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

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Task 21

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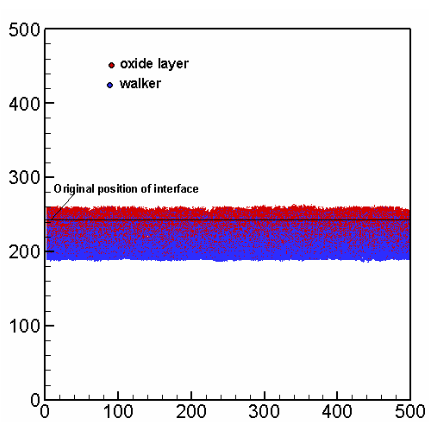
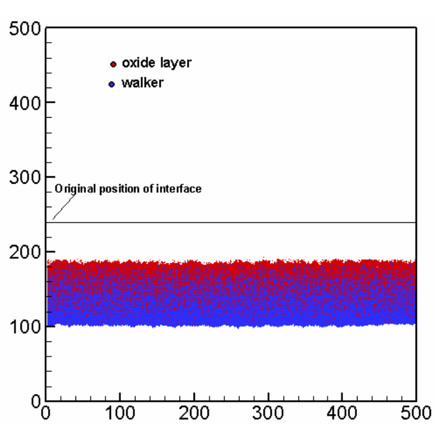
BACKGROUND

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-alloy coolant system. However, in a practical coolant loop, the proper oxygen level depends not only on thermal factors but also on hydraulic factors (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.



(a) time step = 1 x 10^4



(b) time step = 5 x 10^4

Snapshots of the simulated layer in the presence of corrosion with scale removal. They correspond to 2.5 x 10^4 time step. The red dot is the oxide site; The blue dot is the walker site; The upper side of the layer is filled with solvent; The lower side of the oxide layer is pure metal. The corrosion probability of metal is taken as 0.5. The possibility of scale removal of the oxide site close to solvent is 0.004.

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-alloy nuclear coolant systems. The research project, therefore, is aimed at understanding protective oxide layer growth and the optimal oxygen concentration level before lead-alloy nuclear coolants are ready for programmatic implementations and industrial applications.

RESEARCH OBJECTIVES AND METHODS

- 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 oxidation processes on thermal 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.

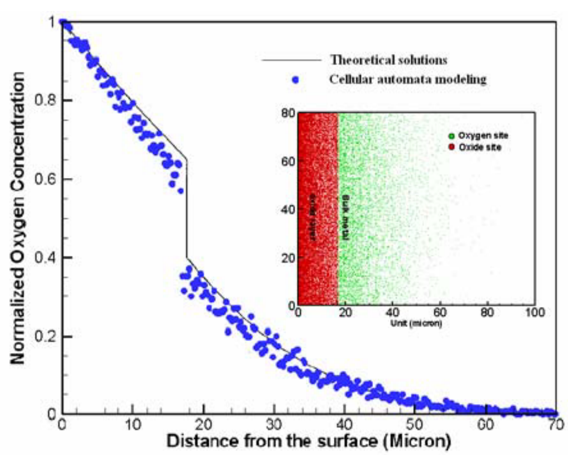
RESEARCH ACCOMPLISHMENTS

Cellular automata modeling of oxide layer growth with scale removal: To simulate the scale removal effect on oxide layer growth, the previous cellular automata model of oxide layer growth has been improved to consider the scale removal effect. In this model, the scale removal rate was taken to be linear. It is assumed that the oxide layer sites which are close to the oxidant site have a probability to move away. The global random walker model is still used to simulate the solid state diffusion of the ionic metal site in the oxide layer. The phenomena of oxide growth which obeyed Tedmon theory was found.

Stochastic modeling with cellular automata method on the surface growth and internal oxidation: The quantitative cellular automata model on the inward oxidation mechanism was developed. The stochastic rule to the population of particles is based on the exclusion principle. Considering the exclusion principle, which permits at most one particle per site, a particle moves to its targeted site if this site is free and simply does not move if the site it targeted was already occupied. To avoid the confliction between particles competing for a single free site, the following rule was adopted. When more than one particle attempts to move to a single open site, only one of the competing particles, which will be chosen randomly from among them, is allowed to move to this open site and the others do not move. A benchmark with the theoretical solution was made, and a good agreement was obtained.

Numerical modeling on the diffusion controlling oxidation model with scale removal: A numerical diffusion-controlling oxidation model with scale removal was developed in oxygen-containing liquid flow. Scale removal effect was considered and the formation mechanism of duplex oxide layer structure was analyzed and employed in the model. The volume expansion effect caused by density difference during the oxidation is coupled with the consideration of increasing weight of reacted oxygen. To solve the diffusion equations with an advection term caused by the moving boundaries, a coordinate transform technique was employed. The governing equations were analogized with the Stefan problem in heat transfer with phase change in the enthalpy formulation and then solved numerically by finite difference method.

Cellular automaton model on the oxide growth coupled with inward oxygen diffusion: To simulate the oxide layer growth of steel in liquid lead alloy environment, a cellular automaton model was developed that considered inward oxygen diffusion. In this model, the lattice sites are occupied by metal, oxide, or lead alloy, while the interstitial sites are route ways for oxygen to occupy and diffuse. The benchmark of cellular automata model for pure oxygen diffusion has been made and a good agreement has been obtained.



Distribution of oxygen concentration at 0.5 hour. Application of quantitative cellular automata modeling of oxidation in metal matrix composite.

ACADEMIC YEAR HIGHLIGHTS

- Y. Chen, H. Chen, and J. Zhang. "Enhancement of oxygen transfer by forced convection in lead alloy system," *International Journal of Heat and Mass Transfer*, 2007, 50 (12): 2139-2147.
- H. Chen, Y. Chen, and H.T. Hsieh. "Stochastic modeling on oxide layer growth with scale removal effect in liquid metal environment," 15th International Conference on Nuclear Engineering (ICONE 15), April 22-26, 2007, Nagoya, Japan.
- H. Chen and Y. Chen, "Cellular automaton modeling on the corrosion/oxidation mechanism of steel in liquid metal environment," 2nd COE-INES International Symposium on Innovative Nuclear Energy Systems, November 26-30, 2006, Yokohama, Japan.
- X. Tan, Y. Chen, H. Chen, T. Tan, and H.T. Hsieh, "Numerical analysis of natural convection induced oxygen transport in liquid lead bismuth eutectic;" T. Tan, Y. Chen, H. Chen, and H.T. Hsieh, "Corrosion precipitation in non-isothermal lead alloy coolant systems;" and, H. Chen, Y. Chen, and H.T. Hsieh, "A stochastic model of oxidation mechanism on high temperature corrosion of stainless steel," 2006 ASME International Engineering Congress and Exposition, Chicago, IL, November 5-10, 2006.

FUTURE WORK

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

- Structure stability analysis on oxide layer under liquid metal environment for different operating conditions.
- Model the erosion-corrosion process mechanisms in oxygen-controlled lead-alloy systems.
- Identify the protective oxide layer growth under corrosion and oxidation mechanisms using stochastic methods.
- Clarify the dependence of the oxidation process on the hydraulics factors (system operating temperature, temperature profile, flow velocity, etc.) and the oxygen concentration distribution and level.
- Predict the optimal oxygen concentration levels in practical coolant system scales.

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