Intergranular Corrosion Imprint of 316LN Stainless Steel Welded Joints

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Abstract

The intergranular corrosion characteristics of 316LN stainless steel welded joints with different heat inputs in boiling nitric acid solution were investigated. The results indicate that: the intergranular corrosion resistance of welded joints deteriorates with increasing welding heat input; the weld zone exhibits higher intergranular corrosion resistance than both the heat-affected zone and base metal zone; the intergranular corrosion process can be divided into several stages including oxidation, passivation, oxide film crack initiation, oxide film crack propagation, and high-speed intergranular corrosion. Regarding acoustic emission signals at different stages: the oxidation signal displays an energy peak at 20 kHz; the passivation signal is a burst-type signal with a prominent energy peak at 40 kHz; the oxide film crack initiation signal is a burst-type signal with a prominent energy peak in the 80-100 kHz frequency band; the oxide film crack propagation signal exhibits a 60 kHz frequency multiplication relationship; and the high-speed intergranular corrosion signal is a burst-type signal of slightly longer duration with a prominent energy peak at 50 kHz.

Full Text

Preamble

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Intergranular Corrosion of 316LN Stainless Steel Welded Joints

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Abstract

This study investigates the intergranular corrosion characteristics of 316LN austenitic stainless steel welded joints with different welding heat inputs in boiling nitric acid solution. The results demonstrate that the resistance to intergranular corrosion of welded joints deteriorates with increasing welding heat input. The weld zone exhibits superior intergranular corrosion resistance compared to both the heat-affected zone and base material. The intergranular corrosion process can be divided into five distinct stages: oxidation, passivation, oxide film crack initiation, oxide film crack propagation, and rapid intergranular corrosion. Acoustic emission signal analysis reveals characteristic frequency signatures for each stage: oxidation signals show an energy peak at 20 kHz; passivation signals exhibit a distinct energy peak at 40 kHz as burst-type signals; oxide film crack initiation signals display energy peaks in the 80–100 kHz range as burst-type signals; crack propagation signals demonstrate harmonic relationships at multiples of 60 kHz; and rapid intergranular corrosion signals appear as burst-type signals with slightly longer duration, showing a prominent energy peak at 50 kHz.

Keywords: materials failure and protection, intergranular corrosion, acoustic emission, 316LN, metallographic analysis

Introduction

The development of nuclear power represents a crucial strategy for adjusting China's energy structure and improving energy supply relationships. Among the factors that can trigger nuclear power plant safety accidents, corrosion-induced failure of critical materials constitutes a significant concern. Defects in main pipeline welds and heat-affected zones substantially increase the probability of failure. Nuclear power plant primary pipelines operate in harsh environments characterized by high temperature, high pressure, and radioactive substances, which dramatically elevates the likelihood of welded joint failure. Intergranular corrosion-induced failures in nuclear power plant primary pipelines account for 25.72% of total incidents, making research on the intergranular corrosion mechanisms, detection methods, failure analysis, and evaluation of these pipelines essential.

Numerous domestic and international experts have employed acoustic emission technology to monitor and study metallic material corrosion. Retting and Felsen et al. discovered that when steel and aluminum wires are placed in corrosive solutions, hydrogen evolution exhibits a linear relationship with cumulative ring-down counts, demonstrating that acoustic emission technology provides a novel approach for investigating and monitoring material corrosion processes. Subsequently, Mazille et al. observed a strong linear correlation between pitting counts and event counts during pitting corrosion studies of 316L stainless steel. Fregonese et al. conducted comprehensive investigations on pitting corrosion in austenitic stainless steel, successfully distinguishing acoustic emission signal characteristics between pitting initiation and propagation stages using parameters such as ring-down count and rise time. Shaikh et al. divided the stress corrosion cracking process of 316LN stainless steel into four stages—initial period, crack initiation, early crack propagation, and later crack propagation—and analyzed the acoustic emission signal characteristics for each stage in detail. Xu et al. studied the acoustic emission characteristics of sensitized 304 stainless steel during intergranular corrosion and stress corrosion cracking, finding that acoustic emission signals from intergranular corrosion were lower in both quantity and amplitude than those from stress corrosion cracking, and that signal counts increased gradually over time throughout the stress corrosion cracking process. Du and Wang Weikui et al. also employed acoustic emission detection technology to investigate the corrosion resistance and stress corrosion process characteristics of 304 stainless steel, comparing results with electrochemical impedance spectroscopy measurements. They utilized average frequency of acoustic emission signals to characterize different corrosion stages of 304 stainless steel, demonstrating that distinct differences in acoustic emission signal characteristics across various corrosion stages facilitate more accurate assessment of stainless steel corrosion conditions.

The severe operating conditions of nuclear power plant primary pipelines predispose them to various failure modes, with intergranular corrosion being particularly difficult to detect. Applying acoustic emission technology to monitor intergranular corrosion behavior in nuclear power plant primary pipelines represents a novel approach. This paper employs metallographic analysis and acoustic emission detection techniques to conduct exploratory research on the intergranular corrosion failure characteristics of 316LN stainless steel—the primary pipeline material used in third-generation AP1000 nuclear power technology.

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1. Experimental Methods

The experimental material was 316LN austenitic stainless steel, the primary pipeline material for the third-generation AP1000 nuclear power technology primary loop, supplied as hot-rolled stainless steel plate. Its chemical composition (mass fraction, %) was: C 0.023, N 0.16, Cr 16.43, Ni 11.33, Mo 2.25, Mn 1.45, Si 0.46, P 0.024, S 0.001. ER316L welding electrodes with a diameter of 3.2 mm were used to complete specimen welding at currents of 120 A, 130 A, and 140 A.

Specimens were divided into three groups based on welding parameters, with four specimens per group tested simultaneously. Two specimens from each group were designated for metallographic observation, while the remaining two were used for acoustic emission signal detection. Metallographic specimens were further categorized as welded joint specimens and base material specimens. Welded joint specimens were prepared by wire cutting with the weld zone as the center, while base material specimens were taken from both ends of the welded joints. Welded joint specimens measured 40 mm × 20 mm × 10 mm, and base material specimens measured 20 mm × 20 mm × 10 mm. All specimens were cleaned ultrasonically, then ground with 400#-2000# metallographic sandpaper to remove surface irregularities and oxidation products. Specimens were polished with 1.5 m diamond metallographic polishing agent, rinsed with water, cleaned with anhydrous ethanol, degreased with acetone, dried, and weighed.

Following the GB4334-2008 standard "Corrosion of Metals and Alloys—Test Method for Intergranular Corrosion of Stainless Steel," high-purity nitric acid conforming to GB/T626 was diluted with distilled water to prepare a 65% concentrated nitric acid solution for boiling nitric acid intergranular corrosion testing. Each experimental group employed four parallel specimens undergoing five 48-hour cycles.

The acoustic emission detection system consisted of an AEwin acoustic emission signal processing system and a SAMOS 24-channel acoustic emission detector. System parameters were set as follows: preamplifier gain of 40 dB, hit definition time (HDT) of 600 µs, peak definition time (PDT) of 300 µs, and hit lockout time (HLT) of 1000 µs. Acoustic emission sensors were mounted on exposed specimen surfaces, and pencil lead break tests using 2B or HB leads with 0.5 mm diameter were performed to ensure good sensor-specimen contact and stable signal acquisition. The heating temperature was set at 115°C with stirring. Acoustic emission signals were collected every 3 hours during each cycle, with three signal sets acquired per collection period, each lasting 3 minutes. After 48 hours, the experimental system was cooled, specimens were removed, corrosion products were brushed off under running water with a soft brush, and specimens were cleaned in an ultrasonic bath with anhydrous ethanol for 30 minutes, dried, and weighed. Specimen surfaces were examined using an LWD300LCS inverted metallographic microscope, and acquired acoustic emission signals were analyzed.

2. Results and Discussion

2.1 Corrosion Rate Analysis

Corrosion rate represents the average degree of metal corrosion per unit time and characterizes the extent and relative speed of metal corrosion. According to the GB4334-2008 standard, corrosion rate is defined as metal corrosion mass loss per unit time per unit area. The calculation formula is:

$$V = \frac{W_f - W_b}{S \times T}$$

where V is the corrosion rate (g/(m$^2 \cdot$ h)), T is the experimental time (h), W_f is the specimen mass before testing (g), W_b is the specimen mass after testing (g), and S is the corroded surface area (m^2).

Metallographic specimens were weighed after each cycle to calculate corrosion rates, with results shown in [Figure 1: see original paper]. The corrosion rate patterns for base material specimens and welded joint specimens were essentially consistent. During the first cycle, all specimens exhibited relatively low corrosion rates. In the second cycle, the three base material specimens showed higher corrosion rates than welded joint specimens, with welded joint corrosion rates increasing correspondingly with welding heat input. In the third cycle, base material corrosion rates remained essentially unchanged from the second cycle, while welded joint corrosion rates increased, with the 140 A welded joint specimen exceeding the base material corrosion rate. The fourth and fifth cycles showed similar corrosion behavior, with base material specimens exhibiting the lowest corrosion rates and welded joint corrosion rates increasing with welding heat input. Since corrosion rates reached maximum values during the fourth and fifth cycles, all metallographic specimens were considered to have entered the high-speed corrosion stage.

These results indicate that the degree of intergranular corrosion in 316LN austenitic stainless steel welded joints correlates with welding heat input: as welding heat input increases, intergranular corrosion becomes more severe. Upon entering the high-speed corrosion stage, welded joint intergranular corrosion rates exceed those of the base material.

2.2 Metallographic Analysis

Metallographic examination revealed no significant differences in microstructure among welded joint specimens prepared with different welding heat inputs. Therefore, this analysis focuses on specimens welded at 130 A. [Figure 2: see original paper] presents metallographic images after the first four boiling nitric acid corrosion cycles.

After the first cycle ([Figure 2: see original paper]a1–c1), the base material zone exhibited single-phase austenitic structure with minor strip-like ferrite, featuring clearly defined grain shapes and boundaries. Small black spots appeared on some specimen surfaces, resulting from grain detachment due to intergranular corrosion. The heat-affected zone also showed single-phase austenitic structure with minor ferrite strips and some strip-like corrosion traces from ferrite corrosion. The weld zone displayed a dual-phase structure of austenite plus ferrite with some corrosion traces, indicating initial corrosion.

After the second cycle ([Figure 2: see original paper]a2–c2), intergranular corrosion became deeper and wider compared to the first cycle, and specimen surface color changed due to oxidation. After the third cycle ([Figure 2: see original paper]a3–c3), corrosion appearance was similar to the second cycle, but cracks appeared across specimen surfaces. After the fourth cycle ([Figure 2: see original paper]a4–c4), severe surface corrosion was observed. The base material zone was completely corroded with significant grain detachment, as continuous grain boundary corrosion caused grains to lose cohesion and detach from the surface. Clear and blurred regions in the metallographic images resulted from height differences, indicating rough surface morphology and continued grain boundary corrosion. The heat-affected zone also showed severe corrosion, but distinct differences existed between the heat-affected and weld zones. The weld zone surface was higher than the heat-affected zone, indicating lower corrosion severity and superior intergranular corrosion resistance. Heat-affected zone corrosion propagated along grain boundaries into the metal depth, consistent with intergranular corrosion characteristics. The weld zone also exhibited some intergranular corrosion.

The fifth cycle showed similar corrosion behavior to the fourth cycle, with only increased depth and width. Corrosion rates for the fourth and fifth cycles were identical, confirming consistent high-speed corrosion behavior.

Post-experiment cleaning with dilute acid to remove surface oxide films revealed no cracks in the substrate, confirming that cracks observed during the third cycle were oxide film cracks. Crack formation resulted primarily from thermal stress between the oxide film and base metal due to their different thermal expansion coefficients during boiling nitric acid corrosion. Crack formation led to oxide film rupture and detachment, exposing fresh metal surfaces. Without timely oxide film repair, this created a “large cathode-small anode” electrochemical corrosion condition, accelerating fresh metal oxidation. This marked a critical transition point into a new accelerated corrosion stage.

Metallographic analysis demonstrates that the weld zone exhibits the lowest corrosion degree and superior intergranular corrosion resistance compared to base material and heat-affected zones. During welding, the weld zone transforms from pure austenite to a dual-phase structure of austenite matrix with dispersed ferrite. This ferrite distribution weakens the directionality of austenite columnar and dendritic structures and interrupts continuous, large-area chromium carbide precipitation near austenite grain boundaries. Additionally, ferrite contains significantly higher Cr content than austenite, and Cr diffusion in ferrite is much faster than in austenite. Consequently, Cr from ferrite can supplement the Cr required for chromium carbide formation at austenite grain boundaries, preventing Cr-depleted zone formation and enhancing intergranular corrosion resistance in austenitic stainless steel weld zones.

Comparative experiments revealed identical intergranular corrosion mechanisms for welded joints prepared with different heat inputs, with essentially identical acoustic emission signal characteristics. Therefore, parameter and waveform analysis focuses on acoustic emission signals generated during intergranular corrosion of 316LN stainless steel welded joints prepared at 130 A.

2.3.1 Total Count and Total Energy Analysis

Variations in total count and total energy during intergranular corrosion experiments are presented in [Figure 3: see original paper] and [Figure 4: see original paper]. Total count represents the cumulative sum of counts collected by the acoustic emission instrument within 3 minutes under identical system settings, while total energy represents the cumulative energy sum collected under the same conditions.

Total count and total energy exhibit similar variation patterns with experimental time. During the early first cycle, high-count and high-energy acoustic emission characteristics appeared, followed immediately by low-count and low-energy features. In the second cycle, total count and total energy began fluctuating but showed significant increases compared to the low values at the first cycle end. Upon entering the third cycle, total count and total energy increased substantially, again displaying high-count, high-energy distribution characteristics. The fourth and fifth cycles showed essentially identical total count and total energy distributions.

Based on these distribution characteristics and previous corrosion morphology analysis, the 316LN austenitic stainless steel intergranular corrosion process can be further divided into five stages:

Oxidation Stage (early first cycle): Intergranular corrosion rates are minimal as specimen surfaces are oxidized by strongly oxidizing nitric acid to form protective oxide films. This stage is very brief, with bubbles appearing on specimen surfaces due to corrosion and nitric acid decomposition producing NO_2 gas, gradually turning the solution reddish-brown.

Passivation Stage (late first cycle): The formed oxide film blocks contact between nitric acid and the 316LN stainless steel, causing intergranular corrosion to enter a low-rate trough with minimal corrosion activity and low-count, low-energy acoustic emission signals.

Oxide Film Crack Initiation Stage (second cycle): Metallographic analysis revealed low corrosion rates and minimal intergranular corrosion during the second cycle, with only localized intergranular corrosion signs observed. However, numerous cracks appeared in the third cycle, indicating oxide film crack initiation during the second cycle, which altered acoustic emission signal total count and total energy parameters.

Oxide Film Crack Propagation Stage (third cycle): Metallographic observation revealed numerous cracks on specimen surfaces after the third cycle, identifying crack propagation as the acoustic emission source generating high-count, high-energy signals. Although corrosion rates remained low, complete oxide film destruction would trigger transition to a high-speed corrosion stage, consistent with metallographic analysis results.

Rapid Intergranular Corrosion Stage (fourth and fifth cycles): Metallographic analysis confirmed that 316LN stainless steel welded joints entered the rapid intergranular corrosion stage during the fourth and fifth cycles. [Figure 3: see original paper] and [Figure 4: see original paper] show identical total count and total energy distributions for acoustic emission signals collected after entering the fourth cycle, identifying rapid intergranular corrosion as the primary acoustic emission source.

2.3.2 Amplitude Analysis

Environmental noise measurements prior to testing were dominated by 25 dB signals, so the threshold was set at 25 dB to minimize noise interference. Amplitude characteristics of acoustic emission signals at different intergranular corrosion stages are shown in [Figure 5: see original paper].

During the oxidation stage, amplitude ranged from 43-45 dB, exhibiting high-amplitude, stable signals consistent with intense surface oxidation by nitric acid, characterized by high count, high energy, and high amplitude. During passivation, amplitude ranged from 25-28 dB, similar to noise signal distribution, but waveform and spectrum analysis confirmed these were not noise signals, indicating very weak corrosion activity. During oxide film crack initiation, amplitude ranged from 25-45 dB with intermittent temporal distribution, consistent with the non-continuous nature of crack initiation processes. During crack propagation, amplitude distribution showed two layers: 25-26 dB (primarily noise) and 32-36 dB (crack propagation signals). During rapid intergranular corrosion (fourth and fifth cycles), amplitude ranged from 25-32 dB.

2.4 Acoustic Emission Waveform Analysis

Environmental noise spectrum analysis revealed dominant frequencies below 250 Hz, so wavelet analysis suppressed signals below this frequency. [Figure 6: see original paper] presents waveforms and spectra after wavelet denoising and reconstruction for different intergranular corrosion stages.

Oxidation signals consist of attenuating signals superimposed with brief burst signals, showing an energy peak at 20 kHz. Passivation signals are burst-type signals with frequency distribution from 10-90 kHz and an energy peak near 40 kHz, consistent with low corrosion rates and low acoustic emission parameters. Crack initiation signals are also burst-type with rapid attenuation, exhibiting low count and energy, and show clear energy peaks in the 80-90 kHz range. The brittle nature of oxide films causes rupture signals to be burst-type, and the appearance of cracks on specimen surfaces confirms crack initiation as the acoustic emission source. Crack propagation signals show high voltage values and long duration, producing high count, high energy, and high amplitude parameters, with clear energy peaks at frequencies harmonically related to 60 kHz.

The numerous cracks observed on specimen surfaces identify crack propagation as the acoustic emission source. Rapid intergranular corrosion signals are burst-type with energy peaks at 50 kHz. The abundance of similar signals collected during the fourth and fifth cycles, combined with corrosion rate distributions and metallographic analysis, confirms that the acoustic emission source during this stage is rapid intergranular corrosion of 316LN stainless steel welded joints.

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3. Conclusions

1. For 316LN stainless steel welding, increased welding heat input degrades intergranular corrosion resistance of welded joints. The weld zone demonstrates superior intergranular corrosion resistance compared to the heat-affected zone and base material zone.
2. Acoustic emission technology can distinguish different stages of intergranular corrosion. Parameters including count, energy, and amplitude characterize variations in acoustic emission signals during intergranular corrosion, enabling division of the 316LN stainless steel welded joint intergranular corrosion process into oxidation, passivation, oxide film crack initiation, oxide film crack propagation, and rapid intergranular corrosion stages.
3. Oxidation signals exhibit energy peaks at 20 kHz. Passivation signals are burst-type signals with distinct energy peaks at 40 kHz. Oxide film crack initiation signals are burst-type signals with clear energy peaks in the 80-100 kHz range. Crack propagation signals show harmonic relationships at multiples of 60 kHz. Rapid intergranular corrosion signals are burst-type signals with slightly longer duration, showing prominent energy peaks at 50 kHz.

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Note: Figure translations are in progress. See original paper for figures.