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Ni-Base Filler Metal 52

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OPTICAL AND ANALYTICAL ELECTRON MICROSCOPY OF DUCTILITY-DIP CRACKING IN NI-BASE FILLER METAL 52—INITIAL STUDIES

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Abstract

Microcharacterization studies were performed on weld-metal microstructures of a Ni-base filler metal. Specimens were taken from the fusion zone and the weld-metal heat-affected zone of transverse- and spot-Varestraint welds. The filler metal was first deposited onto a steel substrate by hot-wire, gas tungsten arc welding before specimen removal. Optical microscopy indicates the crack morphology is intergranular and is along high-angle, migrated grain boundaries. At low magnifications, scanning electron microscopy reveals a relatively smooth fracture surface. However, at higher magnifications the grain faces exhibit microductility. Analytical electron microscopy reveals high-angle, migrated grain boundaries decorated with MC (Ti, Cr) and $M_{23}C_6$ (Cr, Ni, Fe) precipitates ranging from 10 to 200 nm. Auger electron spectroscopy of pre-strained Gleeble specimens fractured *in situ* revealed internal ductility-dip cracks decorated with magnesium aluminate ($MgAl_2O_4$) spinel particles (1000 nm).

Introduction

In 1912, Bengough first reported the tendency for normally plastic metals and alloys to exhibit a ductility minimum in the intermediate temperature range [1], however to date the mechanism is not understood. It has also been known for decades that ductility-dip cracks (DDC) can form on cooling from welding in austenitic weld metals [2,3]. Ductility-dip cracks reportedly have a "smooth" fracture surface at low magnifications, yet at higher magnifications they exhibit microductility. They are most commonly reported in multipass or repair welded austenitic materials, and the fracture surface is not obviously decorated with any one solute, second-phase particle, or impurity element that appears to cause cracking.

Hemsworth *et al.* [3], classified ductility-dip cracking into three categories: cracks occurring in the primary weld metal (fusion zone); cracks occurring in the heat-affected zone (HAZ); and cracks occurring in the weld-metal HAZ. Using optical microscopy, the authors further described cracking as occurring along migrated grain boundaries. Scanning electron microscopy

provided definite proof that the boundaries were free from liquid films.

Several researchers have attempted to study the effects of chemical composition on the cracking behavior of austenitic steels and Ni-base alloys [2-12]. Results of several studies indicate that increased C content appears to lower the cracking tendency of fully austenitic stainless steels [2,3,7]. Abralov and Abdurakhmanov reported that cracking always appeared perpendicular to the applied stress, and that "purer" Ni-base alloys are more crack resistant [4,5]. In other studies, Gooch and Honeycombe, Arata *et al.*, and Matsuda *et al.*, have all reported that additions of Mn and Si to fully austenitic stainless steels result in less cracking [6-10]. A study of Ni-base weld metals by Nakao *et al.*, suggests that maintaining Cr levels at 10 wt% appears beneficial to reducing cracking [11,12]. Their results indicate that increased Cr levels harden the grain matrix thereby localizing strain and cracking at the grain boundary. The authors further noted that decreased levels of Cr resulted in S segregation to the grain boundaries, again promoting the occurrence of cracking.

Rhines and Wray used elevated temperature tensile testing to produce ductility-dip cracking in wrought metals [13]. They tested specimens over a range of temperatures and rapid loading rates, and were able to successfully reproduce the ductility dip phenomenon in cartridge brass and Monel. The authors proposed that ductility dip is a creep-related phenomenon caused by grain boundary shearing at higher strain rates than normally associated with creep. They describe the ductility dip in the following manner: At temperatures below the ductility dip, grain boundary shearing is not possible. At temperatures above the ductility dip, recrystallization is rapid enough, that grain boundaries are constantly reforming, and voids do not have time to link into cracks.

If the study of ductility-dip cracking is to be complete, then consideration must be given to the chemistry and structure of the material. In this paper, simulative weldability testing and analytical microscopy are used to better understand the role of chemical composition on ductility-dip cracking in a Ni-base filler metal. The role of structure (*i.e.*, grain and grain-boundary orientation) will be addressed in the follow-on study.

Table 1. Nominal Composition of Filler Metal 52 (wt%)

Ni	Cr	Fe	C	Mn	Al	Ti	S	P	Si	Cu	Mo	Nb+Ta	Co	B	Mg
bal.	27-31	7-11	0.05	0.5	0.5-1.1	0.2-0.7	0.002	0.01	0.05-0.3	0.1	0.1	0.1	0.02	0.006	0.01
Single values are maximum															

Experimental Procedure

The material used in this study was Filler Metal 52 (FM52), and its nominal composition is given in Table 1. The material was deposited onto a steel substrate until 1-inch thick by hot-wire, gas-tungsten arc welding (GTAW) in a shielding gas of 95% argon-5% hydrogen. Hydrogen aids in scavenging residual oxygen from the weld pool surface. Subsequently, the weld-metal buildup was removed from the steel substrate and machined into several plates each measuring 203 x 76 x 6-mm. Affects from dilution with the steel substrate were considered negligible due to specimen removal near the top of the buildup.

Fusion-zone ductility-dip cracks were generated using the transverse-Varestraint test. The transverse-Varestraint test consists of making a 38-mm long, autogenous GTA weld in 100% Ar shielding gas parallel to the 76-mm dimension of the specimen. When the weld length reached 32 mm, the test specimen was bent around a radiused die block to a specific percent strain at a given bending rate—welding continued until the weld length reached 38 mm. All welds were made using a travel speed of 152 mm/sec, and a bend rate of 254 mm/sec.

Weld-metal HAZ ductility-dip cracks were generated by first making an autogenous GTA weld in a similar fashion to that described above for transverse-Varestraint testing, however, no bending was involved. The autogenous weld provides the required weld-metal HAZ microstructure associated with multipass weldments. Next, spot-Varestraint testing was performed by producing a GTA spot weld in 100% Ar shielding gas at the center section of the preplaced autogenous weld. After a predetermined weld time, the GTA spot weld was extinguished and the specimen was forced to conform to a radiused die block to a specific percent strain at a bend rate of 254 mm/sec.

Gleeble hot-ductility tests were conducted to evaluate the hot-ductility behavior during welding. Additional testing was performed by thermal cycling a specimen to a peak temperature of 1295 °C, cooling to 1000 °C then straining it at a rate of 25 mm/sec to a total strain of 40 percent. All testing was performed in an argon-rich atmosphere. The test was not designed to rupture the specimen, but rather strain it enough to generate internal ductility-dip cracks for later observation in the Auger electron microscope. Optical microscopy verified the presence of internal ductility-dip cracks. Detailed test procedures for transverse- and spot-Varestraint, and Gleeble testing can be found in a study by Lin *et al.* [14].

Optical Microscopy – Each transverse- and spot-Varestraint specimen was examined at magnifications up to 40X using a binocular microscope. These specimens were examined as tested to identify the type, size and distribution of ductility-dip cracks present. Select specimens were metallographically prepared and electrochemically etched at 6 volts in a solution of 5-ml acetic acid, 10-ml nitric acid, and 85-ml distilled water at 21 °C.

Scanning Electron Microscopy – Fracture surfaces from select Varestraint and Gleeble specimens were examined in either a Zeiss DSM 960A or in a Jeol 6300FX scanning electron microscope (SEM) operated between 10-30 kV. Energy dispersive X-ray analysis was also conducted using a PGT system on select microstructural constituents using spot analysis techniques.

Analytical Electron Microscopy - Regions within the fusion zone and weld-metal HAZ were examined, and the size and morphology of major constituents present identified. These specimens were examined in a Philips CM30 transmission electron microscope (TEM) operated at 300 kV. Energy dispersive X-ray analysis was also conducted using a Kevex system on select microstructural constituents using spot analysis techniques.

Auger Spectroscopy - A Model 5600ci Physical Electronics Auger electron microscope was used to examine a Gleeble specimen pre-strained (~40%) on cooling at 1000 °C. The specimen was subsequently fractured *in situ* which revealed internal ductility-dip cracks generated during thermal cycling. Auger analysis of the crack surfaces was performed using a 9 kV incident electron beam coupled with detection of the secondary electrons by a spherical capacitor energy analyzer. The spectrum of kinetic energies from the topmost 20-75 Å of surface atoms and their energy levels were acquired using a multi-technique surface analysis apparatus. The spectrum was differentiated to enhance the peak content.

Results

An on-cooling hot-ductility curve for FM52 is presented in Figure 1. This material quickly recovered its ductility during weld cooling from its nil-strength temperature. There is a ductility dip at temperatures ranging from 800 °C to 1000 °C. The temperature range for this ductility dip is consistent with temperature range associated with the ductility-dip cracking in both transverse- and spot-Varestraint specimens.

Figure 2 documents the appearance of transverse- and spot-Varestraint specimens after welding. Ductility-dip cracks in the fusion zone and the weld-metal HAZ of these specimens are easily differentiated from solidification and liquation cracking. Solidification and liquation cracks are located adjacent to the weld pool at the moment of loading. Ductility-dip cracks are located some distance away from the weld pool, and separated from the solidification and liquation cracking by a band of sound weld metal because they occur at a lower temperature. Cracking extended from approximately 4 to 12 mm from the instantaneous solid/liquid interface. This cracking distance corresponds to a ductility-dip temperature range (DTR) between 1050 °C and 900 °C.

Figure 1. On cooling hot-ductility curve for Filler Metal 52. Note ductility dip between 800 °C and 1000 °C.

The weld-metal buildup, the fusion zone and the weld-metal HAZ microstructures comprised an austenitic, cellular solidification structure, migrated grain boundaries and intercellular particles (Figure 3). These particles range in size from 500 nm to 1000 nm. In Figure 3, it is apparent that the grain boundary in the upper portion of the micrograph has migrated because it cuts across solidification subgrains.

Figure 2. Optical micrographs of: a) transverse-Varestraint specimen; and b) spot-Varestraint specimen, both tested at 7% strain and a bend rate of 254 mm/sec.

The fracture surface morphology of Gleeble specimens tested between 800 °C and 1000 °C exhibit a flat fracture at low magnifications (Figure 4) but have evidence of shallow, micro-dimples at higher magnifications (Figure 5). This dimple morphology implies that fracture occurred in the solid state, and the ductility of the material adjacent to the grain boundary was

relatively high. However, the macroscopic ductility is low as seen by the small reduction in area in Figure 4. From Figure 5, the fracture surface resembles that of a material exposed to an oxidizing atmosphere while at an elevated temperature.

Figure 3. SEM micrograph of the fusion zone microstructure from a transverse-Varestraint specimen. Note intercellular particles and particles along a migrated grain boundary (arrows).

By comparing the fracture surface of a specimen tested in an argon-rich atmosphere (Figure 5) with a specimen-fractured *in-situ* in the Auger electron microscope (Figures 6), subtle differences are revealed. The surface of the specimen-fractured *in-situ* exhibits a similar flat fracture at low magnification and micro-dimple morphology at higher magnification. However, at somewhat higher magnifications fine, octahedral-shaped particles residing in some of the micro-dimples are revealed (Figure 6). Also, the depth of the dimples on the *in-situ* fracture appears deeper. The shallow appearance of the dimples on the oxidized fracture surface is most likely due to the oxide filling the dimples.

The octahedral-shaped particles exposed during *in-situ* fracturing were isolated in the Auger electron microscope, and compared to the surrounding matrix and laboratory fracture. Figure 7 represents the spectrum of kinetic energies of the matrix from within a ductility-dip crack. Comparisons of the matrix energy spectrum with that taken from the laboratory fracture indicates an almost exact similarity between surface compositions. Figures 8 and 9 are energy spectrums from two separate particles on the DDC fracture surface. A total of five particles were analyzed. Mg was always present on the particles, whereas Al was more varied. In Figure 9, a large Mg peak and the absence of Al are observed. The oxygen signal for this particle is very strong.

The particles were also isolated in the TEM and identified by EDS analysis as rich in Mg and Al. These large, square

Figure 4. SEM fractograph of a Gleeble specimen tested at 1000 °C on cooling in an argon-rich atmosphere. Note flat fracture features and relatively large grain size. Fracture was completely intergranular.

Figure 5. Higher magnification SEM fractograph of selected area in Figure 4. The fine details of the surface are obscured by the remnants of a partially oxidizing atmosphere.

shaped particles were found in the matrix, migrated and solidification grain boundaries (Figure 10). From two separate selected-area diffraction (SAD) patterns a lattice constant of 8.06 Å was derived. This corresponds to MgAl_2O_4 , a spinel with a FCC diamond cubic microstructure.

TEM analysis of grain boundaries and their associated particles from the fusion zone of transverse-Varestraint specimens are presented in Figure 11. A bright-field (BF) image exhibits a string of discrete MC-type cubic particles, and an area where the grain boundary has migrated away from some of the articles (Figure 11). EDS analysis identified the particles as rich in Ti and Cr.

Weld-metal grain boundaries from spot-Varestraint specimens are presented in Figures 12 and 13. A centered dark field (CDF) image of a high-angle grain boundary decorated with M_{23}C_6 -type particles is shown in Figure 12. The zone of mismatch for this boundary was about 28° between the two grains. Not all high-angle grain boundaries contained particles (Figure 13). This boundary has migrated as evidenced by its intersection with a solidification subgrain.

Figure 6. Octahedral-shaped particle from DDC fracture surface. EDS and TEM analysis indicate similar particles to be a magnesium aluminate spinel (MgAl_2O_4).

Discussion

The results indicate that ductility-dip cracking is most certainly a solid-state phenomenon. In the present work, optical and scanning electron microscopy verified the presence of ductility-dip cracks along high-angle, migrated grain boundaries. These observations correlate well with the findings of other researchers [2-12]. Moreover, SEM revealed flat fracture morphology at low magnifications, and shallow microdimples on grain faces at higher magnifications. The shallow dimple morphology appears to indicate that strain was localized to the grain boundary region. This strain localization is consistent with the orange-peel effect observed on as-tested transverse- and spot-Varestraint specimen surfaces.

Gleeble testing in conjunction with Auger electron spectroscopy of *in-situ* fractured Gleeble specimen indicate that elemental segregation does not appear to play a role in the formation of ductility-dip cracks on-cooling from welding. This is counter to the findings of Nakao *et al.*, where they observed S segregation to the grain boundaries in Ni-base weld metals [11,12]. However, the current findings are consistent with the premise that if cracking was segregation related, the material would not completely recover its ductility upon cooling below the ductility dip. However, the material ductility does recover at temperatures below 800 °C.

Figure 7. Energy spectrum from DDC matrix surface exposed during *in-situ* fracturing of Gleeble specimen.

Figure 8. Energy spectrum from MgAl_2O_4 particle on DDC fracture surface exposed during *in-situ* fracturing of Gleeble specimen.

Figure 9. Energy spectrum from a MgO particle on DDC fracture surface exposed during *in-situ* fracturing of Gleeble specimen.

Figure 10. TEM BF image of a magnesium aluminate spinel (MgAl_2O_4) particle adjacent to solidification grain boundary. Specimen was taken from the fusion zone of a transverse-Varestraint specimen.

Figure 11. TEM BF image of a string of discrete MC-type cubic particles in the fusion zone of a transverse-Varestraint specimen. Note grain boundary has migrated away from some of the particles.

Auger analysis and analytical electron microscopy both suggest that intergranular, magnesium aluminate (MgAl_2O_4) spinel particles may be acting as stress concentrators, which provide nucleation sites for microvoids. These microvoids would undoubtedly coalesce into microcracks before linking to form ductility-dip cracks. It should be noted that the alloy manufacturer intentionally adds Mg to the master melt before pouring. This addition deoxidizes the final melt.

Figure 12. TEM CDF image of a high-angle grain boundary decorated with $M_{23}C_6$ -type particles. Boundary is in weld-metal HAZ of a spot-Varestraint specimen. Slip band in upper left corner resides ahead of crack generated during specimen preparation.

Whether in the fusion zone or weld-metal HAZ, the size, shape and distribution of particles does not appear to change. Hence, the peak temperature and the thermal cycle experienced by grain boundaries within the ductility dip temperature range do not appear to significantly alter the cracking tendency of this alloy.

Conclusions

In the present investigation, the effect of chemical composition on the ductility dip cracking behavior of a Ni-base filler metal was studied. From the results of optical and analytical electron microscopy, the following conclusions can be drawn:

1. Optical microscopy indicates an intergranular crack morphology along high-angle, migrated grain boundaries;
2. Scanning electron microscopy revealed a fracture surface morphology comprised of microdimples on the grain faces and oxide particles at the base of some microvoids;
3. Analytical electron microscopy revealed migrated grain boundaries decorated with MC (Ti, Cr) and $M_{23}C_6$ (Cr, Ni, Fe) precipitates ranging from 10 to 200 nm;
4. Auger electron spectroscopy of a pre-strained Gleeble specimen fractured *in situ* revealed internal ductility-dip cracks decorated with magnesium aluminate ($MgAl_2O_4$) spinel particles (1000 nm);
5. No evidence was found supporting the role of elemental segregation in the formation of ductility-dip cracks.

Figure 13. TEM BF image of a precipitate-free high-angle grain boundary in weld-metal HAZ. This boundary has migrated as evidenced by its intersection with a solidification subgrain boundary.

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