Review on synergistic damage effect of irradiation and corrosion on reactor structural alloys

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Abstract

The synergistic damage effect of irradiation and corrosion of reactor structural materials has been a prominent research focus. This paper provides a comprehensive review of the synergistic effects on the third- and fourth-generation fission nuclear energy structural materials used in pressurized water reactors and molten salt reactors. The competitive mechanisms of multiple influencing factors, such as the irradiation dose, corrosion type, and environmental temperature, are summarized in this paper. Conceptual approaches are proposed to alleviate the synergistic damage caused by irradiation and corrosion, thereby promoting in-depth research in the future and solving this key challenge for the structural materials used in reactors.

Full Text

Preamble

Review on Synergistic Damage Effects of Irradiation and Corrosion on Reactor Structural Alloys

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Abstract: The synergistic damage effect of irradiation and corrosion on reactor structural materials has emerged as a prominent research focus. This paper provides a comprehensive review of the synergistic effects on structural materials for third- and fourth-generation fission nuclear energy systems, specifically pressurized water reactors and molten salt reactors. We summarize the competitive mechanisms of multiple influencing factors, including irradiation dose, corrosion type, and environmental temperature, and propose conceptual approaches to alleviate synergistic damage caused by irradiation and corrosion. These approaches aim to promote future in-depth research and address this key challenge for reactor structural materials.

Keywords: Irradiation and corrosion; Synergistic effect; Austenitic stainless steels; Nickel-based alloys; Reactors.

1. Introduction

Nuclear energy offers several compelling advantages, including high energy density, cleanliness, and low carbon emissions, making it an effective solution to both fossil fuel depletion and intensifying greenhouse effects. Nuclear power has gained increasing importance in global energy supply, currently accounting for approximately 17% of worldwide electricity production [?]. Ensuring the reliability and safety of nuclear power plant operation has become the foremost consideration worldwide. However, since the 1990s, stress corrosion cracking has been observed in several structural components of the most widely commercialized pressurized water reactors (PWRs) during routine inspections, causing significant concern [?].

According to the United States Institute of Electric Power, 137 stress corrosion cracking events have been recorded in stainless-steel components of PWR main circuits across different countries [?, ?]. The corrosion failure of structural materials in current nuclear power plants has become an increasingly significant issue, posing a major challenge to nuclear power development.

The nuclear power industry has witnessed substantial worldwide growth due to advancements in nuclear technology and increasing energy demand. In the 21st century, extensive considerations and discussions regarding nuclear energy safety have led to the emergence of fourth-generation nuclear reactors that aim to provide cleaner energy with enhanced safety and durability, reduced radioactive waste, improved economics, and higher energy efficiency. However, the operating temperatures and irradiation environments of fourth-generation reactors differ substantially from those of currently operating reactors.

Structural materials face critical challenges when operating at higher temperatures, under stronger neutron irradiation, and in more extreme corrosive environments [?]. The successful development of advanced nuclear energy in fourth-generation reactors hinges upon nuclear structural materials and their transition from experimental reactors to demonstration and commercial applications. Therefore, investigating material behavior under service conditions and comprehending the evolution of material properties in combination with reactor irradiation and high-temperature corrosion environments is of utmost significance.

In addition to possessing excellent corrosion resistance, reactor materials must undergo irradiation testing before deployment. Since the 1960s, researchers have focused on addressing issues such as irradiation hardening, irradiation swelling, and high-temperature helium embrittlement of structural materials [?]. As research progressed, investigations into irradiation-assisted stress corrosion cracking (IASCC) of materials under the combined influence of irradiation and water environment were initiated in PWRs in the early 1980s [?]. The irradiation performance of structural materials, particularly their behavior and properties under operational conditions, has become a key factor affecting reactor lifetime.

The synergistic effect of irradiation and corrosion on materials represents a crucial and universally significant scientific issue in the field of nuclear structural materials research. However, due to the diversity and complexity of influencing factors, unraveling its intricacies remains challenging. To date, numerous experimental and theoretical studies have examined the service behavior of structural materials subjected to irradiation and corrosive environments in PWRs [?]. The rupture of an oxide film under stress represents the first step in stress corrosion cracking (SCC). Therefore, comprehensively understanding the impact of irradiation on material oxidation behavior is essential for accurately predicting IASCC. Furthermore, the effect of irradiation on intergranular and general corrosion of alloys in aqueous environments requires further investigation. For fourth-generation advanced nuclear reactors, the different cooling media raise a widely debated scientific question: does irradiation accelerate or inhibit the corrosion of structural alloys in these unique environments? Studying the evolution laws and mechanisms of material microstructure under combined irradiation and corrosion environments can facilitate better understanding of material performance and failure in nuclear reactor environments, aid in predicting material service life, and inform measures to improve material properties. Given the numerous existing studies, it is essential to comprehensively summarize the synergistic effects of irradiation and corrosion damage on nuclear structural alloys and establish a comprehensive knowledge system for future development and exploration of advanced core structural materials.

This paper provides an overview of reactor environments and materials, summarizes the corresponding corrosion mechanisms, and emphasizes the impact of irradiation on material corrosion, using a PWR and a fourth-generation advanced fission nuclear power molten salt reactor as examples. We present the latest insights into the irradiation-corrosion behavior of structural materials within reactors, outline methods to enhance material corrosion resistance, highlight challenges faced by structural materials in fourth-generation nuclear reactors, and propose strategies for controlling synergistic damage caused by irradiation and corrosion for future applications. These findings aim to enhance our understanding of material failure processes in reactors and expedite the development of new materials.

2.1 Structural Materials of PWRs

In PWRs, austenitic stainless steel and Ni-based alloys are the primary materials used for structural components, in addition to zirconium alloy fuel cladding [?, ?]. Table 1 presents an overview of the main materials used in the primary circuits of PWRs. Austenitic stainless steel (e.g., 304, 304L, 316, 316L) with good corrosion resistance is utilized as in-core structural material. Ni-based alloys, such as 600, 800, and 690, are employed for screws, pins, locating keys, and steam generator tubes in core components due to their remarkable mechanical properties and resistance to intergranular stress corrosion cracking (IGSCC) [?]. Table 2 compares the chemical composition of Ni-based alloys 600, 800, and 690. The Ni-based alloy 690, which has high Cr content, exhibits superior IGSCC resistance and is therefore the preferred material for steam generator tubes in nuclear power plants. Zirconium-based alloys, such as Zircaloy-2, Zircaloy-4, ZIRLO, and M5 alloys, are specifically selected for fuel cladding in PWRs due to their low neutron capture cross-section and excellent corrosion resistance.

2.2 Environment of a PWR

PWRs are the most widely used commercial nuclear power plants, whose core represents an extreme environment composed of high-temperature, high-pressure water and strong radiation fields that affect water chemistry, stress, and the microstructure of core materials [?, ?, ?]. Table 3 lists the water chemistry parameters of the primary coolant water used in PWRs. The temperature range of the water is 280–340 ºC in PWRs. Boric acid is added as a neutron absorber, with acidity offset by lithium hydroxide to maintain a pH value between 6.8 and 7.4. To suppress radiolytic oxidation of water, hydrogen overpressure is introduced by adding 25–50 cc/kg of dissolved hydrogen (DH) in PWRs. Oxygen can be introduced into the primary loop system by adding aerated water, oxygen, or H_2O_2 during plant shutdown processes for PWRs [?, ?]. This stabilizes the oxide, thereby reducing the release of radioactive materials into the coolant.

2.3 Formation of the Oxide Layer

Various corrosion processes of internal components in PWRs during service affect reactor operation [?, ?], including:

- Corrosion of fuel cladding leads to the formation of adherent oxides that affect heat transfer. Hydrogen generated by waterside corrosion that enters zirconium alloy fuel cladding affects the ductility and fracture toughness of the cladding due to precipitation of brittle hydride particles.
- General corrosion of stainless steel and Ni-based alloys. Stainless steel is considered a core component of PWRs, and its major failure mode is IASCC. The practical IASCC threshold for austenitic stainless steel is approximately 3 dpa. No significant degradation in stress corrosion cracking (SCC) resistance is observed when this threshold is not reached.

Ni-based alloys are used in steam generator piping in PWRs and affect heat transfer efficiency by forming deposits. In addition, the diffusion of Ni, Co, and other cations through the oxide leads to their release into the primary water loop. The release of ^{58}Ni into the primary circuit subjected to neutron irradiation increases the overall radioactivity of the circuit in PWRs. The formation of a passivation film on the alloy surface reduces the release of corrosion products such as Ni cations in PWRs.

- Intergranular oxidation of stainless steel and Ni-based alloys. Intergranular stress corrosion cracking (IGSCC) or transgranular stress corrosion cracking (TGSCC) occurs in materials, causing their degradation.

2.3.1 Morphology and Structure of the Oxide Layer

The oxide layer formed in simulated PWR primary water exhibits a double-layered structure. The inner layer is a continuous and dense Cr-rich oxide, while the outer layer is either an Fe-rich oxide (in austenitic stainless steel) or a Ni-rich oxide (in Ni-based alloys). Both layers have spinel structures [**?**]. The thickness of the oxide layer varies from 0.1 to several m. Figure 1 shows a schematic of the oxide layer formed on the surfaces of stainless steel and Ni-based alloys in simulated PWR environments. For austenitic stainless steel, as shown in Table 4, the outer oxide layer is composed of magnetite Fe_3O_4 or iron-nickel spinel oxide, whereas the chemical and crystal structures of the inner oxide layer remain controversial. The inner oxide layer is composed of either a Cr-rich face-centered cubic (fcc) phase [**?**, **?**, **?**, **?**], a hexagonal Cr_2O_3 phase [**?**], or a two-phase mixture [**?**, **?**]. Several studies have observed Ni enrichment at the oxide/alloy interface due to selective oxidation and preferential movement of Cr and Fe from the alloy [**?**, **?**, **?**, **?**]. The Cr content of the alloy determines the thickness of the oxide layer [**?**], as shown in Fig. 2. Generally, the higher the Cr content of the alloy, the thinner the formed oxide layer [**?**]. For Ni-based alloys, the outer oxide layer consists of dispersed nickel ferrite (Ni_1 Fe_2 O_4) and nickel hydroxide. The metastable solid nickel hydroxide and ferric hydroxide formed on the surface could result from precipitation of stable neutral aqueous complexes [**?**]. The inner oxide layer is Fe-rich and contains nickel chromite (Ni_1 Fe Cr_2O_4). As seen in Fig. 1(b), Cr_2O_3 particles are unevenly distributed at the interface between the inner oxide film and the substrate. The number of Cr_2O_3 particles is related to defects on the substrate surface. Some studies have also observed no chromium depletion at the alloy/oxide interface [**?**]. Nevertheless, a Cr-depleted zone was observed beneath the oxide scale, along with a disturbed microstructure of the alloy with small grains and high dislocation density in this Cr-depleted layer [**?**, **?**].

2.3.2 Formation and Growth of the Oxide Layer

The passivation process of metals in solution initiates with the adsorption of anions, particularly OH^-. Previous studies [**?**] have indicated that oxide layer formation on Fe-Cr-Ni alloys such as stainless steel in simulated PWR environments primarily relies on diffusion of alloying elements Fe, Cr, and Ni. The outer oxide layer grows via a dissolution and reprecipitation mechanism, whereas the inner oxide layer grows via a solid growth mechanism [?, ?, ?]. Figure 3 schematically illustrates the oxide formation process on stainless steel exposed to simulated PWR environments. During the initial corrosion stage, when material is exposed to high-temperature and high-pressure water, oxygen in the environment contacts the metal surface directly. Among alloying elements, Cr exhibits the strongest affinity for oxygen to form oxides. Consequently, during the initial stage, a Cr-enriched oxide film gradually develops on the material surface. Due to diffusion rates in oxides following the order Fe > Ni » Cr [?, ?, ?], the comparatively faster diffusion rate of Fe results in its outward diffusion into the aqueous medium or interaction with adsorbed H_2O, eventually precipitating as outer oxide particles. Simultaneously, external oxygen diffuses into the matrix material interior and reacts with Cr to form an inner oxide film. Since the diffusion rate of Ni in the inner oxide film lies between that of Cr and Fe, Ni neither accumulates extensively within the inner layer nor exists in significant quantities within the outer oxide layer. Consequently, the outer oxide layer is predominantly composed of Fe, whereas the inner oxide film is enriched with Cr. Thin Ni enrichment may occur at the metal/oxide interface, depending on the diffusion rate of Ni.

The factors affecting oxide layer growth in simulated PWR environments are summarized in Table 5. Water chemistry parameters (DH [?, ?], DO [?, ?], and pH [?, ?, ?]) are key factors affecting corrosion resistance and oxide properties in PWR primary systems. Among these parameters, DH is favorable for reducing the oxidation rate. In particular, primary water undergoes radiolysis when subjected to irradiation, producing oxidizing species (O_2 + H_2O_2) and making the corrosion potential more positive, which in turn increases the corrosion rate. Irradiation affects material corrosion behavior by changing the water environment, as discussed in detail in Section 2.4.3.

Matthews et al. [?] suggested that the reaction rate at the interface is the principal factor affecting oxide growth, surpassing other contributing factors. Oxide growth depends on the interfacial reaction rate when the same sample is simultaneously exposed to the PWR environment for the same duration. According to the point-defect model proposed by Macdonald et al. [?], in high-temperature and high-pressure water environments, oxygen ion vacancies can be formed at the interface between the inner oxide layer and the base metal through reaction (4) [?, ?]. Metal atoms are converted into metal ions via reactions (4)–(6) [?, ?, ?], where m denotes the metal atom in the base metal (Cr, Ni, and Fe). The oxygen vacancies diffuse to the oxide/matrix interface and combine with metal cations to form the inner oxide layer, resulting in inward migration of the oxide/matrix interface. The oxygen vacancy and cation diffusion rates control the inner oxide growth rate. Further, metal cations formed in the inner oxide layer diffuse into the aqueous medium or interact with absorbed H_2O and eventually precipitate as oxide particles driven by dissolution and redeposition mechanisms [?, ?]. The diffusion rate of cations in the inner oxide layer plays a major role in outer oxide layer growth. Irradiation controls oxide growth by affecting diffusion rates, as described in Section 2.4.

2.4 Effect of Irradiation on Corrosion

Corrosion of core structural materials is affected by irradiation in at least three distinct ways. First, irradiation-induced microstructural changes (formation of dislocation loops, cavities, helium bubbles, and precipitates) in alloys are related to irradiation dose and temperature. These microscale changes influence oxidation kinetics by altering the diffusion rates of oxygen vacancies and metal cations, potentially modifying and expediting oxidation at the oxide/liquid and metal/oxide interfaces. Second, irradiation-induced elemental segregation at grain boundaries, such as depletion of Cr, Fe, and Mo and enrichment of Si, Ni, and P, is believed to be responsible for intergranular oxidation behavior of alloys. Third, irradiation of water causes its decomposition into free radicals and oxidizing substances, which increases the corrosion potential of the aqueous medium. An increased corrosion potential causes dissolution of the Cr-rich inner oxide layer of alloys in high-temperature water, leading to higher corrosion rates.

The general corrosion behavior and intergranular oxidation of irradiated materials have been extensively studied both domestically and internationally [**?**]. However, no consensus exists regarding the effect of irradiation on alloy oxidation. While irradiation aggravates the corrosive environment through water radiolysis and accelerates material corrosion by introducing microstructural damage and microchemical segregation, some experimental studies [**?**] have reported opposite results, suggesting that irradiation inhibits general corrosion behavior and intergranular oxidation due to differences in experimental conditions such as corrosion temperature and time, ion species, and doses. Additionally, no significant difference has been observed in corrosion behavior between irradiated and unirradiated materials [**?**, **?**]. Therefore, this section reviews the role of irradiation on general corrosion behavior and intergranular oxidation from three aspects: radiation damage, radiation-induced segregation, and water environment changes induced by water radiolysis.

The most effective approach for obtaining irradiation-corrosion data involves simulating the irradiated water environment. The autoclave system, capable of operating at temperatures between 280–325 °C and high pressures ranging from 7.1 to 16.2 MPa, has proven successful in simulating the primary circuit. Ion-exchange resins are used to monitor water chemistry in high-pressure sterilizers with recirculation loops. The level of dissolved hydrogen in the medium is assessed by measuring the thermal conductivity of water using an in situ H_2 sensor. Neutron irradiation experiments pose numerous difficulties and limitations for research due to long cycle times, high costs, and high radioactivity. In contrast, ion irradiation offers advantages such as high damage rates, easy parameter adjustment, absence of radioactivity, and cost-effectiveness. Consequently, rapid ion implantation is commonly adopted in laboratory studies to simulate neutron irradiation and facilitate fundamental research endeavors [**?**], thereby establishing radiation damage models and elucidating pertinent mechanisms.

2.4.1 Effect of Irradiation Defects on Corrosion

After irradiation, defects such as voids, helium bubbles, black spots, and dislocation loops are produced inside the material [?, ?]. For comprehensive understanding of the composition and structure of oxides formed on metal surfaces and the depth of inner oxide layers following irradiation, extensive studies have been conducted and summarized in Table 6 [?, ?, ?, ?]. Although Deng et al. [?] reported that a finer oxide grain structure in the inner oxide film formed on 3-dpa proton-irradiated 316L austenitic stainless steel after exposure to simulated PWR primary water for 500 h, it is generally accepted that irradiation does not alter the crystal structure and qualitative chemistry of inner and outer oxides formed on materials, which can be characterized using high-resolution TEM (HRTEM) [?, ?, ?, ?, ?, ?, ?, ?]. Similar to unirradiated samples, the oxide layer maintains its characteristic double-layer structure when stainless steel is irradiated. The outer layer comprises polyhedral oxide particles primarily composed of Fe-rich oxides such as dispersed Fe-Ni spinel or magnetite Fe_3O_4, while the inner continuous and protective oxide film is predominantly composed of Cr-rich oxide known as Fe-Cr-Ni spinel. Both layers have been characterized as spinel structures. Additionally, Ni enrichment at the oxide/matrix interface has been observed [?, ?].

However, available study data [?, ?, ?, ?, ?, ?] indicate that irradiation affects the quantitative chemistry, morphology, and thickness of oxide layers formed on alloys. In irradiated materials, the Cr content in the inner layer has been observed to be high [?, ?, ?]. In contrast, a lower Cr content has been observed in inner oxides of 316L stainless steel irradiated with protons and exposed to high-temperature water, due to dissolution of Cr-rich spinel oxides in irradiated water with added hydrogen [?]. Further, no significant change in Cr concentration in the inner oxide has been reported with increasing irradiation dose [?, ?, ?]. These different oxidation behaviors likely arise from different irradiation and oxidation conditions (dose, temperature, oxidation duration, etc.). In addition to changes in Cr concentration, Kuang et al. [?] observed that the maximum Ni content in the transition zone is higher and wider.

Generally, these irradiation-induced defects accelerate corrosion primarily in two ways [?, ?, ?, ?, ?, ?]: (i) by increasing the size and/or density of outer oxide particles, and (ii) by increasing the depth of the inner oxide layer. The oxidation kinetics model has been employed to elucidate changes in inner oxide layer thickness caused by irradiation-induced defects [?, ?]. Based on the quasi-stationarity assumption [?], the thickness of the oxide scale on nickel-based alloy surfaces in a PWR can be described using Equation (7):

$$X(t) = X_0 + \sqrt{D_{gb} \cdot t}$$

where $X(t)$ represents the thickness of the inner oxide layer, X_0 denotes the thickness of the initial oxide film, t is the oxidation time, D_{gb} is the diffusion coefficient of oxygen vacancies along short-circuit paths (such as grain boundaries in oxides). Only grain boundaries act as channels for oxygen vacancy diffusion in the absence of defects. However, both grain boundaries and irradiation-induced defects contribute to oxygen vacancy diffusion under irradiation. Therefore, D_{gb} is anticipated to increase, indicating a higher diffusion rate of oxygen vacancies and resulting in formation of a thicker inner oxide layer according to Equation (7).

On the other hand, irradiation-induced defects serve as preferred nucleation sites for oxide particles, resulting in a higher number of outer oxide particles. These defects function as fast diffusion channels, facilitating outward diffusion of metal cations. Thus, oxide particles formed on the material surface are larger than those in the unirradiated state. The oxidation kinetics provide insights into oxidation rates. Higher oxidation kinetic values in irradiated regions indicate that irradiation accelerates the corrosion process. For example, Boisson et al. [?] reported a five-fold increase in oxidation kinetics of irradiated 316L austenitic stainless steel compared to unirradiated areas after 24 h of oxidation in simulated PWR water. Similarly, Deng [?] emphasized that radiation enhances oxidation kinetics (up to four times faster) during longer oxidation durations of approximately 500 h in irradiated 304 nuclear-grade stainless steel. Thicker inner oxide layers in irradiated areas indicate faster oxygen diffusion.

However, Perrin et al. [?] conducted an experiment where they irradiated 316L stainless steel with protons and subjected it to corrosion in simulated PWR water at 325 °C for 1024 h. Surprisingly, they found that oxygen diffusion in the inner oxide layer of the irradiated sample occurred at a slow pace. The diffusion coefficients for irradiated and unirradiated samples were 3×10^{-20} cm^2/s and 1×10^{-19} cm^2/s, respectively. The outer oxide particles formed on the irradiated sample were smaller in size, and the inner oxide film was thinner than that of the unirradiated sample, as shown in Fig. 4. These observations indicate that irradiation inhibits corrosion. The growth of the inner oxide film was constrained by slow oxygen diffusion through the inner oxide layer.

Irradiation-induced defects promoted Cr diffusion, resulting in formation of a thicker Cr-rich film. A dense Cr-rich film normally acts as a protective layer that impedes oxygen diffusion and further oxidation. Kuang et al. [?] also suggested that irradiation-induced defects enhance Cr diffusion, resulting in higher protection of the inner oxide layer, which is one reason for enhanced oxidation resistance in irradiated regions. Further, they observed that the continuous Ni-rich zone formed near the oxide/matrix interface reduces the “available space” for oxidation. It should be noted that in the experiment conducted by Perrin et al., the samples were only mechanically polished. Samples in which cold work is induced by polishing [?, ?] tend to exhibit faster oxidation kinetics. However, this cold working diminishes under irradiation [?] due to disappearance of the initial dislocation network. Consequently, the irradiated area without cold work experiences slower oxygen diffusion, resulting in thinner oxides. Therefore, sample polishing must be strictly controlled, and the results obtained by Perrin et al. using mechanically polished samples irradiated with protons (or neutrons) need validation to determine whether polish-induced cold working overcomes the effects of irradiation.

Corrosion of the alloy intensifies with increasing corrosion time, as shown in Fig. 5(a) [?, ?]. Furthermore, the density and/or size of irradiation-induced defects are influenced by irradiation dose or temperature. For a given material, irradiation-induced defect density and/or size increase with increasing irradiation dose (Fig. 5(b)) or elevated irradiation temperature [?, ?, ?]. Consequently, diffusion rates of metal cations and oxygen anions increase, resulting in elevated oxidation rates. It is worth noting that a smoother surface appears in irradiated alloys due to irradiation-induced ion sputtering [?], as shown in Fig. 6(c). During the corrosion process, rough surfaces with large surface areas absorb more dissolved oxygen and impurities, thereby enhancing oxide nucleation rates. In contrast, irradiated alloys exhibit fewer oxide particles. Simultaneously, high-energy particle irradiation causes localized temperature increases on the material surface and formation of Cr-rich oxides (Fig. 6(d)). The thin Cr-rich oxide layer prevents interaction between underlying elements and external oxygen atoms, thus hindering growth and nucleation of iron-rich oxides on the alloy surface during initial corrosion stages (Fig. 6(a)). Additionally, short-term atom diffusion becomes difficult due to defects. However, for long-term corrosion, irradiation-induced defects promote long-range migration of metal atoms, and thus the oxidation rate and corrosion weight gain of irradiated alloys are higher than those of unirradiated alloys (Figs. 6(a) and 6(b)). The defects are principally point defects. Their impact on enhanced diffusion of oxygen and metal atoms during initial corrosion stages is minimal, and material corrosion behavior is almost unaffected by irradiation [?, ?]. Jiao et al. [?] observed that proton-irradiated SUS316 stainless steel corroded for 70 h showed only marginal effects of irradiation on inner oxide film thickness, as shown in Fig. 7. However, no conclusive explanation for this phenomenon was provided. Further empirical research is necessary to investigate the influence of irradiation-induced defects on material corrosion, including exploration of the critical irradiation dose that either promotes, inhibits, or has no effect on material corrosion, as well as the influence and mechanism of irradiation on material corrosion during initial irradiation stages. This will enable a more systematic understanding of irradiation effects on corrosion, which has guiding significance for nuclear material applications in the nuclear industry.

2.4.2 Effect of Radiation-Induced Segregation on Corrosion

Radiation-induced segregation (RIS) refers to microchemical segregation at grain boundaries (GB) due to irradiation, leading to depletion of Cr, Fe, and Mo and enrichment of Si, Ni, and P [?, ?, ?]. Post-irradiation annealing eliminates element segregation at grain boundaries, resulting in no local corrosion of grain boundaries being observed [?]. This indicates that RIS is primarily responsible for intergranular oxidation of materials. Grain boundaries act as preferred diffusion pathways and are therefore more susceptible to oxidation. Intergranular corrosion occurs in both irradiated and unirradiated areas, but deeper oxide penetration in irradiated areas suggests that irradiation intensifies intergranular corrosion [?, ?, ?, ?, ?], which is attributed to Cr depletion at grain boundaries. Lower Cr content at grain boundaries results in increased oxidation penetration depth (Figs. 8 and 9) and decreased intergranular corrosion resistance [?, ?]. Furthermore, unirradiated alloys exhibit Ni-rich regions before the oxide tip at grain boundaries, whereas such regions are not formed in irradiated samples [?]. This observation further supports the notion that irradiation promotes high intergranular corrosion rates. The selective oxidation of Cr and Fe leads to preferential removal of these elements from the alloy, forming Ni-rich regions. In irradiated alloys, rapid corrosion along grain boundaries does not provide sufficient time for selective oxidation to occur [?, ?].

The depth of intergranular oxidation increases with increasing irradiation dose (Fig. 8) [?, ?], possibly due to maximum Cr consumption at grain boundaries at higher doses. Studies [?] have shown that Cr depletion at grain boundaries changes dramatically at low and medium doses (0.1–5 dpa) but no longer changes significantly with further increases in radiation dose, or even remains unchanged at higher doses, as shown in Fig. 9. Fukumura et al. [?] found that the degree of Cr segregation in neutron-irradiated stainless steel exposed to simulated PWR water for 1149 h did not change significantly with dose increases from 25 dpa to 73 dpa, but the oxidation length at grain boundaries increased by approximately 20%, as depicted in Fig. 8(b). It is challenging to quantitatively explain promotion of oxidation at grain boundaries at high irradiation doses through Cr depletion alone. The evolution of Ni and Si segregation at grain boundaries may be implicated in promoting oxidation at these boundaries at high irradiation doses (Fig. 8(b)).

However, the presence of Si segregated at irradiated grain boundaries appears to inhibit intergranular corrosion. Kuang et al. [?] compared intergranular oxidation behavior of solution-annealed (SA) and 2.5 dpa proton-irradiated specimens of 316L in simulated PWR water with 1000 h exposure, finding that the penetration depth of intergranular oxide in the irradiated area was shallower than that in the unirradiated area. This suggests that irradiation enhances intergranular oxidation resistance of stainless steel. Figure 10 shows the intergranular oxidation process at irradiated and unirradiated grain boundaries [?]. Researchers have proposed that Si segregated at grain boundaries of irradiated samples tends to diffuse outward and undergo oxidation due to its high diffusion coefficient and oxygen affinity. The Si-rich oxide at the intergranular oxide tip of irradiated samples serves as a temporary diffusion barrier for oxygen, although it tends to dissolve near the sample surface. The migration efficiency of elements (especially Cr) along grain boundaries of irradiated samples to the oxidation front improves because preferential Si diffusion generates vacancies that promote Cr atom diffusion, leading to increased Cr content at intergranular oxide tips. Irradiation-induced structural defects also contribute to element diffusion such as Cr along grain boundaries. Therefore, although Cr in grain boundaries of irradiated samples is depleted due to RIS at original grain boundaries, Cr is enriched in oxides along grain boundaries of irradiated samples. The co-enrichment of Si and Cr at intergranular oxide tips enhances oxidation resistance, ultimately decreasing the oxidation rate of irradiated grain boundaries. In addition, grain boundary structure affects RIS [**?**, **?**] and intergranular corrosion [**?**]. Therefore, future experimental analyses should focus on selecting the same type of grain boundary across irradiated and unirradiated areas to enable direct comparative studies of intergranular oxidation behavior between the two areas at a single grain boundary, eliminating interference caused by grain boundary structure differences. This is crucial for distinguishing the effects of irradiation on intergranular oxidation.

In summary, irradiation segregation leads to Cr depletion and Ni and Si enrichment at grain boundaries, but its effect on intergranular oxidation of materials remains unclear. Additionally, the impact of RIS on intergranular corrosion has mostly been investigated at low radiation doses, with a lack of research at high radiation doses. Future studies should aim to strengthen understanding of intergranular corrosion mechanisms at high radiation doses to bridge this research gap.

2.4.3 Effect of Irradiation Radiolysis on Corrosion

Irradiation can directly affect material corrosion through irradiation damage, as described in detail in Sections 2.1 and 2.2. Furthermore, radiolysis can affect corrosive media. In PWRs, neutrons interact with water coolant, leading to radiolysis (Equations (4) and (5)). This process results in formation of long-lived molecules (H_2, O_2, and H_2O_2) and short-lived free radical species (OH, HO_2, H, and H_2O_2) that can undergo oxidation or reduction. Literature [**?**, **?**] provides detailed reviews of important reactions associated with these species and explains charge conservation of H^+ and OH^-. The water radiolysis product H_2O_2 affects the electrochemical corrosion potential [**?**, **?**, **?**]. Figure 11 [**?**] shows the change in corrosion potential of 304 and 316 stainless steels as a function of H_2O_2 concentration. As H_2O_2 concentration increases, the corrosion potential also increases until reaching a saturation point. The saturated corrosion potential can extend into the penetration zone of the passivated film, where cracking/dissolution of the oxide film occurs [**?**, **?**]. When subjected to irradiation, the corrosion potential increases significantly. Was et al. [**?**, **?**] performed thermodynamic calculations and radiolysis modeling, finding that at very high irradiation dose rates (4000 kGy/s), the corrosion potential increased by ~0.8 VSHE compared to unirradiated samples, thereby promoting material corrosion. Additionally, in high-temperature water with low dissolved oxygen concentration and no dissolved hydrogen, the corrosion potential increased by more than 0.25 VSHE.

The characteristics of oxides formed on irradiated material surfaces provide insight into alteration of species stability within the oxide film due to the irradiated environment. In experiments, materials were irradiated with a proton beam and simultaneously exposed to simulated PWR water. Although radiolysis and irradiation damage occur on material surfaces simultaneously, in a series of experiments conducted by Was et al. [?, ?, ?], stainless steel samples not directly irradiated but exposed to radiolyzed water exhibited oxide characteristics similar to those of irradiated samples. Hematite was observed on the oxide surface, and Cr was deficient in the inner oxide layer, differing from oxide characteristics of unirradiated samples. This indicated that water radiolysis was the primary mechanism affecting oxide properties and formation. Comparing oxides in irradiated and unirradiated areas, such as the effect of irradiation-induced defects on corrosion, irradiation did not alter the double-layer structure of oxides formed on material surfaces, which still consisted of an outer layer of oxide particles and an inner layer of continuous oxide film. In situ irradiation-corrosion experiments demonstrated significant changes in morphology, composition, and depth of the inner oxide film compared to unirradiated samples [?, ?]. Using austenitic stainless steel as an example, the following observations were made: (i) Oxides formed on irradiated sample surfaces predominantly consisted of hematite with relatively few large planar oxide particles, primarily comprising densely packed small equiaxed crystals; (ii) Irradiated samples displayed a thinner and more porous inner oxide film with lower Cr content, indicating Cr loss from the inner oxide layer after irradiation; (iii) The interface between outer particles and inner oxide was indistinct, and the metal/oxide interface fluctuated significantly in irradiated samples.

As mentioned earlier, long-lived radiolysis products (H_2O_2) generated by irradiation can significantly increase the corrosion potential. This alteration in potential leads to oxidation of Fe^{2+} to Fe^{3+} and subsequent formation of hematite. Cr-rich spinel oxide dissolves due to increased corrosion potential. Consequently, the thickness of the inner oxide layer decreases and Cr loss occurs. Lower Cr content indicates a less protective oxide film, leading to porosity. A higher diffusion rate of oxygen vacancies and cations due to poorer protection by the inner scale results in further corrosion, contradicting the theory that the inner oxide layer is thinner. Hence, growth of the inner oxide film on irradiated alloy in high-temperature and high-pressure water can be characterized as a competing process of solid oxidation and dissolution of the existing film.

3.1 Molten Salt Reactor Environment

In a molten salt reactor (MSR), the operating temperature is usually higher than 700 °C and liquid fluoride serves as both coolant and nuclear fuel carrier. This means structural materials are in direct contact with molten fluoride salt. Consequently, structural components face multiple harsh environmental challenges such as high temperatures, molten salt corrosion, and high-dose neutron irradiation [?]. Corrosion of the UNS N10003 alloy in pure molten salt is chiefly attributed to selective dissolution of the active metal component, chromium (Cr) [?]. Additionally, the complex chemical forms of lanthanides and actinides in fuel salts accelerate alloy performance degradation and affect reactor safety [?]. The fission product Te diffuses into the UNS N10003 alloy, leading to alloy embrittlement under actual in-reactor service conditions [?, ?]. The extent of alloy embrittlement failure intensifies with increasing reactor power and Te concentrations [?].

During MSR operation, the chemical form of uranium is extremely complex, particularly the interaction between U^{4+}/U^{3+} and specific alloy components through the reaction $2UF_4 + Cr = CrF_2 + 2UF_3$, which affects the corrosion process [?].

3.2 Corrosion Effect of Molten Salt

Molten fluoride salt is an ideal coolant and nuclear fuel carrier in MSRs due to its favorable characteristics, including small neutron absorption cross-section, high-temperature stability, high thermal conductivity, high specific heat capacity, high boiling point, and low saturated vapor pressure. Ni-based and Fe-based alloys are considered potential structural materials for molten fluoride salt reactors. Typically, these alloys undergo selective dissolution of active alloying elements at the alloy-molten salt interface. Unlike other high-temperature corrosive environments (such as supercritical water, air, and molten alloys), alloys in molten salt systems cannot easily form protective passive films on their surfaces to inhibit corrosion, primarily because oxides are unstable in molten salt environments. Thus, the corrosion mechanism of alloys in molten salt environments can be microscopically manifested as selective dissolution of active elements, mainly Cr in Ni- and Fe-based alloys. Cr dissolution leads to formation of vacancy defects near the interface and causes internal Cr to diffuse toward the surface. This corrosion-induced Cr loss results in alloy mass reduction (corrosion weight loss). Furthermore, accumulation of vacancy defects caused by corrosion leads to formation of corrosion pits near the alloy surface [?]. As corrosion time increases, both the size and number of corrosion pits progressively increase.

In the 1950s, Oak Ridge National Laboratory (ORNL) proposed using molten fluoride salt as a fuel system. Corrosion tests conducted on 18 different metallic materials in 75% NaF-25% UF_4 molten salt revealed that Ni alloys exhibited better resistance to molten salt corrosion compared to stainless steels [?]. Subsequently, compatibility with fluoride salts became a crucial criterion for selecting structural materials. From 1951 to 1952, ORNL conducted multiple static corrosion tests on various metallic materials, which demonstrated that loops made of stainless steel materials would become blocked, leading to identification of Ni-based Inconel 600 alloy as a candidate material [?]. However, Inconel 600 also had the drawback of low strength. Hastelloy B, a Ni-Mo alloy with low chromium content, exhibited excellent strength and corrosion resistance against molten salt but posed challenges related to processing, oxidation, and stability [?]. Therefore, ORNL attempted to develop a new alloy based on the Hastelloy

B alloying system.

In a 1957 technical report (ORNL-2274), ORNL clearly pointed out that INOR-8 (i.e., UNS N10003 alloy, a 70wt% Ni-16wt% Mo-7wt% Cr solution-strengthened Ni-based superalloy) met all design requirements. The UNS N10003 alloy was used as the main structural material in the Molten Salt Reactor Experiment (MSRE), which achieved criticality in 1965 and was decommissioned in 1969, making it the only alloy successfully employed in an MSR [**?**, **?**].

Under ideal conditions, the UNS N10003 alloy exhibits general corrosion behavior. In the presence of moisture or oxidation impurities in molten salt, the alloy is more prone to intergranular corrosion. The corrosion pattern observed for molten salt is also related to alloy Cr content. Alloys with high Cr content are more susceptible to intergranular corrosion than general corrosion, and Cr dissolution is more pronounced. For example, corrosion pits in 316SS (with 18% Cr content) and Inconel 617 (with 22% Cr content) can become interconnected and penetrate through the material [**?**, **?**]. For low-Cr alloys such as UNS N10003 (7% Cr content), corrosion pits generally remain isolated from one another, except in severe molten salt environments [**?**].

3.3.1 Thermodynamic Driving Force for Corrosion

Coolant salts used in MSRs typically consist of mixtures of alkali metal or alkaline earth metal fluorides such as LiF-NaF-KF or LiF-BeF_2. The driving force for molten salt corrosion originates from differences in Gibbs free energy between molten fluoride salt and metal fluoride. It is widely known that alkali metal (or alkaline earth metal) fluorides have the lowest Gibbs free energies among metal fluorides. Consequently, alloying elements such as Cr, Fe, and Ni cannot be chemically replaced through reactions in alkali metal (or alkaline earth metal) fluorides. Thermodynamic calculations indicate that equilibrium concentrations of metals such as Ni, Mo, Co, Fe, and Cr in corrosion reactions involving LiF, NaF, KF, and BeF_2 are extremely low [**?**]. Generally, when equilibrium concentration of reaction products is very low, the reaction can be considered not to occur. Therefore, intrinsic corrosion of alloys containing these major elements in impurity-free molten salt coolants is almost impossible. First-principles simulations and molecular dynamics simulations have been used to investigate thermodynamic properties of molten salts, revealing that molten salt composition plays an important role in corrosion of Ni-based alloys in molten fluoride salts [**?**]. These studies help efficiently understand the basic fluorine-induced initial corrosion mechanism of Ni-based alloys in molten salt environments.

3.3.2 Impurity-Driven Corrosion

Impurities present in molten salt are the main drivers of corrosion, especially in initial stages, as shown in Fig. 12. Oxidizing impurities (H_2O, HF, NiF_2, FeF_2, metal oxides, oxyacid ions, etc.) can act as oxidants in molten salts, leading to rapid alloy corrosion through various reaction mechanisms (Equations (6)–(10)) [?, ?]. However, as these impurities are consumed, the corrosion rate decreases, transitioning to a slower corrosion stage. In the complex environment of nuclear reactors, alloy corrosion in molten salt environments is affected by multiple factors, including temperature differences [?, ?], variations in molten salt flow rate [?], presence of fission products [?, ?], heterogeneous material corrosion (galvanic corrosion), and irradiation-induced corrosion. This paper focuses on the effect of irradiation on alloy corrosion in molten salt environments and discusses the latest research progress.

3.4 Effect of Irradiation on Molten Salt Corrosion

In nuclear reactors, irradiation damage can lead to atomic redistribution and defect formation in solids [?]. Compared to high-energy incident particles, the binding energy between lattice atoms is small (~10–60 eV). Thus, incident particles can easily strike atoms (primary knock-on atoms, PKA) and displace them from lattice positions [?]. When an elastic collision occurs, the primary colliding atom continues to collide elastically with surrounding secondary atoms, creating a displacement cascade and eventually forming a Frenkel pair (interstitial atom and vacancy pair) [?]. With increasing irradiation time, interstitial atoms and vacancies accumulate and evolve into microscopic defects such as dislocation loops, cavities, bubbles, and stacking faults [?].

From a microstructural perspective, both irradiation and corrosion processes involve atom movement and diffusion within alloys. Irradiation damage enhances atom migration and diffusion by introducing defects such as helium and vacancies within the alloy. These defects can be captured by interfaces, particularly grain boundaries, leading to formation of helium bubbles or cavities and weakening of interface properties [?]. Since interfaces in alloys are important diffusion channels for elements [?], irradiation significantly affects alloy corrosion. However, the synergistic effect of irradiation and molten salt corrosion on alloys remains a topic of debate. Previous tests on UNS N10003 alloy in the MSRE program focused primarily on assessing MSR feasibility and alloy usability [?]. Although mechanical properties of UNS N10003 alloy under working conditions have been investigated, few reports exist on microstructural changes after neutron irradiation. Consequently, the mechanism underlying the synergistic effect of irradiation and molten salt corrosion on alloys remains unclear. However, in recent years, within the framework of fourth-generation nuclear reactors, such studies have gained wide attention. Ion irradiation techniques have been used to simulate neutron irradiation effects [?, ?] and shed light on alloy damage mechanisms in irradiated and corroded environments.

3.4.1 Helium Bubbles A critical problem for Ni-based alloys in MSRs is helium atom accumulation [?, ?]. The ^{59}Ni nuclide in Ni-based alloys has a very high (n, α) reaction cross-section, leading to generation of large amounts of helium. Another important helium source is the (n, α) reaction of thermal neutrons with ^{10}B impurities in the alloy during early reactor operation. These He atoms can easily gather around grain boundaries to form He bubbles or cavities, causing intergranular fracture. This reduces alloy toughness significantly and results in helium embrittlement [?]. Therefore, helium irradiation experiments were conducted on UNS N10003 alloy in molten salt [?]. Results showed that helium irradiation accelerated environmental corrosion of UNS N10003 alloy at certain doses, at which obvious helium bubbles were observed via TEM characterization [?]. The presence of helium bubbles accelerated UNS N10003 alloy corrosion in molten salt environments through: (1) accelerating dissolution of alloying elements [?, ?, ?, ?], (2) coarsening alloy surface and enhancing intergranular corrosion [?], and (3) accelerating hole formation on the alloy surface [?, ?].

An important mechanism by which helium bubbles accelerate molten salt corrosion is migration and diffusion of helium bubbles. Zhu et al. observed that helium bubble size increased from 0.8 nm to 20 nm after alloy immersion in molten salt. Migration of helium bubbles toward the sample interior bulk was observed, which coarsened holes on the alloy surface and enhanced intergranular corrosion of UNS N10003 alloy [?]. Liu et al. reported growth and coalescence of helium bubbles around the peak damaged region, which had no significant effect on Cr loss depth [?]. Lei et al. observed unique outward migration of helium bubbles in helium-ion-irradiated UNS N10003 alloy in a high-temperature molten salt environment, as shown in Fig. 13. Helium bubbles can modify surface morphology and form holes through their outward migration. Hole formation on the surface increases contact area between molten salt and alloy, thus increasing Cr dissolution [?]. Dissolution of alloying elements also creates vacancies for helium bubble growth.

3.4.2 Displacement Damage Defects Fast neutrons in reactors cause displacement damage to alloys through cascade collisions and form irradiation-induced defect clusters inside the alloy. These defect clusters strongly impede dislocation motion, resulting in reduced uniform elongation and fracture toughness and inducing hardening and embrittlement. Evidence also indicates that displacement damage can influence alloy corrosion in molten salt environments.

Researchers from the Massachusetts Institute of Technology reported that irradiation can decelerate intergranular corrosion of Ni-Cr alloys in molten salt via in situ proton beam irradiation-molten salt corrosion experiments [?]. They attributed this inhibition to self-healing of grain boundaries caused by diffusion of Ni and Cr interstitial particles from proton irradiation. Irradiation introduces abundant interstitials into alloys and enhances bulk diffusion. Diffusion of irradiation-induced interstitials to grain boundaries supplies dissolving atoms to the molten salt, thus decelerating intergranular corrosion of Ni-Cr alloys, as shown in Fig. 14. ORNL conducted neutron irradiation-molten salt corrosion experiments on UNS N10003 alloy and 316SS alloy at 800 °C with an irradiation dose of 7.5$×10^{16}$ n/cm^2 [?]. The molten salt composition was NaCl-

$MgCl_2$. Results showed that neutron irradiation did not significantly affect UNS N10003 alloy corrosion but decelerated 316SS alloy corrosion.

Helium ion irradiation can introduce helium bubbles into alloys while causing displacement damage. Researchers from the Shanghai Institute of Applied Physics (SINAP) investigated effects of low and high fluence helium ion irradiation on corrosion behavior of Inconel 617 and GH3535 alloys in FLiNaK molten salt at 700 °C [?]. For substrates of both alloys, corrosion accelerated with increasing irradiation doses. For grain boundaries of 617 alloy, inhibition and promotion of intergranular corrosion were observed at low and high irradiation doses, respectively, which could be ascribed to blocking of interstitial atoms, promotion of molten salt diffusion by helium bubbles, and self-healing mechanisms, as shown in Fig. 15.

3.4.3 Radiation-Induced Segregation (RIS) Particle irradiation can induce element segregation and local microstructural changes in materials, defined as RIS. During irradiation, point defects (vacancies and interstitial atoms) diffuse into nearby traps such as surfaces or internal grain boundaries. Generally, migration rates of these point defects differ. Some migrate to sinks, while others remain away from them. Therefore, RIS causes significant changes in local material composition and affects macroscopic properties.

RIS and precipitation are common in PWR pressure vessel steel. At low doses, Cu segregation leading to precipitation can be observed by small-angle neutron scattering and scanning TEM (STEM). Additionally, segregation of Ni, Mn, P, Si, and other elements at dislocations or grain boundaries has been reported [?]. In Cr-containing austenitic stainless steel and nickel-based alloys, irradiation-induced segregation leads to Cr diffusion to grain boundaries, causing IASCC. At lower temperatures, defect concentration increases, but they migrate to sinks, leading to point-defect recombination. At higher temperatures, thermal diffusion is the dominant mechanism, and material composition tends to be uniform. In the intermediate temperature range (0.3–0.6 T), RIS is most severe due to the "inverse Kirkendall effect." When irradiation dose reaches 1–10 dpa, RIS can lead to local corrosion, grain boundary embrittlement, and decline in mechanical properties. When irradiation dose reaches 10 dpa, RIS leads to phase instability, causing generation of new phases or decomposition of existing phases [?]. In the Ni-Si system, when Si solubility in Ni reaches a certain degree, a new Ni_3Si phase forms (γ' phase) [?]. RIS phenomena have also been observed in alloys used for MSRs. Liu et al. used 30 keV Ar ions to irradiate UNS N10003 alloy at room temperature. When dose reached 48.4 dpa, high Ni concentration was observed at grain boundaries [?]. Huang et al. found that precipitates rich in Mo and Cr appeared in UNS N10003 alloy when dose reached 10 dpa after Xe ion irradiation at 650 °C [?].

In a helium-ion-irradiated and molten-salt-corroded UNS N10003 alloy, Zhu et al. observed significant Si segregation at helium bubble surfaces, as shown in Fig. 16. When Si segregation increased to values above the solubility limit at bubble surfaces [?], precipitation of Ni-Si intermetallic compounds occurred. Si segregation promotes chemical reactions between Si atoms and molten salt, enhancing localized corrosion damage. However, formation of Ni-Si precipitates at large bubbles results in galvanic corrosion due to electrochemical potential differences between the Ni matrix and Ni-Si precipitates. Thus, RIS accelerates localized corrosion damage of Ni-based alloys exposed to high-temperature molten salts.

3.4.4 Effects of the Irradiation Environment In MSRs, fission products can significantly affect corrosion of structural materials. Accumulation of fission products in molten salt, resulting from fission reactions in the core region, can interfere with the corrosion process and potentially promote corrosion due to diffusion of specific fission products into the alloy. One example is deposition and diffusion of the fission product tellurium (Te) into UNS N10003 alloy, which can lead to grain boundary embrittlement and potentially induce stress corrosion cracking. The extent of grain boundary embrittlement caused by Te depends on Te diffusion rate in the alloy. Additionally, occurrence of Te-induced stress corrosion cracking at grain boundaries is influenced by Te concentration and applied stress load [?]. These observations highlight the influence of Te concentration and applied stress on alloy susceptibility to Te-induced stress corrosion cracking in a molten environment. Understanding these factors is crucial for designing corrosion-resistant structural materials and optimizing MSR operating conditions.

4. Methods for Inhibiting Irradiation-Assisted Corrosion

Through comprehensive review of recent studies on irradiation and corrosion effects on nuclear structural materials, it is evident that irradiation effects on corrosion can be categorized into two primary aspects. First, irradiation-induced damage alters alloy microstructure by introducing Frenkel pairs, impurity atoms, dislocations, voids, helium bubbles, and solute atom segregation. These changes can either promote or suppress alloy corrosion. Second, irradiation affects corrosion of structural materials by altering water or molten salt environments. In PWRs, water radiolysis produces oxidizing species, thereby accelerating corrosion. In MSRs, the fission product Te introduced by neutron irradiation is a significant factor influencing alloy corrosion. When Te in fuel salt diffuses to the alloy surface, it forms tellurides, leading to intergranular cracking on the shallow surface of Hastelloy N alloy. ORNL modified Hastelloy N alloy by adding Nb and Ti to address Te-induced intergranular cracking. Research has shown that Hastelloy N alloy modified with 1–2% Nb exhibits favorable resistance to irradiation embrittlement and Te-induced intergranular cracking [?]. Nb addition improves creep strength and fracture strain while mitigating embrittlement. When Nb or Ti is added to metal alloys, intergranular carbides form that can capture helium atoms and impede their diffusion along grain boundaries. Therefore, it is crucial to limit Ti and Nb addition to prevent Ni_3(Ti, Nb) phase formation. However, when Ti is added, the effects of Cr and

Nb are not significant [?].

Various strategies have been explored to enhance irradiation and corrosion resistance of structural materials in PWRs and MSRs.

4.1 Surface Treatment for Irradiation-Resistant Alloys to Improve Corrosion Resistance

Material degradation processes such as corrosion, stress corrosion, wear, and fatigue cracking frequently initiate at material surfaces. Surface modification technologies, which involve altering chemical composition and material microstructure near the surface (typically ranging from one µm to several tens of µm) or applying protective coatings on metal surfaces to mitigate corrosion of structural materials, have garnered widespread attention in materials science. These protective layers can be fabricated by chemical or electrochemical processes. However, it is essential for coatings to possess good adhesion, uniformity, and coverage to provide effective protection to structural materials. Additionally, the presence of cracks resulting from differences in thermal expansion coefficients between materials must be considered, as even small cracks can lead to material corrosion.

Section 2.3 indicates that stainless steel has excellent corrosion resistance, mainly due to formation of dense and continuous internal Cr-rich oxide films such as Cr_2O_3. Cr is widely recognized as highly effective in safeguarding stainless steel and nickel-based alloys against corrosion in PWRs. Table 8 summarizes advantages and disadvantages associated with several surface-coating technologies for obtaining Cr-rich coatings on metal surfaces to improve material corrosion resistance. In conventional electroplating processes, hexavalent Cr electrodeposition has been employed to deposit Cr-rich coatings onto metal surfaces. However, this deposition process generates highly toxic waste that poses significant environmental hazards [?, ?]. The thermal spraying process [?, ?] is economical and simple, but coatings produced with defects such as interlayers, high porosity, and low adhesion are not widely used for preparing complex structural coatings suitable for extreme environments. Laser cladding technology has been widely used in industries owing to advantages of high efficiency, low porosity, and good adhesion. However, many defects exist in coatings prepared by laser cladding. Therefore, to obtain perfect coatings using laser cladding, process parameters must be adjusted repeatedly. Gu et al. [?] investigated morphology, elemental distribution, electrochemical behavior, and oxidation resistance of 316L stainless steel with Cr-rich coating prepared using laser cladding in simulated PWR environments. Their findings revealed that the Cr-rich coating substantially improved corrosion resistance of 316L stainless steel, with the best performance achieved by the Cr coating produced at 220 W.

In fluoride and chloride salt environments, weight loss of structural materials caused by corrosion is also related to alloy carbon content. Carbide phases containing Cr formed at grain boundaries provide favorable pathways for Cr diffusion and dissolution into molten salt. Although it is known that Cr is preferentially corroded in fluoride salts, relatively high Cr and C contents are essential in traditional high-temperature alloys to maintain good high-temperature creep fracture strength and external oxidation resistance. Due to excellent resistance to molten salt corrosion of pure nickel, Luke Olson et al. utilized 800H alloy as substrate and applied Ni-plating treatment with thickness ranging from 40 m to 180 m [?], as shown in Fig. 17. Their experimental results indicated that Ni plating significantly improved corrosion resistance of 800H alloy in molten fluoride salts. During corrosion, Cr diffused from the alloy into the Ni-plated layer. However, if the Ni-plated layer was sufficiently thin, Cr eventually dissolved into the molten salt. The microstructure of Ni-coating after corrosion, particularly void formation, was also related to coating thickness. The protective barrier effect of Ni-plating was further enhanced by introducing a Cr_2O_3 barrier on the alloy surface before Ni-plating.

4.2 Optimizing Corrosion-Resistant Alloys for Irradiation Resistance

Although the effect of irradiation on stainless steel or Ni-based alloys prepared by traditional processing remains debated, most studies indicate that irradiation accelerates material corrosion [?, ?, ?, ?]. Material properties inevitably degrade over long-term irradiation-corrosion under actual operational conditions. Consequently, significant interest exists in the nuclear energy field for developing and fabricating novel structural materials with superior radiation resistance to enhance resistance to radiation-induced corrosion. Research and development efforts in radiation-resistant materials primarily fall into two categories: (1) High-entropy alloys (HEAs) with high lattice distortion and complex compositions can effectively alter formation energy, migration barrier, and diffusion path of irradiation-induced defects. This modulation enables regulation of defect formation and interstitial-vacancy recombination during early irradiation stages [?]. (2) Nano-fine crystalline materials can be obtained or nano-precipitated particles can be introduced through thermo-mechanical processing and powder metallurgy, such as nano-layered metal composites [?, ?] and oxide dispersion-strengthened (ODS) steel [?, ?]. Additionally, additive manufacturing technology allows simultaneous generation of submicron-scale (approximately 0.5 m) cellular sub-crystalline structures and dispersed precipitates [?, ?]. Oxide-matrix interfaces and cellular subgrain boundaries serve as defect sinks that absorb large numbers of point defects (interstitial atoms and vacancies) generated during irradiation, thus improving material radiation resistance.

HEAs have attracted significant attention as potential structural materials for advanced nuclear energy systems [?]. To date, no studies have directly compared irradiation and corrosion behavior of HEAs with other alloys. However, an interesting observation regarding internal oxide film depths formed on HEA surfaces has emerged. Specifically, depths of these films either remain unchanged or decrease following irradiation. Zhang et al. [?] first investigated irradiation-corrosion behavior of two novel HEAs, $Mo_{0 \cdot 5}NbTiVCr_{0 \cdot 25}$ and $Mo_{0 \cdot 5}NbTiVZr_{0 \cdot 25}$, by subjecting them to He ion irradiation followed by exposure to simulated PWR water for 200 h. Their results showed that irradiation up to 10.5 dpa had minimal impact on composition, structure, and thickness of the oxide film formed on $Mo_{0 \cdot 5}NbTiVCr_{0 \cdot 25}$ HEA surface. This phenomenon was primarily attributed to excellent radiation resistance of HEA. At 10.5 dpa, sizes of dislocation loops and helium bubbles formed were small, exerting limited influence on diffusion of oxygen and metal atoms. Furthermore, presence of Ti, Nb, and Cr in the alloy promoted formation of a highly protective oxide film. The thickness of the oxide film formed on $Mo_{0 \cdot 5}NbTiVZr_{0 \cdot 25}$ HEA surface decreased after irradiation. A similar reduction in irradiation corrosion was also observed in Zr-Nb alloys containing 1–2.5% Nb, where Nb plays a role in reducing irradiation corrosion rate.

Nanocrystalline materials, ODS steel, and alloys prepared by additive manufacturing technology exhibit finer grain sizes and increased grain boundary density, which accelerates Cr diffusion. This leads to formation of a thick Cr-rich film during early oxidation stages. This dense Cr-rich film acts as a protective layer, effectively inhibiting further oxidation. Liu et al. [?] compared irradiation-corrosion behavior of traditionally rolled 304L stainless steel and 304L stainless steel formed by selective laser melting (SLM). The SLM 304L exhibited relatively smaller oxide particles and a significantly thinner inner oxide layer (116 ± 7.57 nm for as-built sample, 166 ± 14.21 nm for solution-annealed sample) compared to traditionally rolled 304L stainless steel (348 ± 22.35 nm) after irradiation. Thus, SLM 304L stainless steel demonstrated higher irradiation-corrosion resistance when subjected to simulated PWR primary water. This can be attributed to fewer irradiation-induced defects and the influence of its unique microstructure, which effectively mitigated corrosion. Additionally, corrosion resistance of ODS steel was observed to be influenced by alloying elements and their contents. The appropriate combination of (14–16)% Cr and (3.5–4.5)% Al can efficiently improve corrosion resistance of ODS steel in high-temperature and high-pressure water. Ren et al. [?] studied corrosion of 16Cr-3Al ODS steel in supercritical water. After 1000 h of corrosion, corrosion weight gain of 16Cr-3Al ODS steel was substantially lower than that of 9Cr ODS steel and stainless steel. Formation of dense Al_2O_3 was the main reason for decreased corrosion rate of 16Cr-3Al ODS steel.

Carbide/oxide dispersion-strengthened (CDS/ODS) Ni-based alloys incorporating nanoparticles using powder metallurgy technology have been successfully developed [?]. Compared to SiC nanoparticles, Y_2O_3 nanoparticles exhibit smaller size, greater uniformity, and higher stability at high temperatures due to dissolution and reprecipitation processes. These characteristics provide a theoretical foundation for further improving mechanical properties and resistance to He-induced damage. However, presence of oxides can decrease alloy resistance to molten salt corrosion. To verify this, a comprehensive study examined mechanical properties, irradiation performance, and molten salt corrosion resistance of ODS Ni-based alloys [?], as shown in Fig. 18. Results showed that compared to

Hastelloy N alloy, ODS Ni-based alloy has better tensile strength, yield strength, and resistance to swelling caused by helium bubbles. Additionally, the alloy exhibits excellent resistance to molten salt corrosion, highlighting its significant potential as a core component material in MSRs.

4.3 Adjusting Corrosion Potential of Water and Molten Salt to Slow Alloy Corrosion

The two dominant methods for controlling corrosion in PWRs are Zn injection and dissolved hydrogen optimization. Zn injection was applied to PWR primary coolant systems in the US in the 1990s [?] to reduce irradiation dose. Zn injection promotes formation of thinner and more protective oxides on stainless steel and alloy 600, with Zn substitution in spinel-type oxide forming stable $ZnCr_2O_4$ compound [?]. Furthermore, Zn injection has been found to alleviate primary water stress corrosion cracking (PWSCC) degradation in PWRs. This can be attributed to Zn inhibiting crack initiation [?] and reducing crack growth rate (CGR) [?, ?]. The main reason for PWSCC inhibition is that Zn injection delays stress corrosion crack initiation. Extensive laboratory data supports a 2–10 times reduction in crack initiation and approximately 1.5 times reduction in CGR [?].

In PWR primary water, oxidant generation and corrosion potential shift due to radiation-induced water radiolysis lead to increased corrosion rates, as described in Section 2.4.3. Hydrogen gas is injected into the water supply system to prevent accumulation of oxidant products such as oxygen and hydrogen peroxide and to reduce alloy corrosion potential, thus efficiently suppressing water radiolysis. H_2 recombines with O_2, H_2O_2, and OH and transforms them back into water through chain reactions [?]. Some studies [?, ?, ?] have provided evidence supporting the observation that corrosion potential and corrosion rate of Ni-based alloys decrease as dissolved hydrogen concentration increases in simulated PWR primary water. Fig. 19 illustrates oxide film thickness in simulated primary water with various DH concentrations. Additionally, Cr concentration increases while Ni concentration decreases with higher DH levels. DH weakens stability and protection of Ni spinel oxide films, and susceptibility of Ni-based alloys to PWSCC reaches its peak near the Ni/NiO phase transition. To effectively delay crack initiation time and reduce PWSCC susceptibility, it is crucial to maintain low DH concentration within the stable NiO region.

Only limited research [?, ?, ?] has been conducted on DH concentration effects on stainless steel corrosion in high-temperature water. In Takumi et al.'s study, increasing DH slightly elevated corrosion rate [?]. However, Dong et al. [?] drew the opposite conclusion, finding that oxide films on 316L SS were thinner in hydrogenated PWR water than in deuterated PWR water. Investigations are thus necessary to optimize hydrogen content and increase corrosion resistance.

Addition of redox agents to molten salt can affect salt corrosion potential, thereby decreasing alloy corrosion rate. Corrosion is an irreversible electrochemical process that occurs until equilibrium is reached. Therefore, from a dynamic perspective, it is important to consider redox control effects on metal corrosion. Most oxidants in salt are consumed by redox reactions. In other words, the reducing agent acts as a sacrificial material protecting the metal. Redox control can be achieved through gas-phase control (e.g., HF/H_2), major metal control (e.g., Be(II)/Be), and dissolved salt control (e.g., U(IV)/U(III)) [**?**].

ORNL research studied stainless steel corrosion rate in FLiBe salts with and without beryllium metal. Without beryllium metal addition, 316 SS corrosion rate was approximately 8 m/year after 3000 h of corrosion. Weight loss initially decreased rapidly within the first 1000 h, possibly due to salt impurities, with subsequent corrosion driven by thermal gradients. In contrast, corrosion rate significantly decreased to less than 2 m/year when beryllium was added to the molten salt [**?**].

UF_4 content not only influences salt corrosion potential but also affects molten salt structure. Bessada et al. reported that molten salt structure can be rearranged by adding a few moles of UF_4 to the mixture to increase molten salt basicity, indicating that changing molten salt structure would suppress corrosion [**?**]. In addition to experimental approaches, combination of extended X-ray absorption fine structure (EXAFS) and molecular dynamics simulations have proven valuable for exploring molten salt structure and rich chemistry of molten salt systems.

Regarding fission products, Te induces corrosion. Some studies have shown that Te embrittlement can be mitigated by reducing salt oxidation-reduction potential. To achieve this, the activity ratio of tetravalent to trivalent uranium ions in the salt can be adjusted. Although oxidation-reduction potential can be adjusted to reduce salt oxidizability (e.g., by exposing salt to Be or Cr), UF_3 activity in the salt must be sufficiently low to avoid its disproportionation and uranium alloying with the containment alloy [**?**].

Various studies have also focused on improving alloy resistance to Te embrittlement. One approach involves adding trace elements to the alloy to form nanoprecipitates that decrease Te influence and hinder Te atom diffusion. Han et al. investigated Te vapor effects on intergranular embrittlement of Y-modified Ni-16Mo-7Cr-based alloy at 800 °C. Their results indicated that Y addition significantly reduced Te diffusion depth in the alloy, thus improving resistance to intergranular cracking [**?**]. The YTe phase near the surface reaction layer and surface grain boundaries plays an important role in fixing and reducing Te diffusion. Segregation of Y from the substrate to the surface and formation of YTe phase in the reaction layer and grain boundaries are main factors contributing to reduced Te diffusion depth in Y-modified alloy. Te diffusion depth is ~40 m in Y-modified alloy, whereas it exceeds ~70 m in GH3535 alloy. However, adding more than 0.05 wt% Y does not consistently improve intergranular brittleness and may even degrade mechanical properties.

La addition also efficiently reduces Te diffusion depth in the alloy, thus decreasing susceptibility to intergranular cracking [?]. Although $LaTe_3$ has not been observed during Te-alloy interaction, La segregation in the surface telluride reaction layer and presence of $LaNi_5$ intermetallic compound at grain boundaries facilitate NiTe formation, contributing to reduced Te diffusion depth in La-modified alloys. The optimal La concentration in the alloy should be approximately 0.1 wt%. Excessive La concentration can increase $LaNi_5$ phase formation, which does not improve mechanical properties. Additionally, intergranular alloy crack resistance does not increase with increasing La concentration.

Overall, breakthroughs in materials science, ranging from material development to performance evaluation, are extremely challenging. These challenges require extensive testing and in-depth research on structural materials, particularly construction and development of equipment that closely simulates real working conditions for in situ research. In addition to experimental testing, theoretical calculations and modeling at various scales, ranging from atomic to grain levels, will contribute significantly to profound understanding of material behavior under multiple environmental conditions [?]. Furthermore, integration of new alloy and microstructure design methodologies with novel material processing manufacturing technologies is crucial for achieving optimal system performance under demanding operating conditions.

5. Conclusion and Outlook

The service behavior of reactor structural materials under operating conditions is a key factor affecting reactor lifetime, and understanding synergistic effects of irradiation and corrosion on these materials is a challenging research area. This review focuses on effects of irradiation on corrosion properties of structural materials in PWRs and MSRs, highlighting the following main conclusions:

Austenitic stainless steel and Ni-based alloys are key structural materials for PWR cores. Their corrosion behavior in high-temperature and high-pressure water environments is closely associated with oxide formation on alloy surfaces. Oxide formation is a product of interaction between alloy elements and water medium and is also a key factor affecting IASCC processes. Therefore, this study introduces oxide morphology and structure, along with mechanisms governing its formation and growth. This review summarizes the effect of irradiation on general corrosion behavior and intergranular oxidation of alloys, highlighting three key aspects: (1) The effect of irradiation-induced defects on alloy corrosion rates depends on defect size. (2) Cr presence improves alloy corrosion resistance by forming a dense protective film on its surface. Irradiation-induced sputtering leads to formation of a thin Cr-rich oxide layer on alloy surfaces, efficiently mitigating initial corrosion. However, RIS of Cr at grain boundaries results in Cr depletion, thereby accelerating grain boundary corrosion and cracking. Preferential Si diffusion at grain boundaries creates vacancies for oxides located at grain boundaries, promoting Cr enrichment in oxides and thus enhancing intergranular corrosion resistance. (3) Radiolysis increases corrosion potential and accelerates the overall corrosion process.

In MSRs, structural materials primarily consist of Ni-based alloys that cannot form protective passive films in molten salt environments. The corrosion mechanism of these alloys is mainly attributed to selective dissolution of active elements. In early corrosion stages, alloy corrosion is significantly affected by impurities in molten salt. When impurities are depleted, corrosion is primarily driven by thermodynamics. Irradiation can affect molten salt corrosion of Ni-based alloys through various mechanisms: (1) Irradiation-induced defects accelerate diffusion processes, leading to increased alloy corrosion rates. (2) Alloy grain boundary corrosion is influenced by three effects: blockage, diffusion, and self-healing of interstitial atoms. (3) RIS increases chemical reactivity of alloy elements with molten salts, enhancing galvanic effects and promoting alloy corrosion. (4) Fission products and fuel introduced by irradiation can also affect alloy corrosion.

Considering factors influencing irradiation and corrosion performance of structural materials in PWRs and MSRs, several strategies have been proposed to mitigate corrosion: (i) implementing protective measures for irradiation-resistant alloys, such as enhancing corrosion resistance of surface coatings to slow corrosion rates; (ii) developing new materials with improved irradiation and corrosion resistance; and (iii) reducing oxidation-reduction potential of water and molten salt media, which is also an effective way to decrease corrosion rates.

Overall, to ensure safe reactor operation, it is important to study material service behavior and grasp evolution laws of material properties under combined effects of irradiation and high-temperature corrosion in reactor operating environments. Further theoretical and in situ experimental work is necessary to deepen understanding of relevant mechanisms and develop optimized solutions.

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Data Availability

This is a review paper, and all data is based on published literature, which can be obtained from the internet without any data acquisition issues.

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