodically replenished but at a much reduced frequency than the bulk of the wall region. An interval distance l is introduced in this model, which means the fluid in the thin wall-layer moves for a characteristic distance along the wall until it is disrupted by an eddy which penetrates to the wall. It can be expressed as [4]:

$$x_l = 1740 \frac{\nu}{u^*} \quad (1)$$

where ν is the kinetic viscosity and u^* is the friction velocity. After introducing this theory to the model in ref. [2], the corrected form for the Sherwood number in the LBE loop system will be

$$Sh = 0.019036 Re^{9/10} Sc^{1/3} \quad (2)$$

where Re is the Reynolds number and Sc is the Schmidt number. So the dependence of Reynolds number for corrosion in the LBE loop system is $Re^{0.9}$. This model will be extended to the cases that the thickness of the concentration boundary layer is considerably greater than the thickness of the thin wall-layer.

The equation of mass balance of the corrosion product in the fully developed turbulent pipe flow, neglecting axial diffusion, under steady-state conditions, is a balance between the axial convection and radial diffusion terms:

$$u \frac{\partial C}{\partial x} = \frac{\partial}{\partial y} \left((D_m + \varepsilon_t) \frac{\partial C}{\partial y} \right) \quad (3)$$

where u is the axial velocity, x is the axial coordinate, y is the radial coordinate, D_m is molecular diffusivity, ε_t is the turbulent eddy diffusivity. For a turbulent boundary layer flow, there exist three flow regions: Laminar sublayer, buffer zone, turbulent core. Since the Schmidt number the corrosion product is large ($Sc \approx 150$), the corrosion product (iron) concentration boundary layer exists in the buffer zone. Next, the effect of turbulence diffusion on the corrosion in the LBE Loop system has been checked numerically. In the near wall region ($y^+ \leq 30$), the expression for the turbulent eddy diffusivity is taken as

$$\frac{\varepsilon_t}{\nu} = \frac{0.0007 y^{+3}}{(1 + 0.00405 y^{+2})^{1/2}} \quad (4)$$

where $y^+ = (y/\nu)\sqrt{\tau_w/\rho}$, ν is the kinematic viscosity, τ_w is wall shear stress, ρ is the fluid density.

Our numerical model has been applied to the material test loop system in Los alamos Laboratory. The parameters are used in this numerical procedure: Loop/pipe length $L=29.92\text{m}$, hydraulic diameter $d = 0.0525\text{m}$, kinematic viscosity of LBE $\nu = 1.5 \times 10^{-7} \text{m}^2\text{s}^{-1}$, liquid LBE velocity $V=0.5 \text{ms}^{-1}$, oxygen concentration in LBE $c_o=0.01\text{ppm}$, The difference between maximum and minimum temperature $\Delta T = 350^\circ$. The temperature profile of the LBE test loop can be found in Ref. [2]. Figure1 (a) and (b) show the distribution of the concentration for the whole loop. Figure 2 shows the corrosion rate in along the pipe. From the figure, it can be found the effect of turbulent diffusion term will increase corrosion rate evidently in the test region and the concentration boundary layer becomes thinner than that in the laminar cases.

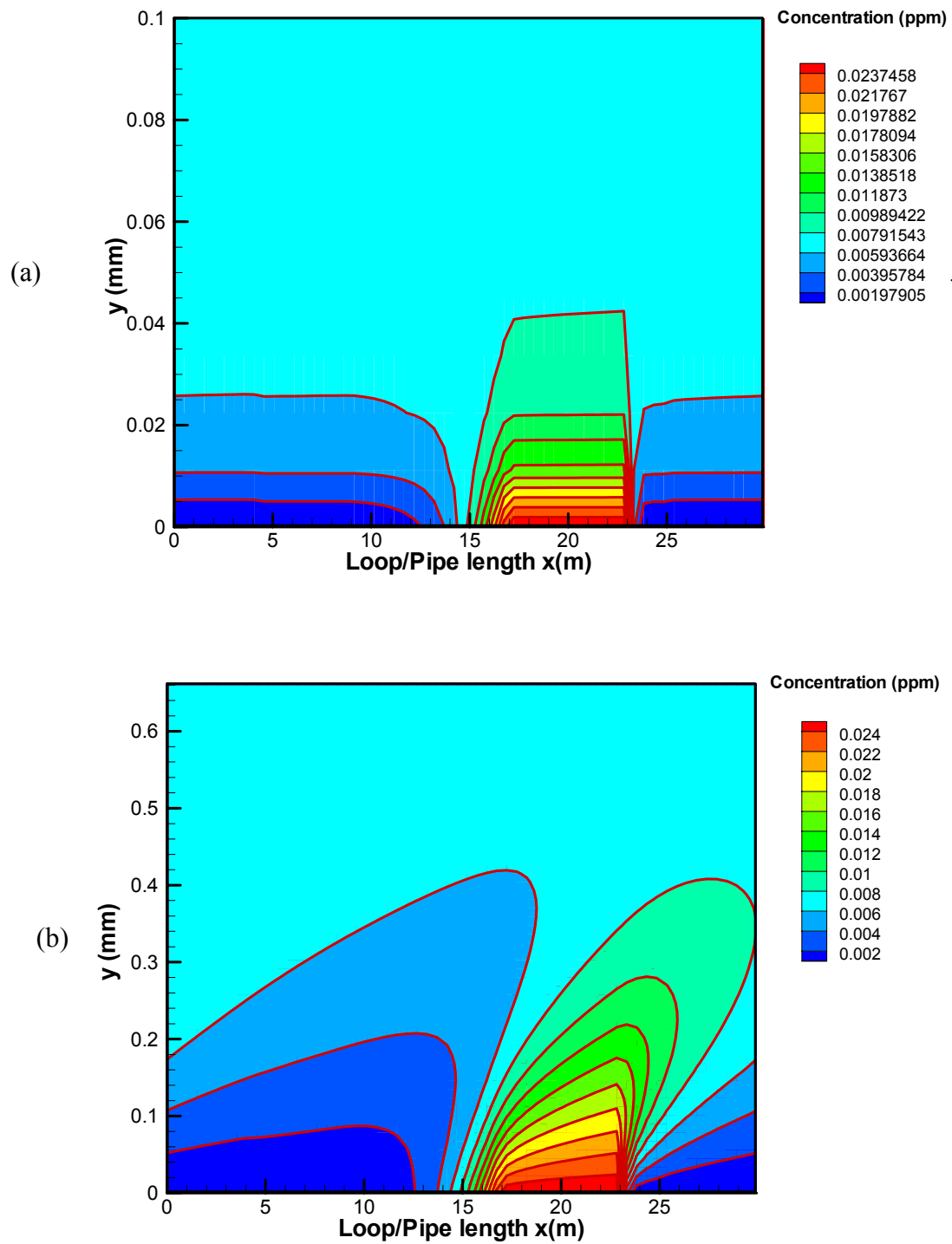


Figure1 Distribution of Concentration of Corrosion product. (a) With turbulent diffusion term. (b) Without turbulent diffusion term.

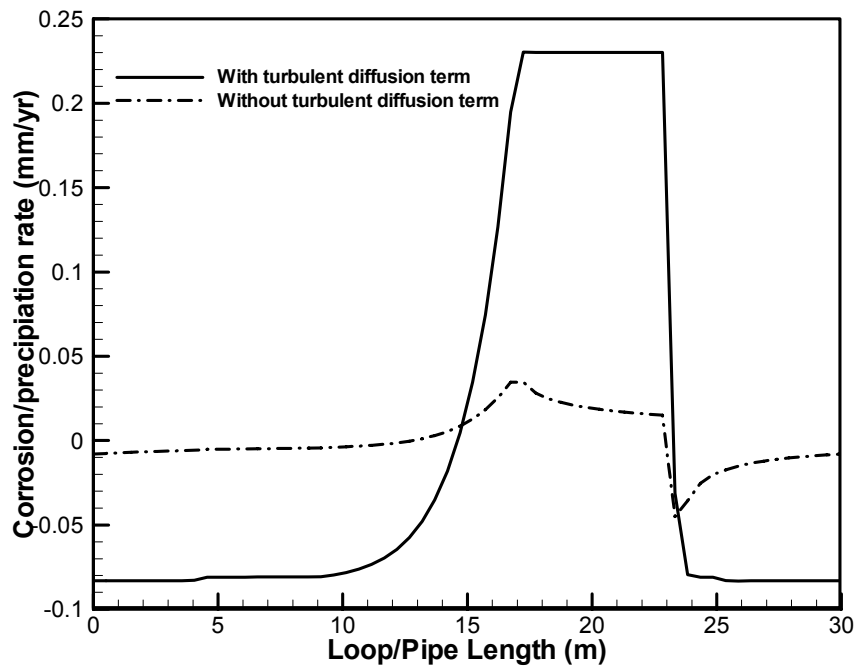


Figure2 Corrosion rate along the Loop

The abstract of “Oxygen Technique in Melt Lead and Lead-Bismuth (LBE) System,” has been accepted by ICONE13 which will be held in Beijing, China, May 16-20, 2005. The draft paper is under preparation.

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