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Capability demonstration of simultaneous proton beam irradiation during exposure to molten lead–bismuth eutectic for HT9 steel

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ABSTRACT

We report the design and assembly of a corrosion station to enable simultaneous proton irradiation of a metallic surface that was also in contact with molten lead–bismuth eutectic (LBE). The capability has been established at the ion beam materials laboratory at Los Alamos National Laboratory (LANL). The engineering design focused on temperature and oxygen content control in the LBE, as well as the ability to achieve doses significantly in excess of 1 dpa in the contact region over the irradiation campaigns. In the preliminary demonstration of capability reported here, a sample made of HT9 steel was placed in contact with LBE at 450 °C and irradiated for 58 h at an average proton beam current of 0.3 μA/mm^2. SRIM [1] calculations indicate that the nominal surface dose ranged from approximately 3–22 dpa. This paper outlines the experimental setup and design constraints. Characterization of the sample will be reported in a subsequent paper.

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1. Introduction

Research and development of lead and lead–bismuth eutectic (LBE) cooled nuclear technology is currently pursued worldwide under the framework of generation-IV nuclear programs or spallation neutron source development. As a nuclear reactor coolant, lead and LBE offer significant advantages over other available alternatives. They have no explosive chemical reactions with air, water and steam, very high boiling points (>1600 °C) and excellent natural circulation cooling capability compared to alternatives such as sodium-based coolants. Lead-based coolants are especially attractive for use in accelerator-driven systems (ADS) as they can be used as a combined spallation neutron source and coolant. Material research facilities such as the proposed material test stand (MTS) at Los Alamos National Laboratory also utilize LBE as a coolant [2]. The main alternatives for fast reactor coolants are sodium-based liquid metal systems or the use of gaseous coolants. Sodium emerged as the main fast reactor coolant technology and has been the coolant choice for all commercial fast reactors that have been constructed so far worldwide.

The major technological hurdles for the widespread adoption of lead-cooled fast reactor (LFR) technology was and continues to be the problem of high temperature corrosion of structural materials [3], liquid metal embrittlement (LME) [4] and liquid metal enhanced creep (LMC) [5]. Discoveries in the Russian lead reactor development program found that careful control of the oxygen content inside a Pb/LBE melt enables the creation of stable protective oxide layers on steel surfaces, which ignited worldwide interest in conceptual and commercial design of lead/LBE cooled reactor concepts [6]. Major LFR projects currently under development are summarized in Table 1. Experiments testing the corrosion behavior in steels at various temperatures, oxygen contents and flow-rates have been set up at a number of institutions worldwide and the corrosion knowledge base for a number of steels is substantial [3]. However, no experimental setup so far has been designed to test the combined effect of corrosion and irradiation in a high temperature environment under chemistry control. Such tests are necessary in the process of building the experimental database for licensing of any steel for use in a LBE cooled nuclear system. Ideally, experiments would be performed in a fast flux region of an existing nuclear test reactor, a high-flux spallation source or in a materials testing Pb/LBE research reactor. These options, however, present major hurdles both in the form of costs and availability.

The first irradiation and corrosion experiment (ICE-I) [7] was an initial attempt at obtaining such data without the use of nuclear

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Table 1
Lead and LBE-cooled nuclear concept characteristics.

Country/region	Concept	Size (MWe)	Cladding	Coolant	Clad. T. (°C)
US	ENHS [17]	50	HT9	LBE	650
	Gen4 Module [a] [18]	25	HT9	LBE	600
	SSTAR [19]	20–400	Si-enhanced FM	Pb	650
	MABR/ABRT [20]	300	HT9	LBE/Pb	600
EU	ELSY [21]	600	T91 (GESA)	Pb	550
	MYRRHA/XT-ADS [22]	20–35	T91	LBE	
	EFIT (Giacomino Bandini et al., [23])	135	T91	LBE	
	ALFRED [21]	120	12R72 (GESA)	Pb	
	ELFR [21]	600	T91 (GESA)	Pb	
Russia	SVBR [24]	75/100	EP823	LBE	650
	BREST [25]	300–1200	Cr12MoVNbB	Pb	
Japan	CANDLE [26]	Variable	HT9	LBE/Pb	528
	PBWFR [27]	150	N/A	LBE	620
	LSPR [28]	53	HT9	LBE	<700
S. Korea	PEACER [29]	300	HT9	LBE	<700
	BORIS [30]	10	HT9	Pb	600
	PASCAR [31]	35	Fe–Cr–Al alloy	LBE	<700
Sweden	ELECTRA [32]	2 (thermal)	12R72(GESA)	Pb	480
India	CHTR [33]	0.1(thermal)	Graphite	LBE	1000+

[a] Previously known as Hyperion Power Module.

reactors or a spallation source, and it utilized a proton particle beam from a conventional ion accelerator as the irradiation source. The ICE-I experiment operated at a temperature of 300 °C, well below the nominal design temperature expected in a commercial nuclear reactor. Also it did not feature oxygen content control and had shielding that limited the beam current to less than 500 nA with concomitant limitation on the dose rate. While the ICE-I experiment did not produce meaningful experimental results, it did form the conceptual basis and outline for the design of the ICE-II station reported here.

2. Degradation phenomena on candidate nuclear materials by lead-based coolants

All of the major elements present in candidate steels for nuclear applications (iron, chromium, and nickel) have significant solubility in Pb and LBE [8]. While this is not a significant issue in isothermal systems due to the eventual saturation of the elements in the melt, it can lead to significant issues in non-isothermal systems such as reactor coolant loops. In such systems, the dissolution of elements in the hot area (cladding) is continuous, as corrosion products precipitate out of the system at cold areas (heat exchanger) due to their reduced solubility at lower temperatures. This brings fresh unsaturated LBE back to the hot area where further dissolution can take place. This process leads to a reduction of wall thickness on the fuel cladding and hot area piping which can result in the subsequent mechanical failure of the components, as well as a clogging of the flow path inside the pipe in the cold section (heat exchanger) because of deposition. Therefore in order to safely operate coolant loops for extended time periods using lead or LBE, the rates of dissolution and deposition need to be minimized.

When oxygen is present in the coolant medium, oxides will form on the steel surfaces of the reactor internal structures because of the fact that the steel constituent oxides (Fe_3O_4, Cr_2O_3, NiO, etc.) have lower Gibbs free energy than the oxides of the melt (PbO, Bi_2O_3, etc.). The oxide layer prevents direct contact between the steel surface and liquid metal, which helps to mitigate the problems of corrosion, LME and LMC mentioned above. The diffusion rate of the alloying elements and the oxygen through the passivation oxide layer is several orders of magnitude lower than in the bulk steel. Since the surface of the steel is not in contact with the corrosion medium if an oxide layer is present, the corrosion process can be slowed down significantly [9]. It has been found that oxidation of steel in LBE is significantly faster than in air at the same temperature, which seems to be based on a combination of leaching and oxidation. Small pore channels enhancing mass transport through the passivation films were discovered, which explain these issues [10]. To form a stable, dense and adherent passivation layer, careful continuous control of the oxygen activity throughout the coolant loop is a necessity. If the oxygen concentration is kept at saturation, the formation of solid lead oxides may lead to clogging of the coolant flow paths. Such an event led to the failure of one of the first soviet LBE cooled nuclear submarine reactors [11]. In addition, high oxygen concentrations will lead to thicker oxide scales, which reduces the structural integrity and the heat transport efficiency and significantly increases the probability of spalling off the oxide layer in the flow. Too low oxygen concentrations may leave bare metal exposed to dissolution in the corrosive medium.

The optimal concentration is a function of temperature in the loop as well as the specific types of the steels being used. Steels with sufficient amounts of strong oxide formers (e.g. aluminum or silicon) will easily form very thin, dense and strong oxide layers, which are excellent diffusion barriers. It has been shown that the Russian martensitic alloy EP823 (containing 1.5 wt% Si) behaves better than similar materials without Si [12] and materials containing 5.5 wt% Al do not show any significant sign of corrosion attack [13].). Surface alloying using the Gepulste Elektronenstrahl Anlage (GESA) process, with protection based on a thin layer of FeCrAlY welded to the surface by an intense electron beam, has been shown to provide excellent corrosion resistance to all steels subjected to the treatment [14].

3. Effect of irradiation on liquid metal corrosion

The main objective of the ICE-II station is to investigate the effects that irradiation induced damage has on the corrosion