Emulating Neutron Irradiation Effect on Stainless steel by Non-Irradiation Method

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1. Introduction

Ensuring the structural integrity of internal components is paramount in the design of nuclear power plants to prevent accidents. Particularly within Pressure Water Reactors (PWRs), the internal environment is characterized by high temperature and pressure, directly influencing susceptibility to damage and corrosion. To counteract these challenges, PWRs primarily employ stainless steel 304 and stainless steel 316, known for their high resistance to corrosion and damage under high-temperature and high-pressure conditions. These materials, facilitated by chromium oxide films, exhibit elevated corrosion resistance and strength at high temperatures. However, actual structural components are exposed to neutron irradiation, making the materials more vulnerable to corrosion and leading to changes in mechanical strength. Hence, understanding the impact of neutron irradiation on materials is essential.

Radiation induces fluxes of defects and elemental fluxes within the material. Depending on the type of elements involved, an abundance of defects may lead to depletion, while a surplus of atom fluxes can result in enrichment. As a consequence, vigorous occurrences at grain boundaries or dislocations with high diffusion rates lead to differences in matrix and microchemical composition, a phenomenon termed Radiation-Induced Segregation (RIS). In the case of stainless steel, RIS causes the depletion of chromium, hindering the formation of chromium oxide films and reducing corrosion resistance. Furthermore, radiation exposure forms defects such as dislocations and voids, resulting in material hardening. Although extensive research is conducted on neutron-irradiated materials, studying neutron-irradiated materials practically is challenging due to the high cost and risks associated with handling radioactive materials. Various emulation methods, including the investigation of neutrons from alternative nuclear isotopes such as protons and heavy ions, are under investigation. [1]

In this study, the goal is to replicate materials irradiated by neutrons at the 5displace per atom(dpa) level through non-irradiation methods. The objective is to emulate both microscopic microstructural changes and macroscopic mechanical strength, providing insights into the material behavior without actual exposure to neutron irradiation. Through heat treatment, the formation of Cr carbides will be induced to mimic chromium depletion around carbides and replicate the RIS phenomenon. Microstructural changes will be analyzed using scanning electron microscopy (SEM), scanning transmission electron microscopy (STEM), and energy-dispersive X-ray spectroscopy (EDS). To emulate irradiation hardening resulting from defects, the material will be subjected to rolling to induce defects, thereby hardening the material. Mechanical property changes will be assessed through tensile testing.

2. Experimental and Results

2.1 Experimental

2.1.1 Materials

Stainless steel 304, a material widely employed in actual nuclear power plants, has been chosen for this study. Specifically, the high-carbon content variant, denoted as H grade, was utilized. The chemical composition is detailed in Table 1.

Table I: Chemical composition (in wt.%) of stainless steel 304H used in this study

Fe	C	Si	Mn	P
Bal.	0.0542	0.0429	1.069	0.0158
Cr	Ni	Cu	Co	
18.25	8.055	0.027	0.022	

In order to replicate Radiation-Induced Segregation (RIS) through neutron irradiation, heat treatment was conducted to induce the formation of Cr carbides. A heat treatment of 1040°C for 2 hours was employed for solution annealing. Referring to the Phase Diagram in Fig. 1, a heat treatment at 700°C was chosen to facilitate the formation of Cr carbides and prevent the unwanted development of the ferrite phase.

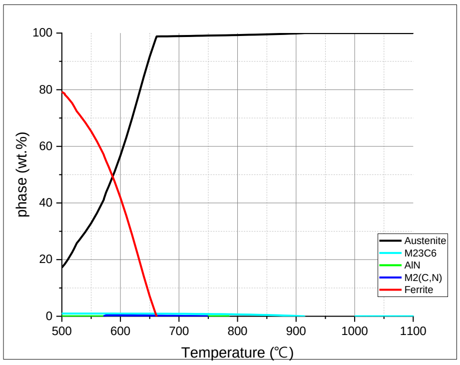


Fig. 1. Phase diagram of Stainless steel 304H

Through a literature review, we investigated the chromium depletion in Stainless Steel 304, a material similar to the one utilized in this study, concerning varying heat treatment durations. According to Butler's research, it was observed that the chromium depletion and the corresponding region of chromium depletion widen with increasing heat treatment time. [2]

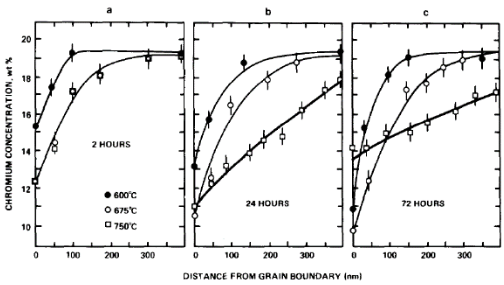


Fig. 2. Chromium concentration profiles obtained from specimens sensitized of (a) 2h, (b) 24h, and (c) 72h at 600℃, 675℃ and 750℃ [2]

The chromium content at the grain boundaries decreases, and the chromium-depleted region widens with increasing heat treatment duration. In this study, we aim to determine the appropriate heat treatment duration to replicate suitable chromium depletion areas and chromium content at grain boundaries. Heat treatments of 1 hour, 3 hours, 7 hours, and 24 hours were conducted to establish the optimal duration. Following the heat treatments, mechanical strength emulation was performed through rolling to induce defects and subsequent work-hardening. To minimize the formation of Martensite phase during rolling, the rolling temperature was elevated to 200°C, and warm rolling at 10%, 20%, and 30% reductions was carried out. [3]

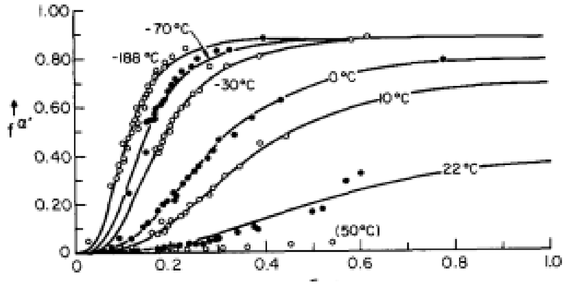


Fig. 3. Martensite fraction by temperature for stainless 304 [3]

2.1.2. Microstructure analysis

All materials were examined for their microstructure using Scanning Electron Microscopy (SEM), while materials subjected to heat treatment were assessed for the formation of Cr carbides. Furthermore, to investigate the chromium-depleted regions around Cr carbides, Transmission Electron Microscopy (TEM) samples were fabricated using Focused Ion Beam (FIB). Scanning Transmission Electron Microscopy (STEM) coupled with Energy Dispersive X-ray Spectroscopy (EDS) was employed to quantify the extent and chromium depletion in the surrounding areas. This data was then compared with neutron-irradiated materials.

Additionally, for materials subjected to rolling, microstructure continuity in different directions was examined. Microstructure specimens were obtained in various orientations, as illustrated in Fig. 5, to confirm any orientation-dependent variations.

2.1.3. Mechanical properties analysis

Tensile specimens were fabricated in accordance with ASTM E8M dimensions, as illustrated in Fig. 4. To minimize variations in mechanical properties induced by the rolling direction, specimens were prepared in the Transverse Direction (TD) from the Rolling Direction (RD), as depicted in Fig. 5. Tensile tests were conducted at room temperature, and the testing was carried out at a rate of 0.015 mm/mm/min. [4]

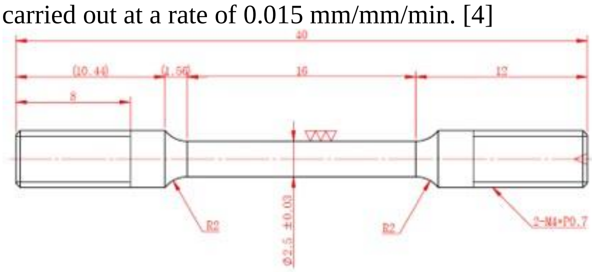


Fig. 4. Tensile specimens

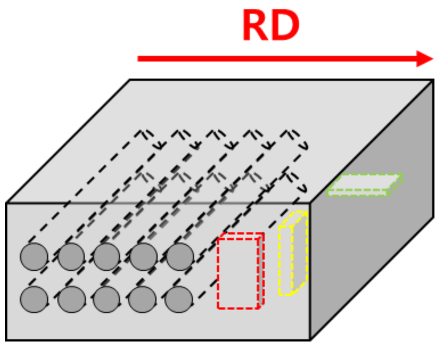


Fig. 5. Rolling direction and specimen direction

2.2. Result

2.2.1. SEM analysis

Observing Fig. 6, it is evident that Cr carbides have formed around grain boundaries with a high diffusion rate. Furthermore, with longer heat treatment durations, an increased presence of chromium carbides within the grain boundaries was observed. To compare the formation levels of Cr carbides over time, the width of Cr carbides was assessed based on the length of grain boundaries. Quantitative evaluation, as depicted in Fig. 7, confirms an increase in the formation of Cr carbides with prolonged heat treatment durations.

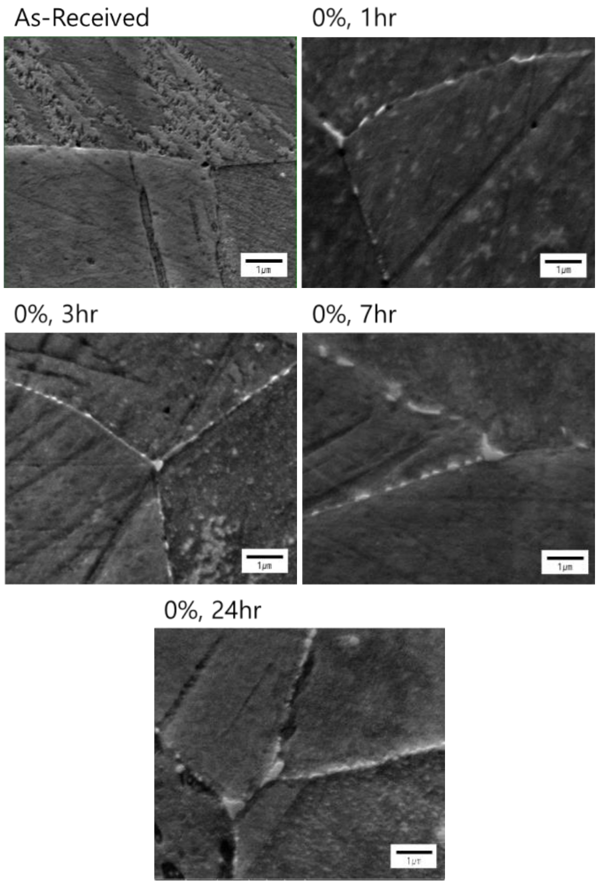


Fig. 6. Microstructure by heat treatment time

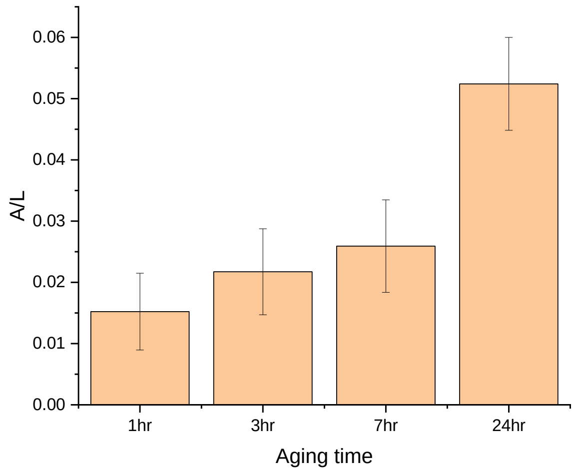


Fig. 7. Cr carbide area per grain boundary by heat treatment time

Additionally, the material underwent rolling, confirming the elongation of grain boundaries along the rolling direction. It was observed that the tendency for grain elongation in the rolling direction strengthened with an increase in the reduction rate.

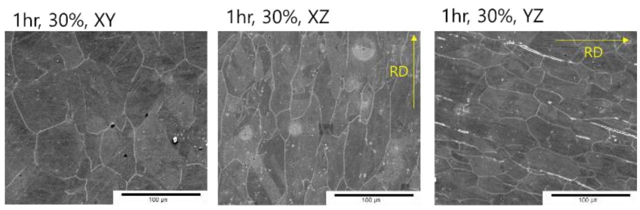


Fig. 8. SEM images for RD

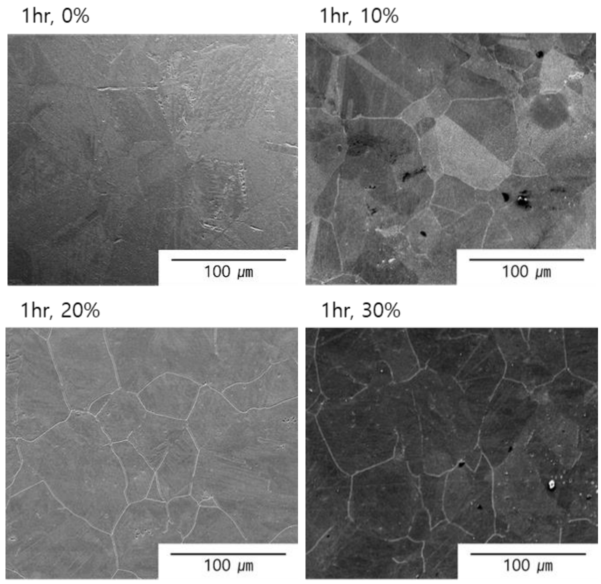


Fig. 9. SEM images for reduction rate

2.2.2. STEM-EDS analysis

To precisely investigate the formation of Cr carbides at grain boundaries and the surrounding depletion, a more precise analysis using Energy Dispersive X-ray Spectroscopy (EDS) in Transmission Electron Microscopy (TEM) was deemed necessary. For the confirmation of Cr carbides, TEM samples were obtained and analyzed, as depicted in Fig. 10.

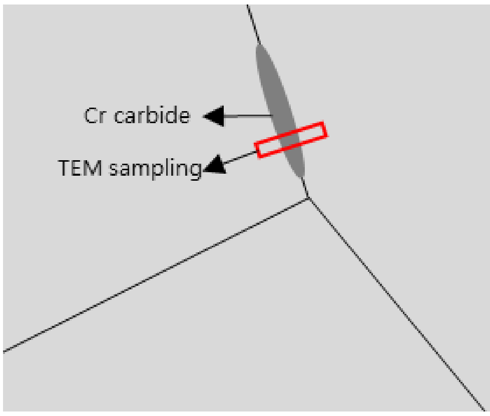


Fig. 10. TEM Sample cutting method

Through Fig. 11, the formation of Cr carbides at grain boundaries and the surrounding depletion zone were confirmed using EDS maps. The measurement of the depletion zone's width and microchemical composition revealed variations in the depletion area and amount of depletion around chromium, as shown in Fig. 12. The differences were observed concerning the heat treatment duration, and the quantitative results are available in Table 2.

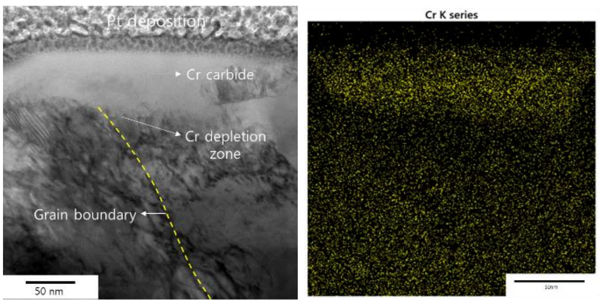
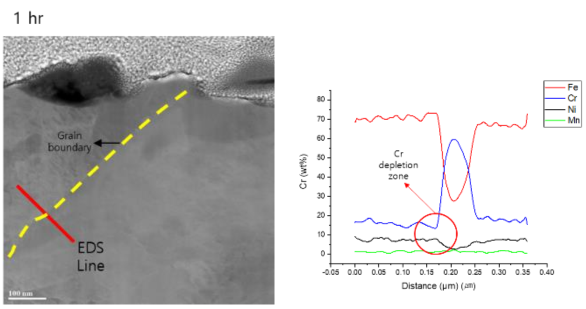


Fig. 11. STEM cross section image and EDS map



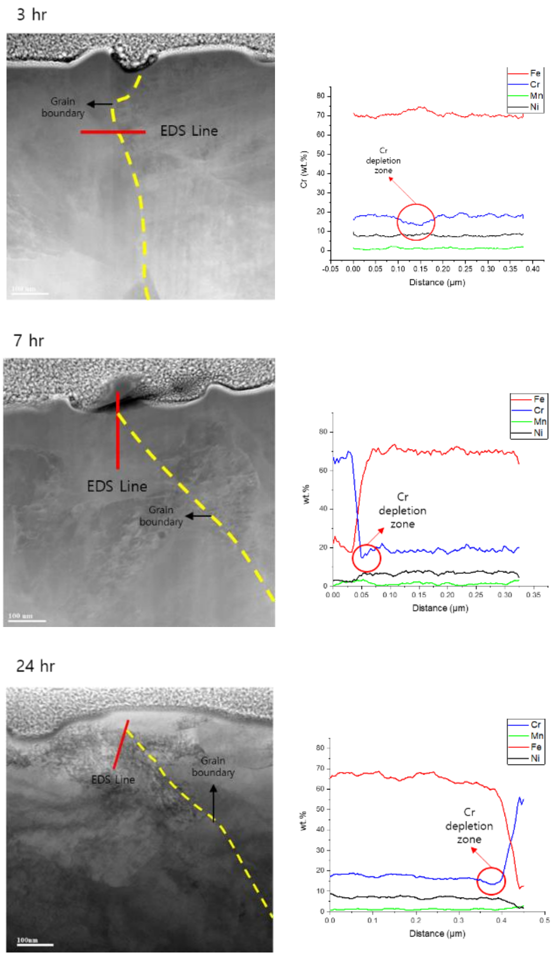


Fig. 12. STEM image and EDS (wt.%) for Cr carbide in heat treatment sample

Table II. Cr min and depletion width

	1 hr	3 hr	7 hr	24hr
Cr min (wt.%)	13.59	13.17	10.28	9.95
Depletion width	0.02 um	0.05 um	0.05 um	0.1 um

According to previous studies, actual neutron-irradiated materials show a reduction of approximately 5 wt.% in chromium concentration compared to the original concentration at 5dpa. Moreover, there is an observed chromium depletion of around 13 wt.% at grain boundaries. [5]

Analyzing the chromium depletion within grain boundaries through heat treatment, it can be concluded, considering the RIS-induced depletion width of approximately 0.1 μm and taking into account the minimum value of chromium, that a treatment duration of approximately 3 to 7 hours is the most suitable for emulation. [6]

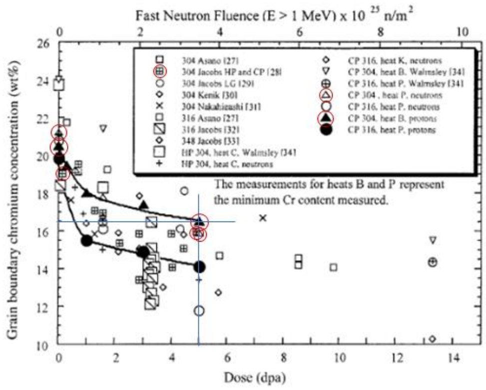


Fig. 13. Cr composition as a function of dose for irradiated [5]

2.2.3. Mechanical properties

When irradiating the material, the hardening effect occurs due to defects such as dislocations and voids. According to the preceding study depicted in Fig. 14, the yield strength, a crucial indicator of mechanical properties, was measured based on the neutron irradiation dose to discern trends. **[7]** At 5dpa, it exhibits a yield strength of approximately 800 MPa. In this study, to simulate the irradiation hardening effect, rolling was conducted to induce defects within the material, thereby increasing the yield strength. Tensile tests were performed to assess the mechanical properties, and the Strain-Stress curve in Fig. 15 provided insights into the mechanical characteristics, yield strength, tensile strength, and elongation. Comparing the yield strength with Table 3 and the actual neutron-irradiated material, rolling with a reduction rate of around 30% yielded the closest resemblance in yield strength.

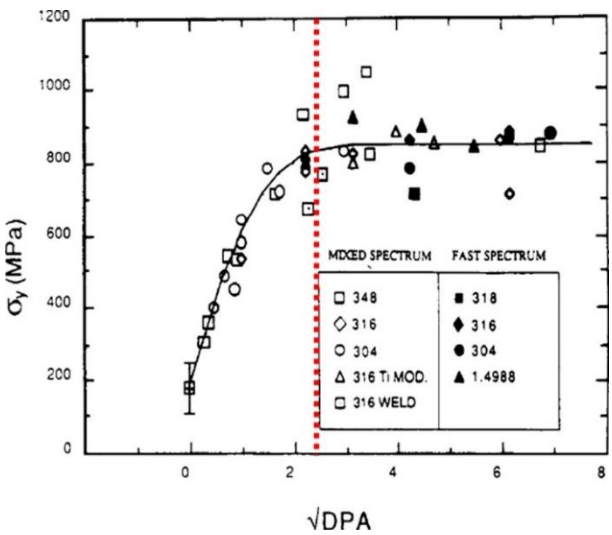


Fig. 14. Yield strength for neutron irradiation dose [7]

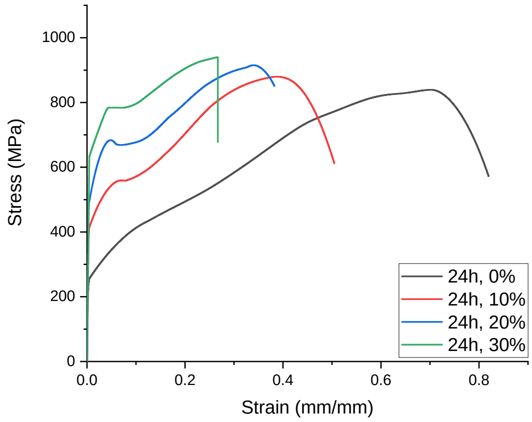


Fig. 15. Strain-Stress curve for reduction rate

3. Conclusions

In conclusion, this study aimed to emulate the microstructural and mechanical changes induced by neutron irradiation in stainless steel 304 at a level of 5 dpa. Through a series of experiments, the effects of heat treatment and warm rolling on the material were investigated to mimic the radiation-induced segregation (RIS) and irradiation hardening observed in actual neutron-irradiated materials.

The material selected for this study was the widely used stainless steel 304, specifically the 304H grade with elevated carbon content. Heat treatment was performed to induce the formation of chromium carbides (Cr carbide) and emulate the chromium depletion phenomenon observed in neutron-irradiated materials. The heat treatment process was optimized based on literature review and phase diagrams to control the formation of unwanted phases.

Microstructure analysis using scanning electron microscopy (SEM) revealed that longer heat treatment times led to increased Cr carbide formation, particularly around grain boundaries. The quantification of Cr carbide area per grain boundary confirmed the time-dependent nature of Cr carbide formation. Warm rolling was employed to induce defects and emulate irradiation hardening. The microstructure analysis along the rolling direction demonstrated a correlation between reduction rate and the elongation of grain boundaries.

Further analysis using scanning transmission electron microscopy (STEM) and energy dispersive X-ray spectroscopy (EDS) provided insights into the chromium depletion phenomenon. The study confirmed that the chromium depletion width and amount varied with heat treatment time, suggesting an optimal range for simulating RIS.

Mechanical properties analysis through tensile testing showed that the emulated irradiation hardening, induced by warm rolling, resulted in an increase in yield strength. The obtained strain-stress curves demonstrated the mechanical behavior of the material, and the comparison with actual neutron-irradiated material's yield strength indicated a good resemblance, especially with a 30% reduction rate in warm rolling.

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