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

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## Task 21

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

Y. Chen, T. Tan, J. Zhang, and J. Li

## BACKGROUND

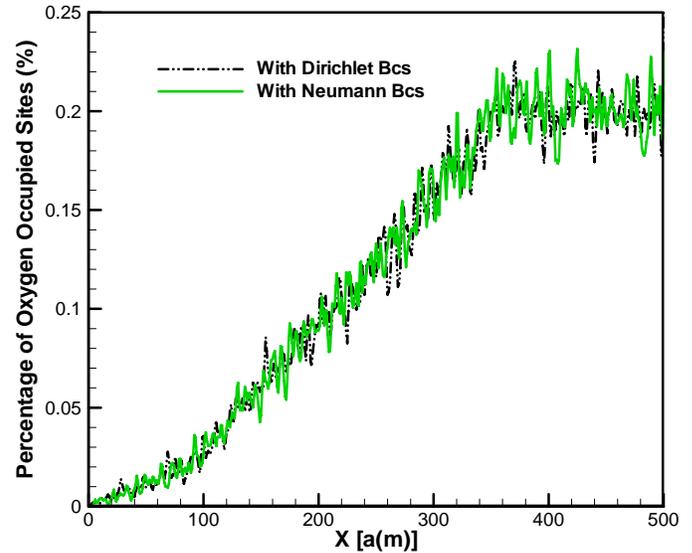
In advanced nuclear energy systems, lead alloys emerge as strong candidates for transmutation and advanced reactor systems as nuclear coolants and spallation neutron targets. However, it is widely recognized that corrosion of materials caused by lead alloys presents a critical barrier to their industrial use. A few experimental research and development projects have been set up by different groups such as at Los Alamos National Laboratory to study the corrosion phenomena in their test facilities and to develop mitigation techniques and materials. One of the central or main techniques 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 lead-bismuth eutectic (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 alloy 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 alloy coolant system is still unclear. The optimal oxygen levels still need to be investigated.

## RESEARCH OBJECTIVES AND METHODS

The goals of this research project are to provide a basic understanding of protective oxide layer behaviors and to develop oxide layer growth models of steels in non-isothermal lead alloys coolant systems, in particular:

- To elucidate the mechanism of the protective oxide layer growth of steels in static, non-isothermal flowing lead alloy coolant systems with oxygen concentration level control.
- To elucidate the mechanism of mass transport of oxygen and corrosion products in the multi-phase system.
- To develop oxidation growth models of steels in lead alloy coolant systems.
- To clarify the dependence of the oxidation process on the hydraulic factors 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 percentage of the oxygen occupied sites for  $K_d=4$ ,  $C_{oxy}=0.2$  and  $P_{act}=0.0005$  at  $N_t=200,000$  with Neumann and Dirichlet boundary conditions at  $y=0$ .

## RESEARCH ACCOMPLISHMENTS

### Boundary conditions study of the self-coded cellular automata (CA) oxidation model coupled with inward oxygen diffusion

In a previous study of the CA model, the oxide layer growth of steel in a liquid lead alloy environment was studied considering the inward oxygen diffusion. The boundary condition of the oxygen concentration at the far away conditions was set as a Dirichlet boundary condition. The oxygen sites were simply eliminated whenever they diffused to the far boundary. This, however, is not a proper physics model. The code of the improved CA oxide layer growth model has been modified with a Neumann boundary condition of oxygen concentration at the far end of the specimen (at  $y=0$ ). The self-coded CA model considers eight Moore neighborhood lattice sites and oxygen diffuses along the boundary of the lattices. The result shows that the oxygen concentration at the far end satisfies the Dirichlet boundary condition as well if the oxygen diffusion rates in the oxide layer and the metal are relatively small compared to the reaction rate. This is true in most of the real cases and the specimen is usually relatively large compared to the oxide layer thickness.

### A simple scale removal model based on the improved CA oxidation model

A stochastic CA oxidation model was developed to consider the scale removal effect. In the simple scale removal model, it is assumed that the oxide layer sites which are close to oxidant sites have a probability to move away. An average removing probability  $\bar{p}_{kr}$  of a lattice oxide cell was assumed, if the oxide cell is contacting with the LBE flow. The simulation results show that the oxide layer growth following the parabolic law, with a thinner

thickness than the case without scale removal.

A new scale removal model considering the hydraulic effect (the flow direction)

In the new scale removal model, the scale removing probability considers the influence of more neighboring cells (the eastern, western, northern, northeastern, and northwestern sites) which surround the objective oxide layer site. The effect of the neighboring cells on the objective oxide cell is based on the estimation of the hydraulic effect (the flow direction). The proposed scale removal model can lay a basis for a future study coupling with alloy components added in the stainless steel.

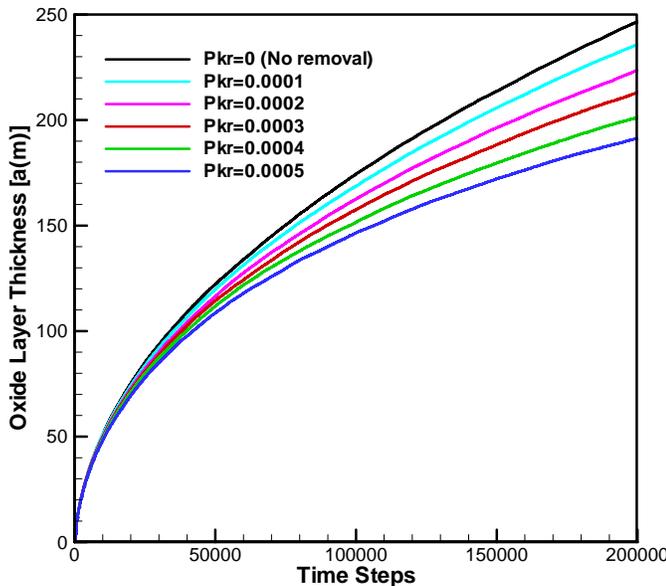
For example, if the flow direction is assumed to be from left flowing to right, more solid sites at the left hand side will make the objective oxide cell more difficult to be removed. Basically, the scale removing probability averaged from the total possible cases should equal to  $\bar{P}_{kr}$  as in the simple model. However, the advantage is that the new  $\bar{P}_{kr}$  model may predict a different roughness on the oxide surface, especially when alloy components are considered, for each cell,

$$P_{kr}^i = f_i \bar{P}_{kr}$$

Where  $f_i$  is a factor reflects the difficulty by which the objective oxide cell will be removed. The more difficult a cell will be to remove, the lower the factor value will be. Thus,

$$\frac{1}{M} \sum_i^M f_i = 1$$

where M is the total number of the possible cases. The scale removal effect on the oxide layer growth has been studied extensively using the self coded CA model. The oxide layer growth was



The oxide layer thickness vs. time steps for cases with  $P_{act}=0.005$  with  $K_d=2$ ,  $C_{oxy}=0.2$ .

**ACADEMIC YEAR HIGHLIGHTS**

- ◆ Taide Tan successfully defended his Ph.D. dissertation titled “Modeling of Protective Oxide Layer Growth in Non-isothermal Lead-Alloys Coolant Systems” on June 20, 2007.
- ◆ T. Tan, Y. Chen, H. Chen, and S. Hsieh, “Modeling of a Diffusion Controlling Oxidation Process with Scale Removal in Oxygen Contained Liquid Flow,” *Proceedings, IMECE*, Seattle, WA, November 11-15, 2007.
- ◆ T. Tan, Y. Chen, and H. Chen, “An Improved Mesoscopic Oxidation Model of Metals in Lead Bismuth Eutectic,” *Computational Material Science*, 43 (2) pp 251-267, August 2008.
- ◆ H. Chen, Y. Chen, J. Zhang, and H.-T. Hsieh, “A Lattice Boltzmann Modeling of Oxygen Transport and Corrosion Behavior in Natural Convection Lead-alloy Flow,” *Nuclear Engineering and Design*, Vol. 237, pp 1987-1998, 2007.
- ◆ H. Chen and Y. Chen, “Cellular automaton modeling on the corrosion/oxidation mechanism of steel in liquid metal environment,” *Progress in Nuclear Energy*, Vol. 50, pp 587-593, 2008.
- ◆ T. Tan and Y. Chen, “Simulations of Metal Oxidation in LBE at a Mesoscopic Level,” *Proceedings, ASME ICONE16*, Orlando, FL, May 11-15, 2008.
- ◆ Y. Chen, T. Tan, and H. Chen, “Oxidation Companioned by Scale Removal: Initial and Asymptotical Kinetics,” *Journal of Nuclear Science and Technology*, 45 (7) pp 662-667, 2008.

found to obey Tedmon’s theory.

Optimization approach in variable charge potential for metal/metal oxide systems in the molecular dynamics (MD) simulations

A new direct approach for finding charge distributions among ions has been proposed in the MD study. Earlier attempts on minimizing the total system energy of metal/metal oxide systems with given charge constraints appear to be indirect and unnecessarily complicated. The energy minimization problem is in fact an optimization problem and hence can be solved by an optimization method. The approach is based on an optimization algorithm, called the Generalized Reduced Gradient (GRG) method. This efficient approach can be readily employed in molecular dynamic simulations for metal/metal oxide systems.

**FUTURE WORK**

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

- To interpret the macroscopic corrosion/oxidation process with the mesoscopic CA model, including scale removal and erosion to the base material.
- To benchmark the CA model with the available experimental corrosion data.
- To perform molecular dynamic simulations of lead/lead-bismuth and iron under varied oxygen concentrations.

**Research Staff**

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