WELDABILITY OF POLYCRYSTALLINE ALUMINIDES

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July, 1995

Report Prepared by

CENTER FOR WELDING, JOINING, AND COATINGS RESEARCH

Colorado School of Mines

Golden, Colorado 80401

Under

Subcontract No. 19X-27421C

for

OAK RIDGE NATIONAL LABORATORY

Oak Ridge, Tennessee 37831

managed by

LOCKHEED MARTIN ENERGY SYSTEMS, INC.

for the

U.S. Department of Energy

under Contract No. DE-AC05-84OR21400

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July, 1995

Research sponsored by the U.S. Department of Energy,
Fossil Energy
Advanced Research and Technology Development Materials Program

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ABSTRACT

To investigate hydrogen cold cracking in iron aluminides and, specifically, to study the effect of fusion zone grain size on cracking susceptibility, welds were produced using magnetic arc oscillation to refine the fusion zone grain structures. Conventional welding produced extremely coarse columnar fusion zone grains (an average linear dimension of 530 micrometers) while welds produced with a magnetically oscillated arc contained equiaxed fusion zone grains averaging 115 micrometers in diameter. Slow strain rate tensile tests were conducted in varying water vapor atmospheres, on weldments with average fusion zone grain sizes ranging between 115 and 530 micrometers. Fracture strength and percent strain to fracture were measured for each fusion zone microstructure. The fracture strength increased in proportion to the inverse square root of the mean grain diameter, and decreased with increasing water vapor concentration. The results of the tensile tests showed that finer, equiaxed fusion zones were less susceptible to hydrogen cracking and more tolerant of high hydrogen concentrations than coarse fusion zone grain structures. Microstructural refinement via arc oscillation was also found to be suitable only for well-controlled fabrication environments.

INTRODUCTION

Interest in developing polycrystalline, long-range ordered iron aluminide alloys for high temperature applications is great because of properties such as superior oxidation resistance. In addition, these alloys exhibit good high temperature strength, relatively low density at relatively low cost, and a high melting point. However, iron aluminides are susceptible to hydrogen cold cracking during gas-tungsten arc (GTA) welding. Brittle fracture in the fusion zone is attributed to excessive grain growth during solidification.

Previous work has shown that iron aluminides can be susceptible to environmental embrittlement when tested in the presence of water vapor [1]. The suggested mechanism is similar to that observed in aluminum alloys: water molecules reacting with freshly exposed aluminum atoms at the crack tip generate high activity atomic hydrogen, which diffuses into the alloy and causes embrittlement [1]. Fine-grained base materials have been shown to be more resistant to environmental embrittlement than coarse-grained materials [2]. The purpose of this paper is to report the effect of fusion zone refinement in weldments by magnetic arc oscillation on environmental embrittlement susceptibility.

EXPERIMENTAL PROCEDURE

Sigmajig Testing in Environmental Atmospheres

A recovered and unrecrystallized microstructure is the most ductile form of iron aluminide alloy FA-129. This poses a problem for welding because the heat affected zone and the fusion zone consist of recrystallized grains after welding. Three different base material microstructures were created to determine the extent of "memory" the fusion zone had for the base material microstructure.

Welding and mechanical testing were conducted in a glovebox to control the atmosphere. Iron aluminide alloy FA-129 is susceptible to hydrogen cold cracking from moisture in the atmosphere. Welding and testing were conducted in atmospheres of argon plus different levels of moisture. Tensile testing was conducted on an apparatus called a sigmajig, which is designed for hot cracking studies but was used here for cold cracking studies.

The material for this investigation was a production vacuum arc remelt (VAR) ingot (HT# D53969-1). This ingot was extruded and hot rolled at a temperature of 900°C to a thickness of 0.125 in. (3.2mm) and then warm rolled at 650°C to a nominal thickness of 0.030 in. (0.75mm). Sigmajig test material was sheared into 2 in. x 2 in. (50mm x 50mm) coupons and divided into three groups. Each group was given a different heat treatment, producing three different base material microstructures. These microstructures are summarized in Table 1.

Table 1: Summary of Heat Treatments for Iron Aluminide Alloy FA-129.

Microstructural State	Thermal Treatment
Unrecrystallized	600°C for 1.5 hours
Fine Grain Size (38 μ m)	750°C for 1 hour
Coarse Grain Size (58 μ m)	750°C for 48 hours

All welding was conducted in a glove box with an argon atmosphere. Tests were conducted in three different argon atmospheres: dry argon, argon + $\sim$ 2000ppm ($\sim$ 0.2 wt. pct.) water vapor, and argon + $\sim$ 19,000ppm ($\sim$ 1.9 wt. pct.) water vapor. In the tests conducted in argon-water vapor atmospheres, the argon was bubbled through warm deionized water in an enclosed glass chamber. The glass bubbler was wrapped with heating tape which was connected to a rheostat, and the water temperature was controlled by changing the temperature of the heating tape.

A Panametrics Model 2000 Hygrometer was used to record the dewpoint of the gas inside the glovebox. This hygrometer measured the dewpoint of the atmosphere by means of an alumina probe, which changed impedance with the percent moisture (humidity) present. This impedance change results in an output voltage which is sensed by the hygrometer. A calibration curve for the probe is supplied by the company and allows the voltage reading to be converted to a parts per million (ppm) moisture value.

Prior to testing in each atmosphere, the glovebox was purged and back-filled twice with Grade 5 argon (99.999%). Argon was then bubbled through warm distilled water. When the atmosphere in the glovebox reached the desired humidity level, the water line was closed, and the glovebox was allowed to equilibrate for one hour prior to testing.

The sigmajig test was developed to study hot cracking susceptibility of thin sheet weldments [3]. When this test is employed to evaluate hot cracking, a transverse stress is applied prior to welding. During welding, a crack will form along the centerline of the specimen. At sufficiently high loads, the specimen will break.

To study the cold cracking susceptibility of FA-129, the specimens were placed in the sigma jig apparatus without applied preload. After making an autogenous weld along the center of the coupon, increments of load were applied until complete fracture occurred. The load at which fracture occurred was converted to a fracture stress. Welding was conducted at 30 in./min. (13 mm/s), or a heat input of 55 J/mm (78 amperes at 9 volts) and at 10 in./min. (4.2 mm/s) or a heat input of 79 J/mm (42 amperes at 8 volts).

Slow Strain Rate Tensile Testing

Conventional welding produced large fusion zone grain structures that were highly susceptible to hydrogen embrittlement. Refined fusion zone structures were produced by employing magnetic arc oscillation. Specimens with these microstructures were tensile tested in water vapor environments to examine their susceptibility to hydrogen embrittlement.

The material for this investigation also came from the production VAR ingot (HT# D53969-1). Coupons 2 in. x 3 in. (50mm x 75mm) were used for welding tensile test specimens and 2 in. x 2 in. (50mm x 50mm) coupons were used to weld samples for metallographic purposes. All samples were annealed at 650°C for two hours prior to welding. The final microstructure consisted of recovered and unrecrystallized grains.

Welding was conducted in the glovebox shown in Figure 1. The atmosphere within the glove box was Grade 5 argon which was recycled through a bed of copper sponge and moisture getter at room temperature to continually clean moisture and oxygen from the argon atmosphere. A Grade 5 argon shielding gas was used in addition during welding of all coupons. The power supply used was a Hobart Arc-Master 500, and welding was conducted using a 225-amp water-cooled GTA torch. The welding speed was 10 inches/min (4.2 mm/s) and the arc gap was 0.060 inches (1.5 mm).

A PowCon two-pole magnetic arc oscillator was used to deflect the arc. The apparatus consisted of a water-cooled electromagnetic probe mounted on the GTA torch, and a control unit as illustrated in Figure 2. The control unit

Figure. 1 Glovebox where oscillated fusion zones were produced.

Figure 2. A double-pole electromagnetic arc oscillator mounted on a 225-amp GTA torch.

consisted of a low-frequency triangle-wave oscillator that generated the current signal relayed to the electromagnetic probe. Individual controls independently adjusted both the frequency and the amplitude of the wave. Other controls included an arc position control, which provided a means of offsetting the arc path relative to the centerline of the torch, and a dwell control for adjusting the relative time that the arc spent on either side of the mean position. The arc position and dwell controls were set at a neutral position (along the centerline). Frequency and amplitude settings were measured from the waveform using an oscilloscope. The frequency to amplitude ratio was determined from the waveform and the amplitude setting was converted to an estimated distance of deflection using travel speed and the relation to frequency. According to the manufacturer, the correlation between amplitude setting and arc deflection will vary depending on the power supply, oscillation head, and the welding substrate.

Slow strain rate tensile tests were conducted on welds using an MTS 810 Material Test System. The tests were strain controlled at a rate of 2.78×10^{-6} /s. An one-inch extensometer was used with a $\pm 1.5\%$ range card. Epoxy beads were placed 0.5 inches on either side of the weld, giving the knife edges of the extensometer a softer material than the base material to grip, allowing for more accurate strain readings. The specimen geometry is shown in Figure 3. The weld acted as a reduced section for these specimens. The microstructures of the welds were so different from those of the base material that the applied strain concentrated in the fusion zone and heat affected zone, and fracture occurred in the same region. Two data points were collected for each microstructure in each water vapor concentration range; however, it was difficult to reproduce the exact same water vapor concentration. Duplicate testing was completed for water vapor concentrations maintained within ± 1000 ppm. At the higher levels of water vapor, control of the atmosphere was limited to ± 500 ppm.

Tensile testing was conducted in a plexiglas environmental chamber designed specifically for the MTS system. Figure 4 shows a photograph of the environmental chamber. The moisture probe was located directly between the grips in the back of the chamber. The chamber was purged with Grade 5 argon

Figure 3. Welding specimen tensile test geometry.

Figure 4. The environmental chamber used to control atmosphere during slow strain rate tensile tests.

until a reading of less than 200 ppm moisture was obtained. Argon was continually passed through the system during testing, and a moisture measurement was recorded both before and after testing. For tests conducted in moisture, the chamber was purged and argon was bubbled through distilled water using an ultra-slow flow rate meter. The atmosphere was allowed to equilibrate prior to testing.

A Nyad Series 300 moisture/oxygen analyzer was used to measure the atmosphere in these tests. The moisture sensor was a capacitive element utilizing a thin aluminum-oxide dielectric which adsorbed water vapor, causing a change in electrical characteristics. The output from the sensor produced a variable frequency output. This signal from the sensor entered the receiver section of the main electronic board which shaped and amplified it, and directed it to the timer port on the microprocessor. After conversion to a frequency value, the corresponding moisture content was determined using memory-resident tables containing the relationship between frequency and engineering units. The moisture content was displayed on the control unit in terms of ppm. The unit was externally calibrated using an ultra high purity calibration gas.

Welding Coupon Preparation

Prior to welding, the material had a heavy oxide layer on the surface from rolling. The oxide layer was removed using a pickling solution of 10 parts water, 1 part nitric acid and $\frac{1}{2}$ part hydrofluoric acid. After pickling, samples were cleaned with methanol.

Metallographic Sample Preparation

The specimens were prepared for metallographic observations using standard metallographic techniques. All samples were ground and polished using an automatic polisher. Specimens were wet ground to 600 grit, polished with $1\mu\text{m}$ diamond paste and finally polished with $0.05\mu\text{m}$ alumina paste. The etchant used consisted of 60ml methanol, 20ml HCl, 40ml HNO_3 . The shelf life of this etchant was about two hours. To etch, the sample was rinsed in hot water and swabbed with etchant for 10-20 seconds.

Grain Size Measurements

The grain size for the base material was measured using an intercept method (ASTM-E112). The intercepts were counted using concentric circles of a total length of 300mm, over three fields. Grain size was calculated by determining the average number of intercepts per 1mm length; this value was multiplied by a conversion factor to determine the average grain size.

Fusion zone grain size measurements were made using a Leco image analyzer. Length, width, and thickness measurements were averaged for 20 grains in the fusion zone. The overall fusion zone grain size was determined by combining average length, width and thickness measurements as stipulated by ASTM-E112, for non-equiaxed grains.

RESULTS

Sigmajig Testing in Water Vapor Atmospheres

Three different base material microstructures, shown in Figure 5, were selected for fusion zone cold cracking studies. Several microstructures were used to determine if the fusion grain size could be controlled by the base material microstructure. An unrecrystallized base material, the most ductile form of this alloy, and two recrystallized base materials were selected for this study. The fine-grained base material, Figure 5b, was expected to produce a finer-grained fusion zone microstructure than the coarse-grained material under the same welding conditions.

After welding, the coupons were tensile tested in the sigmajig apparatus, and a load at fracture was measured. The load at fracture was converted to a fracture strength. All of the weldments tested fractured in the fusion zone or at the fusion line. Fracture strength data are shown in Table 2. Fracture strength generally decreased as water vapor level increased. Fracture strength decreased only slightly with a decrease in heat input, but exhibited a dramatic decrease as the water vapor content increased. Samples welded at 13 mm/s and ~19,000 ppm water vapor exhibited almost immeasurably low fracture strengths. Fracture strength

Figure 5 The base material conditions produced by thermomechanical processing; a) unrecrystallized, b) recrystallized (38- μm grains) and c) recrystallized (58- μm grains).

Table 2: Fracture Strengths for Samples Welded at 4 mm/s and 13 mm/s.

Base Material Condition	Fracture Strength in Water Vapor Atmosphere (MPa)					
	Heat Input of 79 J/mm			Heat Input of 55 J/mm		
	~ 0 (dry) ppm	~ 2000 ppm	~ 19,000 ppm	~ 0 (dry) ppm	~ 2000 ppm	~ 19,000 ppm
Unrecrystallized	376	248	--	--	79	<10
38- μ m Grains	376	265	--	418	193	29
58- μ m Grains	317	224	--	359	183	<10

tests were not conducted for samples welded at 79 J/mm in the high water vapor atmosphere in expectation of similar results.

Table 3 summarizes the measured fusion zone grain sizes for each base material condition and welding speed. None of the samples welded in the moisture-free environment showed cracking in the weld prior to application of the load. Figure 6a shows an example of a weld produced in a pure argon environment. This weldment has a basic columnar structure with no cracking apparent in the fusion zone. Fracture in these weldments initiated along the centerline, and usually propagated into the heat-affected zone (HAZ). The samples welded in a water vapor atmosphere exhibited cracking prior to load

Table 3: Fusion Zone Grain Size.

Base Material Condition	Grain Size (μ m)	
	79 J/mm	55 J/mm
Unrecrystallized	300	421
38- μ m Grains	214	204
58- μ m Grains	220	243

application; this cracking appeared both along and transverse to the welding direction, as shown in Figure 6b. The cracking became more severe as the

Figure 6. Welds with unrecrystallized base material produced in a) pure argon, and b) argon plus 19,000 ppm water vapor.

concentration of water vapor increased. The unrecrystallized samples exhibited the most severe pre-load cracking. In all samples exposed to water vapor, the fracture initiated along the centerline and propagated in and out of the fusion zone and into the HAZ along pre-existing cracks. The fracture morphology for all of the weldments subjected to sigma-jig tests was transgranular cleavage. An example is shown in Figure 7.

Magnetic Arc Oscillation and Fusion Zone Grain Refinement in Iron Aluminide Alloy FA-129 Weldments

Welds made on samples from HT# D5-3966-1 were screened using magnetic arc oscillation to produce refined fusion zone microstructures. The base material for these samples was recrystallized. Table 4 shows the different welding parameters used to produce these welds; oscillation was imposed both transverse

Table 4 Magnetic Arc Oscillation Parameters Used to Produce Refined Fusion Zone Grain Structures

Amplitude Setting (estimated deflection)	Frequency (Hz)	
	250	750
250 (0.8 mm)	X	X
500 (1.2 mm)	X	X
750 (1.5 mm)	X	X

and longitudinal to the welding direction. The microstructures for the welds oscillated longitudinally are shown in Figure 8. Some grain refinement was realized in these fusion zones; however, the weldments still exhibited a columnar grain structure. Figure 9 shows the microstructures for the transversely oscillated fusion zones. Transverse oscillation resulted in much greater fusion zone refinement. Here, the fusion zone morphology changed significantly with changing oscillation parameters. The fusion zone showing the most grain refinement was oscillated with a frequency of 25 Hz and an amplitude setting of 250 (0.8 mm).

Figure 7. Fracture surfaces of sigma-jig test coupons for unrecrystallized base material welded at 79 J/mm in a) pure argon and, b) argon + 19,000 ppm water vapor.

Figure 8. Microstructures for welds oscillated longitudinally to the welding direction at a) 25 Hz and amplitude setting of 250 (0.8 mm), and b) 25 Hz and an amplitude setting of 500 (1.2 mm).

Figure 9. Microstructures for welds oscillated transverse to the welding direction at a) 25 Hz and amplitude setting 250 (0.8mm), b) 25 Hz and amplitude setting 500 (1.2mm), and c) 75 Hz and amplitude 250 (0.8mm).

The results of the initial screening were very promising; consequently, welds were made using the same matrix of oscillation parameters on unrecrystallized material from a second heat, # D5-3969-1. Fusion zones created by oscillation in unrecrystallized base material of the second heat were not refined as extensively as were the fusion zones of the initial heat. Coarse columnar grain morphologies, as shown in Figure 10, were observed. Verification welds on the first heat of material again showed refined fusion zone structures. Differences in the chemical composition and base material grain morphology between the two heats were examined. Both heats of material were chemically analyzed to determine any compositional variations that might have caused a difference in response to magnetic arc oscillation. The results of the chemical analyses are shown in Table 5. In addition, some base material from the second heat (# D5-3969-1) was recrystallized and oscillated welds were produced. No significant fusion zone grain refinement was realized in welds made from recrystallized base material of the second heat.

Table 5 Analysis of Chemical Composition of Heat #D5-3966-1 and Heat #D5-3969-1.

Heat Number	C	Mn	P	S	Si	Ni	Cr	Mo
D5-3966-1	0.056	<0.01	<0.002	0.002	0.12	<0.01	5.32	<0.01
D5-3969-1	0.052	<0.01	<0.002	<0.001	0.16	0.02	5.29	<0.01

Heat Number	Nb	Cu	Al	Zr	Fe	N	O
D5-3966-1	0.94	<0.01	16.0	<0.01	77.6	<0.001	0.007
D5-3969-1	1.03	0.01	16.6	<0.01	76.9	<0.001	0.004

Several welding parameters were subsequently varied to determine if grain refinement could be achieved in an unrecrystallized base material of HT# D5-3969-1. Welding speed was varied to change the cooling rate of the weld pool, and encourage grain refinement. The welding speed was decreased from 10 ipm (73 J/mm) to 6 ipm (122 J/mm) and then increased to 15 ipm (49

Figure 10. Microstructures for oscillated welds for heat # D5-3969-1 at a) 25 Hz and amplitude 250 (0.8mm), b) 25 Hz and amplitude 500 (1.2mm), and c) 75 Hz and amplitude 250 (0.8mm).

J/mm). No difference in the grain refinement was seen. Changes in welding speed still resulted in large columnar grain fusion zones. Welding parameters such as current, arc gap, and oscillation parameters were also varied from those originally used. The amount and characteristics of the fusion zone grain refinement showed some sensitivity to the placement of the oscillation probes. Figure 11 shows fusion zones created using the same welding and oscillation parameters for both heats of material. Both fusion zones exhibited some grain refinement and displayed similar columnar grain structures on the right side of the fusion zone. However, the weldment from the first heat of material (# D5-3966-1) contained a more refined grain structure than the weldment from the second heat (# D5-3969-1). The similar columnar grain structure on the right side of both welds indicated that the probe on the left side deflected the arc while the probe on the right side did not. For equal deflection of the arc, precise leveling of both probes and the electrode tip was required. Careful centering of the electrode between the two probes was also necessary. However, all of these changes resulted in less refined fusion zone structures in the unrecrystallized material as compared to the recrystallized base material of the first heat.

Welds were also produced using different electrode materials sharpened to different tip angles. Pure tungsten electrodes were used on the original oscillated welds. Two percent thoriated electrodes were used in all of the later welds on both heats of material. Both oscillated and unoscillated welds were made to investigate the effect of electrode material and shape on the amount of fusion zone grain refinement. The amount of thorium and angle of the tip had no effect on the grain refinement seen within the fusion zones.

At this point in the study, the welding power supply being used was switched from a Hobart Cyber-Tig to a Hobart Arc-Master 500. Some welds made with the new power supply showed extensive fusion zone grain refinement; these welds were produced using the previous welding and oscillation parameters but they just barely exhibited full penetration. The solution to producing refined fusion zones appeared to be to control the heat input just to the point where full penetration was achieved. These weldments were made using a current of 35

Figure 11. Microstructures showing the sensitivity to probe placement for a) heat #D5-3966-1, and b) heat #D5-3969-1.

amps, 11 volts, and an arc gap of 0.060 inches. The weld was oscillated at 10 Hz and an amplitude setting of 250 (0.8mm).

The process of grain refinement was found to be extremely sensitive to heat input, and the power supply was susceptible to power surges which caused current fluctuations of up to $\pm 5\%$ of the current setting. Differences of one amp were found to be large enough to push the heat input out of the operable range for grain refinement. Unfortunately, ± 1 amp was as much sensitivity as the power supply allowed. To limit the power surging problem, all welding was conducted at night when the power usage within the building was at a minimum. In addition, thickness variations in the sheet exceeding ± 0.002 inches caused sufficiently large changes in the temperature and shape of the weld pool to affect the fusion zone morphology. Consistent fusion zone grain structures were produced by welding at night and using coupons with thickness variations of ± 0.001 inches.

Slow Strain Rate Tensile Testing in Water Vapor Atmospheres

An experiment was developed to test the resilience of the refined fusion zone microstructures to hydrogen embrittlement in water vapor atmospheres. For these tests welding was conducted in pure argon while tensile testing was conducted in argon plus varying levels of water vapor. Four different weldments were produced using different welding and oscillation parameters, shown in Table 6. Different heat inputs and oscillation parameters were used to produce four unique microstructures that were tested to determine their resistance to hydrogen embrittlement. The unoscillated microstructures produced are shown in Figure 12, and the transversely oscillated fusion zone microstructures are shown in Figure 13. The welds of the unoscillated Microstructure A, shown in Figure 12a, consisted of large columnar grains. The second unoscillated microstructure (Microstructure B), shown in Figure 12b, also exhibited a columnar grain structure and a well defined centerline. In this lower heat input weld, however, the grains were not as large as grains in the higher heat input weld. The transversely oscillated fusion zone (Microstructure C), Figure 13a, looked similar to the unoscillated fusion zone

Figure 12. Unoscillated microstructure for weldments used in slow strain rate testing. a) Microstructure A (H.I.=81 J/mm), and b) Microstructure B (H.I.=73 J/mm).

Figure 13. Oscillated microstructures for weldments used in slow strain rate tests. a) Microstructure C (90 degrees at Freq=10 Hz, Amp.=0.8mm), and b) Microstructure D (45 degrees at Freq.=10 Hz, Amp.=0.8mm).

Table 6 Welding Parameters Used to Produce Fusion Zones of Differing Grain Sizes

Microstructure Type	Current (amps)	Arc Gap (inches)	Heat Input (J/mm)	Oscillation Frequency (Hz)	Oscillation Amplitude Setting	Oscillation Orientation to Welding Direction
A	38	0.060	80.9	--	--	--
B	31	0.060	73.3	--	--	--
C	31	0.060	73.3	10.0	250 (0.8mm)	90 degrees
D	31	0.060	73.3	10.0	250 (0.8mm)	45 degrees

(Microstructure B) in Figure 12b; however, in this microstructure there was no clear centerline. The grains in this microstructure were still elongated. Finally, the fusion zone created by oscillating the arc at 45 degrees to the welding direction (Microstructure D) resulted in a nearly equiaxed, fine-grained fusion zone (Figure 13b). Also, no clear centerline or fusion line was apparent in this weldment.

Fusion zone grain size measurements were made using a Leco image analyzer system. The results of the grain size measurements are shown in Table 7. The average grain size and aspect ratio of each microstructure are reported as well. As was apparent from the microstructures (Figure 12a), the weld with the highest heat input had the largest average grain size and the largest aspect ratio. The fine-grained microstructure (Microstructure D) produced by oscillating at a 45° angle to the welding direction produced the smallest average grain size with the smallest aspect ratio.

Slow strain rate tensile tests were conducted on welds of all four microstructures in pure argon and argon plus four water vapor levels. Load at fracture and strain at fracture were recorded for each sample. Summaries of the tensile data from these tests are shown in Tables 8 to 11. The data points shown in Table 8, 9, and 11 are for those welds which broke in the fusion zone. Welds of Microstructure C (Table 10) broke at the fusion line. The fracture surfaces were either transgranular cleavage or a mixed mode of intergranular and

Figure 14. Fracture surfaces for welds tested in $\sim 6000\text{ppm}$ water vapor a) Microstructure A, b) Microstructure B, c) Microstructure C, and d) Microstructure D.

Figure 15. Fracture surfaces for Microstructures B tested in atmospheres containing a) pure argon, b) 1350 ppm H_2O , c) 3200 ppm H_2O , and d) 5770 ppm H_2O .

Table 9 Slow Strain Rate Tensile Data for Microstructure B (300 μ m grain size)

Water Vapor in Atmosphere (ppm)	Percent Strain at Fracture	Fracture Strength (MPa)
227	1.15	608
1342	0.77	591
2739	0.59	574
3195	0.72	548
5770	0.85	582
7080	0.62	548

Table 10 Summary of Slow Strain Rate Tensile Data for Microstructure C (172 μ m grain size).

Water Vapor in Atmosphere (ppm)	Percent Strain at Fracture	Fracture Strength (MPa)
252	1.49	637
2094	0.71	576
2912	0.76	560
6310	0.54	530

Table 11 Summary of Slow Strain Rate Tensile Data for Microstructure D (115 μ m grain size)

Water Vapor in Atmosphere (ppm)	Percent Strain at Fracture	Fracture Strength (MPa)
197	1.39	671
1247	1.04	632
2747	0.79	629
7082	1.01	656
13000	0.88	578

Fracture strength versus moisture content was plotted for welds representing each fusion zone grain size, as shown in Figure 16. A linear regression was performed on the data for each grain size. The correlation coefficient (R) is shown and describes the accuracy of the fit. (If the line perfectly fits the data, R is equal to one.) The slope of the line for the coarsest ($530\ \mu\text{m}$) fusion zone is slightly steeper than that of the finest ($115\ \mu\text{m}$) fusion zone. The data for the middle grain sizes plot nearly on top of one another. Percent strain at fracture versus the moisture content in the atmosphere was also plotted for each of the fusion zone grain sizes, see Figure 17. These data were not as well defined as the fracture strength data; however, the basic trends were the same. The finest fusion zone ($115\ \mu\text{m}$) shows the highest percent elongation at fracture and the coarsest fusion zone ($530\ \mu\text{m}$) shows the least amount of elongation at fracture.

Figure 18 is a Hall-Petch style plot in which each range of the water vapor levels tested is shown. Since testing occurred over a range of water vapor concentrations, points were read from the linear regression fits of the data in Figure 18 at the specific levels plotted. A linear regression was conducted on each of these data sets and the slope of each line was determined. The correlation coefficient (R) is shown for each line. The slopes of the lines in Figure 19 correspond to the k_F value in the Hall-Petch equation. k_F was plotted as a function of water vapor concentration in Figure 19. The data fit a square root function showing $k_F = 70 + 0.25M^{1/2}$ ($\text{MPa mm}^{1/2}$), where M is the water vapor concentration in parts per million. The σ_o term is also a function of water vapor concentration, where $\sigma_o = 472 - 1.5M^{1/2}$ (MPa). The plot of σ_o versus water vapor concentration is also shown in Figure 19.

Welds in Varying Water Vapor Atmospheres

Further tests of the hydrogen cold cracking behavior using refined fusion zone microstructures were conducted on welds made with shielding gas containing varying levels of water vapor. Tensile coupons were cut from the welding coupons, as shown previously in Figure 3. Welds were made using shielding gas of

Figure 16. Fracture strength versus water vapor content in the atmosphere showing the effect of fusion zone grain size on the mechanical properties of the weldment.

Figure 17. Percent elongation at fracture for weldments of different fusion zone grain size tested in atmospheres of varying levels of water vapor.

Figure 18. Hall-Petch plot for iron aluminide alloy FA-129 weldments tested in varying water vapor atmospheres. The correlation coefficient (R) for each line is shown.

Figure 19. k_F and σ_o as a function of water vapor concentration for aluminide alloy FA-129 weldments.

pure argon and with argon containing either 93 ppm or 560 ppm water vapor. Figure 20 shows the microstructures for the unoscillated fusion zones welded in pure argon and argon plus water vapor. The microstructures revealed that all of the weldments contained columnar fusion zones and showed little variation in morphology with changes in water vapor concentration. The microstructures for the oscillated fusion zones are shown in Figure 21. The oscillated fusion zones welded with shielding gas of argon plus water vapor exhibited no significant grain refinement. Further tests confirmed that water vapor additions to the shielding gas were the cause of the less extensive grain refinement seen in the oscillated fusion zone microstructures.

Tensile tests conducted using a MTS Material Test System enclosed in a glovebox revealed that, for those coupons welded in water vapor, the fracture strengths were approximately the same, as shown in Table 12. In other words, magnetic arc oscillation did not increase the fracture strength of the weld. This was not surprising since limited grain refinement occurred in these weldments.

Figure 20. Microstructures for unoscillated fusion zones welded in a) pure argon, b) argon + 93.3 ppm H_2O , and c) argon + 556 ppm H_2O .

Figure 21. Microstructures for oscillated fusion zones (10 Hz, amplitude setting 250 (0.8mm)) welded in a) pure argon, b) argon plus 93 ppm water, and c) argon plus 560 ppm water shielding gas.

Table 12 Tensile Data for FA-129 Welded in Varying Water Vapor Atmospheres

Sample Number	Frequency (Hz)	Amplitude	Atmosphere (ppm H ₂ O)	Fracture Load (kg)	Fracture stress (MPa)
7-10	10.0	250.0	0.0	939	427
7-11	10.0	250.0	0.0	1220	565
7-17	--	--	0.0	1045	483
7-19	--	--	0.0	1089	510
7-20	10.0	400.0	0.0	871	434
7-25	10.0	400.0	0.0	1099	496
7-26	10.0	400.0	0.0	1126	462
7-27	10.0	400.0	0.0	1087	455
7-29	--	--	93.9	1089	455
7-30	10.0	250.0	93.9	1231	531
7-31	10.0	250.0	93.9	1257	552
7-32	10.0	400.0	93.9	1222	531
7-33	10.0	400.0	93.9	1100	469
8-17	--	--	556	1277	503
8-18	--	--	556	1232	503
7-35	100	250	556	1284	537
7-36	100	250	556	1256	490
8-60	100	400	556	1295	503
8-61	100	400	556	1134	531

DISCUSSION

Sigmajig Testing in Environmental Atmospheres

The results of the sigmajig welding studies shown in Tables 2 and 3 suggest a correlation between the base material grain size and the fusion zone grain size and, therefore, the fracture strength. The unrecrystallized base material produced much larger fusion zone grain sizes than the recrystallized base material, as shown in Table 3. The reason for this is unclear; however, some carbides exist in the recrystallized material which are believed to pin the grain boundaries. The recrystallized base material was annealed prior to welding, allowing precipitates to form uniformly along the grain boundaries. As the arc moves through unwelded base material the region adjacent to and in front of the arc is exposed to a high temperature (1260°C) for a very short period of time. The precipitates along the grain boundaries retard the grain growth, as was seen in the grain growth study [2]. On the other hand, the unrecrystallized base material only underwent a recovery step prior to welding, leaving the precipitates along the elongated grain boundaries. Again, the unwelded base material in front of and adjacent to the arc is exposed to a high temperature for a very short period of time. The unrecrystallized grains in the region of the torch recrystallize; however, the cooling rate is so fast that the solute remains in solution and no precipitates form at the grain boundaries. Without precipitates along the grain boundaries to retard the grain growth, the grains in the heat-affected zone grow larger. Since columnar fusion zone grains grow epitaxially from grains in the heat-affected zone, if the grains in the heat-affected zone are larger, the grains in the fusion zone will also be larger. Hence, the unrecrystallized base material produced larger fusion zone grain structures than either recrystallized based material.

The difference in fracture strength of the weldments observed for the three base material conditions was relatively small, but the fracture strength did decrease with increasing fusion zone grain size. During welding, a water vapor concentration of 19,000 ppm resulted in a fracture strength which was almost immeasurably low (Table 2). In comparison, at a water vapor concentration of

2000 ppm, the fracture strengths were approximately half those measured for samples welded in pure argon. These results clearly indicate that the welding environment must be controlled to contain less than 2000 ppm moisture if hydrogen embrittlement effects are to be avoided. The coarsest fusion zone microstructure, produced in the unrecrystallized base material at a heat input of 55 J/mm, showed the greatest decrease in fracture strength when tested at ~ 2000 ppm water vapor. All the other weldments, with similar fusion zone grain size, performed about the same. This indicated that fusion zone grain size must also be controlled to reduce cold cracking effects.

Control of the fusion zone grain size is important for increased fracture resistance and decreased susceptibility to cold cracking. Refined fusion zone microstructures are expected to exhibit superior mechanical properties compared to non-refined microstructures. Fe_3Al , whether ordered or disordered, becomes completely ductile at temperatures above 150°C . In both conditions, the slip mode at this temperature changes from a mixture of planar and wavy glide to wavy glide alone [4]. Cross-slip influences the rate at which multiple slip can spread from one grain to another along the intersection of the glide plane and the grain boundary. If cross-slip is easy, slip can easily spread from one grain to another and the material will be ductile. Since cross-slip is difficult in iron aluminides on the $\{110\}$ slip planes, dislocations glide on $\{112\}$ and $\{123\}$ planes as well [4]. This results in inherently low tensile ductilities [5].

In 1951 and 1953, Hall and Petch [6,7] developed a relationship that showed yield strength to be proportional to the inverse of the square root of grain size. Many researchers since have shown that the fracture stress of many structural materials, including ordered alloys, exhibit Hall-Petch type behavior. Cottrell [8] found that steel, both above and below the ductile-to-brittle transition temperature, followed the Hall-Petch relationship. Low [9] tested a low carbon steel at -196°C . Figure 22 shows the effect of grain size on the yield and fracture tensile stress for Low's data. For large grain sizes, fracture occurred at the yield stress. This line corresponded to the Hall-Petch equation ($\sigma_{\text{flow}} = \sigma_o + k_F d^{1/2}$). At a critical grain size ($2.3 \text{ mm}^{-1/2}$), the slope of the fracture line changed. The

Figure 22. Effect of grain size on yield and fracture tensile stresses, and on strain to fracture of low-carbon steel at -196°C [7].

second line, which the data followed for the smaller grain sizes, corresponded to the Griffith crack equation. The Griffith line passed through the origin and was dotted for grain sizes above the critical value. Fracture does not follow the Griffith equation in this range because a microcrack has not yet nucleated. When the grain size was sufficiently refined, the material accommodated strain before fracture occurred. The flow stress line is dotted in this region because the microcrack formed at the yield stress but continued to grow slowly to fracture.

Schulson and Barker [10] showed that the yield strength of NiAl obeyed the Hall-Petch equation. Chan [5] confirmed the work of Schulson and Barker and showed the existence of a critical grain size below which tensile ductility could be imparted to brittle intermetallics such as NiAl. Vasudevan [11] showed a strong dependence of TiAl yield stress on grain size. The yield strength of iron aluminide alloys has also been shown to follow the Hall-Petch relationship [12]. The observations of brittle materials following the Hall-Petch equation have been explained on the basis that the stress required to nucleate microcracks, and thus initiate unstable crack propagation, depends on dislocation pileup length [8]. Fusion zone microstructures of an Fe₃Al alloy, refined through arc oscillation or arc pulsation, should manifest an increased fracture toughness.

Effect of Magnetic Arc Oscillation on Solidification

Magnetic arc oscillation is known to increase stirring and change the thermal gradients within the weld pool, thus changing the fusion zone grain structure. The increased stirring helps to break off partially melted grains at the fusion line (grain detachment) and encourages dendrites in the mushy zone to break off (dendrite fragmentation). The partially melted grains and dendrites are pulled by convection into the center of the weld pool and, instead of melting back into the weld pool, continue to solidify and grow.

Magnetic arc oscillation also affects the solidification mechanics of the weld pool by changing the competitive grain growth process. During solidification in an unoscillated fusion zone, shown in Figure 23a, grains tend to grow in the direction perpendicular to the solid/liquid interface, since this is the direction of the

Figure 23. The effect of oscillation on the thermal gradient of the weld pool for a) non-oscillated, b) longitudinally oscillated, and c) transversely oscillated arcs.

maximum temperature gradient and thus the direction of maximum driving force for solidification. However, grains have a preferred easy growth direction ($\langle 100 \rangle$ in cubic materials). Therefore, during solidification, grains with an easy growth direction parallel to the direction of the maximum temperature gradient (as indicated by the arrows) will grow more easily and crowd out grains whose easy growth direction deviates significantly from the direction of maximum temperature gradient. If the arc is oscillated longitudinally to the welding direction, as indicated in Figure 23b, the welding speed is effectively decreased. Grain growth occurs epitaxially and the solidified fusion zone consists of parallel, columnar grains. On the other hand, if the arc is oscillated transverse to the welding direction, as shown in Figure 23c, the weld pool responds to movement of the arc, which is determined by the magnetic field as well as by the moving torch. The arrow locates a point perpendicular to the isotherm, in the preferred growth direction. In this case, the preferred growth direction constantly changes. An individual grain can only grow a short distance before it is no longer oriented in the direction of the highest thermal gradient. An adjacent grain, however, may be more suitably oriented and will consequently impinge on the first grain as it grows. The new grain in turn can grow only a short distance before it is no longer oriented preferentially, and is trapped by a faster growing neighboring grain. The solidified fusion zone that results from transverse oscillation is a smaller, more equiaxed grain structure without a well defined centerline.

The preceding scenario was confirmed. Figure 9 shows iron aluminide alloy FA-129 welds that were oscillated longitudinally. All of these welds exhibited columnar grain structures. For the welds that were oscillated transversely, shown in Figure 10, no clear centerline was evident. Depending on the oscillation parameters, varying degrees of grain refinement occurred in each of these welds.

Slow Strain Rate Tensile Testing in Water Vapor Atmospheres

Iron aluminides are embrittled when tested at strain rates slower than 10^{-3} /s [13]. A slow strain rate test (10^{-6} /s) was developed to tensile test the weldments in controlled water vapor atmospheres. The slowest reasonable strain

rate was selected to maximize the time for hydrogen to diffuse. For samples tested in pure argon, the maximum test duration was designed to be 3 hours (10.8 kiloseconds).

Slow strain rate tensile testing revealed that welds containing fine-grained fusion zones resisted hydrogen embrittlement in the presence of water vapor significantly better than welds of coarse-grained fusion zones. Four widely varying fusion zone microstructures (different grain sizes and aspect ratios), as well as the welding parameters which produced them, are shown in Figures 12 and 13.

Microstructure D, the smallest and most equiaxed fusion zone, exhibited the highest fracture strength and the most ductility at fracture. The trends were very clear for the fracture strength data, and a first order polynomial equation was fit to these data. The values of the correlation coefficient (R) for each fusion zone grain structure are shown in Figure 16. These values ranged from 0.7 to 0.99. Variations in moisture level over the duration of the test and experimental error can account for some of the scatter seen in the fracture data. However, the linear regression fits accurately describe the trend of these data.

Using points from the linear fits in Figure 10, the fracture strength was plotted as a function of the inverse square root of fusion zone grain size, as shown in Figure 18, for four water vapor levels. Again, a first order polynomial equation was fit to the data for each water vapor concentration, and the correlation coefficient (R) is shown. The data for the largest grain size fusion zone, Microstructure A, fall below the linear regression line. Microstructure A had a very large aspect ratio and an obvious centerline. The effect of the directional grains and the centerline was lessened by averaging of the grain size in three directions. Hence, the linear regression predicts a higher fracture strength for Microstructure A ($530\ \mu\text{m}$) than was measured, as shown on Figure 18. The data points of Microstructure C ($172\ \mu\text{m}$) also fall below the linear regression line. In this case, failure occurred in the heat affected zone, causing the measured fracture strength to be less than that of the actual weldment.

The slopes of these lines, shown in Figure 18, increased slightly as water vapor concentration increased, indicating that k_F is a weak function of water vapor

concentration. The points shown in Figure 18 are single data points so no error bars are shown. The slope varied from 77 in pure argon to 97 MPa mm^{1/2} in argon containing 10,000 ppm water vapor. This change in slope is small, and the experimental scatter of the data could account for the difference. Additional tests are needed to determine if the observed increase in k_F with increasing hydrogen was real or if it reflected the experimental error in the data points.

The y-intercept decreased as water vapor concentration increased, indicating that the flow stress σ_o was also a function of water vapor concentration. The flow stress ranged from 448 MPa in pure argon to 327 MPa in argon containing 10,000 ppm water vapor; thus, a 30% decrease in flow stress occurred over the range of water vapor concentrations tested. Figure 19 shows that σ_o was a much stronger function of water vapor concentration than k_F . The decrease in flow stress was sufficiently great that the change could not be accounted for in the experimental error.

Hydrogen Embrittlement and the Effect of Fusion Zone Grain Size

The basic form of the Hall-Petch equation for brittle fracture is:

$$\sigma_F = \sigma_o + k_F d^{-1/2} \quad (1)$$

where σ_F is the fracture stress, d is the grain size, σ_o is the flow stress within a single crystal, and k_F is the slope of the straight line obtained when σ_o is plotted against $d^{-1/2}$. The value k_F (the slope in the Hall-Petch plot) has been called the locking parameter, which measures the relative hardening contribution of the grain boundaries. When considering microcrack nucleation at dislocation pile-ups, k_F has been expressed as:

$$k_F = (\pi \mu \gamma_B)^{1/2} \quad (2)$$

where μ is the shear modulus and γ_B is the surface energy [14]. Using the values for σ_o and k_F determined experimentally in the slow strain rate tests, the

Hall-Petch equation becomes:

$$\sigma_F = (473 - 1.5M^{1/2}) + (70 + 0.25M^{1/2}) d^{-1/2} \quad (3)$$

where M is the water vapor concentration of the atmosphere in parts per million (ppm). The results of the slow strain rate tests showed that fracture strength was a function of not only grain size, d , but also of the water vapor concentration in the test atmosphere.

The critical stress concentration necessary to nucleate a microcrack is proportional to the square root of grain size in the fusion zone [7]. For a given applied stress in a small-grained material, a dislocation pileup will create a lower stress concentration than a large-grained material would. Hence, under the same loading conditions, a large grained material, such as Microstructure A, will reach a critical stress concentration and nucleate a microcrack before a smaller-grained material such as Microstructure D. Experimentally, a fine-grained fusion zone (Microstructure D) failed at a significantly higher applied stress than a coarse-grained fusion zone (Microstructure A), following the hypothesis of the brittle microcrack model.

Figure 24 shows how hydrogen is expected to affect the Hall-Petch plot. The bold lines indicate when fracture is predicted. Two lines are shown, each representing fracture for different water vapor concentrations. In an atmosphere containing no hydrogen, large grained material fractures at the flow stress, meaning that as soon as a microcrack is nucleated, it propagates unstably to fracture. As the microstructure is refined, the flow stress increases. Below a critical grain size, the material no longer fractures at the yield stress. In this case, the microcrack nucleates at the flow stress, but does not grow unstably. The microcrack grows slowly with additional strain until it reaches a critical length for unstable propagation. For this case, fracture follows the brittle fracture line, which goes through zero and is dotted for large grain sizes. This line corresponds to the Griffith equation.

Figure 24. Schematic of Hall-Petch plot showing the effect of hydrogen on the flow stress (σ_o) and the fracture stress (σ_F).

When hydrogen is added, the behavior is more complicated. Figure 24 suggests that σ_o is decreased by diffusible hydrogen, and that the slope of the line (k_F) does not change when hydrogen is introduced. The Griffith line also shows a decreased slope when hydrogen is present, corresponding to a reduction in the surface energy term, γ . Hydrogen may reduce the surface energy of the crack by lowering the cohesive strength of the atomic bonds at the crack tip. The change in value of σ_o and the slope of the σ_F lines show that, when hydrogen is present, additional grain refinement is necessary to achieve similar mechanical properties to those in hydrogen-free environments.

Mechanistically, hydrogen is thought to affect the fracture strength of a material in several ways. The kinetics of hydrogen transport as well as the mechanisms of failure must be considered. In general, two sources of hydrogen may be available to initiate failure: internal or solute hydrogen and external or atmospheric hydrogen [16].

Internal hydrogen reaches the point at which failure occurs by diffusion and/or dislocation sweeping. Hydrogen diffusion is characterized by high diffusivities and low activation energies; BCC metals have the highest hydrogen diffusivities [16]. Hydrogen has also been shown to migrate to stress concentrations such as cracks [17]. In addition to transport of hydrogen by diffusion in response to stress gradients, it has been suggested that moving dislocations can transport hydrogen over large distances as solute atmospheres [16]. This mechanism, based on a hypothesis by Cottrell [8] requires a large binding energy between the hydrogen solute and the dislocation traps. Experimental evidence for deformation-enhanced mobility has been obtained during studies of the effects of plastic strains on hydrogen evolution. These results suggest that at low strain rates, the range of dislocation transport can exceed that due to diffusion by a factor of 10^4 [16].

In the case of external hydrogen environments, hydrogen may be produced at the propagating crack tip either by adsorption and dissociation of H_2O molecules or by producing dissolved hydrogen through a chemical reaction. For the case of iron aluminides embrittled by water vapor in the atmosphere, the

hydrogen enters through cracks in the oxide layer at the surface and then moves internally to the stress concentrations. However, hydrogen entry at points of stress concentration is not a limiting factor in the observation of hydrogen embrittlement [16]. The mechanism by which fracture occurs is independent of the hydrogen source.

For the hydrogen embrittlement seen in iron aluminide alloy FA-129 weldments, two types of mechanisms of fracture can be argued. The first mechanism of hydrogen embrittlement is the decohesion mechanism. This mechanism postulates that brittle fracture occurs when the local stress exceeds the atomic bond strength and that the presence of hydrogen as a solute decreases the atomic bond strength [16]. The energy required to reversibly break the atomic bonds across an atomic plane while maintaining all other atom spacings at their equilibrium values is the thermodynamic surface energy, γ . Adsorption of hydrogen decreases this surface energy; the decohesion mechanism postulates that hydrogen produces a decrease in the maximum bonding force between atoms across an atomic plane as they are being reversibly separated. The basic assumption is that brittle fracture occurs at a critical bond distance where the local applied stress exceeds the cohesive stress; it is assumed that the cohesive stress decreases as the local hydrogen concentration is increased [16].

The second hydrogen embrittlement mechanism concludes that hydrogen changes the plastic deformation characteristics of a material. Bond et al. [18] studied hydrogen embrittlement mechanisms in Ni_3Al alloys and found that transgranular crack growth by hydrogen resulted from enhanced dislocation mobility. These results were consistent with the "hydrogen-enhanced localized plasticity" mechanism [19,20], in which hydrogen enters the material near the crack tip through the surfaces produced by slip. This results in a reduction of the flow stress which, in turn, leads to a highly localized transgranular failure by ductile processes. Because of this localization, the resulting fracture surface will appear to be macroscopically brittle. As discussed previously, these tests found that the friction stress, σ_0 , decreased as hydrogen content increased and that the locking parameter, k_p , increased slightly with hydrogen content. These results are

consistent with Bernstein [21], who made Hall-Petch measurements at room temperature on zone-refined iron under dynamic charging conditions. Bernstein found a lower friction stress and an increased Hall-Petch slope with increasing hydrogen concentration. Beachem [22] torsion tested hydrogen-charged 1020 steel and found a reduction in the yield and flow stress compared to uncharged material. However, Adair [23] precharged zone-refined iron specimens of various grain sizes and tested them at 242K. An analysis of fracture stress versus inverse square root of grain size indicated an increased friction stress and a decreased k_F value slope for charged specimens. These results suggest that no single hydrogen mechanism can explain all embrittlement phenomena.

The softening behavior, evidenced by the decreasing friction stress, is associated with enhanced motion of dislocations and possibly with enhanced injection of dislocations at free surfaces [17]. A number of possible explanations have been proposed for the solid solution softening. The first is a reduction in the magnitude of the Peierls-Nabarro stress field by the solute [24]. The second theory suggests that hydrogen increases the mobility of screw dislocations by enhancing the rate of double kink nucleation [25]. Finally, softening has been said to be a result of a reduction in the effectiveness of solutes as dislocation pinning points resulting from the formation of solute clusters [26,24].

The hardening behavior, indicated by an increasing Hall-Petch slope, is more difficult to explain. In steel, increases in k_F have been related to carbon and nitrogen strengthening the grain boundaries. Bernstein [21] suggested that hydrogen might play a role similar to carbon and nitrogen at the grain boundaries. He compared the relative strengthening effect of carbon and hydrogen and found them to be comparable, although the hydrogen value was low. Interactions between dissolved hydrogen and grain boundary solutes such as oxygen and sulfur were suggested to result in further strengthening. A slightly strengthened grain boundary would provide resistance to microcrack nucleation.

Engineering Aspects of Magnetic Arc Oscillation

As stated previously, precise control of the welding parameters was

necessary to produce consistently repeatable refined fusion zone grain structures in iron aluminide alloy FA-129. The initial screening was conducted on recrystallized base material of HT# D5-3966-1, and subsequent welds were made on unrecrystallized base material of HT #D5-3969-1. The weldments produced on unrecrystallized base material from the second heat showed that the operating range in which fusion zone grain refinement occurred was very narrow.

The possibility of unfavorable microstructural characteristics associated with the unrecrystallized base material was investigated. The unrecrystallized base material was recrystallized prior to welding. Welds using several oscillation parameters were produced on recrystallized material of the second heat. There was no difference in the response to the arc oscillation for the unrecrystallized and recrystallized base material of the second heat.

A chemical analysis was conducted on both heats of material to determine if there were some variations in composition that might cause a difference in response to the arc oscillation. The results of the analysis are shown in Table 5. Both heats were analyzed for compositional and trace elements. The composition of both heats varied only slightly from each other in all of the elements found. The largest differences were found in the iron and aluminum contents. Oxygen levels in the first heat of material were almost twice that of the oxygen levels in the second heat of material. Oxides forming in the molten weld pool may act as sites for heterogeneous nucleation and encourage more extensive fusion zone grain refinement. Sulfur levels were also greater in the first heat. The actual cause of the different response to arc oscillation between the two heats of material remains unclear.

All of the welding and oscillation parameters were systematically varied to determine an appropriate operating range for achieving different levels of fusion zone grain refinement. The key to successfully refining the fusion zone grain structure was found to be controlling the heat input of the weld, primarily by controlling the current. The power supply was known to vary the output up to $\pm 5\%$ depending on usage rate. Fluxuations of this level were enough to move the heat input of the weld outside the established grain refining range. More accurate

control of the output for the power supply was not available. Welding was conducted at night, when power usage was at a minimum, so the power supply would see a more uniform input level. In addition, thickness variations exceeding 0.003 inches (10 percent of the total thickness) changed the weld penetration. For each welding condition, the thickness variations between all of the welding coupons was limited to ± 0.001 inches. By precisely controlling the welding variables, weldments exhibiting extensive fusion zone grain refinement were produced consistently.

Welds were made with water vapor added to the shielding gas to investigate the resistance of the refined fusion zones to hydrogen cold cracking. As greater amounts of water vapor were added to the shielding gas, a lack of extensive fusion zone grain refinement in the oscillated weldments became apparent. Figure 21 shows the oscillated fusion zone microstructures. At water vapor levels as low as 93 ppm, the fusion zones exhibited columnar grain structures similar to the unoscillated fusion zones shown in Figure 20. The welds made with water vapor in the shielding gas were obviously 'hotter', and the bead size and amount of penetration were significantly different from these characteristics in welds made with pure argon shielding gas. The addition of water vapor to the argon shielding gas apparently increased the ionization potential of the arc, and therefore, increased the heat input. The increase in the cross-sectional area of the weld made with water vapor present in the shielding gas was assumed to be proportional to the change in heat input from welds made in a pure argon shielding gas. This change in heat input was then correlated to a change in the arc efficiency. The water added to the shielding gas was then related to a change in the ionization potential of the arc. The equation used to calculate heat input is:

$$\text{Heat Input} = \frac{V i}{s} \eta \quad (4)$$

where η is the arc efficiency, s is travel speed, V is voltage, and i is the current.

An attempt was made to calculate the ionization potential of the arc with the water vapor added to the shielding gas, then correct the heat input with appropriate changes in the current and voltage. The experimental equipment was a constant current system; therefore, the only way to change the voltage was by changing the arc gap. Changes in the arc gap ranging from 0.025 inches to 0.120 inches were unable to significantly change the voltage, and as current decreased the voltage increased. In the end, adjustments made in current and arc gap were not sufficient to compensate for an increased ionization potential caused by water vapor. Welding speed was not varied to prevent changing the weld pool shape.

The results show that arc oscillation requires tight control of the welding process, since optimal refinement parameters are sensitive to welding water vapor concentration, material composition, and heat input. Magnetic arc oscillation was shown to be an effective method for refining fusion zone grain structures in iron aluminide alloy FA-129; however, it was found to be best suited for well-controlled fabrication environments. Other techniques such as inoculation should be investigated to refine the fusion zone grain structures in field situations where precise parameter control is not available.

CONCLUSIONS

1. Conventional GTA welding produced coarse fusion zone grain structures susceptible to cracking in water vapor environments, even in fine-grained base material. Reductions in fracture strength were significant at water vapor concentrations as low as 2000 ppm.
2. Fusion zones created in unrecrystallized base material were coarser than those created in recrystallized base material. Recrystallized base material grain size did not have a significant effect on the fusion zone grain size.
3. Arc oscillation was shown to be an effective method for refining fusion zone microstructures in iron aluminide alloy FA-129 by changing the thermal gradients in the weld pool and affecting the competitive growth process.
4. Slow strain rate tensile tests showed fine-grained iron aluminide fusion zones to be less susceptible to hydrogen cracking and more tolerant of high hydrogen concentrations than coarse fusion zone grain structures. The fracture strength data was found to obey the Hall-Petch relationship.
5. Microstructural refinement by arc oscillation is suitable for well-controlled fabrication environments. Optimal refinement parameters were sensitive to numerous processing variables including welding water vapor concentration, material composition, and heat input.
6. Fusion zone inoculation or another method of fusion zone grain refinement should be considered to refine weldments produced in field fabrication situations.

ACKNOWLEDGEMENT

The authors thank the DOE Fossil Energy Program for financial support under Lockheed Martin Energy Systems, Inc. Subcontract Number 19X-27421C. The assistance and cooperation of many individuals and work groups at Oak Ridge National Laboratory are also gratefully acknowledged.

REFERENCES

1. Liu, C.T., McKamey, C.G., Lee, E.H., Scripta Metall., 24, 1990, p. 385-390.
2. Fasching, A.A., Edwards, G.R., David, S.A., "Grain Growth Kinetics of Iron Aluminide Alloy FA-129", ORNL/SUB/85-27421/05, February, 1993.
3. Goodwin, G. M., "Development of a New Hot-Cracking Test - The Sigmajig", Welding J., 66(2), 1987, p. 335-385.
4. Stoloff, N.S., Davies, R.G., "The Mechanical Properties of Ordered Alloys", Progress in Materials Science, Vol. 13, Pergamon Press Ltd., 1966.
5. Chan, K.S., "Theoretical Analysis of Grain Size Effects on Tensile Ductility", Scripta Metall., Vol. 24, 1990, p. 1725-1730.
6. Hall, E.O., Proc. Phys. Soc., (London), Vol. B64, p. 747-753, (1951).
7. Petch, H.J., Iron Steel Inst., Vol. 174, p. 25-32, (1953).
8. Cottrell, A.H., Dislocations and Plastic Flow in Crystals, Oxford Press, Oxford, 1953, p. 136.
9. Low, J.R., Symposium on Relation of Properties to Microstructure, ASM, 1954, p.163.
10. Schulson, E.M., Barker, D.R., "A Brittle to Ductile Transition in NiAl of a Critical Grain Size", Scripta Metall., Vol. 17, 1983, p. 519-522.
11. Vasudeyan, V.K., Court, S.A., Kurath, P.P., Fraser, H.L., "Effect of Grain Size and Temperature on the Yield Stress of the Intermetallic Compound TiAl", Scripta Metall., Vol. 23, 1989, p. 467-469.
12. Prakash, U., Buckley, R.A., Jones, H., Sellars, C.M., "Structure and Properties of Ordered Intermetallics Based on the Fe-Al System", ISIJ International, Vol. 31, 1991, p. 1113-1126.

13. Stoloff, N.S., Duquette, D.J., "Moisture and Hydrogen Induced Embrittlement of Iron Aluminides", *Journal of Metals*, December, 1993.
14. Lawn, B., Fracture of Brittle Solids, 2nd Ed., Chambridge Press, Cambridge, GB, 1993, p. 202-204.
15. Physical Metallurgy Principles, 2nd Ed., R.E. Reed-Hill, PWS Publishers, Boston, MS, 1973, p. 755-765.
16. Birnbaum, H.K., "Hydrogen Related Failure Mechanisms in Metals", Environmental Sensitive Fracture of Engineering Materials, ed. Z.A. Foroulis, Metall. Soc., AIME, Warrendale, PA, 1979, p. 326.
17. Hirth, J.P., "Effects of Hydrogen on the Properties of Iron and Steel", *Metall. Trans. A*, Vol. 11, 1980, p. 861-890.
18. Bond, G.M., Robertson, I.M., Birnbaum, H.K., "On the Mechanisms of Hydrogen Embrittlement of Ni₃Al Alloys", *Acta Metall.*, Vol. 34, (1989), p. 1407.
19. Robertson, I.M., Birnbaum, H.K., *Acta Metall.*, Vol. 34, (1986), p. 353.
20. Bond, G.M., Robertson, I.M., Birnbaum, H.K., *Acta Metall.*, Vol. 36, (1988), p. 2193.
21. Berstein, I.M., "The Effect of Hydrogen on the Deformation of Iron", *Scripta Metall.*, Vol. 8, 1974, p. 343-350.
22. Beachem, C.A., *Met. Trans. A*, Vol. 3, 1972, p. 437.
23. Adair, A.M., *Trans. TMS-AIME*, Vol. 236, 1966, p. 1613-1615.
24. Matsumoto, T., Eastman, J., Birnbaum, H.K., "Direct Observations of Enhanced Dislocation Mobility Due to Hydrogen", *Scripta Metall.*, Vol. 15, 1981, p. 1035-1037.
25. Birnbaum, H.K., "Hydrogen Effects on Deformation-Relation Between Dislocation Behavior", *Scripta Metall.*, Vol. 31, 1994, p. 149-153.
26. Matsumoto, T., Eastman, J., Birnbaum, H.K., *Scripta Metall.*, Vol. 15, 1981, p. 1033.

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