Influence of Proton Irradiation on Corrosion in Liquid Lead

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INTRODUCTION

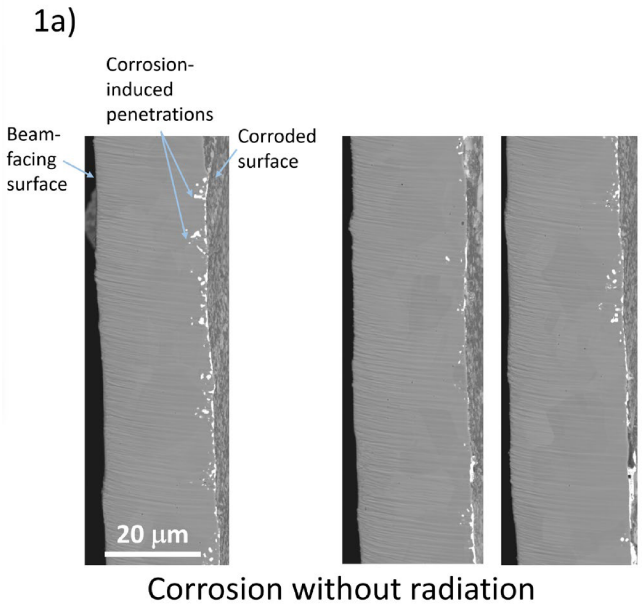
The next-generation Gen IV nuclear reactors are designed to operate under increasingly challenging environments, aiming for higher thermal efficiency while adhering to strict physical and safety constraints. These harsh conditions, characterized by elevated temperatures and accelerated corrosion rates, coupled with the presence of high radiation damage rates, necessitate a thorough understanding of the complex interaction between radiation and corrosion. However, experiments that incorporate radiation into the corrosion evaluation of structural materials, particularly in liquid metal environments, are scarce and challenging to conduct.

To address this research gap, we have developed a unique experimental apparatus that enables simultaneous irradiation and corrosion testing using proton beams as the radiation source. In this setup, a foil sample is exposed to liquid lead on one side, while protons are directed from the opposite side, resulting in a central region within the foil that experiences both irradiation and liquid lead corrosion. By comparing the behavior of this central region with the surrounding areas, we can observe the specific effects introduced by the additional proton beam on the corrosion process. This facility provides valuable insights into the rates and mechanisms of radiation-altered corrosion in lead and lead-bismuth eutectic (LBE) environments, ultimately contributing to improved material selection, design optimization, and enhanced corrosion resistance in next-generation reactor systems.

EXPERIMENTS AND RESULTS

Originally designed for testing in a molten salt medium [1], substantial efforts have been made to adapt the apparatus for liquid lead experiments. Several different materials, including SS316L and Fe-Cr-Si alloys, were evaluated at temperatures ranging from 650 ℃ to 700 ℃ under high vacuum conditions. Additional oxygen getters were incorporated to suppress oxygen content during the experiments. Preliminary testing was conducted using liquid lead containing 4 wt.% bismuth, which was purified beforehand using an argon-hydrogen mixture to remove oxygen from the oxides. The SS316L foils were commercially available, while the Fe-Cr-Si alloy foils were rolled in-house. Proton beams with an energy of 3 MeV and a beam current of 80 nA were directed onto a 5 mm-diameter circular region, which resulted in a damage rate on the order of 10^{-7} DPA (displacements per atom)/s.

Preliminary results using SS316L revealed that proton irradiation altered the localized corrosion attack, causing the corrosion to spread over a larger area while decreasing the average depth of attack. Cross-sectional analysis of SS316L samples after irradiation and corrosion experiments, as shown in Figure 1, indicates a distinct corrosion appearance in regions subjected to both radiation and corrosion compared to regions undergoing corrosion alone. In Figure 1, it can be observed that SS316L corrodes in a localized fashion, as indicated by the penetration of liquid metals. However, the distribution of the localized penetration shows significant variation between the two regions. Corresponding scanning electron microscopy-energy-dispersive X-ray spectroscopy (SEM-EDX) elemental mapping confirmed lead penetration accompanied by significant nickel and chromium depletion during the liquid lead corrosion attack.



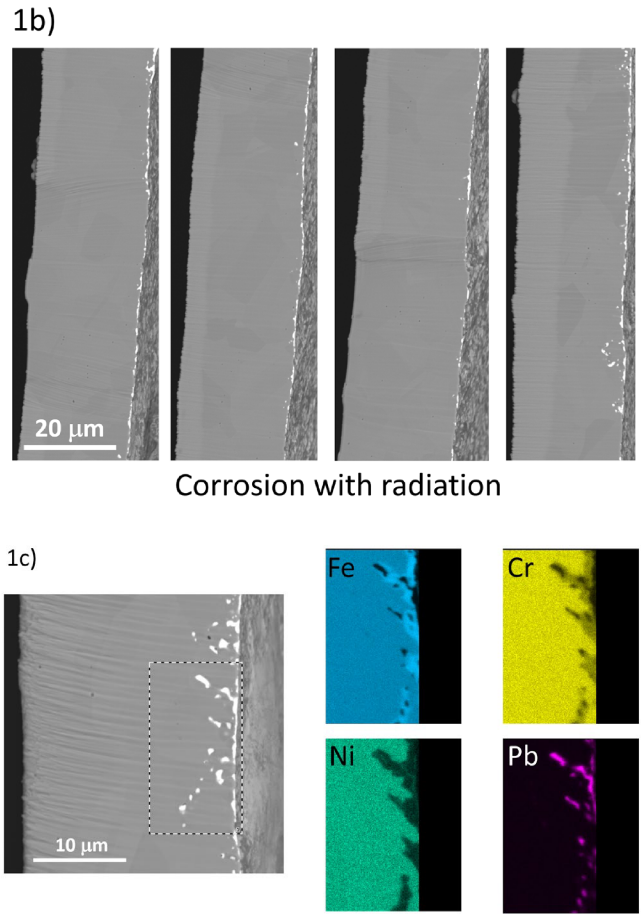


Fig. 1. Characterization of cross sections after radiation and corrosion experiments. The sample used is SS316L, corroded at 675 °C for 4 hours. (a) Cross sections from regions not subjected to proton irradiation. (b) Cross sections from regions under proton irradiation (0.4 uA/cm²). (c) Representative elemental behavior from corrosion.

To quantify the delocalization effect, we employed a similar image processing method used in irradiation corrosion experiments with molten salt [2], and Figure 2 presents the corresponding distribution of corrosion depths. The process involved obtaining areas of cross sections around 1 mm wide, in which the penetration depths were identified along the depth direction. To limit uncertainty resulting from sample preparation and SEM imaging, the obtained penetration depths were normalized by the sample thickness. The data across the areas were then plotted using a cumulative distribution function notation, enabling a comparison between the two regions. The intersection of the two curves at small normalized corrosion depths (near the lead-facing surfaces) indicates that proton irradiation caused the corrosion attack to be shallower but extended over larger areas.

Furthermore, alterations in the wetting behavior of the lead-contacting surface were observed in the irradiation region. The exact mechanism behind this change, whether it is a direct effect of radiation on wetting or an indirect effect resulting from different corrosion modes that lead to surface roughness and subsequent changes in wetting behavior, is yet to be fully understood. Ongoing efforts are dedicated to investigating the changes in corrosion depth, area, and wetting behavior.

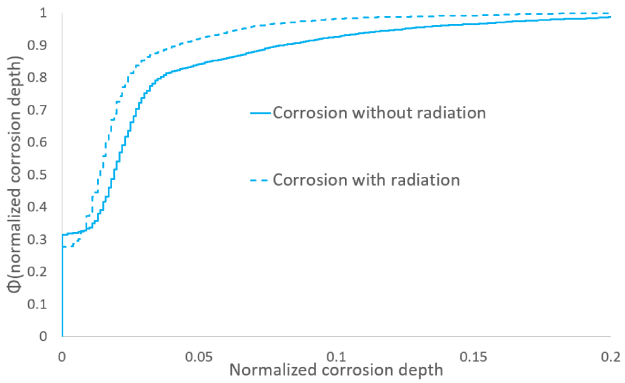


Fig. 2. Corrosion depth distribution illustrated by the cumulative distribution function of the sample shown in Figure 1. The method used to convert the cross-sectional images into depth distribution is based on our previous work on irradiation and corrosion in molten salt [2].

The utilization of this advanced experimental facility marks a critical advancement in addressing the knowledge gaps associated with the combined effects of radiation damage and corrosion in Gen IV coolants. By unraveling the intricate interplay between radiation and corrosion, this research offers a promising pathway towards the development and deployment of corrosion-resistant materials in lead-cooled fast reactors (LFRs). While the preliminary testing conditions may exceed the current design region of LFRs in terms of temperature and oxygen content, understanding material behavior under abnormal conditions is crucial. The efforts presented also lay the foundation for adapting to long-term studies under conditions close to standard operation of LFRs.

REFERENCES

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2. W. Zhou, et al. "Proton irradiation-decelerated intergranular corrosion of Ni-Cr alloys in molten salt." *Nature communications* **11.1**, 3430 (2020).