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Investigation on Compatibility of Structural Materials in Liquid Gallium Environments

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1. Introduction

Liquid Metal Fast Breeder Reactor is one of promising candidates among Gen IV nuclear energy systems. Until now the coolants considered as for the Gen IV nuclear systems are water, gas such as helium and carbon dioxide, and liquid metals including sodium, lead, and lead-bismuth eutectic, etc. Among liquid metal coolants, sodium is a spotlighted coolant for the designing fast breeder reactor. However the disadvantage of sodium, high activity with water and air, is the factor that searches for alternatives. In liquid metal environment, structural metals and alloys interact with the liquid metal, and it causes liquid metal embrittlement. Therefore, materials that bear on liquid metal corrosion are required.

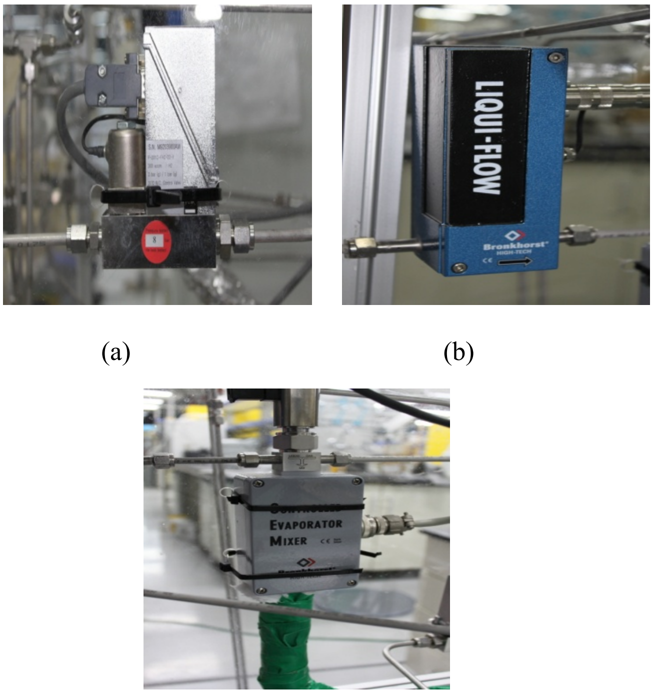
This study suggests gallium as potential coolant for the next generation reactor because of its low melting point (30°C), high boiling point (2204°C), and high safety against explosion. In 1950s, researchers studied the use of gallium as coolants, but the corrosion at elevated temperature in some metals and alloys were observed significantly. The most competitive materials interacted with gallium were known as refractory metals such as tungsten at high temperature [1]. But the difficulty in manufacturing process of refractory metals was one of issues for the use of refractory metals as structural materials. Meanwhile, stainless steels could be good at this point if there are no corrosion problems. For the prevention of liquid gallium corrosion, especially mass transfer type, an active control of oxygen potential pressure to form protective oxide scales on the surface of metals, which has been extensively studied for preventing lead-bismuth corrosion with structural alloys could be one way which can be adopted in liquid gallium environment.

2. Experiment

In this study, active control of oxygen partial pressure in gallium environment as a coating technique to prevent corrosion by forming a protective oxide scale on structural material alloys. This can offer the specific oxygen potential via H_2/H_2O ratio.

2.1 Experimental system setup

To control oxygen partial pressure in liquid gallium environment at high temperature, experimental system which is similar to ones used previous studies [2][3] is designed and assembled in this study. Many experiments in lead-bismuth environment using this system were conducted to form oxide scales between interfaces [2][3]. This system consists of mainly three components, i.e., mass flow control system for gases, liquid flow meter for water, and controlled evaporation & mixing system as shown in Fig. 1. Detail schematic diagram for the experimental system is shown in Fig. 2.



(a) (b)

(C)

Fig. 1 Main components of oxygen control system: (a) mass flow control system, (b) liquid flow meter, (c) controlled evaporation & mixing system

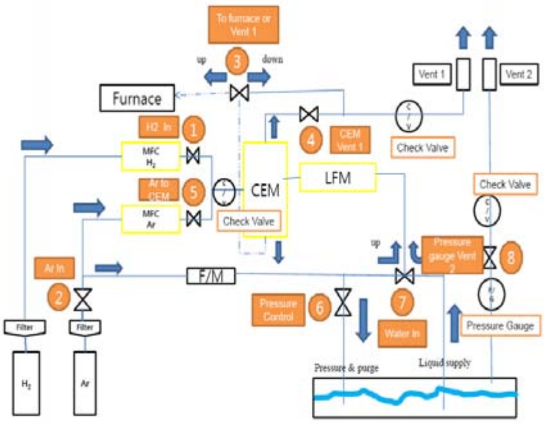


Fig. 2 The schematic diagram of oxygen control system in liquid gallium environment

2.2 Thermodynamic condition

In this study, to form chromium oxide scale on the surface of structural material as a protective scale can help to prevent corrosion occurred at the interface. When a metal oxidizes, there need energy called Gibbs free energy change in thermodynamic view point.

These value as a function of temperature can be plotted, which is well known as Ellingham diagram as shown in Fig. 3

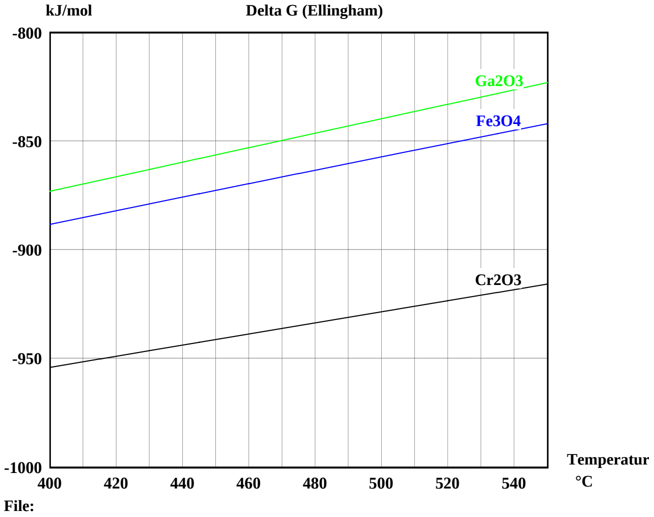


Fig. 3 Ellingham diagram of Ga_2O_3 and Cr_2O_3.

For the gallium environment, this system must fulfill next condition to prevent Ga_2O_3 precipitation and support Cr_2O_3 formation:

$$\frac{2}{3}\Delta G^0_{Ga_2O_3} > RT\, ln p_{O_2} > \frac{2}{3}\Delta G^0_{Cr_2O_3} \quad (1)$$

where the p_{O2}, oxygen partial pressure, is defined as follow:

$$p_{O_2} = \frac{p^2_{H_2O}}{p^2_{H_2}} \exp\left(\frac{2\Delta G^0_{H_2O}}{RT}\right) \quad (2)$$

To obtain optimized oxygen partial pressure, the term p_{H2O}/p_{H2} ratio is controlled via oxygen control system. Thermodynamic conditions at 500°C are evaluated and shown in Table I:

Table I: The thermodynamic conditions at 500°C

Oxides	ΔG (J/mol)	H_2/H_2O ratio	p_{O_2}(atm)
Ga_2O_3	-840,000	$1.2818*10^5$	$1.483*10^{-38}$
Cr_2O_3	-987,502	$2.6863*10^8$	$3.71*10^{-42}$

Using this system, the possible thermodynamic conditions are shown in Table II:

Table II: The possible thermodynamic conditions

H_2/H_2O ratio	p_{O_2}(atm)
450,000 (Max.)	$6.4642*10^{-39}$
180 (Min.)	$4.0401*10^{-32}$

Formation of Cr_2O_3 on the surface of structural material can be expected thermodynamically.

3. Conclusion

In this study, the formation of protective oxide scale as chromium oxide by active oxygen partial pressure control in liquid gallium environment at high temperature is currently in process and it is believed to prevent the liquid metal embrittlement as barriers between interfaces.

Refractory metals such as tungsten have low solubility in gallium, which can be an important index of the resistance of structural metals to the liquid gallium. One of issues for the use of refractory metals as structural materials is generally caused by difficulty in manufacturing process. To improve mechanical properties of refractory metals, alloying with other minor elements such as rhenium in tungsten will be considered in the future work.

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