

DEGRADATION OF A Ni-Cr-Fe ALLOY IN A PRESSURISED-WATER NUCLEAR POWER PLANT

DEGRADACIJA ZLITIN Ni-Cr-Fe V TLAČNOVODNIH JEDRSKIH ELEKTRARNAH

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In the early days of pressurized-water nuclear-power-plant design Ni-based alloys were selected because of their good mechanical properties and corrosion resistance. Alloy 600 was used for some reactor-coolant pressure-boundary components and Alloy 82/182 was used for welds. Industrial experience in the past three decades has shown that Alloy 600 components and Alloy 82/182 welds are susceptible to primary-water stress-corrosion cracking (PWSCC). PWSCC is the intergranular or transgranular cracking due to the combined action of stresses, temperatures and components in contact with the primary water (reactor coolant). PWSCC leaks and cracks were detected on the reactor-coolant pressure-boundary components. In this work some characteristics of the Alloy 600 and Alloy 82/182 welds and their PWSCC degradation are presented.

Key words: Ni-Cr-Fe alloy, stress corrosion cracking, nuclear power plant, repair

V začetni fazi konstruiranja tlačnovodnih jedrskih elektrarn so bile zaradi dobrih mehanskih lastnosti in korozijske odpornosti izbrane zlitine na osnovi Ni. Za nekatere komponente na tlačni meji reaktorskega hladila je bila izbrana zlitina z oznako Alloy 600, za zavarjene spoje pa zlitini z oznako Alloy 82/182. Izkušnje industrije z uporabo zlitine Alloy 600 in Alloy 82/182 v zadnjih treh desetletjih pa so pokazale, da so le-te občutljive za napetostno korozijsko pokanje. Napetostno korozijsko pokanje je pojav interkristalnih ali transkristalnih razpok, ki nastanejo zaradi skupnega vpliva napetosti, temperature in stika komponente z reaktorskim hladilom. Zaradi napetostnega korozijskega pokanja so bile na komponentah tlačne meje reaktorskega hladila odkrite razpoke in netesna mesta. Namen prispevka je predstavitev lastnosti zlitin Alloy 600 in Alloy 82/182, njihova uporaba v jedrskih elektrarnah ter napetostno korozijsko pokanje.

Ključne besede: zlitina Ni-Cr-Fe, napetostno korozijsko pokanje, jedrska elektrarna, popravilo

1 INTRODUCTION

Alloy 600 was originally selected for use in smaller-diameter piping penetrations in pressurized-water-reactor (PWR) nuclear plants because of its good corrosion resistance and a coefficient of thermal expansion similar to that of low-alloy steel vessels and piping material. Most of these penetrations are attached to the vessel or piping with Alloy 82 or 182 (nickel-chromium-iron) J-groove welds. Alloy 82/182 weld materials have also been used for field-butt welds.

Primary-water stress corrosion cracking (PWSCC) of Alloy 600 nozzles and Alloy 82/182 weld metal became a source of concern in non-steam generator tubing in the mid-1980s. Some significant PWSSC events in the past twenty years are listed below¹:

- 1991 – A through wall crack leak to the top of the reactor vessel was detected during a pressure test at the nuclear power plant Bugey 3 in France.
- 2000 – Cracks were discovered in Alloy 182 welds joining the low-alloy-steel reactor vessel hot-leg nozzles to stainless-steel pipes at Ringhals 4 (Sweden) and VC Summer (United States).
- 2002 – The most severe event was the NPP Davis-Besse case. PWSSC and the significant boric acid corrosion of carbon steel material caused a serious degradation of the reactor vessel closure head.

- 2003 – A small leak was discovered on a pressurizer relief nozzle at Tsuruga 2 (Japan) due to an axial crack in the Alloy 182/82 butt weld between the low-alloy-steel nozzle and the stainless-steel relief-valve pipeline.
- 2005 – Calvert Cliffs NPP (United States) identified crack indications in an Alloy 182/82 dissimilar metal weld on a 2-in. (51-mm) diameter hot-leg drain nozzle. Two axial crack indications were contained entirely within the weld and butter area.

The list of components that are known to contain Alloy 600/82/182 in at least some nuclear-power-plant designs includes:

- reactor vessel heads,
- reactor vessel hot-leg and cold-leg nozzles,
- reactor vessel bottom-mounted instrument penetrations,
- steam generator primary nozzles and tubes,
- pressurizer and heat exchangers,
- reactor coolant loop-pipe branch connections.

Some of the generic locations of Alloy 600/82/182 are shown in **Figure 1**.

The main types of affected welds are:

- J-groove welds of the control rod drive mechanism (CRDM) penetrations (**Figure 2**), reactor vessel

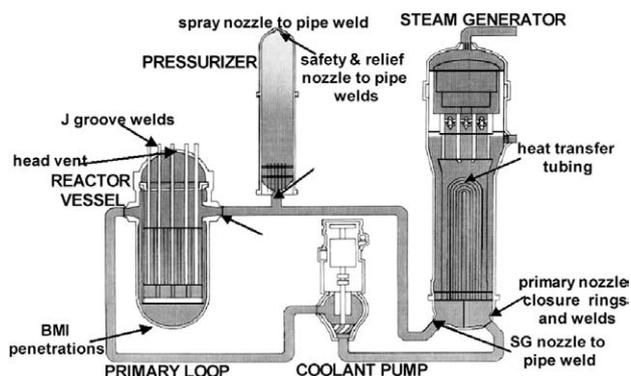


Figure 1: Generic locations of Alloy 600 components and Alloy 182/82 welds

Slika 1: Splošne lokacije komponent iz zlitine 600 in zvarov zlitin 82/182

bottom-mounted instrumentation penetrations (BMI), steam generator (SG) drain lines, pressurizer instrument nozzles and hot-leg instrument nozzles;

- butt welds or full-penetration dissimilar-metal welds of reactor pressure vessels and pressurizer nozzles (Figure 3).

Alloy 600

Inconel Alloy 600 with UNS N06600 or W. No. 2.4816 is a standard nickel-chromium-iron engineering material for heavy-duty applications. The limiting chemical composition² of the Alloy 600 is shown in Table 1.

Table 1: Alloy 600 chemical composition in mass fractions (w%)

Tabela 1: Kemijska sestava Alloy 600 v masnih deležih (w%)

Ni	Cr	Fe	C	Mn	S	Si	Cu
>72	14–17	6–10	≤0.15	≤1.0	≤0.015	≤0.5	≤0.5

Alloy 600 is a stable, austenitic solid-solution material. The high nickel content gives the alloy a good corrosion resistance in many organic and inorganic compounds. Chromium provides the resistance to sulphur compounds and the resistance in oxidizing conditions at high temperatures. The alloy can be hardened and strengthened only by cold work.

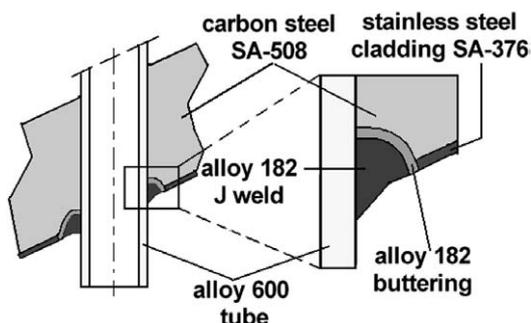


Figure 2: J-groove weld on a reactor-vessel head

Slika 2: J-zvar na glavi reaktorske posode

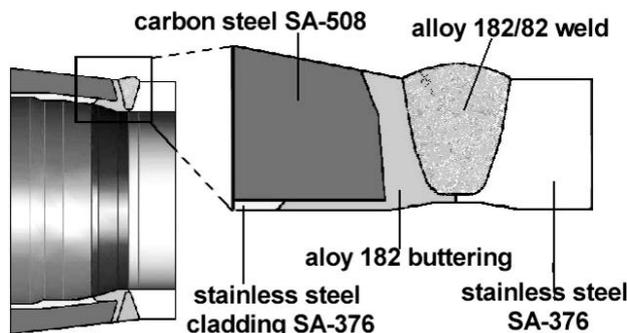


Figure 3: Nozzle-to-pipe dissimilar-metal weld

Slika 3: Zvar med cevnim nastavkom in cevovodom iz različnih materialov

The phases precipitating in the microstructure are titanium nitrides, titanium carbides and chromium carbides. The nitride particles are stable at all temperatures below the melting point and are unaffected by any heat treatment. At temperatures between 540 °C and 980 °C the chromium carbide precipitates from the solid solution at the grain boundaries and in the matrix. Figure 4 shows the austenitic microstructure of Alloy 600 with carbide particles visible in the polished and etched metallographic specimen.

Alloy 600 components can be fabricated by press forging, hammer forging, hot rolling, forming and machining from a bar product (ASME II SB-166) or cold drawing and hot finishing from a pipe and tube product (ASME II SB-167).² The typical mechanical properties³ for various forms and conditions are listed, for information only, in Table 2.

The alloy's strength and oxidation resistance at high temperatures make it useful for many applications for

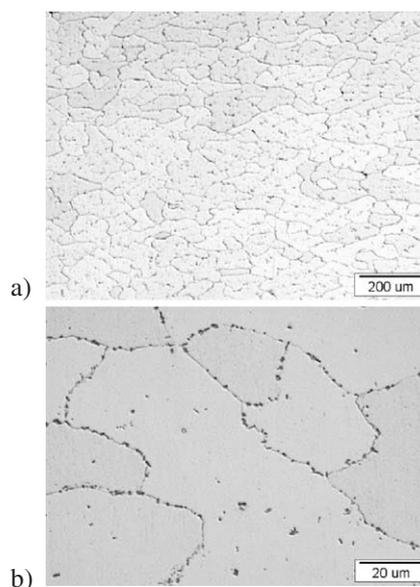


Figure 4: Alloy 600 austenitic microstructure (a) with chromium carbide particles at the grain boundaries (b)

Slika 4: Avstenitna mikrostruktura zlitine 600 (ao) z zrni kromovega karbida na mejah kristalnih zrn (b)

Table 2: Mechanical properties of Alloy 600 (for information only)**Tabela 2:** Mehanske lastnosti zlitine 600 (samo za informacijo)

Product Form	Condition	Tensile strength (MPa)	Yield strength (MPa)	Elongation (%)
Rod and Bar				
Cold drawn	Annealed	550–690	170–345	55–35
	As drawn	724–1034	550–860	30–10
Hot finished	Annealed	552–689	205–345	55–35
	Hot finished	586–827	240–620	50–30
Plate				
Hot rolled	Annealed	55–725	205–345	55–35
	As rolled	580–760	240–450	50–30
Tube and Pipe				
Hot finished	Hot finished	520–690	170–345	55–35
	Annealed	520–690	170–345	55–35
Cold drawn	Annealed	550–690	170–345	55–35

furnace components in the heat-treatment industry. In the aeronautical industry Alloy 600 is used for a variety of engine and airframe components that must withstand high temperatures, for example, exhaust liners and turbine seals.

In pressurized-water reactor (PWR) nuclear power plants, Alloy 600 has been used for steam-generator tubes, control rod drive mechanism (CRDM) nozzles, reactor vessel bottom mounted instrument (BMI) penetrations, pressurizer heater sleeves and other pressure-retaining components.

The Alloy 600 was selected for use in nuclear power plants because of:

- Its good mechanical properties, similar to those of austenitic stainless steels.
- Its good general corrosion resistance in high-temperature water environments and resistance to caustic stress-corrosion cracking better than austenitic stainless steels.
- It can be welded to carbon, low-alloy and austenitic stainless steels.
- It is a single-phase alloy requiring no post-weld heat treatment, also when submitted to post-weld heat treatments required for low-alloy steel parts to which it is welded. The resulting sensitization (decreased chromium levels at grain boundaries associated with the precipitation of chromium carbides at the boundaries) does not result in a high susceptibility to chloride attack exhibited by austenitic stainless steels exposed to such heat treatments.
- Its thermal expansion properties, between those of carbon/low-alloy steels and austenitic stainless steels, make Alloy 600 a good transition metal between these steels.

2 PRIMARY-WATER STRESS-CORROSION CRACKING (PWSCC)

PWSCC is the initiation and propagation of intergranular cracks through the material in a seemingly

brittle manner, with little or no plastic deformation of the bulk material and without the need for cyclic loading. Generally, it occurs at stress levels close to the yield strength of the bulk material, but does not involve significant material yielding.

An analysis of damaged components showed that PWSCC has a tendency to occur most quickly in parts which were¹:

- fabricated from more susceptible or higher-strength materials,
- machined or cold worked prior to welding,
- installed using methods that can produce high residual stresses, such as welding or roll expansion,
- operating at high temperatures,
- not stress relieved after installation.

Over the past three decades a significant number of laboratory studies and industry events reported on the PWSCC failure mechanism for components from Alloy 600 with Alloy 182/82 welds. The results of the tests and data from industry have shown that the occurrence of the PWSCC depends on the simultaneous contribution of:

- susceptible material,
- corrosive environment (primary water (reactor coolant)),
- high stresses including residual stress and operating stress.

2.1 Material

The fabrication process, heat treatments, and chemical compositions affect the formation of the material's microstructure and are the main contributors to the PWSCC susceptibility. Alloy 600 bars, rods, plates, pipes and stripes are usually heat treated to reduce the yield strength and increase the material toughness to an acceptable level. A higher heat-treatment temperature and a longer duration result in a lower yield strength. The fabrication process, heat treatment, and chemical compositions affect the formation of the material's

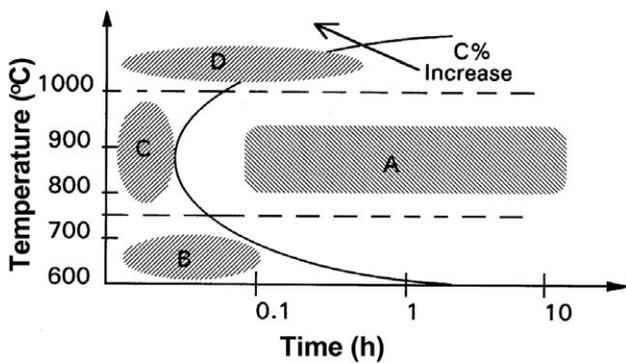


Figure 5: Carbide-precipitation diagram for Alloy 600
 Slika 5: Diagram izločanja karbidov za Alloy 600

microstructure and are the main contributors to the PWSCC susceptibility.

The heat-treatment temperature, annealing time and the carbon content are interrelated and affect the alloy's microstructure. A carbide-precipitation diagram for the Alloy 600 material shown in Figure 5 could be used to assess the effect of the heat-treatment temperature and time on the microstructure.^{4,5} The kinetics of precipitation depends on the velocity of the diffusion processes, which is greater at high temperature. The amount of carbide precipitates depends on the annealing time.

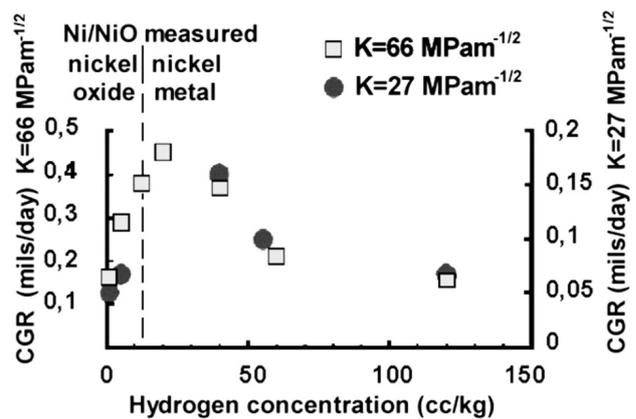
A high-temperature (1066 °C) heat treatment for a sufficient time (zone D in Figure 5) could form an intergranular carbide-particles network without the de-chromization of the area of the adjacent grain boundary. Stress-relief annealing in the range 700–800 °C in zone C in Figure 5 also limits the intragranular carbide formation and improves the material's resistance to PWSCC. A post-fabrication heat treatment in zones B and A will result in PWSCC-susceptible material.

The mass fraction of carbon-content ($w(C)/\%$) is an important factor in the microstructure formation of Alloy 600 due to heat treatment. The lower $w(C)$ moves the x -curve of the carbide-precipitation diagram in Figure 5 to the right and improves the conditions for intergranular carbide formation.

Generally, in Alloy 600 a good grain-boundary carbide network increases the PWSCC resistance. However, the susceptibility of Alloy 600 also depends on the surface cold work due to machining, grinding and reaming. A surface layer with a high cold work of the highly susceptible material, for example, components that were machined from bar stock and with weld root grinding, is considered to be highly PWSCC susceptible.

2.2 Environment

There are several environmental parameters that are influential on the PWSCC initiation and growth. The most significant is the primary-water temperature. The results of laboratory tests indicated that PWSCC is a



Note: 1 mil/d is $\approx 1.0 \cdot 10^{-6}$ m/h or $\approx 29 \cdot 10^{-10}$ m/s

Figure 6: Influence of hydrogen concentration on the crack growth rate of PWSCC in Alloy 600 at 338 °C

Slika 6: Vpliv koncentracije vodika na hitrost napetostnega korozijskega pokanja za zlitino 600 pri temperaturi 338 °C

thermally activated process and that the crack initiation and growth rate are strongly temperature dependent.⁶

Like many temperature-dependent processes, the correlation between the PWSCC growth rate and the temperature can be expressed with the Arrhenius equation⁷:

$$\dot{a} = C \exp\left(-\frac{Q}{RT}\right) \quad (1)$$

where:

\dot{a} = growth rate

Q = activation energy for the crack growth phase

R = ideal gas constant

T = temperature (K)

C = constant

The other influential environmental parameters are chemical additives to the primary water. The chemistry of the primary water (reactor coolant) is maintained by the chemical and volume control system, which is designed to allow the operators to regulate the water's chemical composition. The major use of this system is to control the primary-water boron content as a function of the nuclear reactor's power level. With the addition or removal of lithium hydroxide the reactor coolant's pH value is controlled. The system is also designed to allow the addition of hydrogen during normal operation. Hydrogen gas is dissolved in the reactor coolant to scavenge all the dissolved oxygen, which may be present in the primary water. The effect of the lithium concentration and pH value on the PWSCC is minimal.⁸

Tests using crack growth rate (CGR) specimens have shown that crack growth tends to be faster when the water's electrochemical potential, depending on hydrogen concentration, is close to the potential where the Ni/NiO phase reaction occurs.^{8,9} Higher or lower values of the hydrogen concentration decrease the crack growth rates (Figure 6).

2.3 Stress

Allowable stresses for a nuclear power plant component are specified in ASME Boiler and Pressure Vessel Code Section III. These requirements apply to operating loadings, such as internal pressure, differential thermal expansion, dead weight, and seismic loading. On the other hand, the industry design standards do not typically address residual stresses that can be induced in the parts during fabrication. These residual stresses are often much higher than the stresses in operation. In most cases the residual stresses are ignored by the standards since they are considered as secondary and self-relieving. However it is the combination of operating-condition stresses and residual stresses that lead to PWSCC.¹⁰ For the case of penetrations attached to the reactor-vessel heads by partial penetration J-groove welds, high residual stresses are caused by the surface machining prior to installation. This machining causes a thin, strongly deformed layer on the surface, increasing the material yield and the tensile strength near the machined surface.

The second source of residual stresses in the J-groove weld is shrinkage, which occurs when welding the nozzle into the high-restraint vessel shell, pulls the nozzle wall outward. This creates yield-strength level residual-hoop stresses in the nozzle base metal and higher-strength cold-worked surface layers. These high residual-hoop stresses contribute to the initiation of axial PWSCC cracks on the cold-worked surface layer and to the subsequent growth of these axial cracks in the lower-strength nozzle base material. Residual stresses in the nozzles and welds can lead to crack initiation from the inside surface of the nozzle, opposite from the weld and from the outside surface of the nozzle near or from the surface of the J-groove weld.

Based on the industrial experience and laboratory tests data, the crack growth rate model taking into consideration the temperature dependence and stresses was developed.¹¹⁻¹⁶ The recommended crack growth rate model of detected PWSCC flaws in thick-walled Alloy 600 components exposed to the primary water is:

$$\dot{a} = \exp \left[-\frac{Q_g}{R} \left(\frac{1}{T} - \frac{1}{T_{ref}} \right) \right] \alpha (K - K_{th})^\beta \quad (2)$$

where:

\dot{a} = crack growth rate at temperature T (m/s)

Q_g = 130 kJ/mole activation energy for crack growth

R = universal gas constant $8.314 \cdot 10^{-3}$ kJ/(mole K)

T = absolute operating temperature at the location of the crack

T_{ref} = absolute reference temperature used to normalize data 598.15 K

α = crack growth amplitude 2.67×10^{-12} at 325 °C

K = crack tip stress-intensity factor (MPa m^{1/2})

K_{th} = crack tip stress-intensity factor threshold 9 MPa m^{1/2}

β = exponent 1,16

2.4 Dissimilar metal welds

The conventional welding processes can be used to produce nickel alloy joints. Some of the characteristics of nickel alloys require the use of slightly different welding techniques than normally used for stainless-steel welds.

Weld Alloys 82 and 182 have been commonly used to weld Alloy 600 to itself and to other materials. These alloys are also used for nickel-based alloy weld deposits (buttering) on weld preparations and for cladding on areas such as the insides of reactor-vessel nozzles and steam-generator tube sheets. Alloy 82 with UNS N06082 or W. Nr. 2.4806 is a bare electrode material and is used for gas tungsten arc welding (GTAW), also known as tungsten inert gas (TIG) welding. Alloy 182 with UNS W86182 or W. Nr. 2.4807 is a coated electrode material and is used in shielded metal arc welding (SMAW). The compositions of the two alloys are different², leading to different susceptibilities to PWSCC. Alloy 182 has a lower chromium content (13–17 %) than Alloy 82 (18–22 %) and has a higher susceptibility to PWSCC, probably as a result of the lower chromium content. A comparison between these weld metals is shown in **Table 3**.²

Table 3: Comparison between Alloy 182 and Alloy 82 weld metals (w/%)

Tabela 3: Primerjava kemijske sestave Alloy 82 in Alloy 182 (w/%)

	Ni	Cr	Fe	Ti	Ni + Ta	C
Alloy 182	≥59	13–17	≤10	≤1.0	1.0–2.5	≤0.1
Alloy 82	≥67	18–22	<10	≤0.75	2.0–3.0	≤0.1
	Mn	S	Si	Cu	P	Co
Alloy 182	5–9.5	≤0.015	≤1.0	0.5	≤0.03	≤0.12
Alloy 82	2.5–3.5	≤0.015	≤0.50	0.5	≤0.03	≤0.10

The location of dissimilar-metal welds between low carbon and austenitic steel tubing and piping are shown in **Figure 1**. Such transition joints are necessary because of the corrosion resistance of stainless steel, while low-carbon steels are commercially more appropriate.¹⁷ For example, the reactor pressure vessel and steam

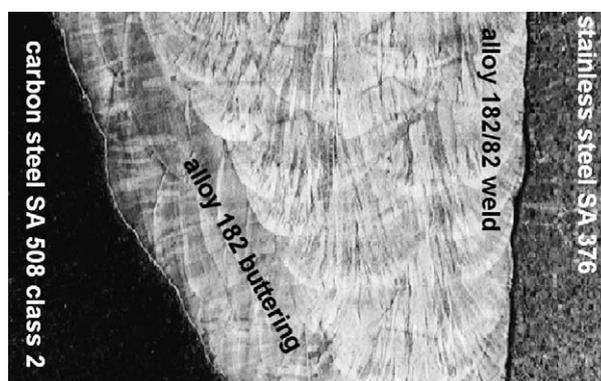


Figure 7: Dissimilar-metal weld cross-section

Slika 7: Prečni prerez zvara med različnima kovinskima materialoma

generators are made of low-carbon steels, whereas the primary piping is made of stainless steel. Therefore, to join the low-carbon steel components to stainless-steel piping Alloy 182/82 welding consumables were used (**Figure 3**). The macrostructure of a typical multiple-passes dissimilar-metal weld is shown in **Figure 7**.

A distinguished columnar pattern of dendrites can be seen through many weld passes. The dendrites, growing in the opposite direction to the heat flow, tend to be perpendicular to the base material at the weld–base-material boundary and tend to become vertical (root-to-crown direction) as the weld thickness increases. The dendrites in the centre of the weld are mainly vertical.

The boundaries between these similarly oriented dendrites are called solidification subgrain boundaries (SSGBs) and tend to have low angular mismatches, as well as low energy, and are believed to form paths relatively infrequently for PWSCC only. Where different sheaves of dendrites intersect or overlap, larger angular mismatches often occur between the grains. In this case, the resulting grain boundaries, termed solidification grain boundaries, can be high energy and are believed to be more common paths for PWSCC.

The EPRI study¹⁸ concluded that PWSCC crack growth rates for the alloy 82/182 weld metal behave in accordance with the following relationship:

$$\dot{a} = \exp\left[-\frac{Q_g}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] \alpha f_{alloy} f_{orient} K^\beta \quad (3)$$

where:

\dot{a} = crack growth rate at temperature T in m/s

Q_g = 130 kJ/mole activation energy for crack growth

R = universal gas constant 8.314×10^{-3} kJ/(mole K)

T = absolute operating temperature at location of crack, K

T_{ref} = absolute reference temperature used to normalize data 598.15 K

α = power law constant 1.5×10^{-12} at 325 °C (598,15 K)

f_{alloy} = 1.0 for Alloy 182 and 0.385 for Alloy 82

f_{orient} = 1.0 except 0.5 for crack propagation that is clearly perpendicular to the dendrite solidification direction

K = crack tip stress-intensity factor (MPa m^{1/2})

β = exponent 1.6

3 MITIGATION, REPAIR AND REPLACEMENT

3.1 Mitigation

Since PWSCC became a serious issue a number of techniques have been evaluated to delay or mitigate the occurrence of degradation processes. These techniques can be divided into three categories¹⁹:

1. Mechanical surface enhancement (MSE),
2. Environmental barriers or coatings,
3. Chemical or electrochemical corrosion potential (ECP) control.

MSE techniques represent processes that reduce surface tensile residual stresses or induce compressive surface stresses on a component or weld surface. Examples of MSE techniques are shot peening and electro-polishing. Environmental barrier or coating techniques represent processes that protect the material surface in aggressive environments. Coating examples include nickel plating and weld-deposit overlays. Chemical or ECP control techniques represent changes to the environment that alter the corrosion process or produce corrosion potentials outside the critical range for PWSCC. Examples of chemical or ECP control include zinc additions to the primary water and modified primary-water chemistry (e.g., dissolved hydrogen levels, lithium concentrations, and boron concentrations). In some nuclear power plants a component temperature reduction has also been applied.

3.2 Repair and replacement

Pressure boundary-components repair or replacement is the alternative to mitigation techniques, especially when a leakage is detected. ASME Code XI specifies that the flaws, detected during in-service inspection, must be removed or reduced to an acceptable size in accordance with Code-accepted procedures.²⁰ For PWSCC in Alloy 600/182/82 components, several approaches have been used, such as flaw removal, flaw embedment and weld overlay.

For relatively shallow or minor cracking, the flaws may be removed by grinding. This approach is going to eliminate flaws and return the component to ASME Code compliance. However, there will still be susceptible material exposed to the PWR environment that originally caused the cracking. Simple flaw removal is thus not meant to be a permanent solution, unless in future component replacement is planned.

One of the approaches to repair is to embed the flaw under a PWSCC-resistant material, typically Alloy 52 weld metal deposited over the susceptible Alloy 182/82

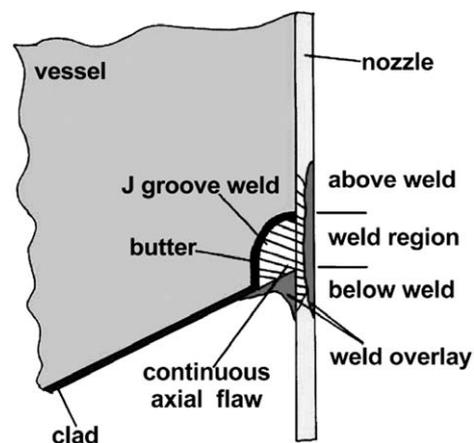


Figure 8: J groove weld-flaw embedment repair

Slika 8: Popravilo J-zvara s prekritjem napake

weld surface. The embedment must satisfy all ASME BPVC Section XI flaw-evaluation requirements.

Another form of repair that has been used extensively to repair cracked and leaking pipe welds is the weld overlay (**Figure 8**). The weld-overlay procedure has been recognized as a Code-acceptable repair in ASME Section XI.

In many cases mitigation techniques and/or repairs were successful. To satisfy Code and regulatory requirements, maintenance and inspection programs were specially developed and implemented for PWSSC-susceptible components. Additional maintenance and inspection activities mean more inspection costs for improved techniques to detect PWSSC and extra costs due to outage-time extension. The replacement of the components offers many advantages when considering the long-term operation of nuclear power plant. The new components are designed to eliminate all the susceptible materials, replacing them with PWSSC-resistant materials, such as Alloy 690 and the associated weld metals Alloys 52 and 152. The main components that have been replaced are steam generators, reactor vessel heads and pressurizers.

4 CONCLUSION

Alloy 600 component items were used in pressurized water reactors (PWRs) due to the material's inherent resistance to general corrosion in a number of aggressive environments and because it has a coefficient of thermal expansion very close to that of low-alloy steel. Over the past thirty years, primary-water stress-corrosion cracking (PWSSC) has been observed in Alloy 600 components and in Alloy 182/82 welds.

In some cases PWSSC cracks caused relatively quick and simple plugging of the leaking, small-diameter tubes in steam generators. On the other hand, many times PWSSC degradation resulted in long plant outages to replace leaking pressurizer heater sleeves, a leaking reactor-vessel hot-leg outlet nozzle weld and several CRDM nozzles. Many plants around the world have replaced their steam generators and reactor-vessel closure heads. The replacements were performed because of the high cost of repairs, the risk of leakage or the high cost of inspections necessary to ensure a satisfactory level of safety and reliability.

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