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An improved mesoscopic oxidation model of metals in lead bismuth eutectic

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Abstract

The oxidation process of metals in lead bismuth eutectic (LBE) environment is studied at a mesoscopic scale. An improved stochastic cellular automaton model based on an improved Moore neighborhood is proposed to investigate the development of a continuous oxide layer of metals in LBE. The ionization of metal and the oxidation reaction were simulated with consideration of the transport of oxygen along the grain boundaries and the diffusion of metallic ions. The growth of oxide layer in two directions is observed and the volume expansion effect can be realized by changing the volume control parameter. The model was benchmarked with a diffusion process, both with the analytical solution and with the previous work. Significant agreement was reached between the data. The developed model is also mapped with the experimental data from an LBE loop. A parametric study was conducted in order to check the importance of the main explicit parameters of the mesoscopic model.

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1. Introduction

Lead bismuth eutectic (LBE) has been determined to be a potential coolant candidate in reactors and accelerator driven systems (ADS) because of its favorable thermal–physical and chemical properties [1–3]. However, the corrosiveness of LBE presents a critical obstacle and challenge for safe applications in advanced nuclear reactors and ADS. One of the effective ways to protect the materials is to form and maintain a protective oxide film along the structural material surfaces by active oxygen control technology [3–7].

The corrosion and oxidation of stainless steel in molten lead or LBE have been studied for years [3–11]. The oxide layer structures from experimental results were summarized for steels in molten lead and LBE in Refs. [8,11]. Based on the observations, the mechanism of the oxide layer growth of stainless steel in liquid lead and lead alloys were analyzed [8,11]. However, to study such a problem at a microscopic level, such as from an atomic or molecular point of view, has always been a great challenge for the scientists and researchers in the field of fluid mechanics, heat transfer, computational fluid dynamics (CFD), etc. It is difficult and time consuming to study so complex a macroscopic phenomenon at a microscopic level. There is no favorable microscopic theory available to explain and calculate all macroscopic fluid mechanics phenomena as a whole. With the development of modern computers, it becomes possible to simulate a simple fluid problem at a microscopic scale, but still not for such a complex problem in which fluid mechanics, heat transfer, mass transfer, chemical reactions and other phenomenon all are involved.

Therefore, to build a mesoscopic model for the oxidation of metals in lead or LBE environment will be significant and beneficial for the future study of oxidation

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Nomenclature

a	width of a square lattice in CA model in unit of meters
B	Pilling–Bedworth ratio
C	concentration of oxygen
$\overline{C_{\mathrm{Oxy},j}}$	mean concentration of oxygen sites in jth line for interstitial sites
$\overline{C_{\mathrm{wa},j}}$	mean concentration of "walkers" in jth line for lattice sites
D_{eff}	effective diffusion coefficient
D_{S}	tracer self-diffusion coefficient
D_{B}	diffusion coefficient along the grain boundary
i	node number of the calculating domain
$\mathrm{Inte}_{i,j}(t)$	state variable of the interstitial sites
$\mathrm{Inte}_{i,j}^{\mathrm{Nb}}(t)$	state variable of the neighbor interstitial sites of the lattice (i,j)
Inte_0	a "vacancy" interstitial site
Inte_1	an "occupied" interstitial site by oxygen
$\mathrm{IO}_{i,j}$	oxygen state value for an interstitial site
$\mathrm{IW}_{i,j}$	walker state value for a lattice site
k	iteration step number
$k_{N,p}$	parabolic rate coefficient based on CA model
k_p	parabolic rate coefficient for real oxide layer growth
K_{d}	ratio of transport steps of an oxygen site and diffusion steps of a walker
L_0	original thickness of the specimen
$\mathrm{Lat}_{i,j}(t)$	state variable of the lattice site
$\mathrm{Lat}_{i,j}^{\mathrm{Nb}}(t)$	state variable of the neighbor lattice sites of the lattice (i, j)
Lat_2	an atomic metal lattice site (solid phase)
Lat_3	a LBE lattice site (liquid phase)
Lat_4	an oxide lattice site (solid phase)
Lat_5	a lattice site of oxide with overlapping with ironic metal (solid phase)
N_{OT}	transport steps of oxygen in a calculating step
N_{WD}	diffusion steps of walker in a calculating step
N_{t}	calculating steps
N	lattice number in x-direction
P_{act}	reaction probability of oxygen sites and metal sites
t	time
Δt	a short time difference
x	coordinate in longitude direction
Y_0	original interface of metal/LBE
$Y_{\mathrm{Ox\_Min}}(i)$	minimum position of the oxide site at ith column
$Y_{\mathrm{Ox\_Min}}(i)$	maximum position of the oxide site at ith column

Greek symbols

α	number of metallic ions in an oxide molecular
β	number of oxygen ions in an oxide molecular
δ	thickness of laminar sub-layer
$\delta_{N,\mathrm{in}}$	mean inner layer thickness
$\delta_{N,\mathrm{out}}$	mean outer layer thickness
$\delta_{N,\mathrm{tot}}$	mean total thickness of the oxide layer
Δ	difference
$\Delta t(s)$	the time for forming thickness a (m) of oxide layer
$\Delta\delta_{\mathrm{in}}$	increase of inner oxide layer
$\Delta\delta_{\mathrm{out}}$	increase of outer oxide layer
$\Delta\delta_{\mathrm{tot}}$	increase of the total oxide layer
ϕ	fraction of the diffusion along the grain boundary
Φ_{Lat}	local evolution rules for a lattice site
Φ_{Inte}	local evolution rules for an interstitial site
φ_{Lat}	control variables for an interstitial site
φ_{Inte}	control variables for a lattice site
ε	volume control parameter
ω	number of oxygen atoms at an interstitial site

problems at a mesoscopic scale. To the best of knowledge, there has been no such mesoscopic work yet done, with consideration of the diffusion of metal and transport of oxygen separately. Thus, this is one of the main motivations for this part of the research. Usually, the corrosion and oxidation involve several macroscopic processes, e.g., corrosion, precipitation, passivation/depassivation, growth of oxide layer, mechanical stress, etc. Each of these processes may be the result of several processes at a microscopic scale. In addition, all these phenomena take place in ill-defined conditions resulting from the environmental evolutions [12]. At the same time, the combination of these processes may generate different kinds of interfaces.

In the domain of interface growth, there has been much theoretical investigation based on numerical simulations [13]. The Eden model [14] and the Diffusion-Limited Aggregation (DLA or LDA) [15] model represent two basic starting points from which a theoretical description of the growth processes can be elaborated. The Eden model is the simplest model, which describes a growing cluster of particles. It is a lattice model in which particles are added one at a time randomly to sites adjacent to occupied sites [14]. A particle on the perimeter of the cluster acquires a new particle on its empty nearest-neighbor sites with equal probability. The DLA model was first proposed by Witten and Sander [15] and further explored in Refs. [16,17]. It is a variant of the Eden model. In the DLA model there are screening effects, which differentiate the exposed perimeter particles from the shadowed ones by assigning them different probabilities. The connection between the Eden model in finite dimensions and the DLA model has been discussed by Plischke and Racz [18]. The DLA model and the Eden model should coincide in infinite dimensions since the excluded-volume effects disappear in a DLA model. This