

FRACTURE BEHAVIOR OF ALLOY 600, ALLOY 690, EN82H WELDS AND  
EN52 WELDS IN WATER

W. J. Mills, C. M. Brown and M. G. Burke

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## ABSTRACT

The cracking resistance of Alloy 600, Alloy 690 and their welds, EN82H and EN52, was characterized by conducting  $J_{IC}$  rising load tests in air and hydrogenated water and cooldown testing in water under constant-displacement conditions. All test materials displayed excellent toughness in air and high temperature water, but Alloy 690 and the two welds were severely embrittled in low temperature water. In 54°C water with 150 cc H<sub>2</sub>/kg H<sub>2</sub>O,  $J_{IC}$  values were reduced by 70% to 95%, relative to their air counterparts.<sup>(1-4)</sup> The toughness degradation was associated with a fracture mechanism transition from microvoid coalescence to intergranular fracture. Comparison of the cracking response in water with that for hydrogen-precharged specimens tested in air demonstrated that susceptibility to low temperature crack propagation (LTCP) is due to hydrogen embrittlement of grain boundaries. The effects of water temperature, hydrogen content and loading rate on LTCP were studied. In addition, testing of specimens containing natural weld defects and as-machined notches was performed to determine if low temperature cracking can initiate at these features. Unlike the other materials, Alloy 600 is not susceptible to LTCP as the toughness in 54°C water remained high and a microvoid coalescence mechanism was operative in both air and water.

Cooldown testing of EN82H welds under constant-displacement conditions was performed to determine if LTCP data from rising load  $J_{IC} / K_{P_{max}}$  tests predict the onset of LTCP for other load paths. In these tests, bolt-loaded CT specimens were subjected to 288°C water for up to 1 week, cooled to 54°C and held in 54°C hydrogenated water for 1 week. This cycle was repeated up to 6 times. For two of the three welds tested, critical  $K_I$  levels for LTCP under constant-displacement conditions were much higher than rising load  $K_{P_{max}}$  values. Bolt-loaded specimens from a third weld were found to exhibit LTCP at  $K_I$  levels comparable to  $K_{P_{max}}$  values. Although work to date indicates that rising load tests either accurately or conservatively predict the critical conditions for LTCP under constant-displacement conditions, the potential for LTCP at  $K_I$  levels less than  $K_{P_{max}}$  has not been fully evaluated.

Annealing at 1093°C reduces or eliminates LTCP susceptibility. The microstructure and mechanical properties for susceptible and nonsusceptible EN82H welds were characterized to identify the key material parameters responsible for LTCP in the as-welded condition. The key microstructural feature associated with LTCP appears to be fine Nb- and Ti-rich carbonitrides decorating grain boundaries. In addition, the higher yield strength for the as-fabricated weld also promotes LTCP because it increases stresses and local hydrogen concentrations ahead of a crack.

# **FRACTURE BEHAVIOR OF ALLOY 600, ALLOY 690, EN82H WELDS AND EN52 WELDS IN WATER**

## **Objective**

Characterize the fracture behavior of Alloy 600, Alloy 690, EN82H and EN52 welds in water.

## **Expected failure process:**

Crack initiation and propagation due to high temperature SCC or corrosion fatigue.

Stable or unstable tearing when crack depth reaches a critical size, Controlled by fracture toughness in water.

## **Parameters studied:**

Temperature

Hydrogen content of water

Loading rate

Natural welding defects

As-machined notches

Load path

Heat Treatment



## CONCLUSIONS

High temperature (>150°C) water:

Fracture toughness of wrought and weld metals is exceptionally high in air and high temperature water.

Fracture is not a primary concern.

Low temperature water:

In low temperature water, EN82H, EN52 and Alloy 690 experience a severe degradation in fracture resistance.

Degradation in low temperature fracture resistance is associated with transition from ductile dimple rupture to intergranular cracking.

LTCP in water is due to hydrogen embrittlement mechanism.

Cracking resistance is recovered at loading rates above 26,000 MPa√m/h (300 mm/h),

insufficient time to embrittle grain boundaries ahead of crack.

LTCP does not initiate at as-machined notches, but can initiate at sharp weld defects.

Alloy 600 is not susceptible to LTCP, even after 10-16% cold work.

Cooldown testing - EN82H under constant displacement conditions:

$K_{P_{max}}$  conservatively predicted critical  $K_I$  level for LTCP in two welds.

$K_I$  for LTCP in a third weld was consistent with rising load  $K_{P_{max}}$ .

Annealing at 1093°C reduces or eliminates LTCP susceptibility.

LTCP susceptibility in welds is correlated with the presence of fine Nb- & Ti-rich carbonitrides decorating grain boundaries.



## EXPERIMENTAL PROCEDURES

### Materials:

- Alloy 600 – 2 heats (as-received & cold worked)
- Alloy 690 – 2 heats
- EN82H – 7 GTA welds (3 manufacturers: 'A', 'B', 'C')
- EN52 – 3 GTA welds (2 manufacturers: 'A', 'B')

### Materials were tested in:

- as-received or as-welded condition &
- annealed at 1093°C & furnace cooled condition.

### Test Environments:

24°-338°C Air

24°-338°C Water:

pH of 10.1 to 10.3

150, 50 & 15 cc H<sub>2</sub>/kg H<sub>2</sub>O

3-17 ppb O<sub>2</sub>

### J<sub>IC</sub> Fracture Toughness Testing

ASTM E1737-96 & J<sub>IC</sub> normalization procedure<sup>(5)</sup>

0.6T CT Specimens (20% side groove)

Precracked,

As-notched, Weld root defects.

### K<sub>Pmax</sub> Testing of Hydrogen-Precharged (45-70 ppm) CT Specimens

Precharged in 99.999% H<sub>2</sub> at 360°C for 6 weeks.

### Cooldown Testing Under Constant Displacement Conditions

Bolt-loaded 0.6T CT cooled from 288°C to 54°C.

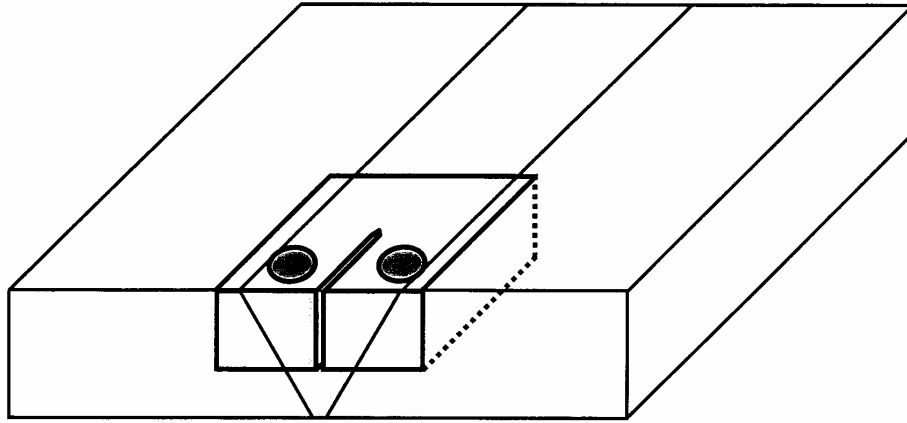
### Characterization of Microstructure:

Analytical Electron Microscopy (AEM)<sup>(6)</sup>

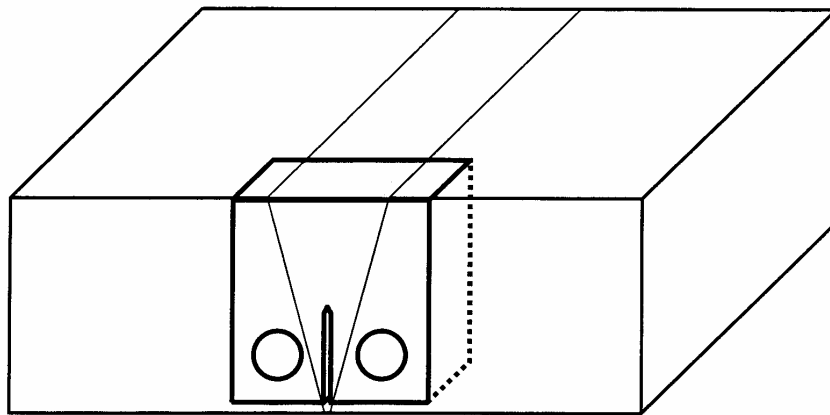
Auger Electron Spectroscopy (AES)



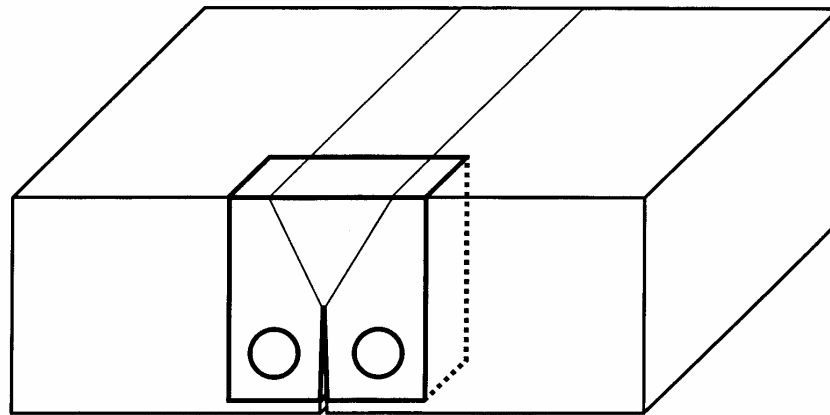
# WELD SPECIMEN ORIENTATION



Longitudinal CT



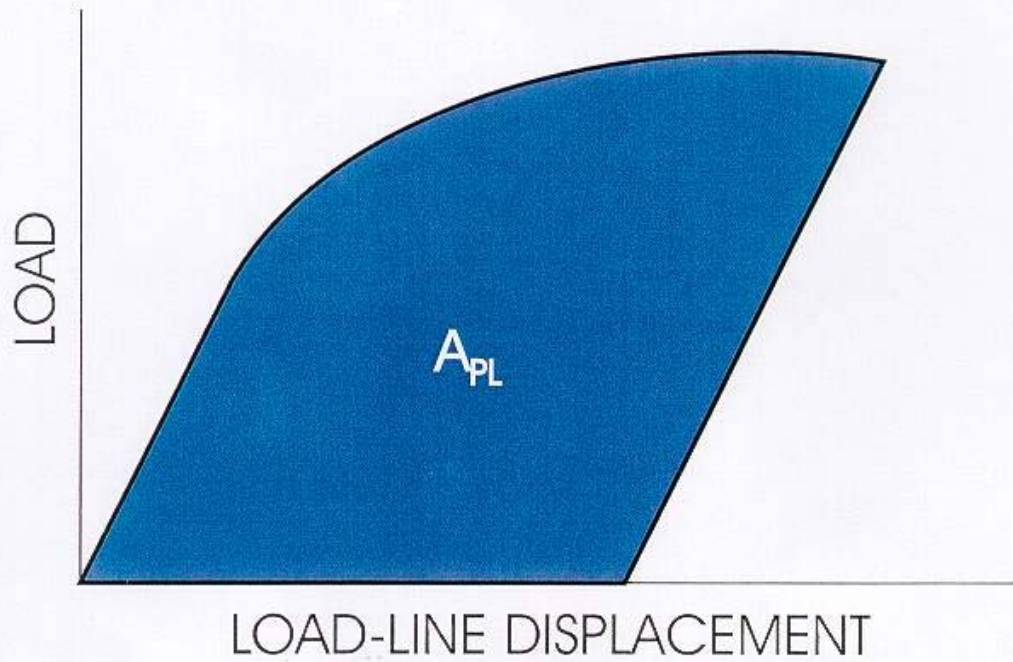
Transverse CT



Transverse CT



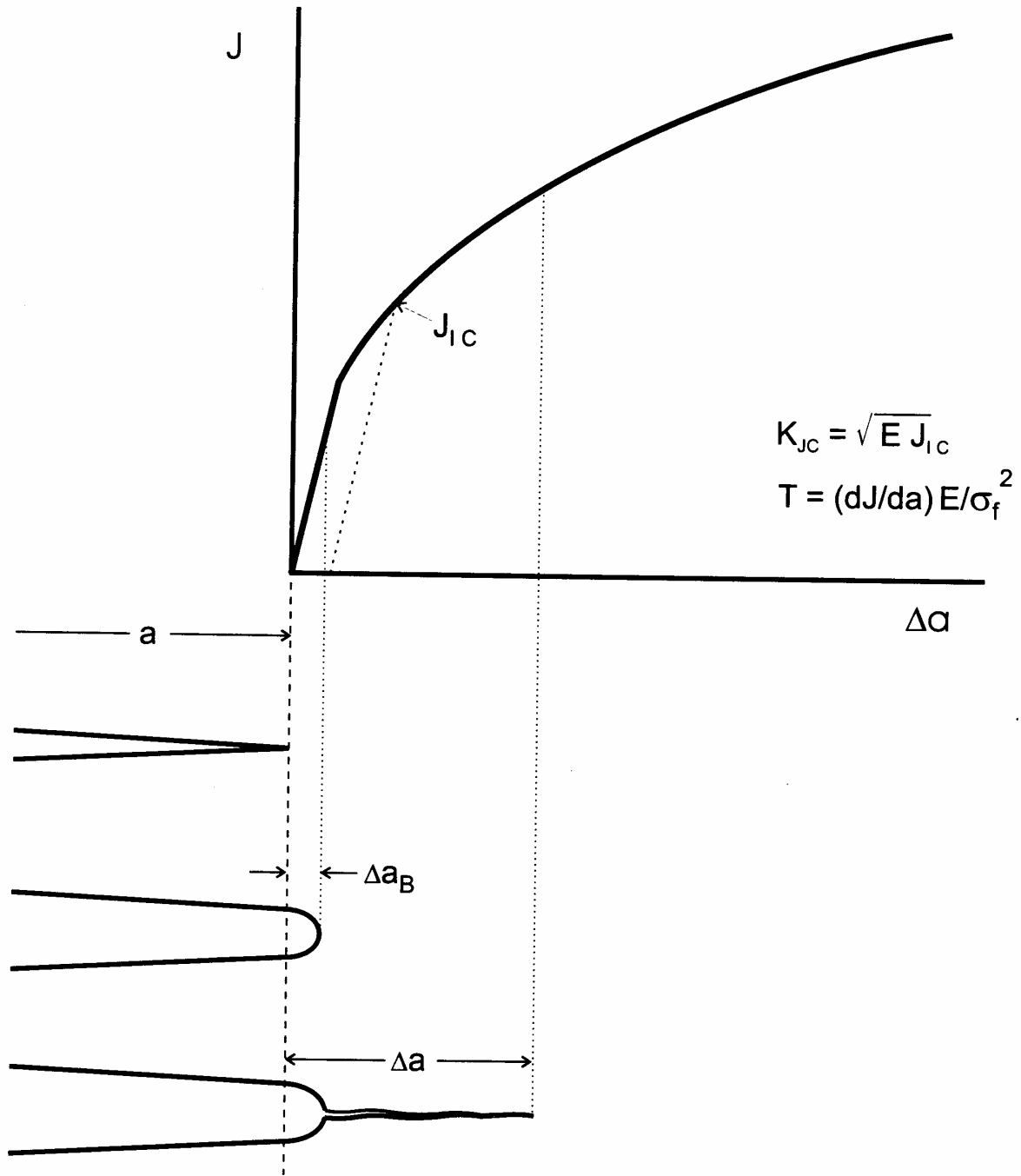
# Calculation of J



$$J_{PL} = \frac{\eta A_{PL}}{B_N b}$$

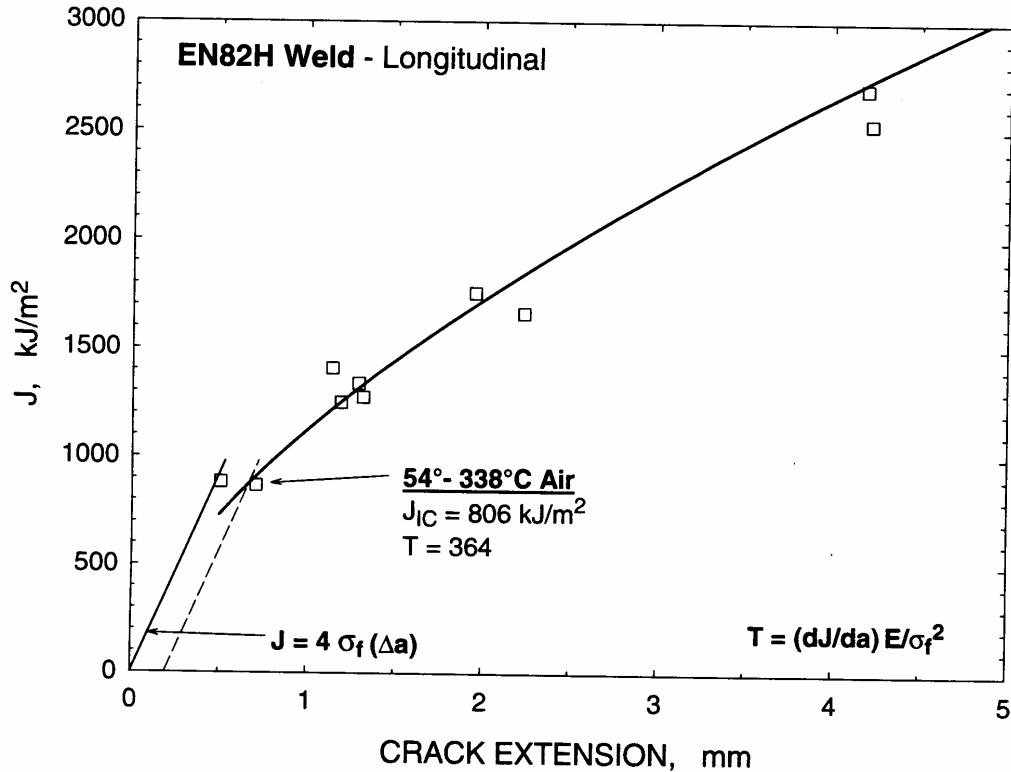
$$J = J_{PL} + \frac{K^2 (1 - \nu^2)}{E}$$

# Schematic Diagram of J-R Curve & Corresponding Cracking Behavior





## J-R Curve for EN82H Weld Tested in 54° & 338°C Air



Category I:  $J_{IC} < 30 \text{ kJ/m}^2$  ( $K_{IC} < 75 \text{ MPa}\sqrt{\text{m}}$ ),  $T < 10$

Low toughness material where failure can occur below yield strength loadings for relatively small flaw sizes.

Category II:  $30 < J_{IC} < 150 \text{ kJ/m}^2$  ( $75 < K_{IC} < 150 \text{ MPa}\sqrt{\text{m}}$ ),  $10 < T < 100$

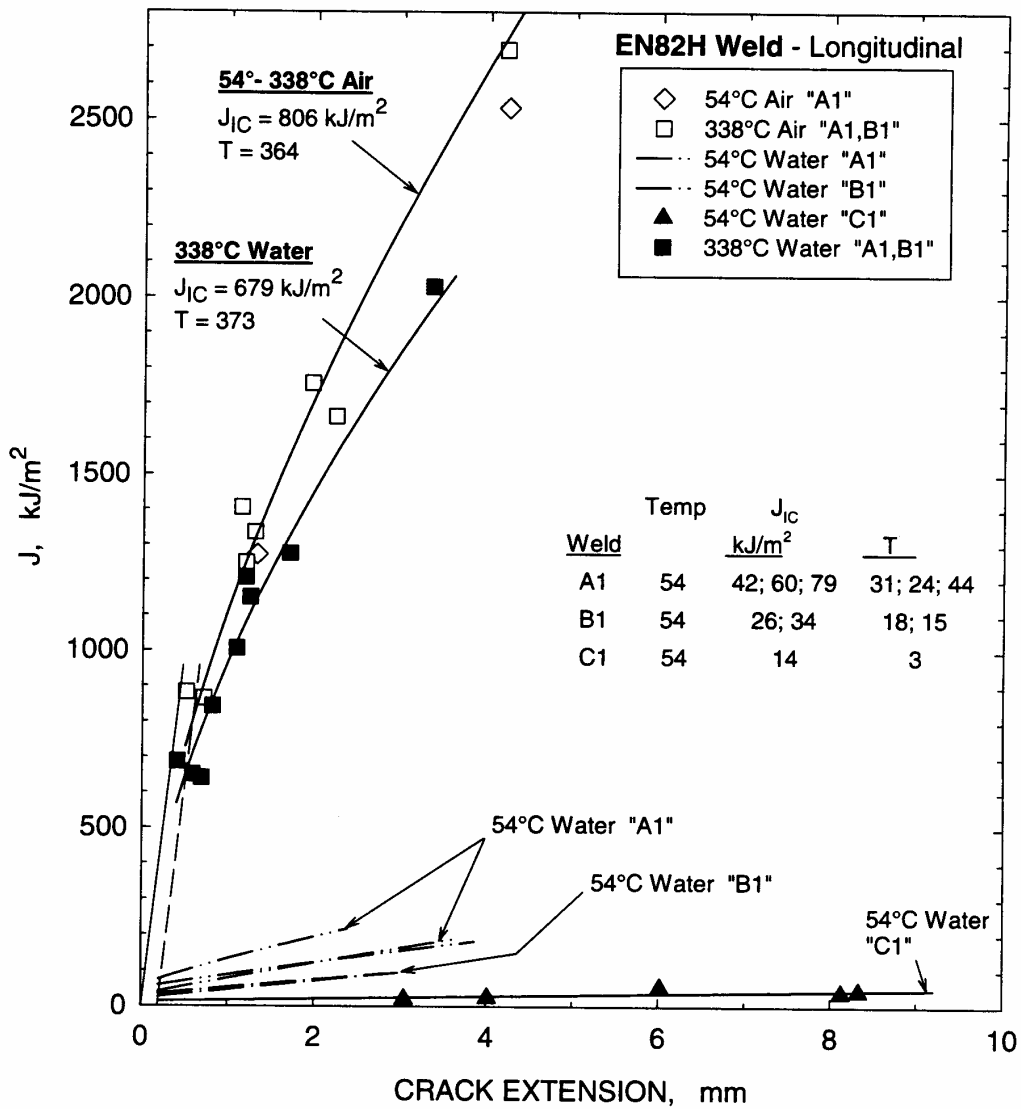
Intermediate toughness material where unstable or stable fracture can occur at approximately yield strength loadings for small to medium flaw sizes.

Category III:  $J_{IC} > 150 \text{ kJ/m}^2$  ( $K_{IC} > 150 \text{ MPa}\sqrt{\text{m}}$ ),  $T > 100$

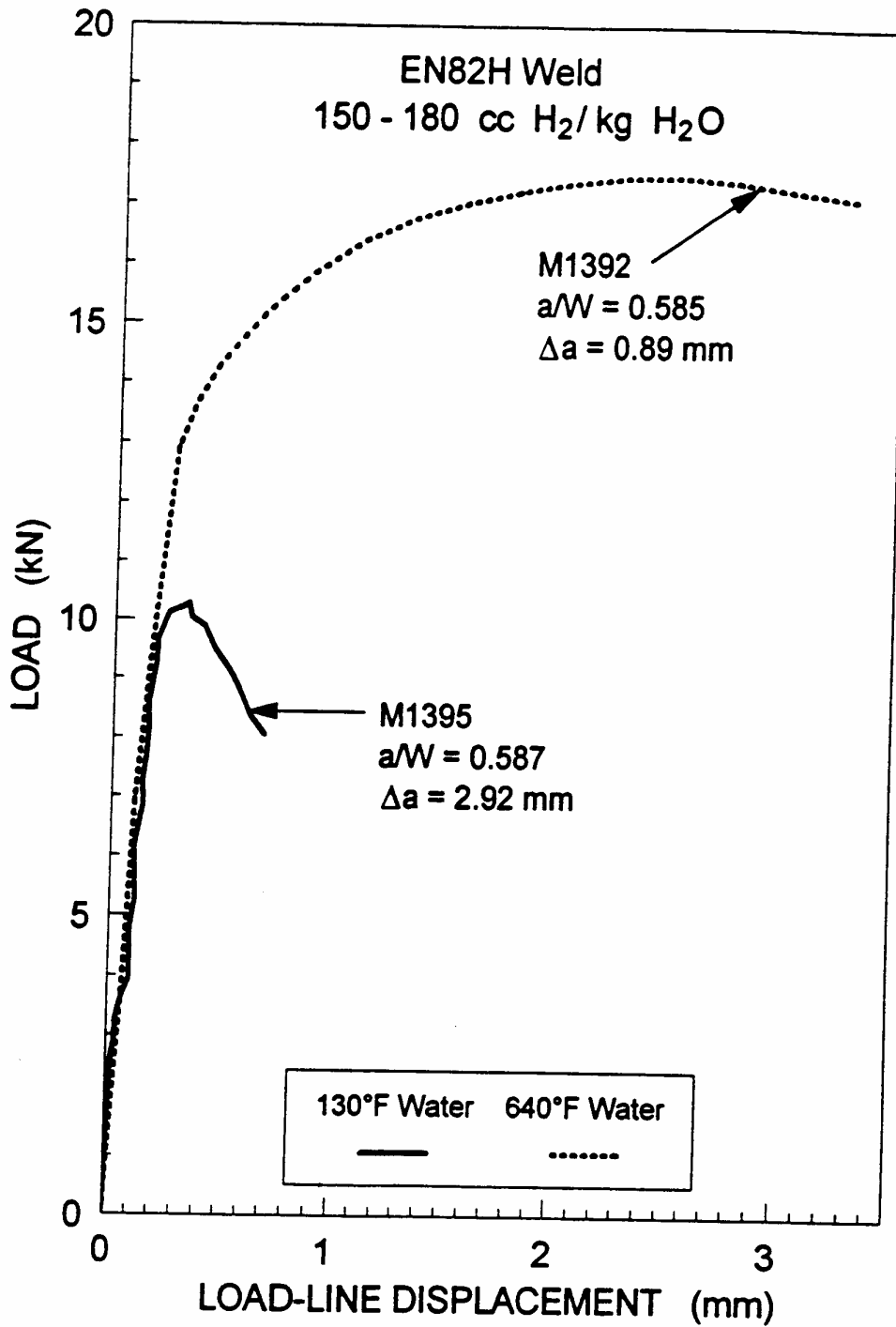
High toughness material where fracture involves stable tearing at stresses well above yield strength. Tearing instabilities are unlikely except after gross plastic deformation.



# Effect of Low and High Temperature Water on J-R Curve for EN82H Welds

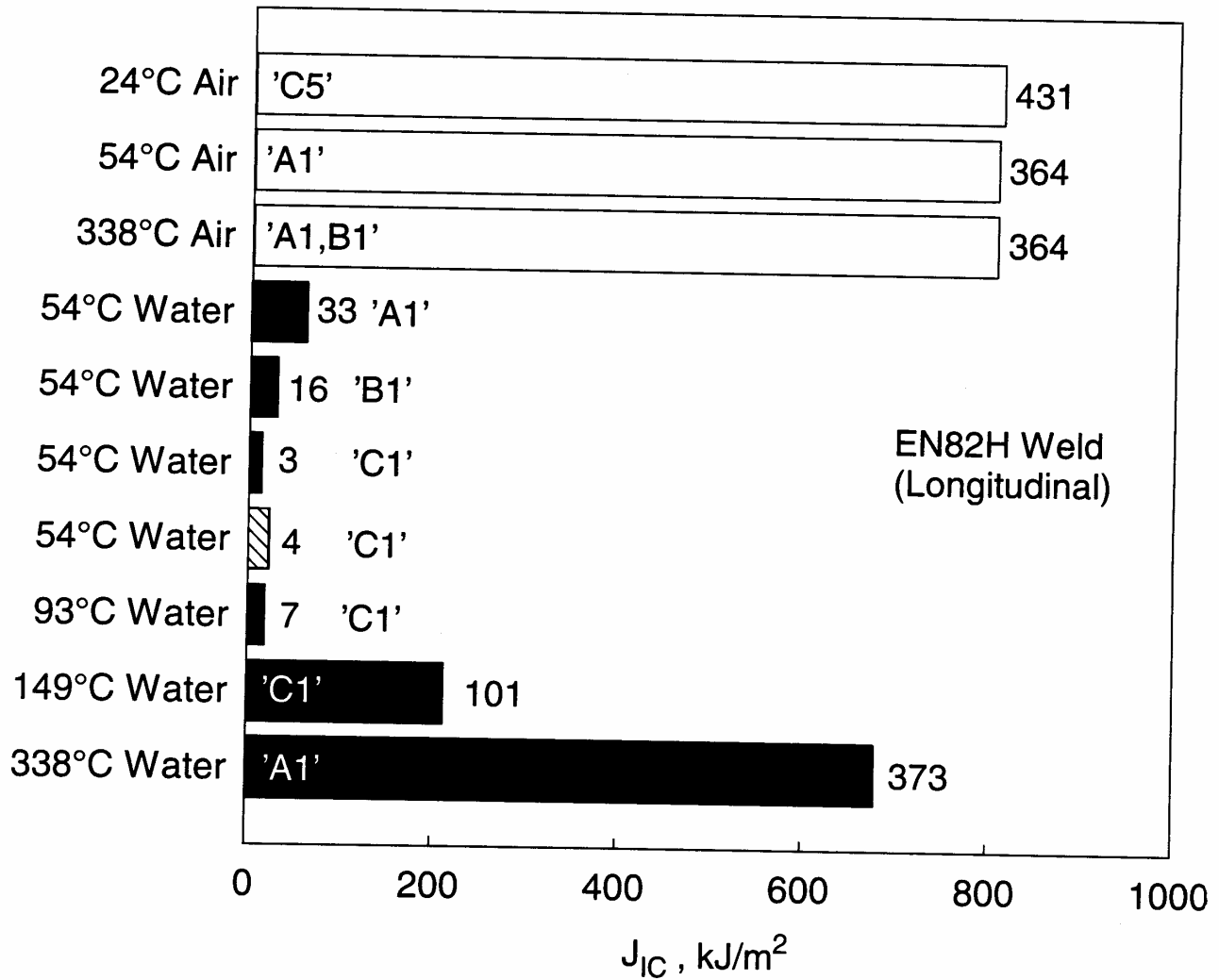


# Comparison of Load-Displacement Curves for EN82H Weld Tested in 54°C & 338°C Water



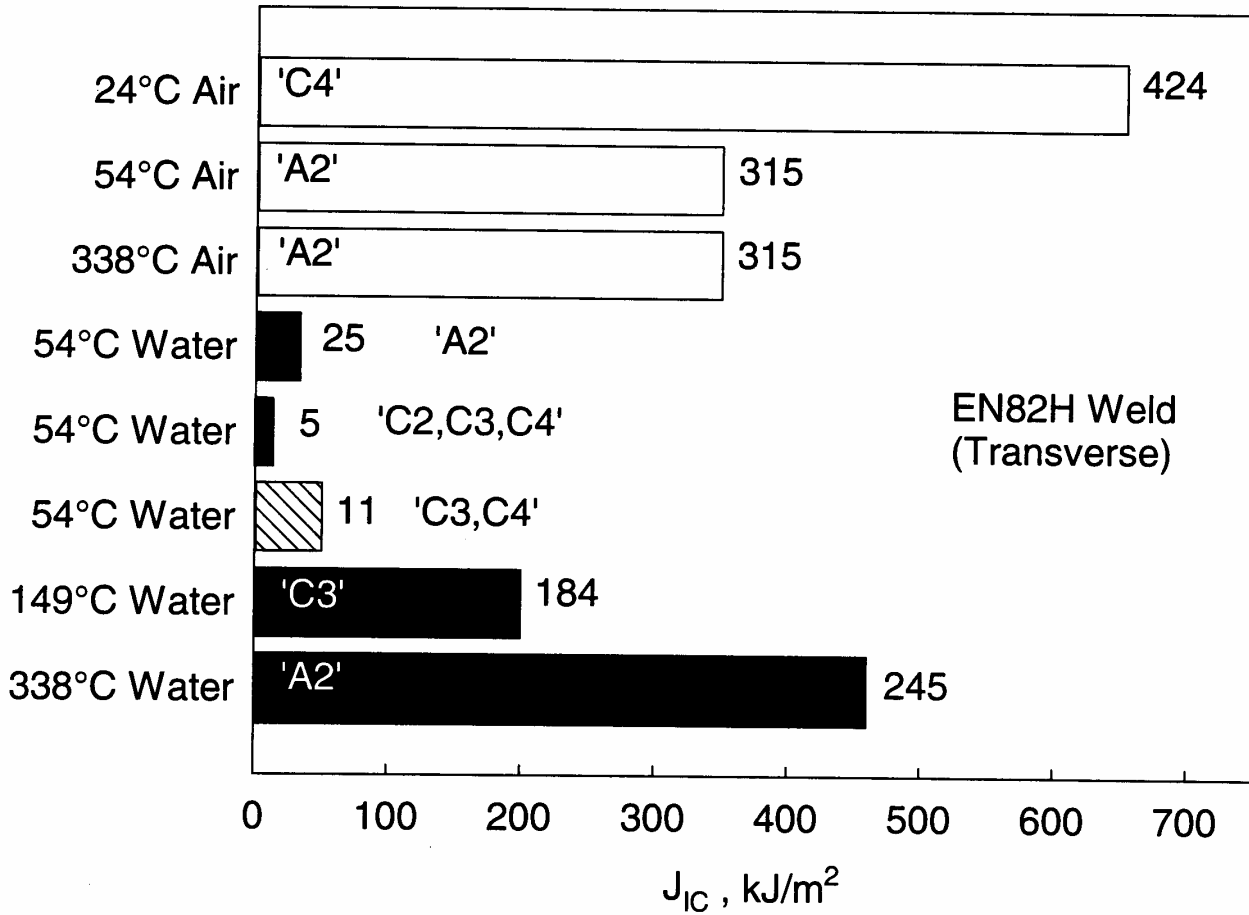
# Fracture Toughness of EN82H Weld (Longitudinal Orientation) in Air & Water

(Values of T are provided beyond each bar)



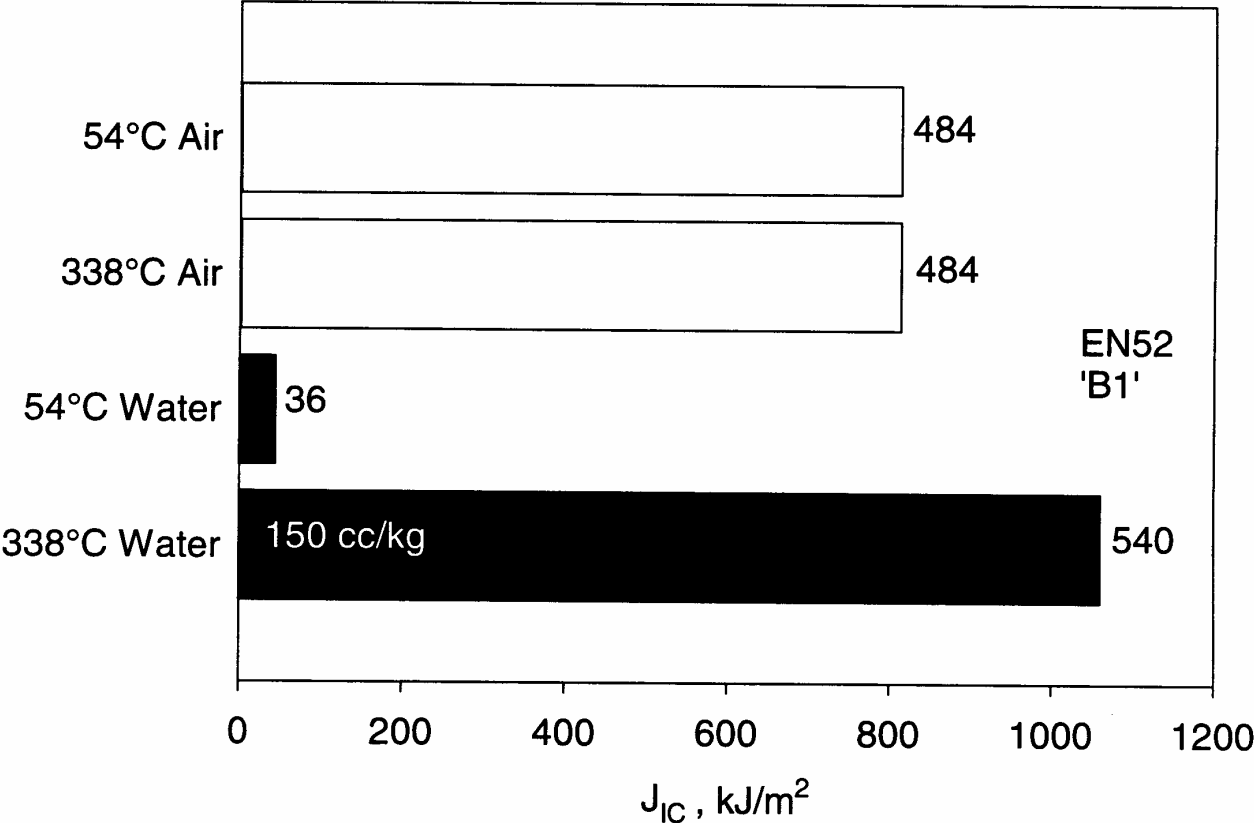
# Fracture Toughness of EN82H Weld (Transverse Orientation) in Air & Water

(Values of T are provided beyond each bar)

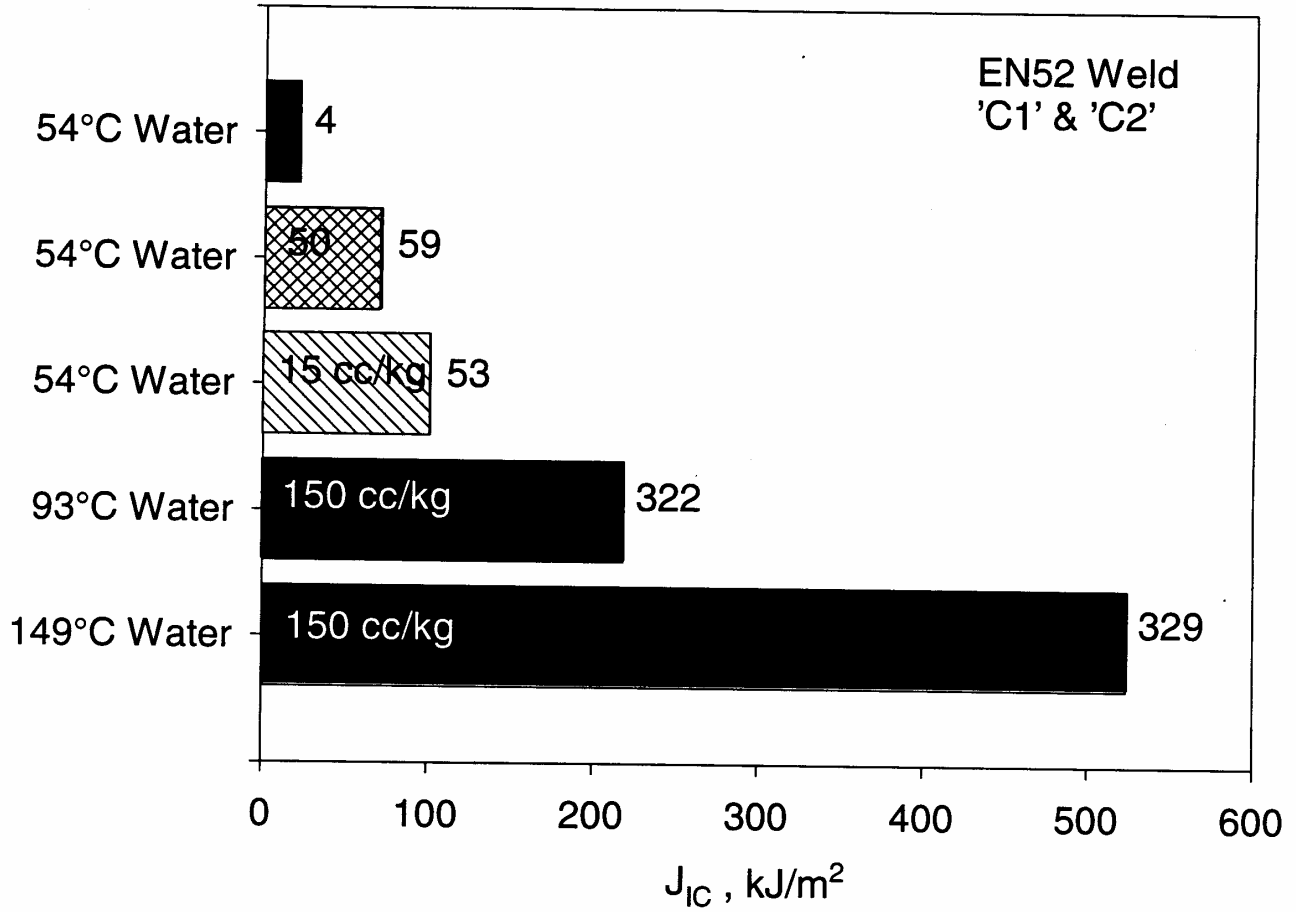


# Fracture Toughness of EN52 Weld "B1" in Water

(Values of T are provided beyond each bar)

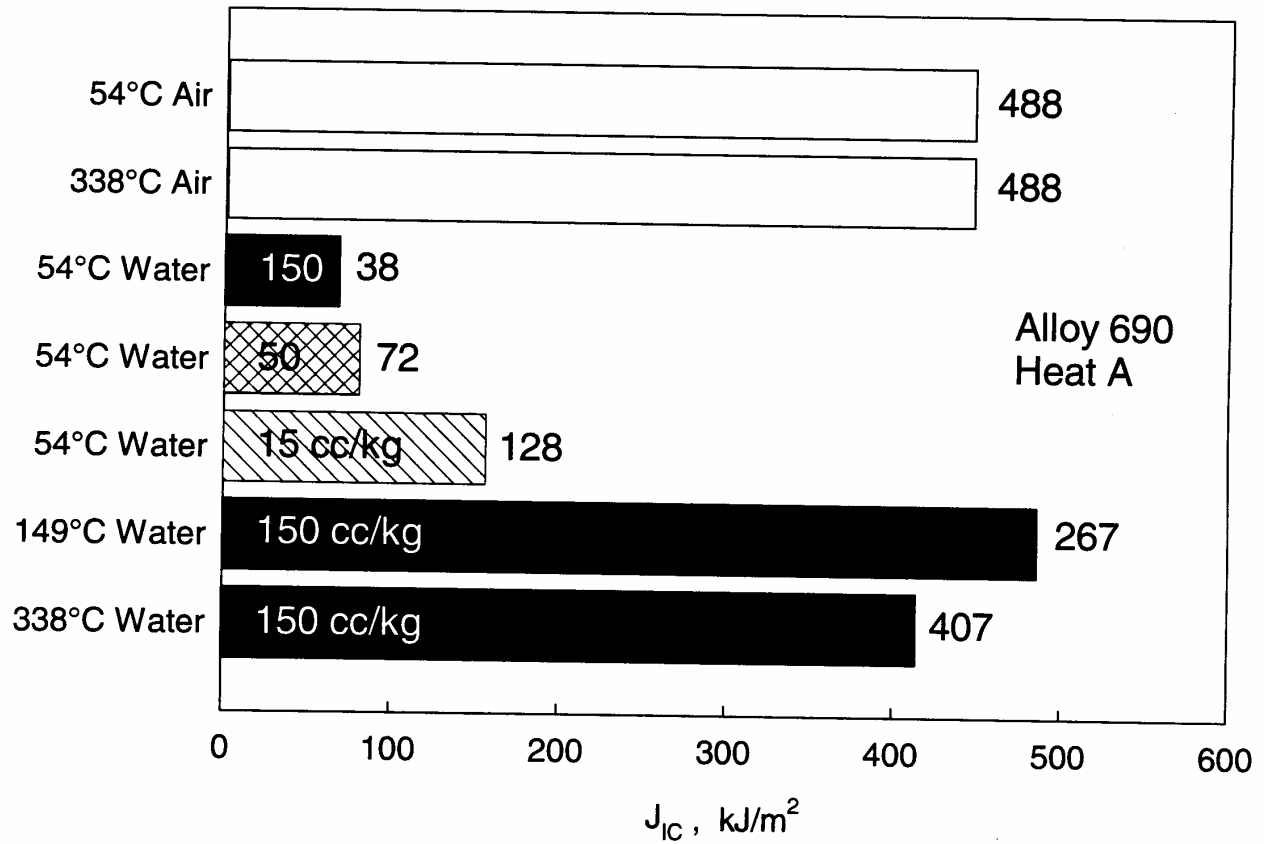


Fracture Toughness of EN52 Welds "C1" & "C2" in Water  
(Values of T are provided beyond each bar)



# Fracture Toughness of Alloy 690 (Heat A) in Air & Water

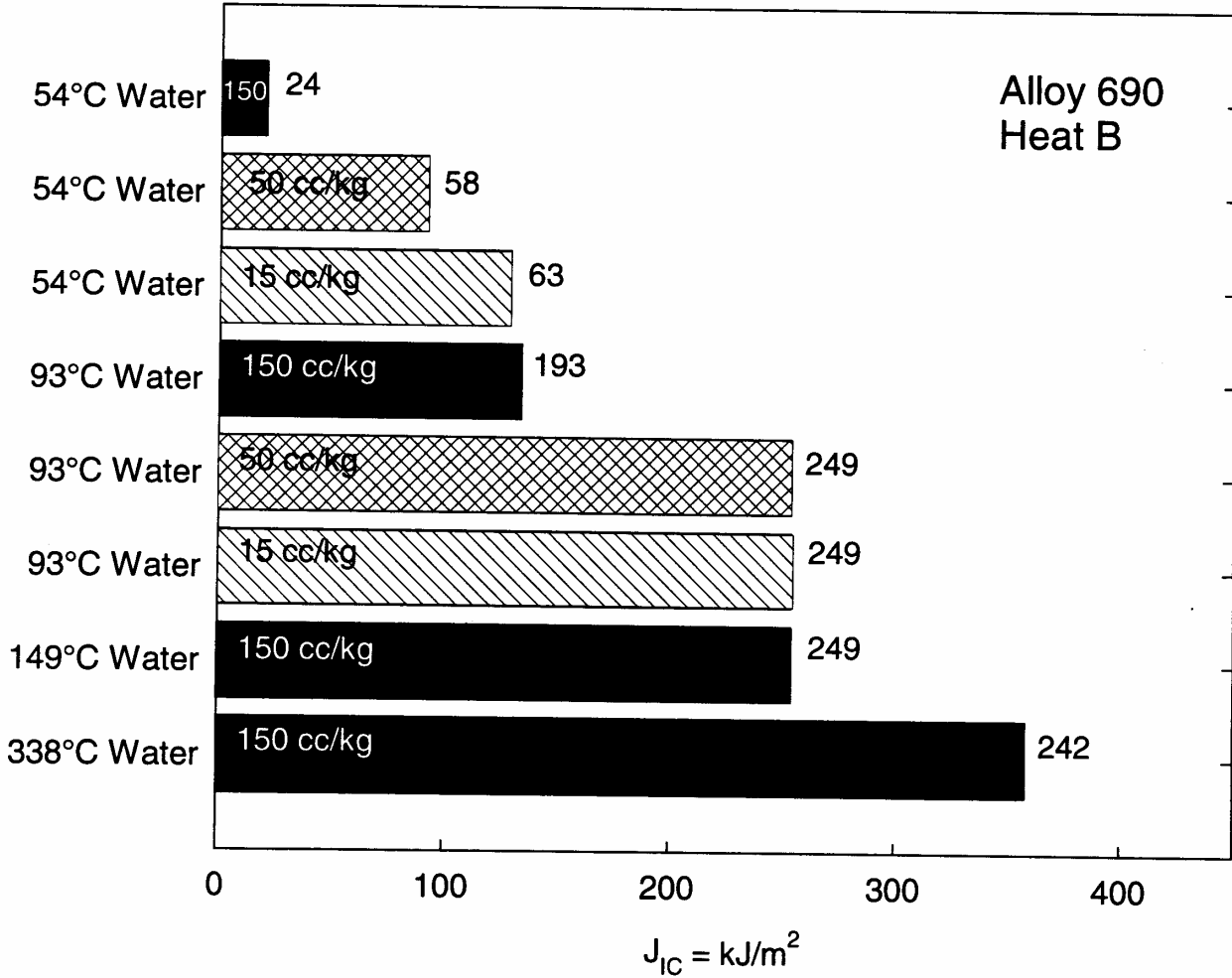
(Values of T are provided beyond each bar)





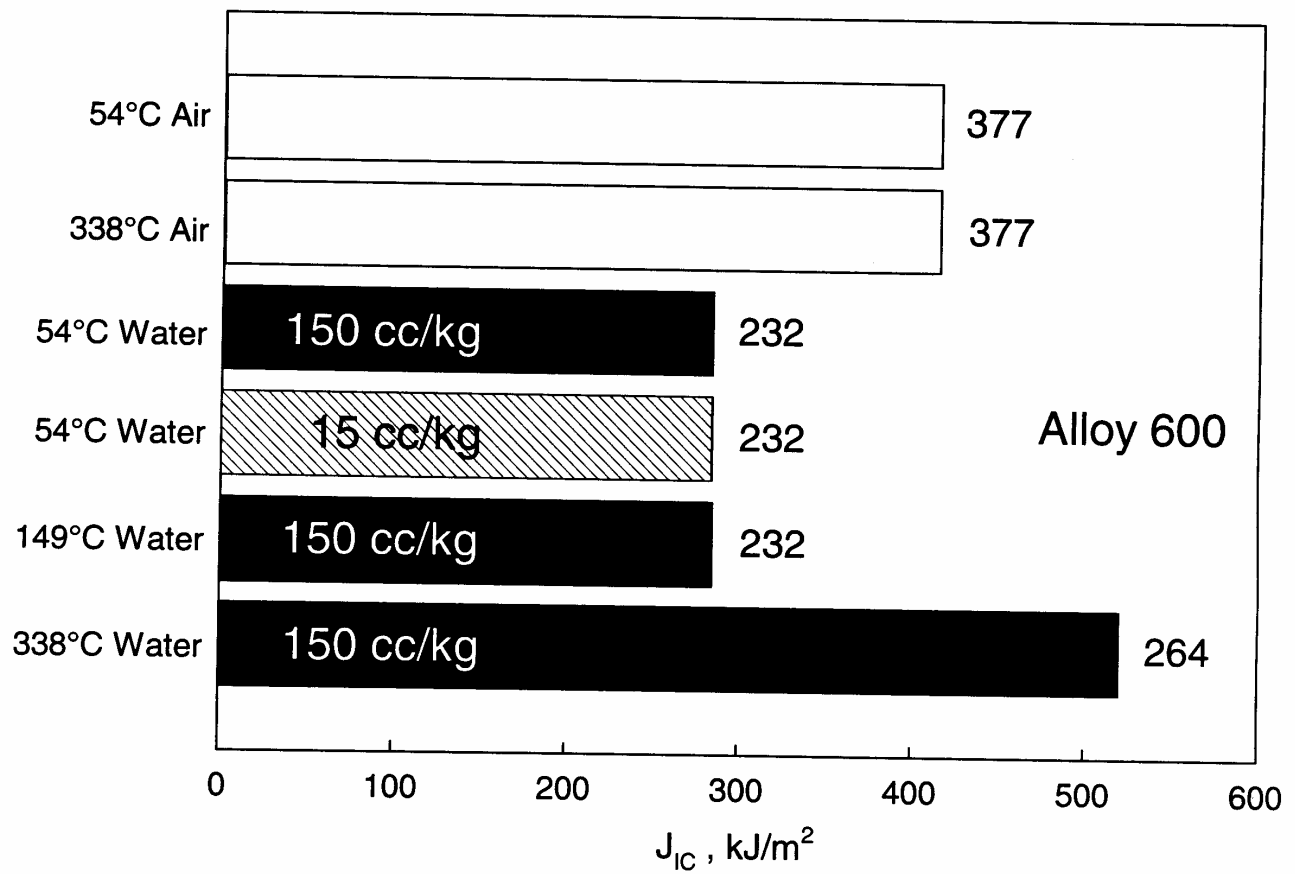
## Fracture Toughness of Alloy 690 (Heat B) in Water

(Values of T are provided beyond each bar)

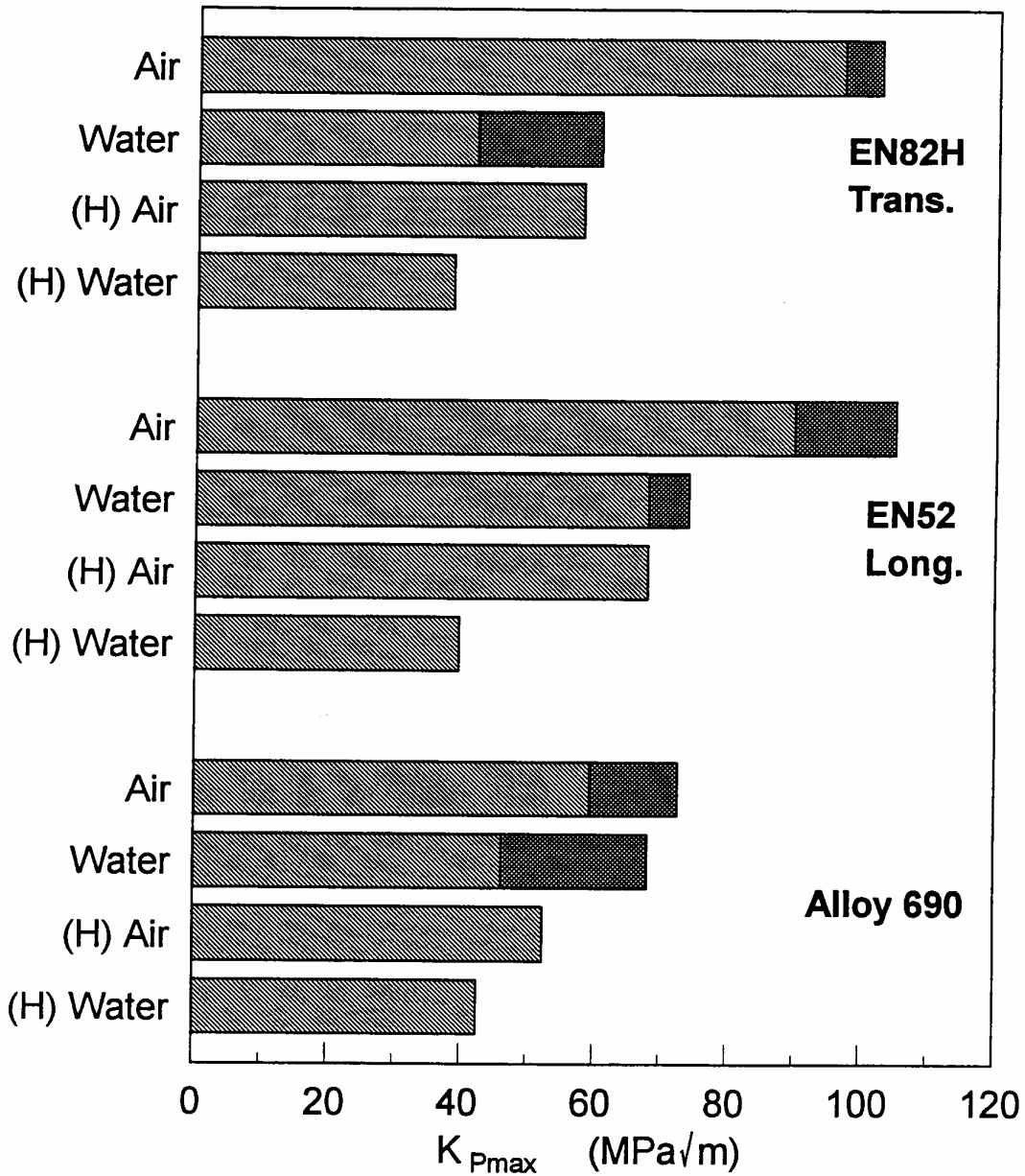


## Fracture Toughness of Alloy 600 (Heat A) in Air & Water

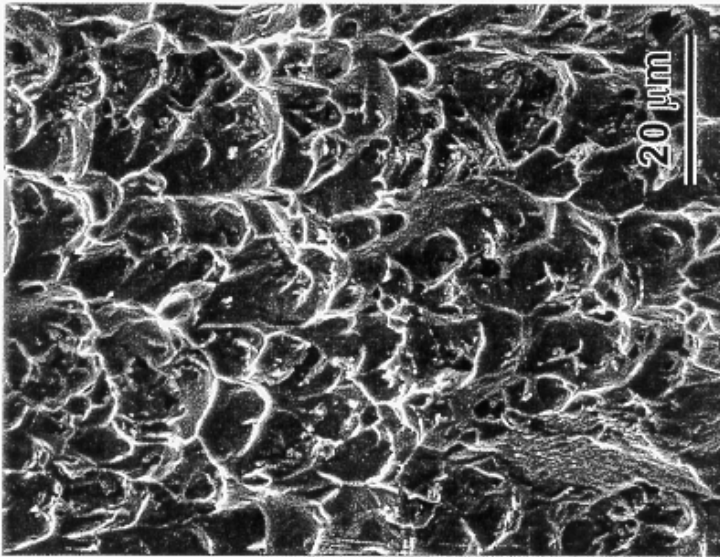
(Values of T are provided beyond each bar)



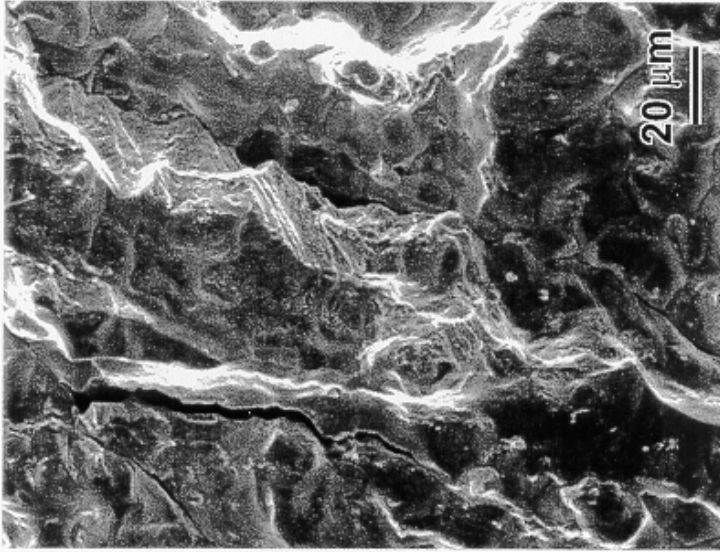
$K_{Pmax}$  values for EN82H, EN52 & Alloy 690  
 Non-precharged & Hydrogen-Precharged (H) Specimens  
 Tested in 24°/54°C Air & Water



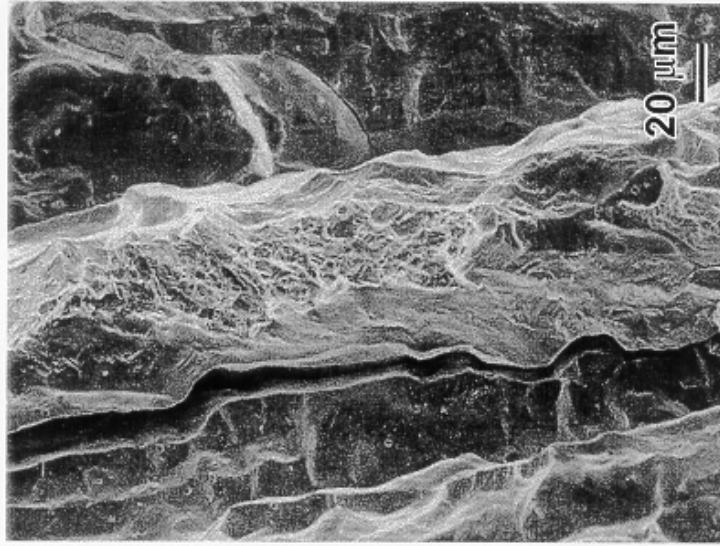
# EN82H Weld



54°C Air  
Non-precharged

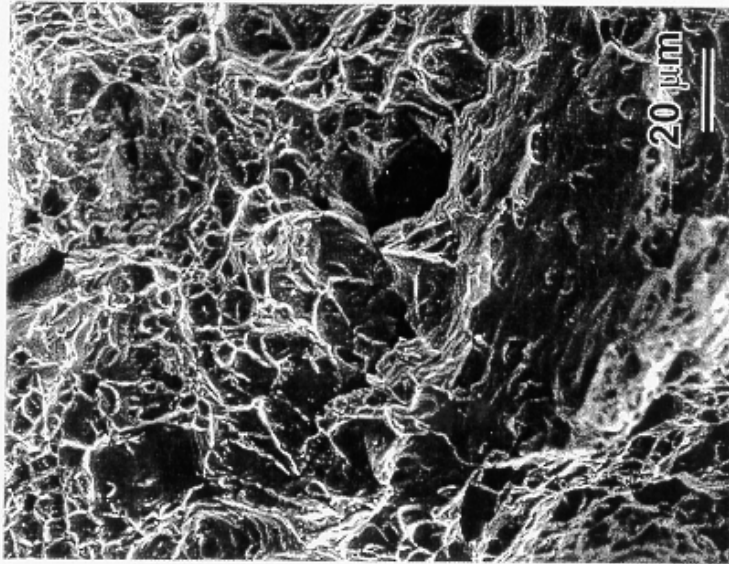


54°C Water  
Non-precharged

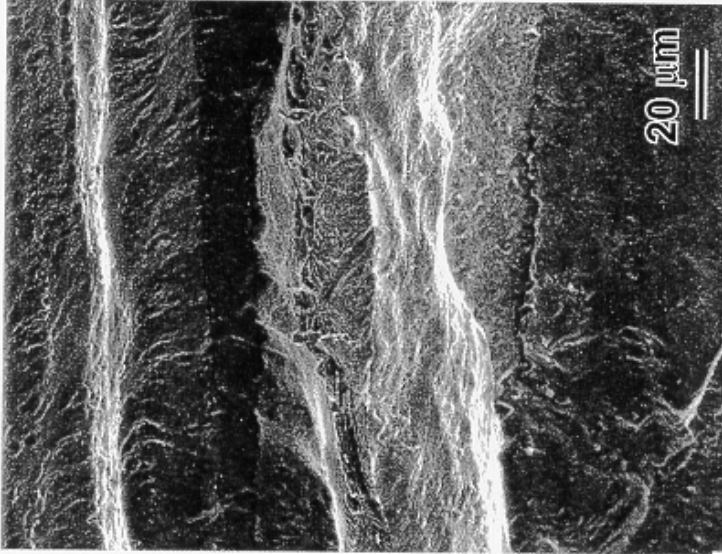


24°C Air  
Hydrogen-precharged

# EN52 Weld



54°C Air  
Non-precharged

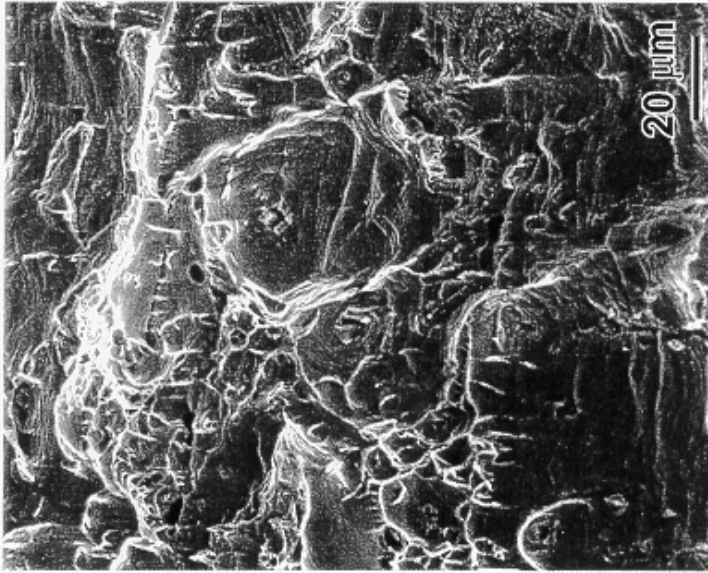


54°C Water  
Non-precharged

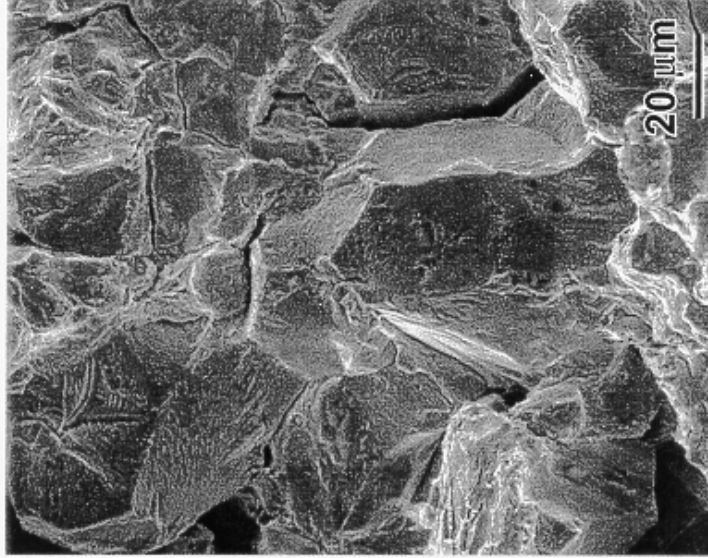


24°C Air  
Hydrogen-precharged

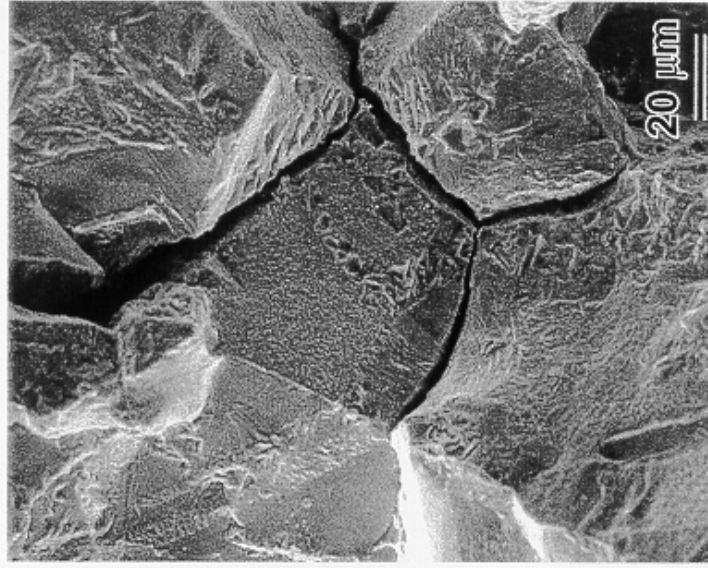
# Alloy 690



54°C Air  
Non-precharged

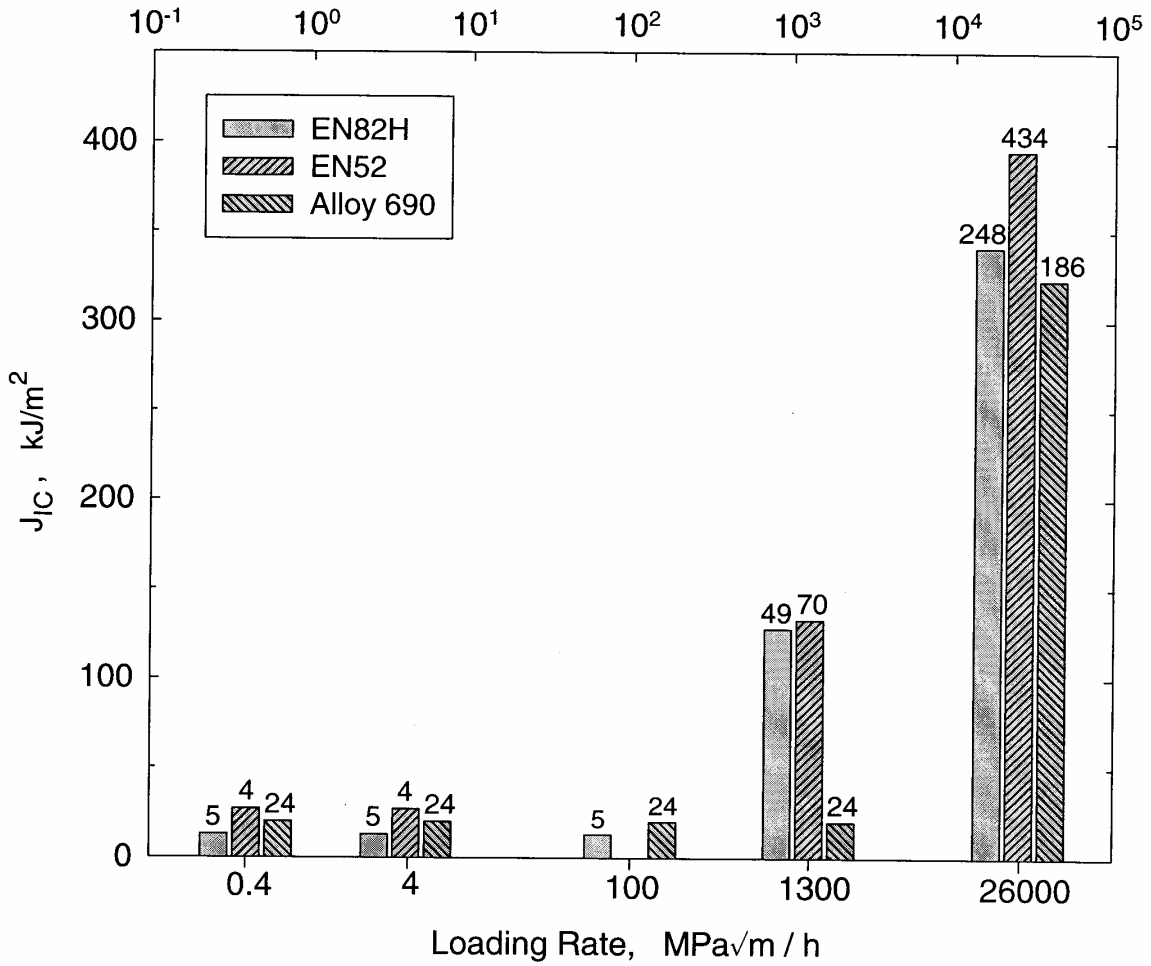


54°C Water  
Non-precharged

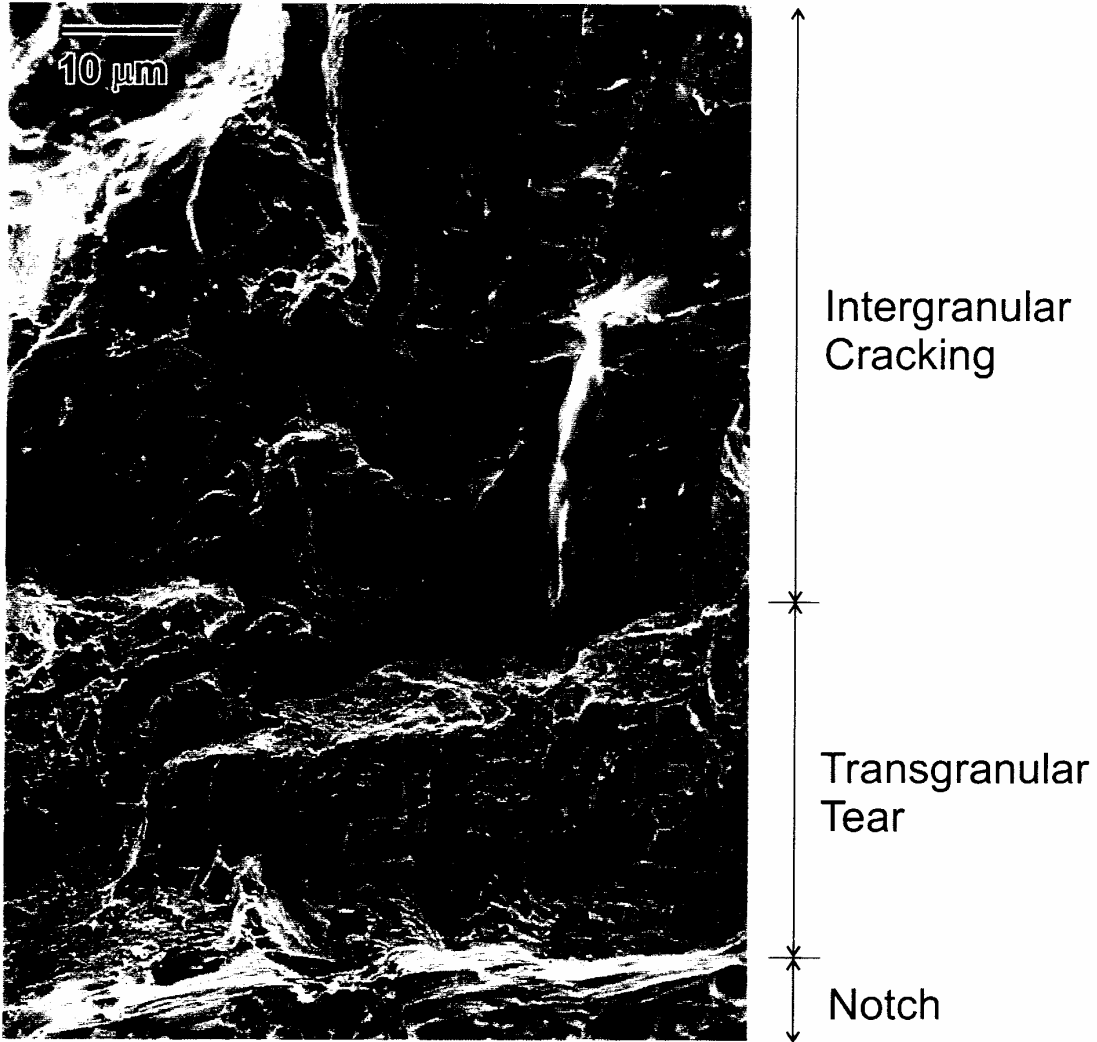


24°C Air  
Hydrogen-precharged

**Effect of Loading Rate on Fracture Toughness  
of EN82H, EN52 and Alloy 690 in 54°C Water**  
(Values of T are provided above each bar)

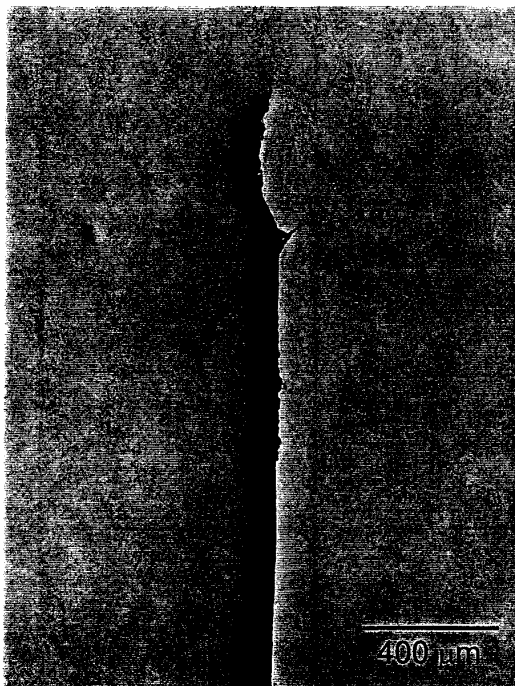
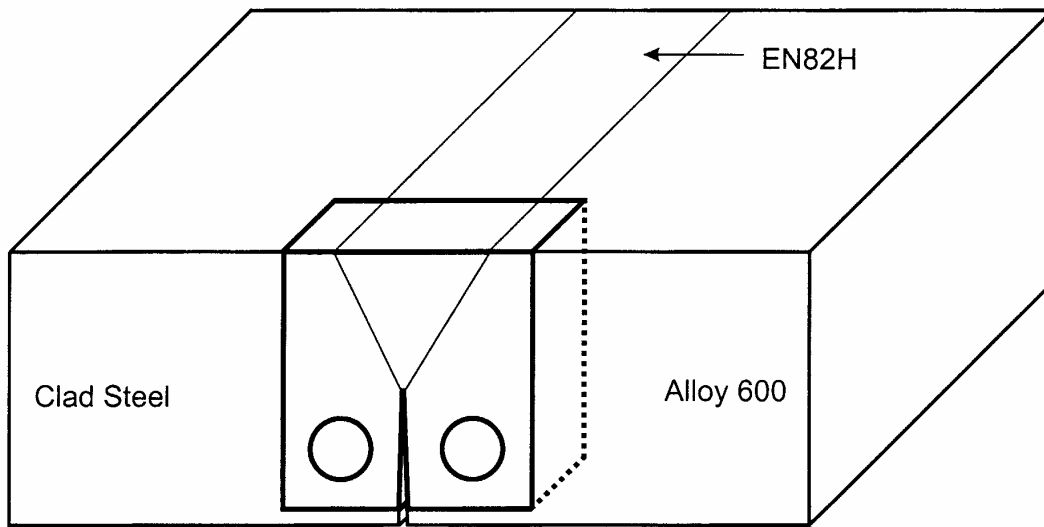


LTCP does not initiate at a notch. However, once a tear forms at a notch, it serves as a sharp crack from which intergranular LTCP initiates.

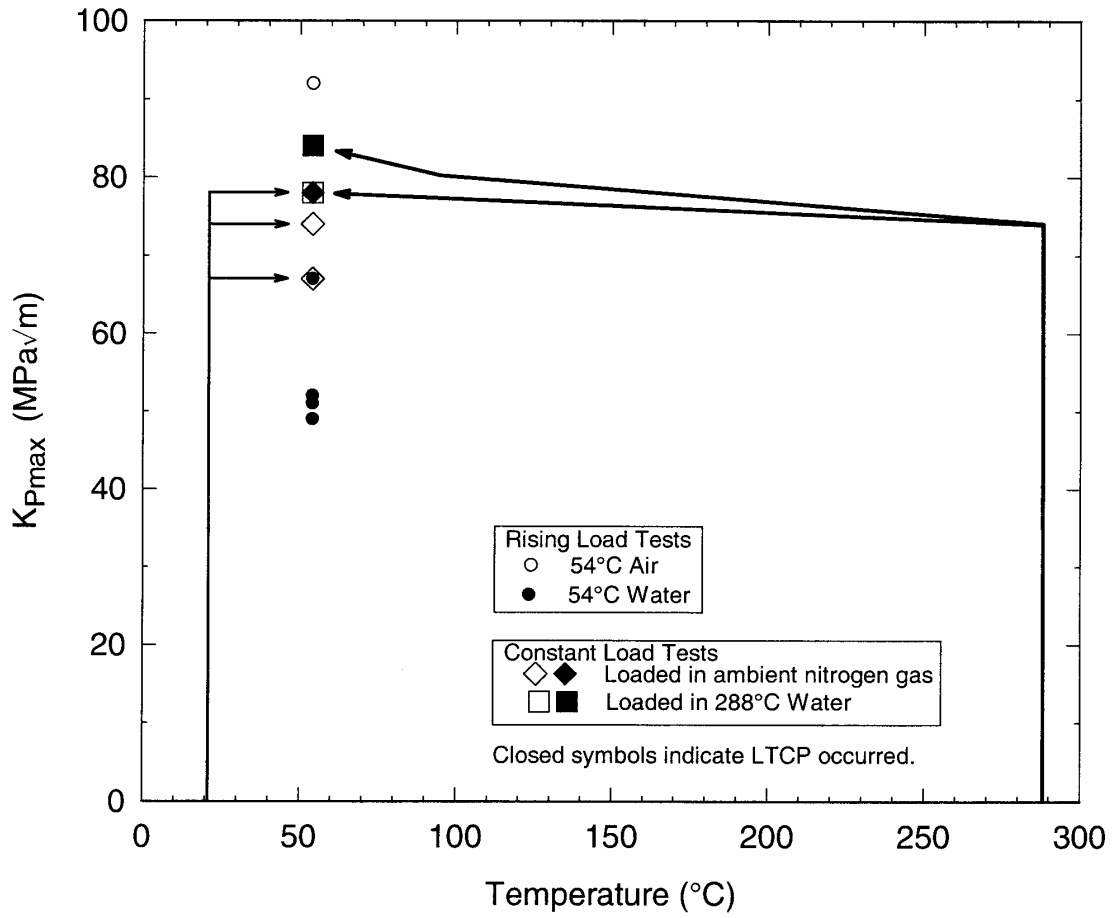




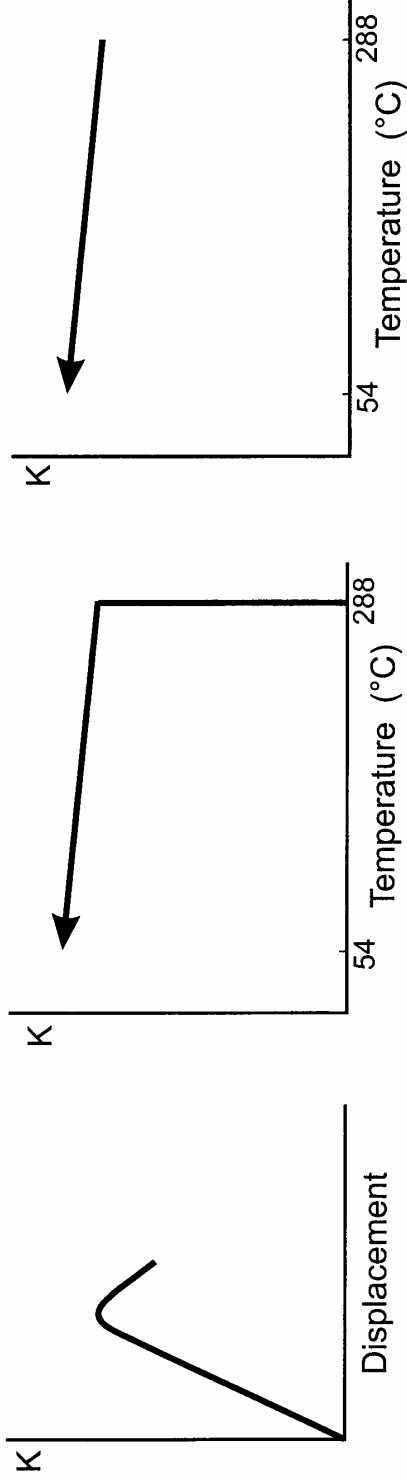
Specimens with fatigue precracks and natural weld root defects exhibited similar LTCP properties in water.



## Effect of Load Path on LTCP Behavior for EN82H Weld in Water with 150 cc H<sub>2</sub>/kg H<sub>2</sub>O



# TEMPERATURE & LOAD PATH HISTORY EFFECTS IN HYDROGENATED WATER



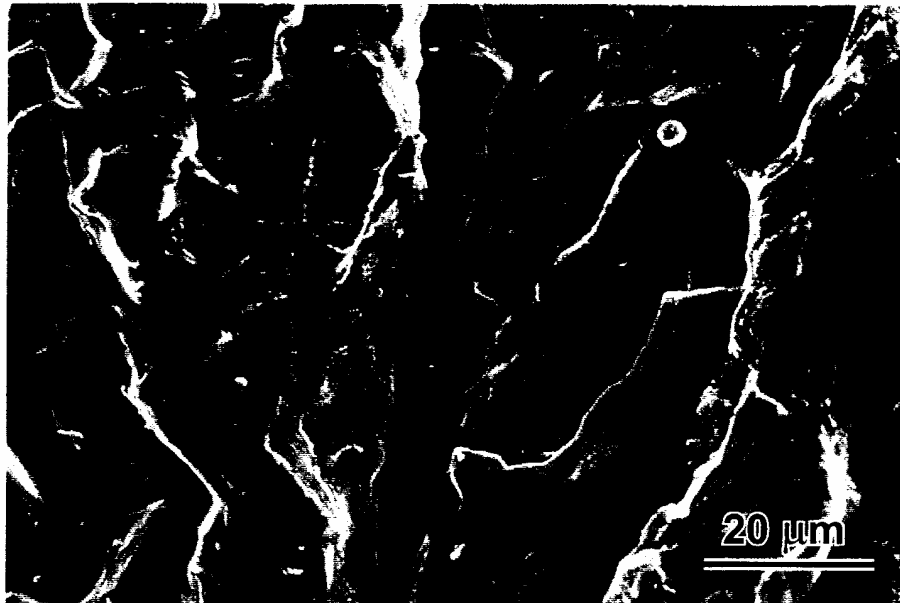
| Weld | $K_{Pmax}$<br>MPa√m | Cracking<br>Response | MPa√m      | Cracking<br>Response |
|------|---------------------|----------------------|------------|----------------------|
| C4   | 49-67               | No LTCP              | 47, 49, 57 | No LTCP*             |
| A2   | 43-56               | LTCP                 | 62, 67, 78 | No LTCP*             |
| C2   | 48-60               | LTCP                 | 78, 82     | No LTCP*             |
|      |                     |                      | 73         | LTCP initiated**     |
|      |                     |                      | 46, 58     | No LTCP*             |
|      |                     |                      | 53, 59, 73 | LTCP**               |

288°C for 1 week / 54°C for 1 week  
 \* 6 cycles  
 \*\* 2-3 cycles

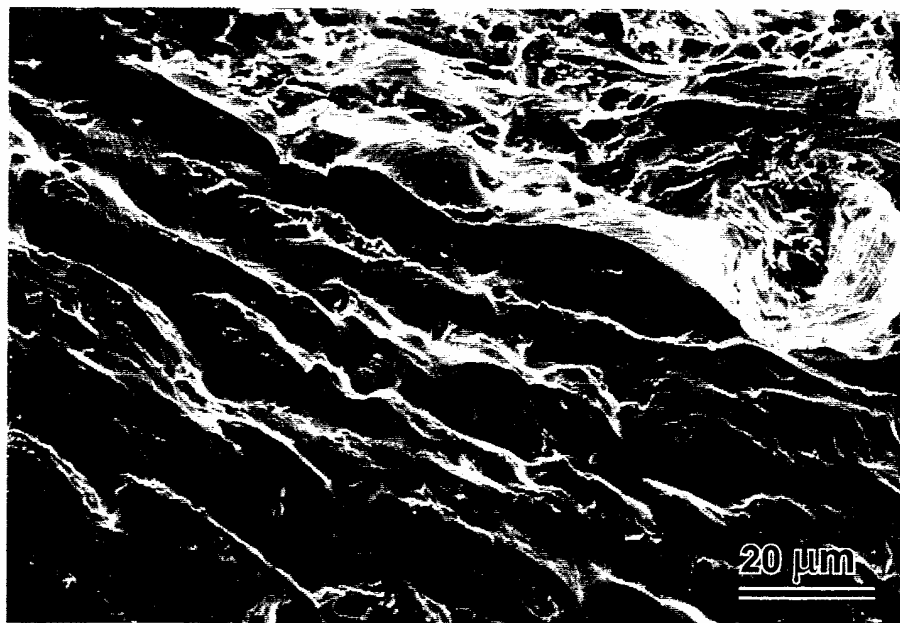


# As-welded EN82H Tested in Water with 150 cc H<sub>2</sub>/kg H<sub>2</sub>O

Predominantly intergranular cracking  
in bolt-loaded specimen

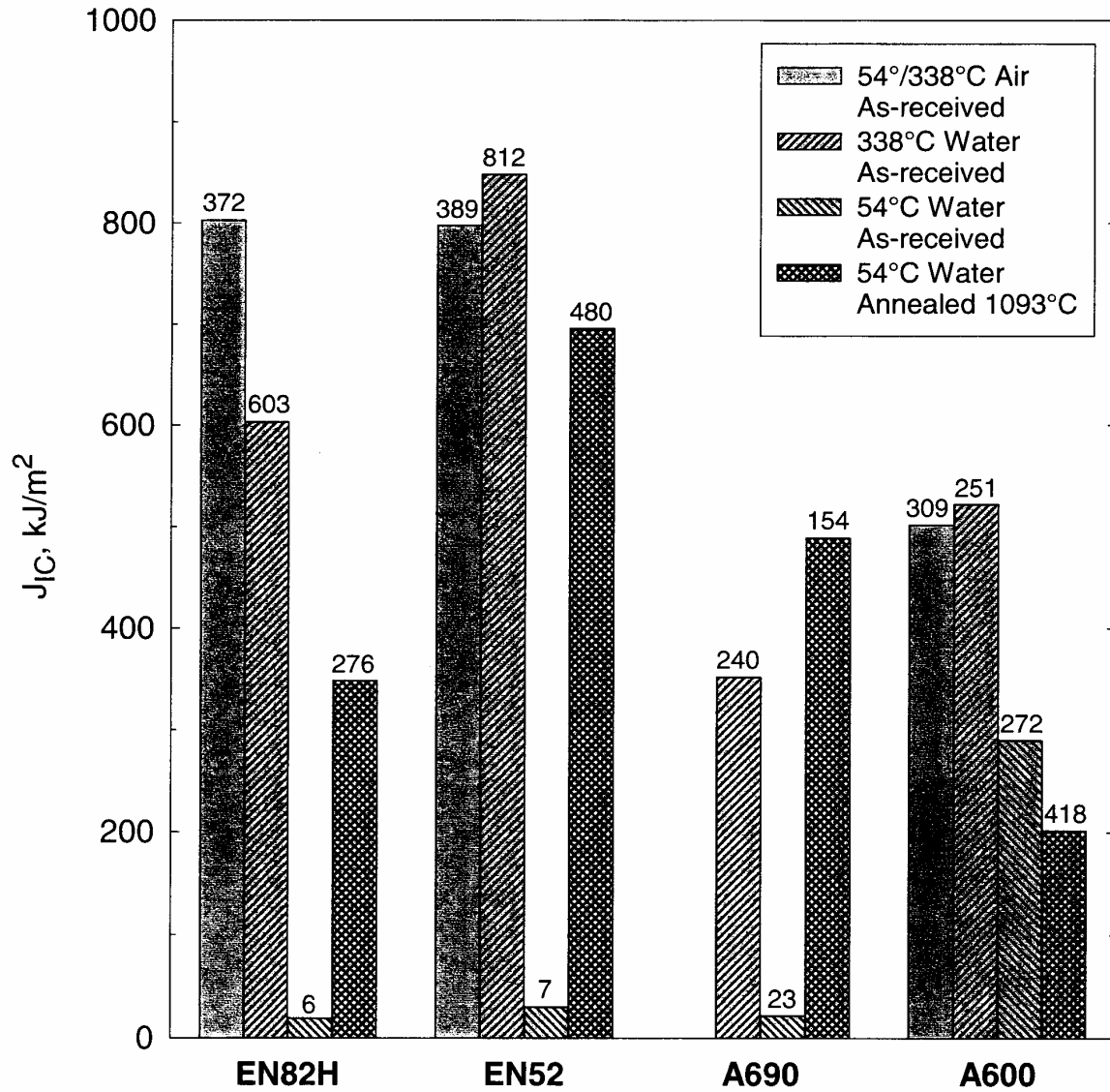


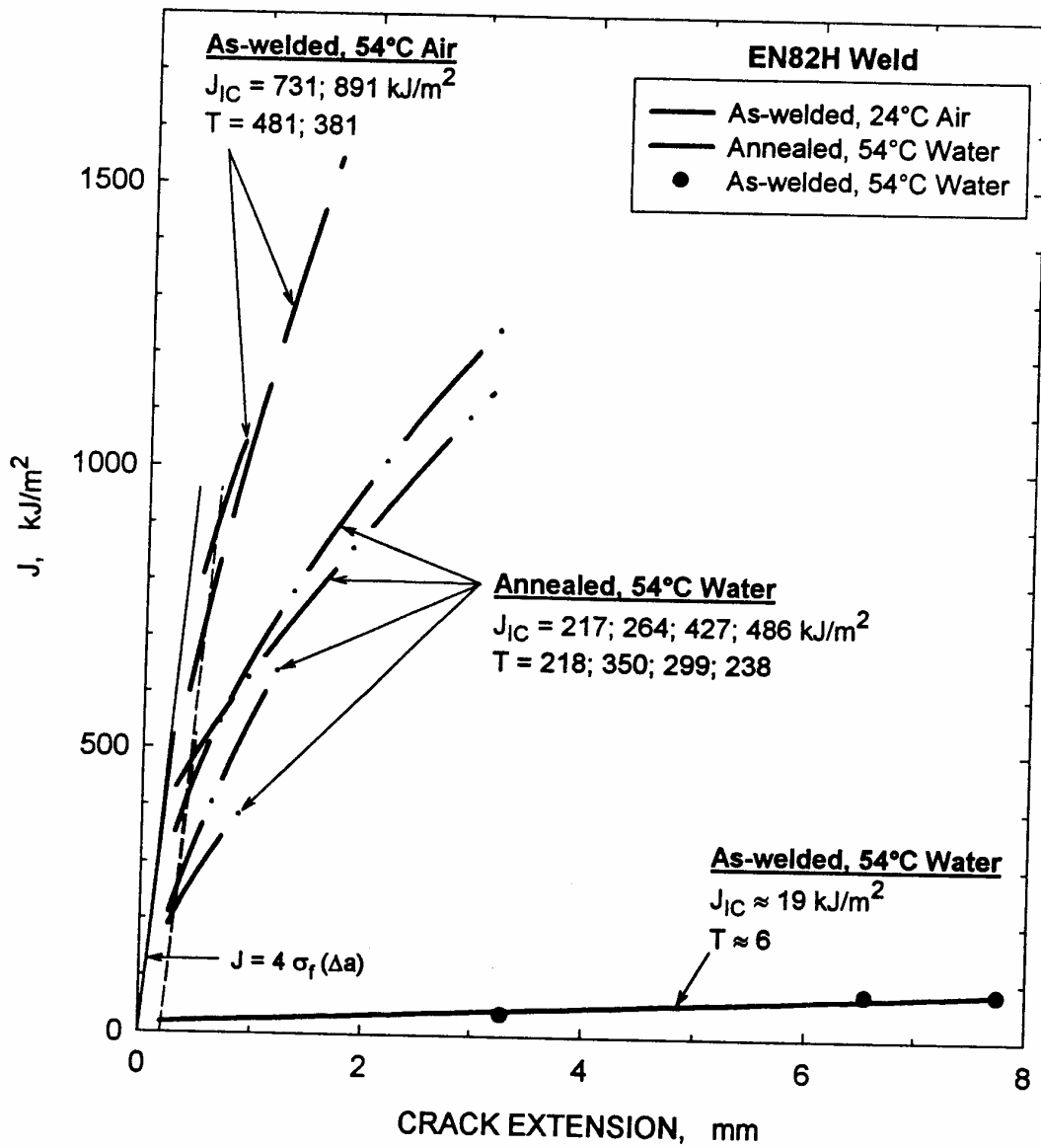
Intergranular cracking and dimple rupture  
in load-controlled specimen



### Annealing at 1093°C:

Fully restores the fracture resistance of EN52 & Alloy 690  
 Restores significant fracture resistance for EN82H  
 Has little effect on the fracture resistance of Alloy 600  
 (Values of T are given above each bar.)



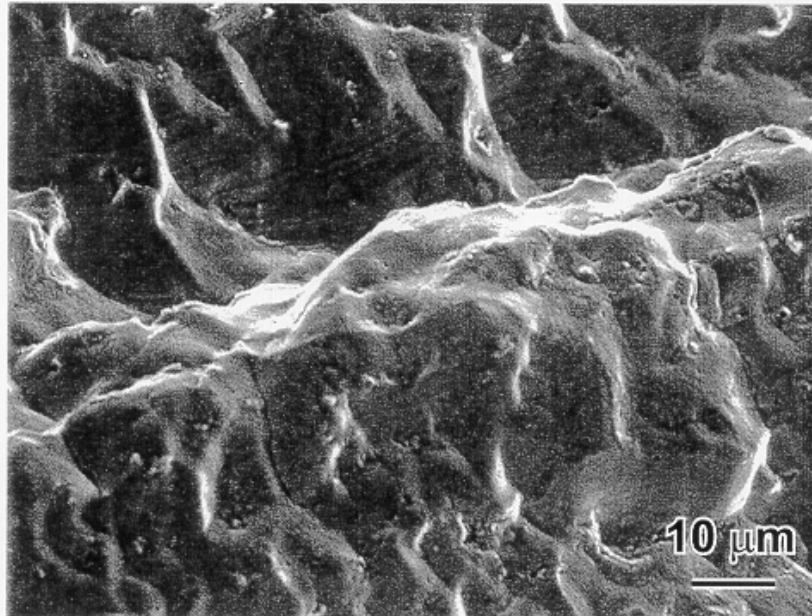


J-R curves for as-welded and annealed EN82H in 24°C air and 54°C water with 150 cc H<sub>2</sub>/kg H<sub>2</sub>O.

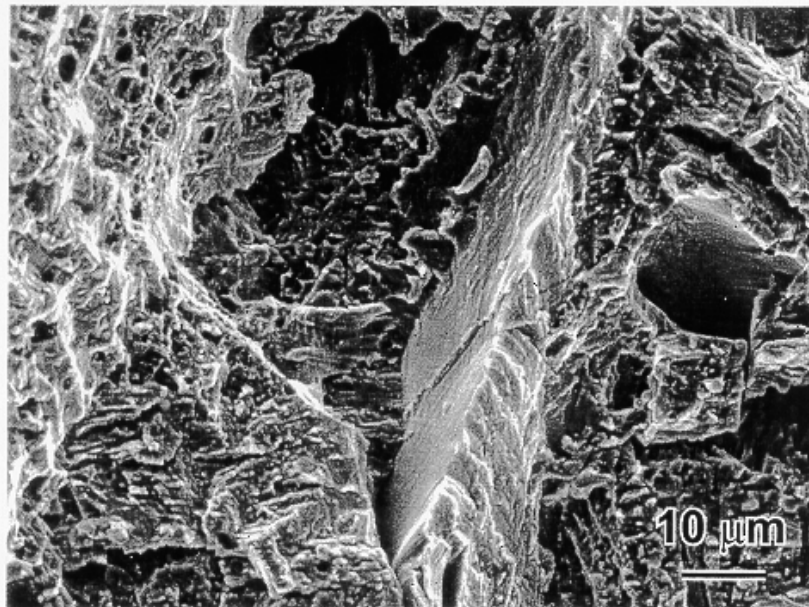


## EN82H Weld Tested in 54°C Water

**As-welded EN82H**  
Intergranular cracking



**Annealed EN82H**  
Transgranular facets & poorly defined dimples



## Microstructural & Mechanical Properties for As-welded & Annealed EN82H Weld

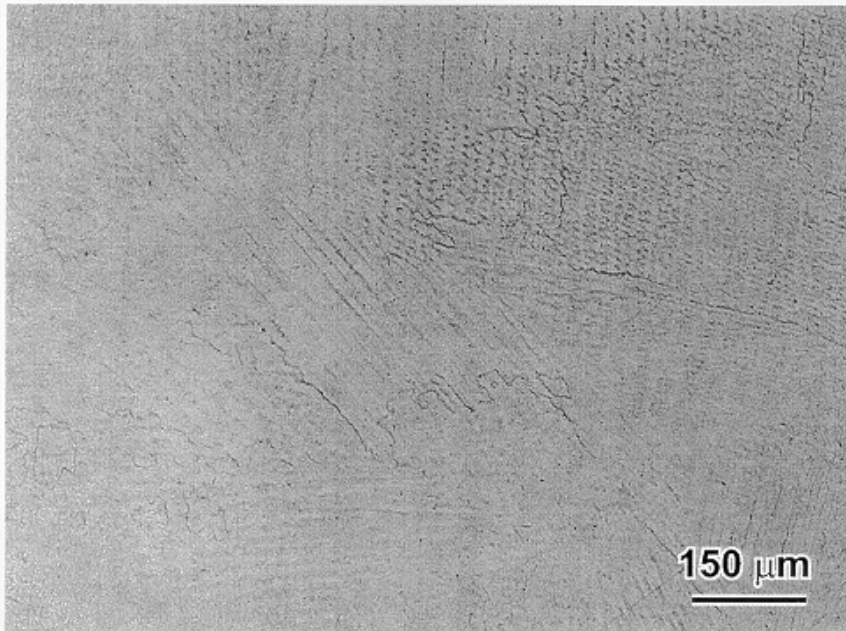
| As-welded  | Annealed  |
|--|---|
| <i>General Microstructure</i>  |   |
| Coarse dendritic grains. Cored structure—enriched Nb & Mn in interdendritic regions.                     | Recrystallized grains; some dendritic grains. Reduced Nb & Mn enrichment.   |
| <i>Grain Boundary Segregation</i>  |   |
| <b>No S &amp; limited P segregation.</b>   | <b>No segregation.</b>  |
| <i>Intergranular Precipitates</i>  |   |
| <b>Nb, Ti(C,N) [3-16 nm] on most GBs.</b><br>Few TiN inclusions on GBs.<br>Very few MgS on GBs.          | Nb, Ti(C,N) limited to nonrecrystallized GBs.<br><b>Cr-rich M<sub>7</sub>C<sub>3</sub> &amp; M<sub>23</sub>C<sub>6</sub> on GBs.</b><br>Few TiN/Nb(C,N) inclusions on GBs.<br>Extremely few MgS on GBs. |
| <i>Intragranular Precipitates &amp; Inclusions</i>   |   |
| Nb, Ti(C,N) ppt [3-16 nm] on dislocations.<br>TiN inclusions<br>AlMgSi-rich oxides                       | Nb, Ti(C,N) ppt [10-130 nm] in local regions<br>Multiphase TiN/Nb(C,N) inclusions<br>AlMgSi-rich oxides   |
| <i>Dislocation Content</i>   |   |
| High density of dislocation tangles & networks   | Low density of dislocations, except in isolated nonrecrystallized regions.  |
| <i>Mechanical Properties</i>   |   |
| $\sigma_{ys}$ = <b>360-480 MPa</b><br>$\sigma_{UTS}$ = 680-710 MPa<br>Elong. = 27-57%<br>R-in-A = 47-59% | $\sigma_{ys}$ = <b>240-280 MPa</b><br>$\sigma_{UTS}$ = 680-710 MPa<br>Elong. = 41-42%<br>R-in-A = 65%   |



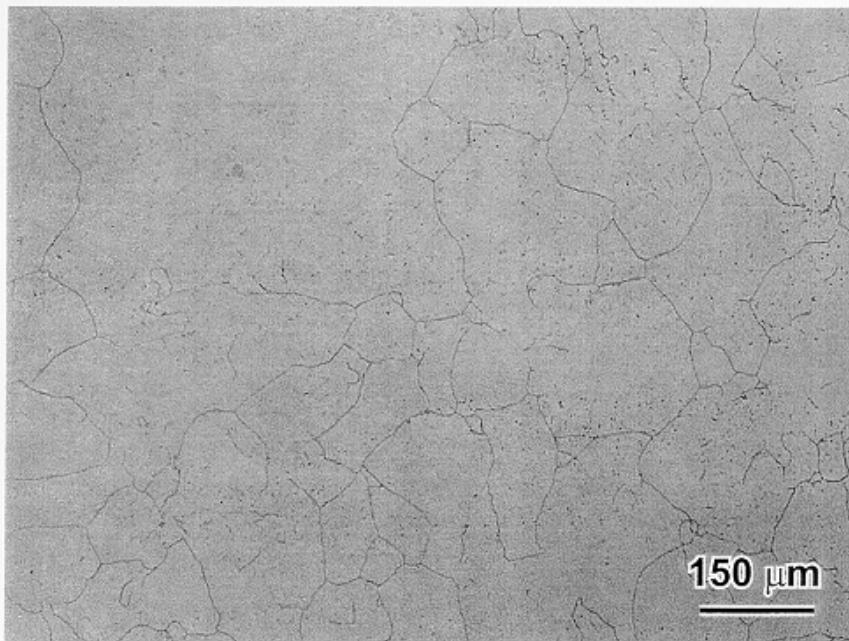


## Microstructure of EN82H Welds

**As-welded EN82H**  
Coarse dendritic grains

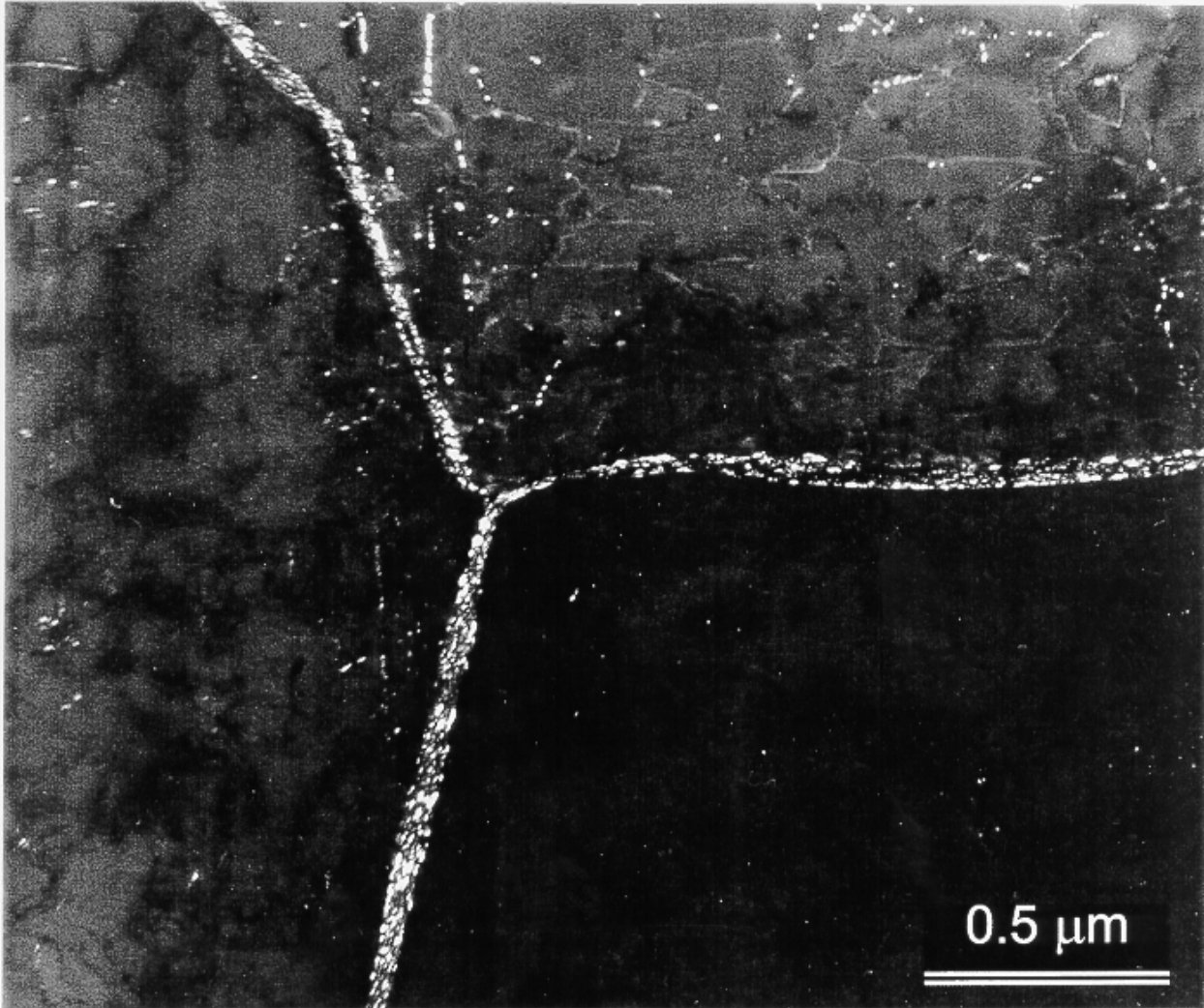


**Annealed EN82H**  
Recrystallized structure with equiaxed grains  
Localized regions with nonrecrystallized grains



## AS-FABRICATED EN82H WELD

Dark-field TEM Micrograph of Fine Nb,Ti(C,N)  
on Grain Boundaries and Dislocations



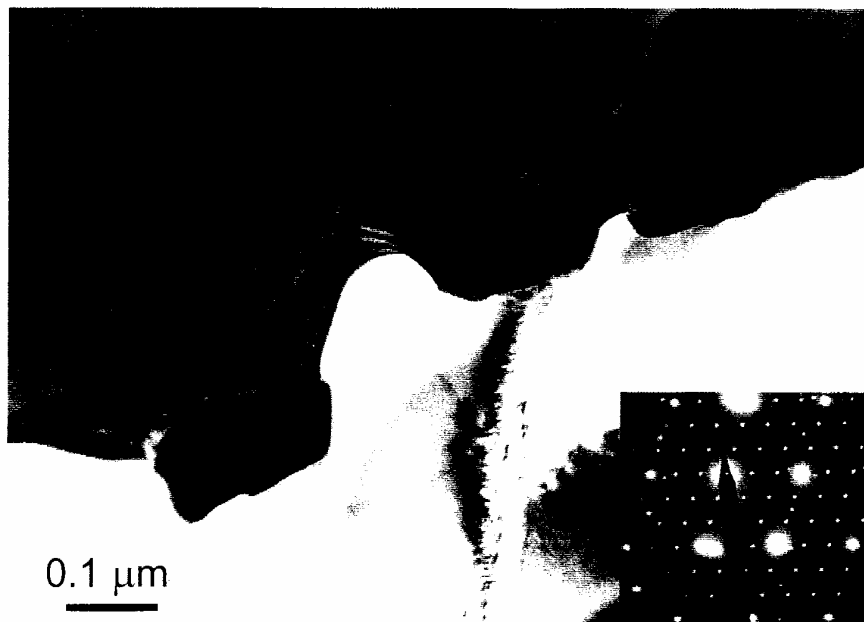
## Annealed & Furnace Cooled EN82H Weld

AEM analysis confirmed presence of Cr-rich  $M_7C_3$  and  $M_{23}C_6$  precipitates decorating grain boundaries.

Secondary electron image

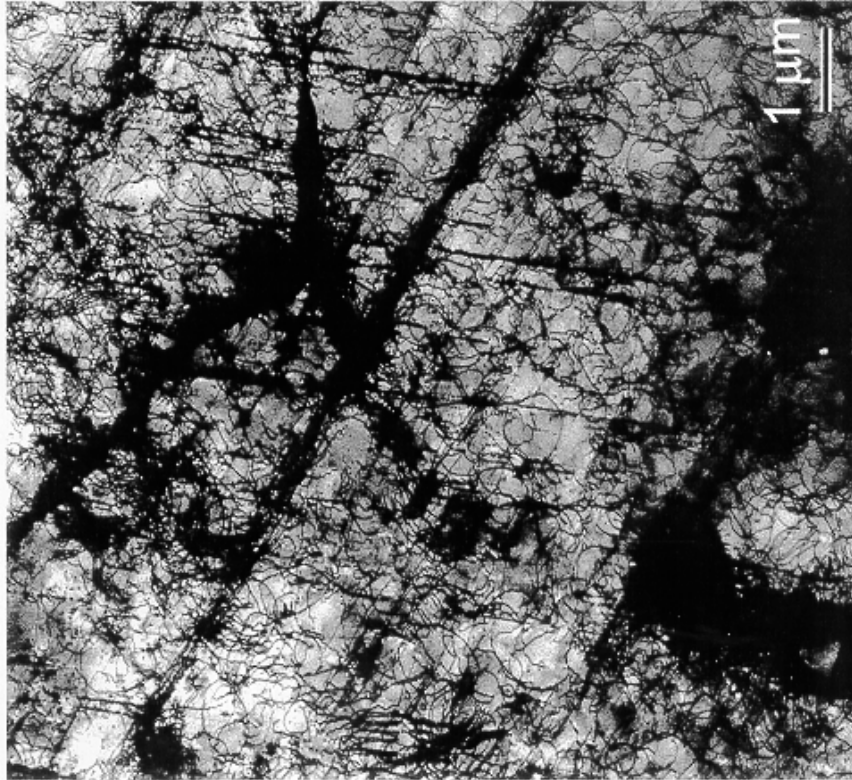


TEM image showing  $M_{23}C_6$



## DISLOCATION STRUCTURE IN EN82H WELDS

Extensive Dislocation Structure  
in As-Welded Condition

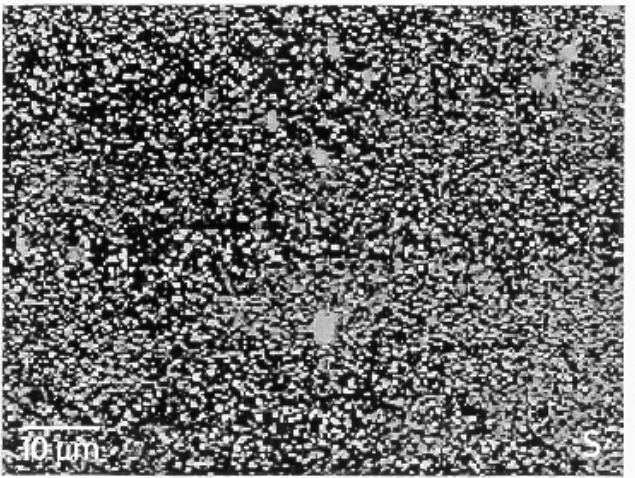
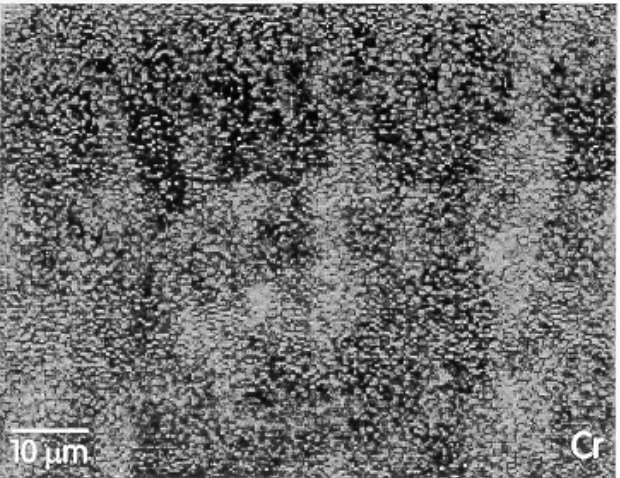
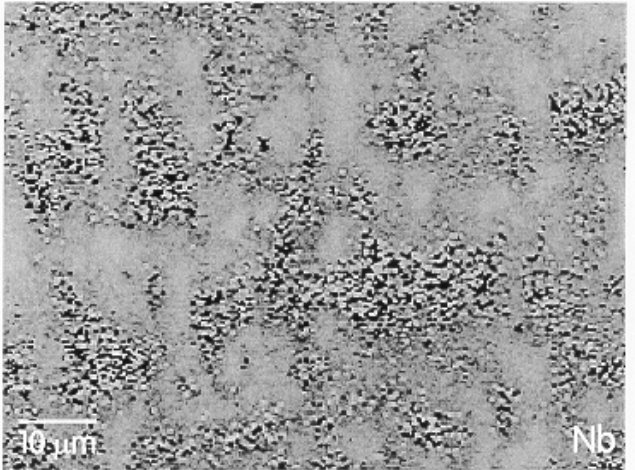
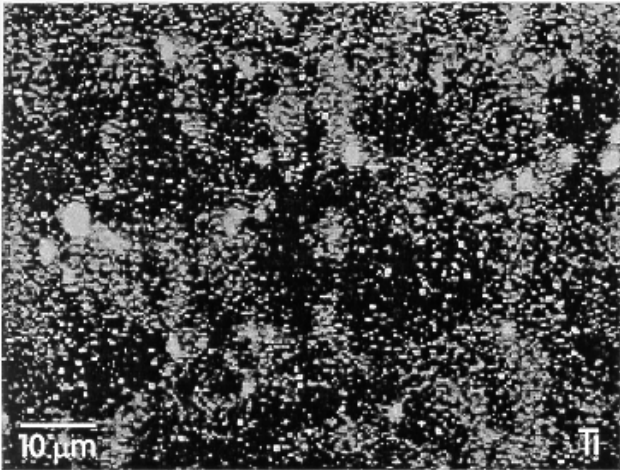
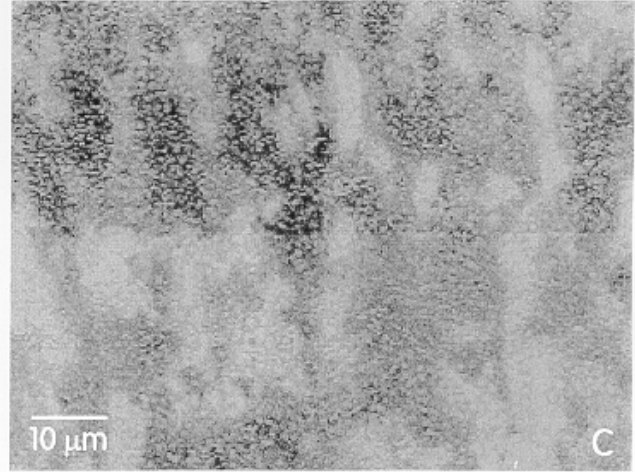
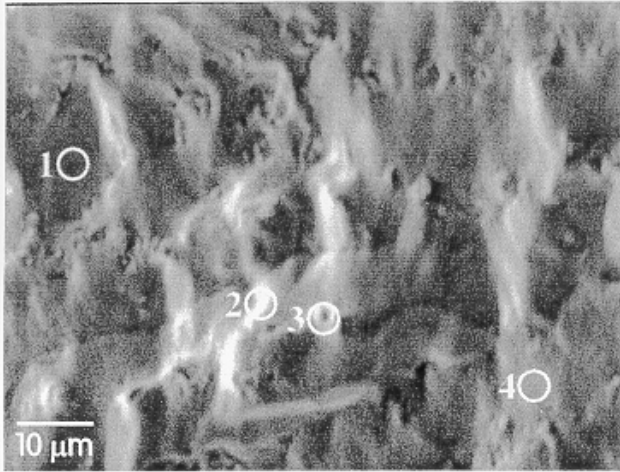


Significantly Reduced Dislocation  
Structure in Annealed Condition



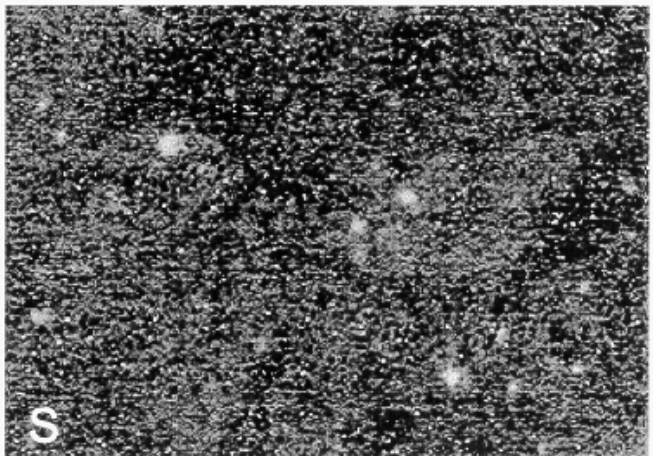
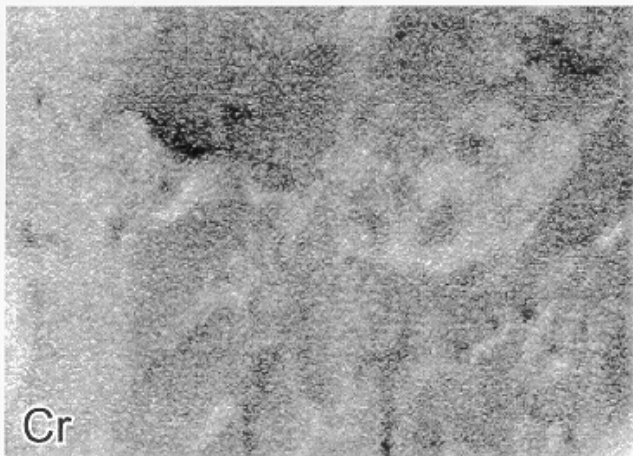
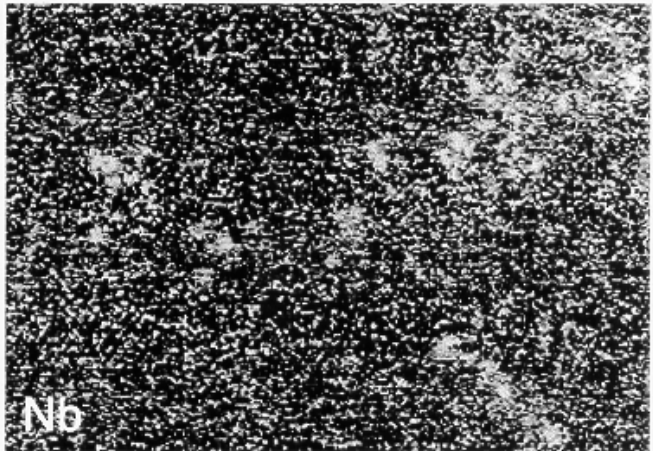
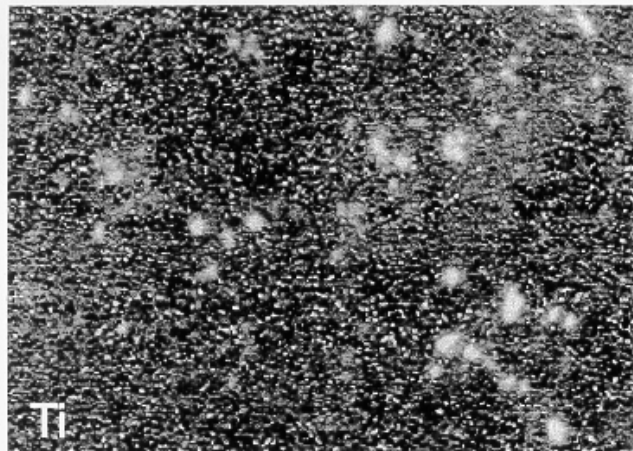
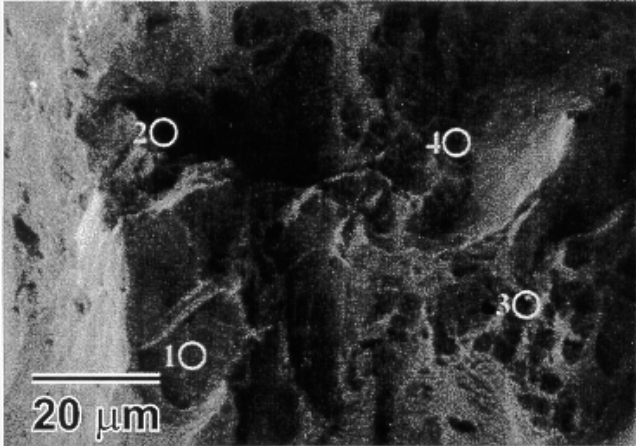
## AES Elemental Distribution Maps of As-Welded EN82H

- Nb, Ti(C,N) decorating most GBs.
- TiN inclusions & Limited sulfide (MgS) inclusions.



## AES Elemental Distribution Maps of Annealed EN82H

- Limited intergranular cracking.
- Nb, Ti(C,N) confined to nonrecrystallized GBs (upper right).
- Intergranular Cr-rich carbides.
- Multiphase TiN / Nb(C,N) inclusions & MgS inclusions.



## Grain Boundary Composition (a/o $\pm$ $\sigma$ ) for As-fabricated and Annealed EN82H Welds.<sup>†</sup>

Auger Electron Spectroscopy of EN82H welds revealed:

No S segregation in as-welded or annealed condition.

No P segregation in annealed condition.

Slight P segregation for half of the GBs in as-welded condition:

6 of 13 GBs with no P segregation ( $0.6 \pm 0.2$  a/o),

7 of 13 GBs with slight P segregation ( $1.4 \pm 0.3$  a/o).

| Material Condition   | Areas Studied       | B         | N               | Al        | P               | S         | Ti        | Cr        | Mn        | Fe        | Ni        | Nb               |
|----------------------|---------------------|-----------|-----------------|-----------|-----------------|-----------|-----------|-----------|-----------|-----------|-----------|------------------|
| As-welded            | 13 GBs <sup>‡</sup> | 1.3       | $2.5 \pm 0.4^+$ | 0.9       | $1.4 \pm 0.3^*$ | 0.7       | 1.2       | 15.2      | 1.8       | 2.0       | 62.2      | $19.2 \pm 2.0^*$ |
|                      |                     | $\pm 0.7$ | $1.5 \pm 0.5^+$ | $\pm 0.3$ | $0.6 \pm 0.2^*$ | $\pm 0.4$ | $\pm 0.3$ | $\pm 2.2$ | $\pm 1.2$ | $\pm 1.3$ | $\pm 7.0$ | $7.6 \pm 2.4^*$  |
| Annealed             | 5 GBs               | 1.0       | 0.9             | 0.8       | 0.8             | 0.7       | 1.2       | 18.6      | 0.9       | 1.9       | 66.7      | 6.8              |
|                      |                     | $\pm 0.7$ | $\pm 0.8$       | $\pm 0.5$ | $\pm 0.5$       | $\pm 0.3$ | $\pm 0.2$ | $\pm 2.2$ | $\pm 0.5$ | $\pm 0.9$ | $\pm 5.3$ | $\pm 4.7$        |
| As-Welded & Annealed | 11 TG <sup>‡</sup>  | 1.3       | 0.7             | 1.0       | 0.5             | 0.5       | 1.0       | 19.5      | 0.8       | 2.1       | 69.2      | 3.8              |
|                      |                     | $\pm 0.5$ | $\pm 0.5$       | $\pm 0.5$ | $\pm 0.3$       | $\pm 0.2$ | $\pm 0.1$ | $\pm 1.4$ | $\pm 0.4$ | $\pm 0.9$ | $\pm 2.1$ | $\pm 1.4$        |

\* Mean and standard deviation values were determined for high and low concentration groups.

+ Separate N concentrations were determined in regions with high and low Nb concentrations.

† Note that values below about 0.5 to 1 a/o are probably not significantly different from 0.

‡ GB = Grain Boundary; TG = Transgranular Region



## CONCLUDING REMARKS

High temperature water:

Fracture is not a concern because toughness is exceptionally high.

Low temperature water:

Fracture is a concern for EN82H and EN52 due to a severe degradation in cracking resistance, caused by a hydrogen embrittlement mechanism.

Although the fracture toughness of Alloy 690 is degraded, modest cracking resistance is retained.

LTCP is not a primary concern.

LTCP is not an issue for Alloy 600.

Decreasing hydrogen content of water to 15 cc H<sub>2</sub>/kg H<sub>2</sub>O: small to modest increase in toughness for EN82H, substantial increase in toughness for EN52 and Alloy 690.

LTCP does not initiate at as-machined notches.

Failure scenario: Crack initiation and growth by HTSCC or fatigue  
LTCP causes final failure ( $K_{JC}$  or  $K_{Pmax}$ ).

LTCP can initiate at a sharp weld defect.

Cooldown testing - EN82H under constant displacement conditions:

Tests to date show that rising load  $K_{Pmax}$  values accurately or conservatively predict the critical  $K_I$  for LTCP.

Annealing at 1093°C reduces or eliminates LTCP susceptibility.

Dissolution of fine intergranular Nb,Ti(C,N) in welds appears to improve LTCP resistance.





## References:

1. C. M. Brown and W. J. Mills, "Effect of Water on Mechanical Properties and Stress Corrosion Behavior of Alloy 600, Alloy 690, EN82H Welds and EN52 Welds," *Corrosion*, Vol. 55, 1999, pp. 173-186.
2. W. J. Mills and C. M. Brown, "Fracture Behavior of Nickel-Based Alloys in Water," *9<sup>th</sup> International Conference on Environmental Degradation of Materials in Nuclear Power Systems- Water Reactors*, TMS, (Report B-T-3240), 1999.
3. W. J. Mills and C. M. Brown, "Fracture Toughness of Alloy 600 and EN82H Welds in Air and Water," Bettis Atomic Power Laboratory, Report B-T-3264, 1999.
4. C. M. Brown and W. J. Mills, "Fracture Toughness of Alloy 690 and EN52 Welds in Air and Water," Bettis Atomic Power Laboratory, Report B-T-3265, 1999.
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6. M. G. Burke, R. J. Wehrer and C. M. Brown, "Real World Microscopy: Understanding the Environment-Sensitive Behavior of Ni-Base Welds," *Microscopy and Microanalysis*, Vol. 4, 1998, p. 528.

