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Corrosion characterisation of laser beam and tungsten inert gas weldment of nickel base alloys: Micro-cell technique

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ABSTRACT

The electrochemical studies using micro-cell technique gave new understanding of electrochemical behaviour of nickel base alloys in solution annealed and welded conditions. The welding simulated regions depicted varied micro structural features. In case of tungsten inert gas (TIG) weldments, the weld fusion zone (WFZ) showed least corrosion resistance among all other regions. For laser beam (LB) weldments it was the heat-affected zone (HAZ) that showed comparatively high stable anodic current density. The high heat input of TIG welding resulted in slower heat dissipation hence increased carbide precipitation and segregation in WFZ resulting in high stable anodic current density.

1. Introduction

Majority of failures that take place in components made from nickel base alloys used in nuclear industry are due to pitting corrosion, intergranular corrosion (IGC) or stress corrosion cracking (SCC) [1–3]. The initiation of localised corrosion is strongly dependent on the stability of surface passive film. The macro electrochemical studies understand pitting behaviour of alloys and provide information with respect to overall corrosion behaviour of area exposed to environment whereas the nucleation sites would be a microscopic region such as inclusion, precipitates and grain boundaries [4]. The electrochemical studies if carried out on these heterogeneities would give a better insight into the mechanistic aspect of localised corrosion [5,6]. A lot of research is going on in this direction and quite a number of techniques have developed for assessing corrosion at a local area [7–9]. These methods have an advantage of measuring the currents at resolution of pico ampere to femto ampere (pA and fA) range [10,11]. The nickel base alloys presently studied are generally used for high temperature and corrosion resistant applications such as steam generator tubing and other structural components in nuclear reactors. Since these alloys show active–passive behaviour they are prone to localised attack such as pitting and IGC, which may then result in SCC initiation [12–18]. One of the most challenging subjects of nickel-base super alloys in nuclear application is SCC of dissimilar material welds. Nickel-base alloys (e.g., Alloy 82/182 and Alloy 52/152, the filler materials for Alloy 600 and Alloy 690, respectively) are often used to weld carbon steels (e.g., A 533 Grade B and A508) to stainless steel (e.g., Types 304 and 316) in various locations in nuclear reactors. The present study aims at understanding the passive film stability and pitting behaviour of nickel base alloys such as Alloy 600 and Alloy 690 in the (a) coarse grained condition and (b) their weldments using the micro-cell technique. Presently no literature is available on using micro-cell technique for corrosion evaluation of weldments of nickel base alloys.

2. Experimental procedure

Alloy 600 and Alloy 690 plates were solution annealed (SA) at 1100 °C for 30 min and water quenched to get a carbide free microstructure. These sheets were analysed for chemical composition as per ASTM E1473 [19]. The chemical compositions for the alloys are summarised in Table 1. Specimens of 1 cm^2 area were cut and subjected to further heat treatment so as to obtain coarse grains. The alloys were heated to 1100 °C for 1 h in a quartz tube (filled with argon atmosphere) and water quenched. Grain size was measured using the linear intercept method described in ASTM E112. The metallographic studies on Alloy 600 and Alloy 690 in heat treated and welded conditions were done after electrolytic etching in 10 wt.% oxalic acid for 60 s at 1 A/cm^2.

The autogenous welding of solution annealed (1100 °C for 30 min and water quenched) plates of 1.5 mm thickness were carried out using tungsten inert gas (TIG) welding and laser beam (LB) welding techniques. Both the welding was done by scanning an

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Table 1
Chemical composition of Alloy 690 and Alloy 600 plates (wt.%).

Element (wt.%)	Ni	Cr	Fe	C	Si	Ti
690	61.14	29.56	8.02	0.02	0.93	0.33
600	71.2	17.03	8.71	0.06	0.8	0.35

arc/laser beam over the length of plate so as to get through thickness melting of plate. This generates different microstructures that are observed in a weldment. The TIG process was performed in a pure argon shielding gas using the following welding parameters: 130–135 A, 16–18 V and welding speed 80–90 mm min^{-1}. The LB welding process used a Neodymium–doped yttrium aluminium garnet (Nd–YAG) laser with a constant mean power of 800 W and a travel speed of 600 mm min^{-1}.

The electrochemical studies using the micro-cell technique [20,21] had a standard three-electrode cell configuration. It has a saturated silver–silver chloride (Ag/AgCl) (in saturated potassium chloride (KCl)) as a reference electrode, platinum as counter electrode and transverse section of the weldment or top surface of the coarse-grained alloy (1100 °C for 1 h and water quenched) mounted in a cold setting resin as a working electrode. A micro-electrochemical cell comprises of a pulled micro-capillary filled with electrolyte. The tip diameter of capillary was in the range of 100–110 μm. A layer of silicone rubber is applied as a sealant between the front end of the micro capillary and the surface of interest. The micro-electrochemical cell assembly is fixed at one of the revolving turrets, replacing an objective of a microscope while the specimen is mounted on the microscope stage. The electrochemical measurements were done using a low current measuring potentiostat at a scan rate of 1 mV s^{-1}. The different regions of interest in the weldment were studied by placing the micro capillary tip at base metal, heat affected zone (HAZ) and weld fusion zone (WFZ) locations. The microstructure of the weldment was developed by electrolytic etching in oxalic acid and HAZ, WFZ and base metal regions were identified. The demarcation of HAZ was done by optical microscopy. The region immediately next to weld fusion line had coarse grains and HAZ had finer grain size. Subsequently the base metal had slightly higher grain size than that in HAZ. A transparent sheet was placed over the etched weldment and the three regions were marked. For micro-corrosion studies of a given region of interest, the remaining regions of the polished specimen identified by using the transparent sheet were masked with a lacquer exposing the region of interest for the study. In micro-cell studies the tested area would change but not very drastically as the area change depends on the flexibility of the silicone sealant applied at the tip of the micro-cell. Therefore, after each test the exposed area was measured. The exposed area for microcell experiment was defined, after the test, by measuring the attacked area under an optical microscope (using a grid).

For the experiments to establish the behaviour of grain boundary and grain matrix the microstructure was developed by electrolytic oxalic acid etching and the bigger grains were identified by measuring the co-ordinates from the sample edge. Measurements were done at different locations at those co-ordinates after polishing to a fine finish using a diamond paste to get the best result which gave the maximum grain matrix or grain boundary for examination by the micro-cell. Confirmation of whether the test was actually done inside a grain or at a grain boundary was made by examination under an optical microscope after the test. The electrochemical polarisation caused attack at grain boundaries thus revealing these features. In micro-cell studies for grain boundary the focus was more on length of the grain boundary that gets exposed under the micro-cell. If the ratio of the grain boundary to the grain matrix is high then there would be more contribution to measured current from the grain boundary rather than that from the grain matrix. As the measurements inside a grain was made after coarsening the grain by a high temperature annealing treatment, the micro-cell probe could be placed inside a grain without intercepting the grain boundary.

The macro electrochemical studies describe bulk electrochemical measurements (as per G5, ASTM) as opposed to the "microcell". Macro cell studies were carried out using the conventional electrochemical Greene cell. The specimens were mounted in a cold setting resin with an approximate area of 1 cm^2 ground on silicon carbide papers and finally polished with a diamond paste of 1 μm size. Precautions were taken to avoid any crevice formation at the specimen/mount interface by applying a lacquer. Platinum disc was used as a counter electrode and saturated calomel electrode (SCE) as a reference electrode. The electrochemical studies on the weldments were done using 0.5 M H_2SO_4 + 0.25 M NaCl solution freely exposed to air at room temperature at a scan rate of 1 mV s^{-1}.

3. Results and discussion

3.1. Microstructural examination

The examination of microstructures for Alloy 600 and Alloy 690 in the as-received condition showed grains, which were small. The measured average grain size values are tabulated in Table 2 for as received and all heat-treated conditions. The data in Table 2 shows an increase in grain size due to grain growth, with increased exposure time when annealed at 1100 °C. Micrographs show microstructure in as received; solution annealed and welded conditions of Alloy 690 and Alloy 600 in Figs. 1 and 2 respectively. The WFZ of the TIG and LB welded samples showed characteristic dendritic features for Alloy 690 and Alloy 600 (Figs. 1d, f and 2d, f respectively). In the case of TIG, weld dendrites showed large lamellae width (Figs. 1d and 2d). In the LB weld, WFZ showed distinctly three different modes of solidification such as equiaxed region at centre surrounded by a lamellar region and an outermost region of a chilled zone (Fig. 1f). The WFZ of LB-welded samples were narrow in width (around 2 mm) since the heat input for LB weld was less and therefore experienced high cooling rates. On the contrary, TIG welds were wider, (~6 mm). Similar was the case with respect to HAZ regions in both the type of weldments (2–3 mm wide for the TIG weldment and 0.5 mm wide for LB weldment). The heat input calculated for TIG, LB weldments were 1147 J/mm, and 68 J/mm respectively based on the heat input equations given by Fuerschbach [22,23].

3.2. Grain matrix and grain boundary corrosion

Conventional electrochemical techniques lack the ability to distinguish the kinetics of electrochemical corrosion processes taking place on different zones of a typical weldment and grain matrix versus that at grain boundary. The current scientific interest is to pin point these heterogeneous electrochemical behaviour and identify a microstructure that results in the most intense localised

Table 2
Average grain size tabulated for Alloy 600 and Alloy 690 using the linear intercept method.

Material grain size (μm)	As received (μm)	Solution annealed 1100 °C/½ h (μm)	Solution annealed 1100 °C/1 h (μm)
Alloy 600	60	63	106
Alloy 690	33	73	126