BNL-NUREG-31814

Conf-821037--39

BNL-NUREG--31814

DE83 002102

TESTS WITH INCONEL 600 TO OBTAIN QUANTITATIVE STRESS-CORROSION CRACKING DATA FOR EVALUATING SERVICE PERFORMANCE*

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September 1982

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*Work Supported by the U. S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research.

EXTENDED ABSTRACT

Inconel 600 tubes in pressurized water reactor (PWR) steam generators form a pressure boundary between radioactive primary water and secondary water which is converted to steam and used for generating electricity. Under operating conditions the performance of alloy 600 has been good, but with some occasional small leaks resulting from stress corrosion cracking (SCC), related to the presence of unusually high residual or operating stresses. The suspected high stresses can result from either the deformation of tubes during manufacture, or distortion during abnormal conditions such as denting. There have also been a few minor leaks that were not positively identified as stress corrosion cracks because the tubes were merely plugged without removal for examination. It is not yet certain what long-term effects are to be associated with lower stress levels that would be encountered during usual temperature cycles of a steam generator in service.

A primary to secondary leak causes contamination of the environment to some degree, and for this reason the Nuclear Regulatory Commission (NRC) is involved in a licensing decision whenever leaks are reported. In order to assist the NRC in making decisions concerning the licensing of a plant with SCC defects, data are necessary that could be used for predicting or estimating failure times when abnormal conditions are encountered. A program is active at present at Brookhaven National Laboratory to examine the factors involved in the SCC of Inconel 600 in high temperature deaerated water, with the objective of developing a model that will relate life expectancy of tubing to factors such as stress, strain, strain rate, environmental conditions, microstructure and cold work of the material. Such a model is intended to form the basis for determining a predicted life expectancy for an "unknown" by extrapolating one or two accelerated data points to operating conditions. This could then be used in various cases to assist in determining tube plugging or inspection criteria.

The present experimental program addresses two specific conditions, i.e., 1) where deformation occurs but is no longer active, such as when denting is stopped and 2) where plastic deformation of the metal continues, as would occur during denting. Laboratory media consist of pure water as well as solutions to simulate environments that would apply in service; tubing from actual production is used in carrying out these tests. The environments include both normal and "off" chemistries for primary and secondary water.

The results reported here were obtained in several different tests. The main ones are 1) split tube "reverse" U-bends, 2) constant extension rate tests (CERT), and 3) constant load. The temperature range covered is 290-365°C.

U-BENDS:

Split tube type U-bends have their inside surfaces exposed in tension. Using these specimens, a first series of experiments suggested a possibility that the carbon level of the Inconel influences the crack initiation/temperature relationship. Specifically, activation energy seemed to increase with increasing carbon content. These data are based on cracks observed in

the temperature range 325°C to 365°C, with most of the data from the highest temperature levels. Since the temperature range is small, it obviously remains important to verify that this effect persists down to operating temperatures. There is also some scatter in stress corrosion data in general, and the present tests are no exception; therefore, a statistical analysis was deemed necessary and tests were started in the first half of 1981 in which a larger number of replicate samples are exposed in water at 290 and 315°C. These U-bends had been in test for 60 weeks at the previous inspection without any observed cracking; it should be noted that little or no cracking is expected during such a short exposure. Figure 1 shows the data points in the U-bend tests, and Figure 2 shows the tentative activation energies plotted against carbon contents. For the 0.02% material, one test at 290°C and one at 315°C have now exceeded, without SCC, the times at which SCC would have been predicted by extrapolation. The expected failure times are shown in Table 1. At least for 0.02%C, therefore, the activation energy may be greater than 36 Kcal/mole over the lower temperature range, which includes operating temperatures. The other materials (.01, .03 and .05%C) have yet to reach points of intersection with the extrapolations of the high temperature portions of the curves. When sufficient data are available, we will use statistical methods (such as Weibull) to develop the final, quantitative model. The heats that are now exposed at 290 and 315°C, cover the range of 0.01 to 0.03% carbon, and a few (8) of the original specimens with 0.01, 0.02, 0.03 and 0.05%C also remain in the 290°C test. The latter 8 samples have reached 192 weeks without SCC.

CERT:

CERT data so far have shown a distinction between the initiation and propagation stages. Cracks do not initiate at the start of plastic deformation, but take a finite time (after that) to develop, and initiation times are much shorter than in the U-bend exposure. Extrapolations were made to determine the onset of SCC in CERT at temperatures of 325°C, 345°C and 365°C. These curves, shown in Figure 3, were obtained with specimens made from production tubing, flattened before cutting tensile specimens. For the present, corrections based on these curves also are used for calculating crack propagation rates in undeformed materials; similar initiation corrections are being developed for as-received tubing. Cracking in the CERT was achieved readily in cold worked or as-received material at strain rates in the vicinity of 2×10^{-7} sec^{-1}. As will be shown below in more detail, the activation energies for cold worked and non-cold worked Inconel 600 were found to be identical, suggesting that the mechanism is the same in the two cases, although the crack growth velocities for the types of specimens are different.

Figures 4 and 5 show the straight line Arrhenius plots of CERT data obtained to date. Several sets of points provide parallel curves that correspond to an activation energy of 33 Kcal/mole. Some additional observations based on the CERT data can be detailed as follows:

1. The slopes of the lines remain consistent with an activation energy of 33 Kcal/mole regardless of whether the material is cold worked, aged (365°C), or mill annealed.

2. Crack growth rates are faster in cold worked material due to a change in the constant k of the Arrhenius equation:

$$\text{Rate} = k.\exp\left(-\frac{Q}{RT}\right)$$

3. Environmental conditions may also affect the quantitative aspects of SCC. Hydrogen (added to pure water) increases the number of cracks as well as SCC growth rate of as-received material while H_3BO_3 does not appear to have this effect. Other combinations of the ingredients of primary and secondary water are in test now.

4. An "unknown" tube can now be tested in one accelerated test to establish its initiation and propagation rates, and data for other temperatures can be calculated from this determination.

5. Cold worked (flattened) specimens gave crack growth rates in simulated AVT and primary water consistent with rates observed in pure water. Tests with as-received material will be completed this year.

6. Specimens aged (furnace) at 365°C for several weeks before exposure in pure water (CERT) gave crack growth rates similar to fresh material.

7. Crack growth velocities in our work are in the same ranges as were found in published work for tests is sodium hydroxide solutions at elevated temperatures. (See Figure 4.)

8. Strain rates in the range 3×10^{-8} to 1×10^{-6} sec^{-1} were used for producing SCC, and it seems necessary to adjust the rate downwards in order to see SCC in the more resistant materials.

9. Temperature exerts a much greater influence on crack velocity in CERT than strain rate. The latter, within the range used, has had an effect of less than a factor of 2, and there are no plans now to examine the effects of strain rate any further. Most of the present tests are done at about 2×10^{-7} sec^{-1}.

10. Extrapolation of data from cold worked samples shows initiation at approximately 10% strain at operating temperature, as shown in Figure 6. This number appears to be in good agreement with what has been observed in the field when denting led to stress corrosion cracks in deformed Inconel tubing. A comparison of SCC times based on the laboratory data (using susceptible Inconel) with the field observations show considerable promise that the laboratory data can indeed be used to predict service performance. (See 11. below.)

11. An example of an extrapolation is as follows: Laboratory tests for as-received material in pure water indicate a crack velocity at 300°C of about 5×10^{-8} mm sec^{-1}. In order to achieve observable cracking in the CERT, strain rates at these low temperatures appear to be of the order of 1 to

5×10^{-8}. Assuming that a strain rate of 2.5×10^{-8} is observed, we can use this to show that it will take almost two months to reach 10% strain at which time cracks will initiate. At this temperature (for tubing that has not been cold worked), the crack velocity will be approximately 5×10^{-8} mm sec^{-1} based on presently available data, so that it would take approximately four to six months for cracks to propagate 60% through wall. A series of more accurate calculations will be made within the next few months when more refined data are available. However, it is evident that reasonable predictions can already be made for the case of active deformation.

12. A point to keep in mind is that the actual conditions of stress, strain and strain rate under operating conditions would have to be known, or calculated, in order to use the quantitative SCC data predictions to best advantage.

13. Figure 7 shows a comparison of the stress-strain curve for heat #2 in the as-received, mill annealed condition, with another that had first been subjected to a heat treatment of 20 hours in Ar at 700°C. This latter treatment is equivalent to the latest commercial method used to induce chromium carbide precipitation, which is believed to provide resistance to SCC in deaerated high temperature water. The as-received specimens showed intergranular failure, whereas the material after 700°C treatment showed a ductile fracture with only extremely shallow intergranular penetration at one point on the surface. This is an encouraging result, because the laboratory heat treatment was but a single step following after a processing procedure that obviously was quite "adverse" in terms of SCC resistance. In future production; we believe that the prior processing may be arranged to optimize the effects of the final 700°C heat treatment, and may well produce even greater resistance to this kind of SCC.

More tests are planned with samples of commercial (700°C treated) tubing, and these will include a range of strain rates to obtain comprehensive data, including primary coolant conditions.

<u>CONSTANT LOAD:</u>

For the case where denting or active deformation is no longer occurring, it is necessary to obtain data that relate the time to failure to the stress present in the surface of the material, i.e., the load on that part of the tube. These stress patterns can consist of residual plus operational stress, and may be complex. In the present test series, a first attempt at relating load to SCC failure time is made by means of tensile specimens under applied load. This will be compared with simulated dents in order to find out how the quantitative values compare for this type of failure in Inconel. Figure 8 shows the curves for stress versus failure time on logarithmic scales including results for as-received and cold worked material. In the equation $T_F = k. \sigma^b$, the slope of the two parallel log-log curves correspond to a value of $b = -4.0$, in the range that has been studied so far. This is a much more reliable number than the previously reported value for b which was based on fewer data. Figure 9 shows 2 points of data obtained in simulated primary water at 365°C, where the slope agrees with the pure water plots. Figure 10 is taken from the work of Theus (B&W) in caustic for comparison with our results.

In the cold worked material, the cold work resulted from the flattening of the tube specimens during the preparation of the tensile pieces. These cracked more readily than the as-received material, in agreement with the findings in CERT, but the stress dependence is the same.

One test has shown SCC at a stress level below the yield point in as-received Inconel 600, and relates to the important question whether the quantitative equation can be applied to stresses well below the yield point.

It is intended to combine the CERT data with the U-bend and or constant load results in one equation for translating exposure under known operating conditions into future performance, taking into account the spread to be expected within extrapolations.

Since cold work is an important parameter in accelerating SCC , although it is obviously not a prerequisite for cracking to occur, it will be examined in more detail. In practice, tubing is shaped, e.g., into U-bends, rolled into tube sheets, straightened without subsequent annealing during manufacture, and there are certain to be many other sources of residual stresses. Little is known about the influence of the degree of cold work on SCC, and for this reason it is included in the present BNL program. We are comparing the as-received condition with 5, 10, and 20% cold work in tests that include direct load and CERT. Environments include pure deaerated water as well as oxygen-free simulated primary and secondary water.

Capsule tests in which denting is being reproduced as well as tests in which cyclic stresses are applied to the specimens are due to resume in the near future. No new results are available for these experiments at present. They will be important in covering certain practical conditions.

HEAT TREATMENT:

Attempts have been made during the past two years to generate our own susceptible heats of Inconel by means of high temperature annealing of heavily cold worked Inconel 600. Annealing temperatures were chosen to simulate those that may possibly exist in tube mills. Earlier work had indicated no sucess in the temperature range of about 1600°F to 1850°F (approximately 870°C to 1,000°C), for times ranging from 15 to 30 minutes. In more recent work, the material has been held at an annealing temperature for relatively short times, and some success has been achieved by holding at temperature for about 2 minutes. At shorter or longer times than this we did not achieve susceptibility, as determined in CERT (by the presence of cracks and a maximum loss of ductility at 365°C in pure deaerated water) and also in U-bend tests (where cracking occurred only in the specimens that had been heated for about 2 minutes) as shown in Figure 11. It is stressed that these results may only apply to the specific heat that we used (0.03% carbon) and it is not suggested at this time that the specific temperature-time combination would be generally applicable to any heat of Inconel 600.

STRUCTURE:

The various heats of mill annealed Inconel 600 tubing used in this program are typical of nuclear grade production; however, only about half of these heats have shown evidence of intergranular SCC when U-bend specimens were exposed to pure deaerated water at high temperatures. It is difficult to establish what differences exist between these heats that account for the fact that some are susceptible while others appear to be immune. A susceptible structure is generally associated with carbide-free grain boundaries, while semi-continuous grain boundary precipitates are beneficial in preventing SCC in caustic and pure water environments. Electrolytic etching in phosphoric acid showed that all of the materials used in this program were relatively free of carbide precipitates in the grain boundary regions. The susceptibility of this alloy, therefore, cannot be judged on microstructural analysis alone. Small variations in processing history which occur within a mill or different mills must play an important role.

H_2 IN PURE H_2O:

A definite accelerating effect of H_2 has been observed on SCC in high temperature water in U-bends as well as tensile specimens. In the latter case, a heat (#11, 0.03%C) of commercially produced tubing did not crack in the as-received surface condition in pure water at 365°C in the CERT test as well as U-bends - although a basic tendency towards cracking was found in U-bends that were first pickled in HNO_3/HF. When tested as-received (resistant in water at 365°C) in CERT at 365°C in $H_2O + H_2$, intergranular SCC occurred.

Confirmation of a H_2 effect came from a comparison of 9 heats tested in pure H_2O and $H_2O + H_2$ as U-bends at 365°C. In water alone, as shown in Table 2, only 2% failures occurred in 12 weeks, compared to 83% in $H_2O + H_2$.

Table 1

Calculated Failure Times for Lab. U-Bend SCC

%C	Projected Weeks at: 315°C	290°C
0.05	150	1500
0.03	120	700
0.02	30*	150*
0.01	80	240

*Exceeded by ongoing tests, without visible SCC.

Table 2

Effect of the Presence of H_2 in H_2O at 365°C

Test duration:	12 weeks
Test medium:	Pure, deaerated water (with and without H_2)
Test temperature:	365°C
Test specimens:	U-bends
# Heats:	9

RESULTS

	Cracked	# Tested	% Failed
Pure H_2O	1	45	2
Pure H_2 + H_2O	15	18	83

(H_2 = amount found in primary H_2O).

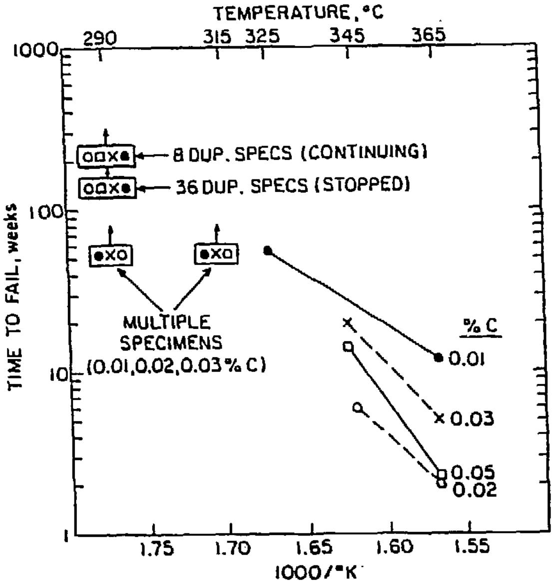

Fig. 1 SCC in Pure Water. U-Bends of Commercial Tubing

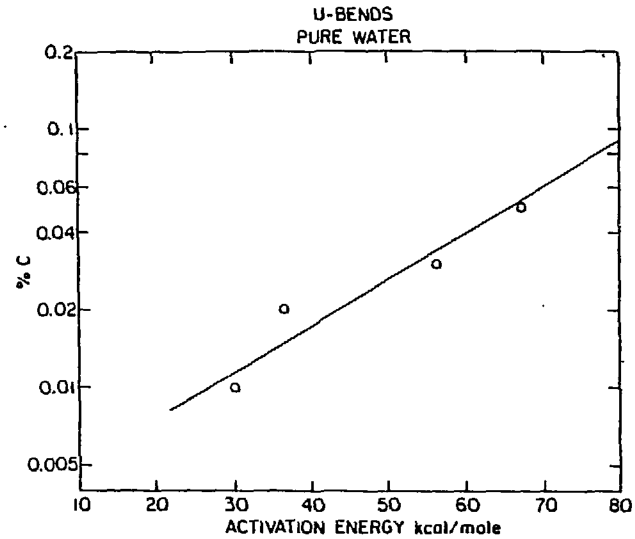

Fig. 2 Tentative Activation Energies (based on higher temperature data)

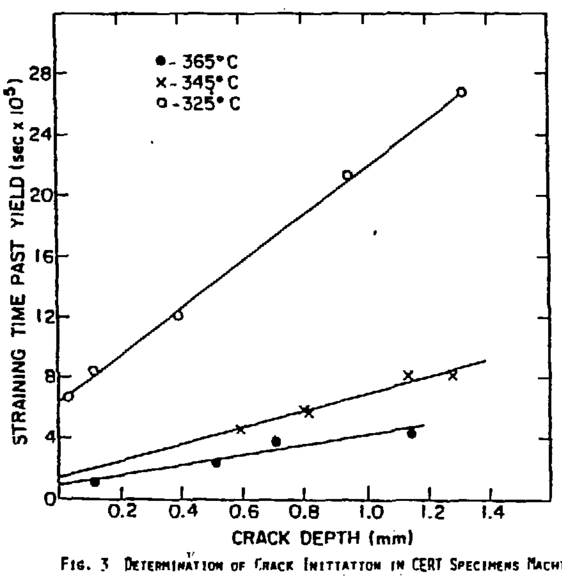

Fig. 3 Determination of Crack Initiation in CERT Specimens Machined from Heat #4 (.01%C) - Flattened Tube Tensile Pieces

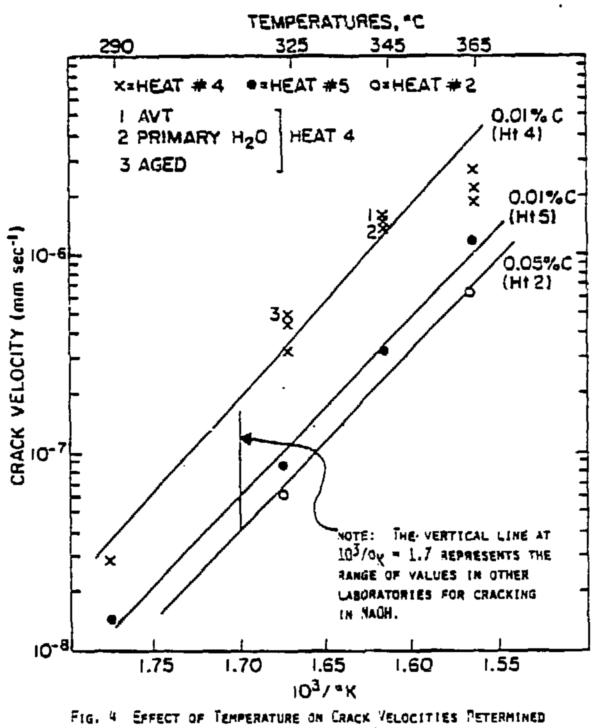

Fig. 4 Effect of Temperature on Crack Velocities Determined Using Cold Worked Inconel in CERT.

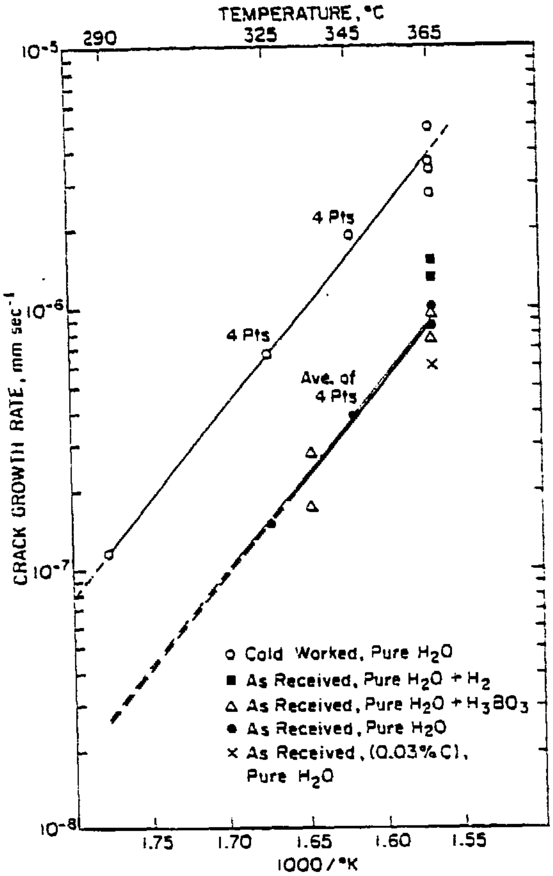

Fig. 5 Crack Growth Rates, CERT Experiments, 0.01%C

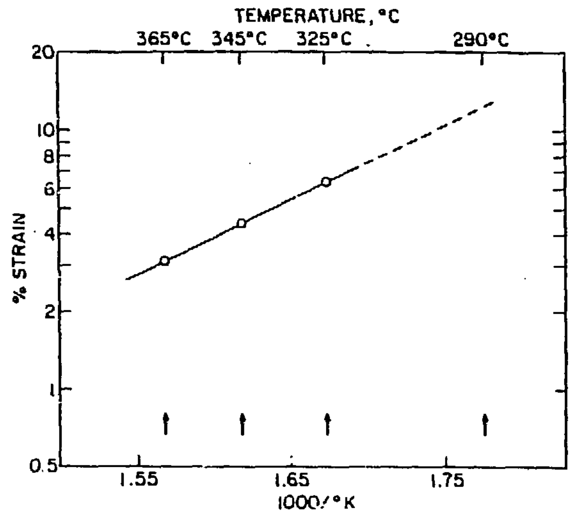

Fig. 6 Extrapolation of SCC Initiation Strain Values from Fig. 3

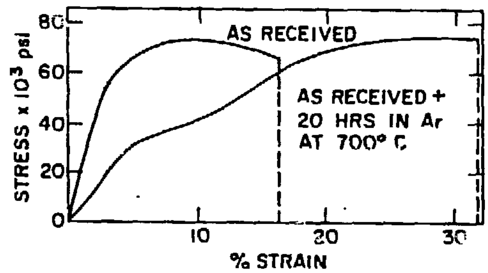

Fig. 7 Comparison of Stress Strain Curves for Heat #2 with and without Heat Treatment at 700°C for 20 Hours

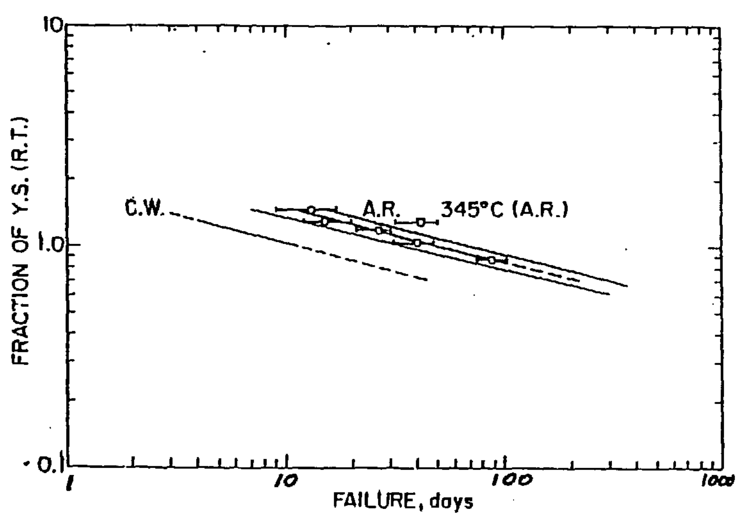

Fig. 8 Failure Times vs. Stress, Constant Load in Pure H_2O, 365°C

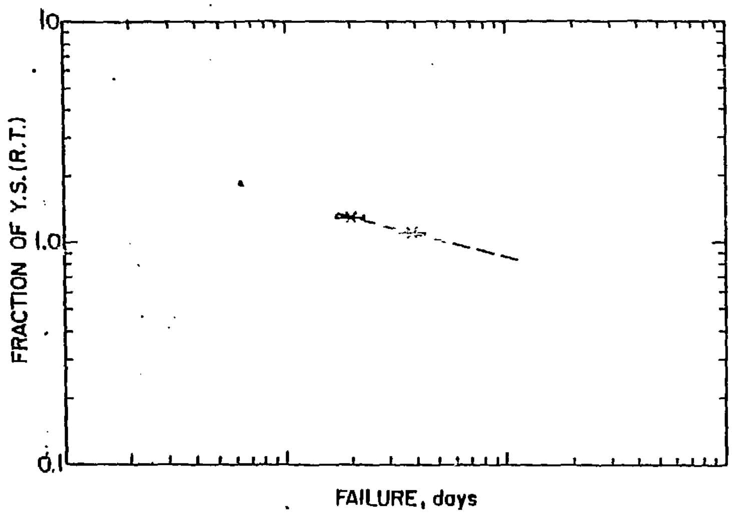

Fig. 9 Failure Times vs. Stress. Constant Load in Simulated Primary H_2O, 365°C

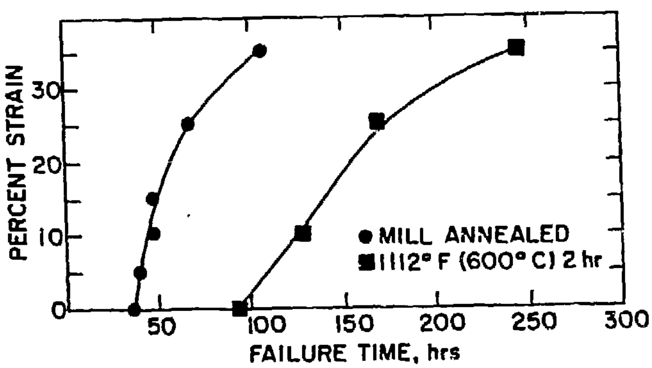

Fig. 10 Percent Strain versus Time to Fail - 550°F (10% NaOH) (from G. Theus, B&W)

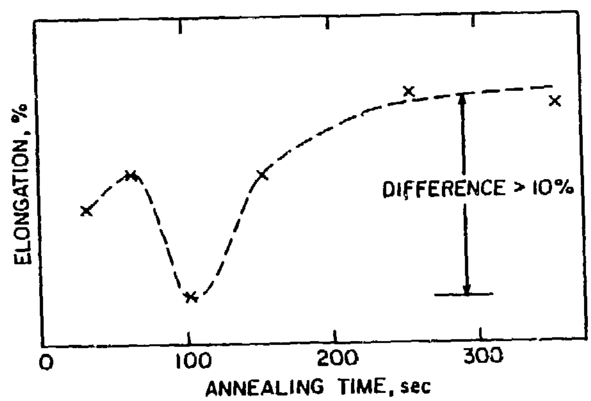

Fig. 11 CERT Test: Annealing Response of Cold Worked Inconel in the Range 1700°F to 1775 F, Inconel 600, 0.03%C, 365°C, Pure H_2O, Deaerated.

Note: The only U-bends to crack in a similar test medium were also the ones heated for 100 seconds