



Model of liquid gallium corrosion with austenitic stainless steel at a high temperature



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ABSTRACT

The purpose of this study is to model the interaction between austenitic stainless steel and liquid gallium at a high temperature to predict the weight loss for potential application as a target material or coolant in an advanced nuclear system. In the present study, models for liquid gallium corrosion with austenitic stainless steel are studied and discussed. This paper presents a mathematical analysis of liquid gallium corrosion, especially the surface recession due to solubility and diffusion, in a static cell, as well as the mass exchange at the liquid/solid interface. Also, a mathematical analysis of liquid gallium alloy (Ga–Sn–Zn) corrosion is conducted in order to study its effect on the diffusion behavior after the addition of alloying elements. The results show that the predicted corrosion behavior agrees well with experimental data and the weight loss of austenitic stainless steel are significantly reduced in gallium alloy compared to those in pure gallium at high temperatures.

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1. Introduction

Liquid–metal fast-breeder reactors (LMFBRs) such as the sodium-cooled fast reactor (SFR) and the lead-cooled fast reactor (LFR) are promising candidates among Gen-IV nuclear energy systems. Also, the MYRRHA (multi-purpose hybrid research reactor for high-tech applications) design uses liquid lead–bismuth (Pb–Bi) as the coolant and the spallation target.

Among the various liquid metals that can be used as a primary coolant, sodium is a widely used coolant in the design of fast breeder reactors. However, sodium has a disadvantage in that it reacts violently with water and air. This factor drives the search for alternatives. Therefore, heavy liquid metals, including pure lead and lead–bismuth eutectic (LBE) alloy, have been extensively studied for their applicability to fast reactors and as a spallation target. However, issues such as corrosion with structural alloys at a high temperature can arise with these materials, and Po-generation can occur in LBE-cooled nuclear systems.

This study focused on another liquid metal, gallium (Ga), as a potential coolant for next-generation nuclear reactor systems such as ultra-long cycle fast reactor [1]. The element of gallium possesses several unique properties. It is very stable in air or water, has a very low melting point, and a very high boiling point. It melts at 29 °C, and its boiling point is as high as 2204 °C, which gives a designer a larger margin before coolant boiling, a severe accident, occurs [2,3]. When gallium is alloyed with tin and zinc at a certain

ratio, the melting point becomes 21 °C. Tin and zinc are materials which have both a lower absorption cross-section than gallium and both make ternary eutectic point with gallium [4].

When viewed in terms of compatibility as a coolant, which is the main interest of this study, gallium has relatively high affinity for many metals and alloys, especially steels. Therefore, relatively high corrosion of these structural metals and alloys occurs due to the dissolution of their constituents by liquid gallium [4–7].

However, an assessment of gallium or its alloys for use in advanced nuclear reactor as a primary coolant is needed due to the fact that the chemical safety and design margin of gallium in a liquid state at room temperature are attractive to designers of nuclear reactors. While lead or lead–bismuth alloy and sodium corrosion models are widely studied for their application to advanced nuclear reactors [8,9], a model-based study for long term corrosion behavior of structural materials in high temperature gallium condition was not widely studied and it is needed to be performed prior to the further research and development for the application. The interaction between austenitic stainless steel, AISI 316L, and gallium liquid metals at a high temperature, for the potential application to advanced fast reactor coolants was investigated in author's previous work [10]. In this study, therefore, a model of gallium corrosion is scrutinized to predict the lifetimes of structural materials at high temperature.

2. Methodology

Zhang et al. [8] made a review of different corrosion models in liquid metals. This review emphasizes the model of Brush [9]. Their study presents a mathematical analysis of liquid metal corrosion

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Nomenclature

c'_i	concentration of species i in liquid	ρ_m	density of the metal
$c'_{i,l}$	concentration of species i at the interface in liquid side	ρ_r	density of the reaction layer
c_i	concentration of species i in solid	R	recession rate or bulk corrosion rate
$c_{i,0}$	bulk or initial concentration of species i in solid	Δw_i	weight loss
$c_{i,l}$	concentration of species i at the interface between metal and reaction layer	$\vec{N}$	normal vector
$c_{i,m}$	concentration of species i in the metal	τ	dimensionless time
$c_{i,r}$	concentration of species i in the reaction layer	D'_i	diffusion coefficient species i in liquid
$c_{i,r,l}$	concentration of species i at the interface in reaction layer side	D_i	diffusion coefficient species i in solid
$c_{i,m,0}$	bulk or initial concentration of species i in metal	$D_{i,m}$	the diffusion coefficient of species i in the metal
$c_{i,m,l}$	concentration of species i at the interface in metal side	$D_{i,r}$	the diffusion coefficient of species i in the reaction layer
ρ_L	density of the liquid	J'_i	mass flux of species i in the liquid
		$J_{i,m}$	mass flux species i in the metal
		$J_{i,r}$	mass flux species i through the reaction layer

with Na, Pb and Pb–Bi, including species transport in solid steels, in flowing liquid metals, and the mass exchange at the liquid/solid interface. The model developed by Brush [9] is extended and applied to gallium and its alloys in corrosive environments. The present study focuses on the corrosion of gallium, which shows different corrosion behavior compared to the Na, Pb or Pb–Bi cases.

In this study, models of the weight loss of iron, chromium, nickel, and iron–chromium–nickel alloy (74Fe–16Cr–10Ni in wt.%), which simulates AISI 316L stainless steel, are predicted using concentration data and the diffusion coefficients from author's previous study [10].

2.1. Governing equation in a static cell

In order to theoretically model and to analyze the corrosion behavior of metals and steels in gallium, mass transfer equations through Eqs. (1)–(5) are introduced as follows.

The mass transfer equation can be expressed as

$$\frac{\partial c}{\partial t} + \nabla J + q = 0 \quad (1)$$

where c is the concentration of species, J is the flux and q is the homogeneous reaction rate of disappearance of species in the bulk material. The mass flux equation of Ref. [8] for species i in a liquid can be simply described in a static cell as

$$J'_i = -D'_i \nabla c'_i \quad (2)$$

where c'_i is concentration of species i in liquid, D'_i is the diffusion coefficient of species i in the liquid. For species i in solid, the mass flux in a solid can be expressed as

$$J_i = -D_i \nabla c_i + R(t) c_i \quad (3)$$

where D_i is the diffusion coefficient of species i in the solid, c_i is concentration of species i in solid, and $R(t)$ is the corrosion rate as a function of time. The mass transport equation in the coordinate for constant diffusion coefficient can be expressed as

$$\frac{\partial c'_i}{\partial t} = D'_i \left(\frac{\partial^2 c'_i}{\partial x^2} + \frac{\partial^2 c'_i}{\partial y^2} + \frac{\partial^2 c'_i}{\partial z^2} \right) + q'_i \quad (4)$$

for the species i in a liquid ($x > 0$) and as Eq. (5) of Ref. [8] can be extended as

$$\begin{aligned} \frac{\partial c_i}{\partial t} + R(t) \frac{\partial c_i}{\partial x} &= D_{i,m} \left(\frac{\partial^2 c_{i,m}}{\partial x^2} + \frac{\partial^2 c_{i,m}}{\partial y^2} + \frac{\partial^2 c_{i,m}}{\partial z^2} \right) + D_{i,l} \left(\frac{\partial^2 c_{i,l}}{\partial x^2} + \frac{\partial^2 c_{i,l}}{\partial y^2} + \frac{\partial^2 c_{i,l}}{\partial z^2} \right) \\ &+ D_{i,r} \left(\frac{\partial^2 c_{i,r}}{\partial x^2} + \frac{\partial^2 c_{i,r}}{\partial y^2} + \frac{\partial^2 c_{i,r}}{\partial z^2} \right) + q_i \end{aligned} \quad (5)$$

in a solid ($x < 0$), where $D_{i,m}$ is the diffusion coefficient of species i in the metal, $c_{i,m}$ is concentration of species i in the metal, $D_{i,l}$ is the diffusion coefficient of species i in the interface between the metal and the reaction layer, $c_{i,l}$ is concentration of species i in the interface between the metal and the reaction layer, $D_{i,r}$ is the diffusion coefficient of species i in the reaction layer and $c_{i,r}$ is concentration of species i in the reaction layer. In Eq. (5), the term of $\frac{\partial^2 c_{i,m}}{\partial x^2} + \frac{\partial^2 c_{i,m}}{\partial y^2} + \frac{\partial^2 c_{i,m}}{\partial z^2}$ becomes zero, because of no concentration gradient along the metal. This is caused by the fast weight loss by the liquid metals.

If the reaction term in a solid is less than zero, the reaction is species consumption while if the reaction term in a liquid is greater than zero, the reaction is species production.

2.2. Corrosion rate of metal in static cell

In pure gallium, the dissolution reaction will be an early step. In addition, for long-term operation in a steady state, the net corrosion will be controlled by either dissolution at the surface of metal or diffusion of species through the reaction layer which formed on the surface of the metal. In these cases, the diffusion rate of species through the reaction layer is fairly large, but it is less than the mass transfer rate. There will be surface recession of the metal in this case. The recession rate can be calculated through the mass balance at the interface between the metal and the reaction layer. The Eq. (16) of Ref. [8] can be applied to the interface between the metal and the reaction layer as

$$R = - \frac{\rho_r}{\rho_m - \rho_r \sum_i c_{i,r,l}} \left(\sum_i D_{i,r} \frac{\partial c_{i,r}}{\partial \vec{N}} - \frac{\rho_m}{\rho_r} \sum_i D_{i,m} \frac{\partial c_{i,m}}{\partial \vec{N}} \right) \quad (6-a)$$

$$R_i = - \frac{\rho_r}{\rho_m - \rho_r c_{i,r,l}} \left(D_{i,r} \frac{\partial c_{i,r}}{\partial \vec{N}} \right) \quad (6-b)$$

where ρ_r is density of reaction layer, ρ_m is density of the metal, $c_{i,r,l}$ is concentration of species i at the interface in reaction layer side and $\vec{N}$ is normal vector. In Eq. (6) (a), the term, $\frac{\partial c_{i,m}}{\partial \vec{N}}$, becomes zero due to no concentration gradient in the metal, then Eq. (6) (b) is introduced for the calculation of the surface recession of each species.

2.3. Transport in a solid

With a constant recession rate, the governing equation for transport in the solid can be expressed as follows:

$$\frac{\partial c_i}{\partial t} + R \frac{\partial c_i}{\partial x} = D_{i,m} \frac{\partial^2 c_{i,m}}{\partial x^2} + D_{i,l} \frac{\partial^2 c_{i,l}}{\partial x^2} + D_{i,r} \frac{\partial^2 c_{i,r}}{\partial x^2} \quad (7)$$

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