ENVIRONMENTALLY ASSISTED FRACTURE OF CAST AND WROUGHT MILD STEELS IN BRINE

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ABSTRACT

Two test methods, slow strain rate (SSR) tests and corrosion fatigue tests, are being used to investigate the susceptibility of wrought and cast mild steels to environmentally assisted fracture. The SSR test provides an assessment of relative cracking susceptibility, and is most useful for comparing the effects of different environments on a given material, and for measuring the relative susceptibility of different materials to environmentally assisted cracking in a given environment. The corrosion fatigue test measures the stress intensity (which is proportional to stress times the square root of crack length) below which an existing flaw or crack will not grow. The test may also prove useful in measuring the stress intensity that provides an acceptable rate of crack growth.

The SSR test has been used to investigate the effects of strain rate, temperature, and solution oxygen content on fracture properties of mild steel. The results of currently available corrosion fatigue tests are limited to crack growth rate measurements at one frequency in air and in brine. These measurements will be part of an overall program to evaluate allowable stress intensities, but this evaluation cannot be made until measurements are taken at different frequencies and at lower stress intensities. This report, then, discusses the background of SSR testing, experimental procedures, results, and conclusions.

INTRODUCTION

The deformation and fracture behavior of wrought and cast mild steels is being investigated by Pacific Northwest Laboratory (PNL)[a] under the Waste Package Program, which is funded by the Office of Nuclear Waste Isolation (ONWI). The objective of this study is to determine the potential for the environmentally assisted fracture of candidate overpack materials for use in salt repositories.

The environmentally assisted propagation of cracks from preexisting defects or flaws is potentially one of the most damaging failure modes for waste package materials. Cracks can propagate in susceptible materials below the general yield stress even when general corrosion is insignificant. This failure mode is difficult to quantify because of the generally long initiation times, the difficulty of detecting minor crack growth, and the unpredictability associated with failures of this type.

Environmentally assisted fracture is often affected by strain rate. At high strain rates, the influence of the environment is limited by the time of exposure. At very low strain rates, the oxide film is sometimes able to repassivate during stress application so that interaction between the metal and the environment is again limited. The lowest ductility is often found at intermediate loading rates where interaction occurs as the passivating film is broken.

SSR tests are being used to accelerate the environmentally assisted fracture process and thus to measure the crack propagation resistance of candidate overpack materials.

In the SSR tests, a tensile specimen is loaded to failure at a constant strain rate in the presence of an environment simulating that of the service environment. The load and strain of the specimen are monitored. Susceptibility of metals to fracture is determined by three factors:

- if ductility is lower in the service environment than in an innocuous environment
- if ductility is influenced by strain rate
- if fractographic evidence of environmentally enhanced fracture mode(s) is observed.

Fracture surfaces of low-ductility specimens are examined to see whether a change of fracture mode has occurred. If the fracture mode has changed, the diminished ductility may be attributable to stress corrosion cracking; if it has not, the specific mechanism of environmentally assisted fracture may be difficult to determine.

EXPERIMENTAL METHODS

The SSR tests were conducted using specimens fabricated from 0.6-cm-thick wrought plate or from ~13-cm-thick castings. The wrought plate (AISI 1025 steel) was tested in the mill-annealed condition; the cast steels (ASTM A27 gr. 60-30 in unirradiated tests and ASTM A216 WCA in irradiated tests) were tested in the as-cast condition because of the inherent difficulties in heat-treating thick-walled overpacks. The differences between these materials are insignificant in SSR testing. A216 steel is the reference cast steel in the Waste Package Program, and was used in the more recent irradiated tests. These materials are usually normalized or annealed before use.

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[a] Operated for the U.S. Department of Energy by Battelle Memorial Institute.

The tensile specimens used in the SSR tests were inscribed with gage marks at 2.54-cm intervals to determine elongation after the test. The specimen width was slightly reduced in the center of the gage section to induce fracture between the gage marks.

In nonirradiated SSR tests the reservoir of brine solution (Table I) was continuously sparged with argon, oxygen, or a mixture of argon and 20% oxygen. The brine solution, designated Permian Basin Brine No. 2, was derived from dissolution of salt cores from a Permian Basin salt horizon considered representative of a bedded-salt-site repository. The recipe was modified to mitigate precipitation of solids (mostly carbonates) due to inverse solubility effects. The modified recipe was obtained by holding the brine in an autoclave for several days, then performing an analysis of the supernatant fluid existing in quasi-equilibrium with the dissolved solids. Solution flow rate was maintained at approximately 35 ml/h. Constant extension rates were applied to the specimens using a gear-driven loading device (Fig. 1). Load was measured on a load cell that was external to the autoclave, and strain was measured on a linear-variable differential transformer (LVDT) between the autoclave and the loading rod. The load, strain, temperature, and pressure signals were recorded on a programmable data logger. The load-versus-strain data were plotted for each test so that deformation characteristics could be determined.

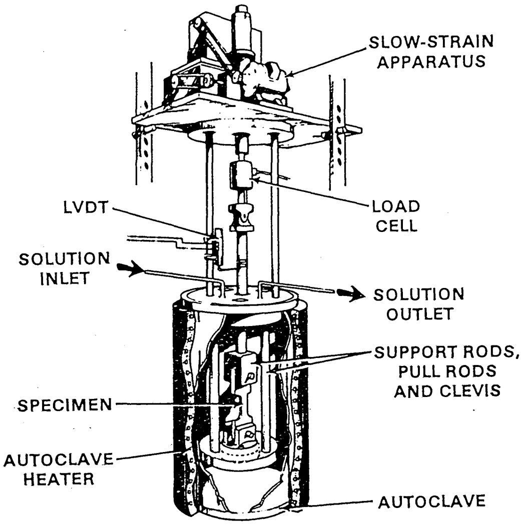

Fig. 1. Slow strain rate test system.

Reduction of area was determined by measuring the dimensions of the specimen gage section before the test and the dimensions of the fracture surface after the test. The energy absorbed by the specimen during deformation and fracture was determined graphically by measuring the area under the load/elongation curve. The energy absorbed during uniform deformation (before maximum load) was measured, as was the energy absorbed during nonuniform deformation (between maximum load and fracture).

Irradiated SSR tests were performed in much the same manner, except that a long test assembly was used to hold the autoclave in a field of high irradiation intensity (3×10^5 rad/h) from a Co^{60} source. The irradiated SSR facility[a] is relatively new, and tests to date have only been conducted under nonpressurized conditions at 30°C and 90°C. The brine used in these tests had the same composition as that used in the unirradiated tests, except that the solution was static and was not sparged.

TABLE I

Composition of Permian Basin Brine, No. 2[a]

Ion	Concentration, mg/l
Na^+	123,000
Ca^{2+}	1,110
Mg^{2+}	122
K^+	39
Sr^{2+}	35
Zn^{2+}	7.9
Cl^-	191,000
SO_4^{2-}	1,910
HCO_3	23
Br^-	24
F^-	1.0

[a] pH = 7.05

The test system used for the irradiated SSR tests is shown in Fig. 2. A microprocessor-controlled stepping motor is used to operate a series of harmonic drive units and drive shafts. The rotary motion is turned into axial motion by a roller screw assembly, and this axial strain is applied to the specimen through a seal. Load is measured with a load cell outside the autoclave and above the high-intensity gamma irradiation.

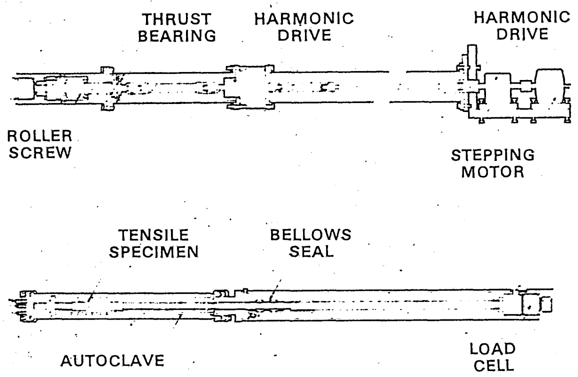

Fig. 2. Slow strain rate test system for irradiated testing.

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[a] Operated by Westinghouse Hanford Company for the U.S. Department of Energy.

RESULTS

Test data for wrought mild steel in brine and in air are given in Fig. 3 for the transverse orientation (stress applied perpendicular to the rolling direction of the plate). Similar results were obtained for the longitudinal orientation (stress applied parallel to the rolling direction).

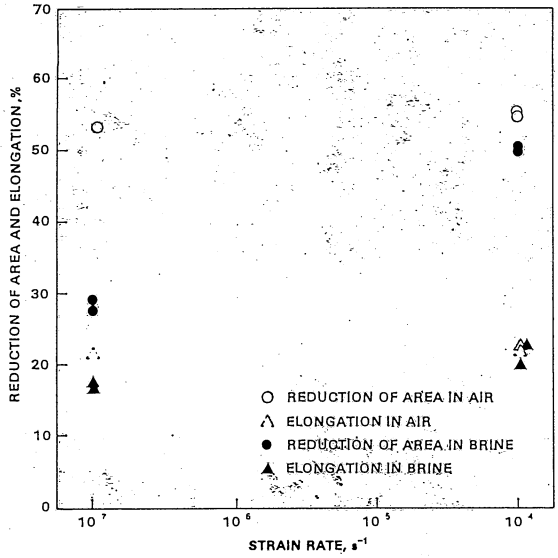


Fig. 3. Results of SSR tests of 1025 wrought steel in brine and air at 150°C; reduction of area and elongation versus strain rate.

The ductility of this material (for both elongation and reduction of area) was unaffected by the brine solution when tests were performed at the higher strain rate (10^{-4}/s). At the lower strain rate, however, both reduction of area and elongation were lower for brine tests. Ductility was not affected by strain rate when tests were performed in air.

The cast mild steel (Fig. 4) was similarly affected; reduction of area was substantially lower in the brine environment. Elongation was not measured in these tests because gage marks were found to provide sites for fracture initiation and were removed. The low reduction of area measured on one air-tested specimen was probably the result of a casting defect, and is indicative of the unhomogeniety of castings in comparison to wrought materials. The as-cast mild steel has a lower ductility than wrought steel, and the effect on ductility is about the same for each material.

The load/strain curves from tests of cast mild steel were analyzed to determine the effect of the brine environment on steel deformation. In each case the energy absorbed before maximum load, the energy absorbed after maximum load, and the total energy absorbed were measured. The results are shown in Fig. 5. Both energy to maximum load and energy after maximum load were lower at the lower strain rate. The significant lack of deformational energy absorbed after maximum load means that cracks propagated readily after initiation, but that the energy required to cause crack initiation was not affected by the brine environment.

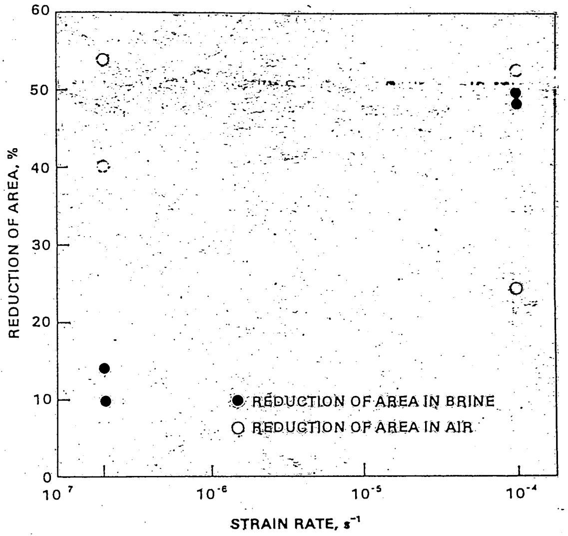


Fig. 4. Results of SSR tests of A27 cast steel in brine and in air at 150°C; reduction of area versus strain rate.

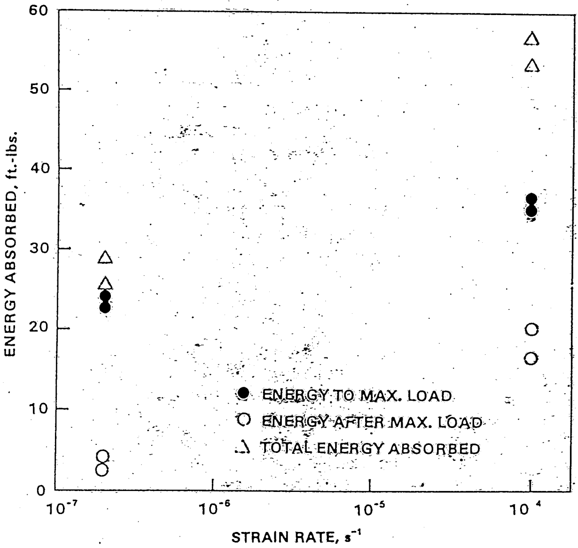


Fig. 5. Results of SSR tests of A27 cast steel in brine at 150°C; energy absorbed versus strain rate.

The significant ductility reduction observed in SSR tests of cast mild steel was investigated by varying the oxygen content of the inlet solution. It was thought that the high oxygen content (relative to expected repository conditions) may have increased the aggressiveness of the brine. Tests were conducted at 150°C and a strain rate of 2 x 10^{-7}/s at both higher and lower solution oxygen contents to investigate the possible effects of oxygen on ductility diminution. In these tests the inlet solution was sparged with argon or with oxygen to attain solution oxygen contents lower and higher than those used in previous tests.

The test results (Fig. 6) clearly show that the relative oxygen content does not have a significant influence on the reduced ductility of cast mild steel over the range of oxygen contents studied. In each case, the post-maximum-load deformation was severely limited and the energy absorbed before maximum load was approximately the same.

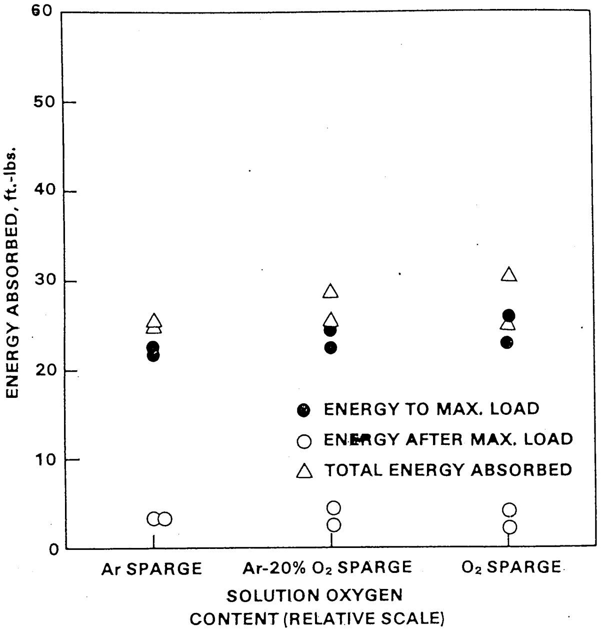
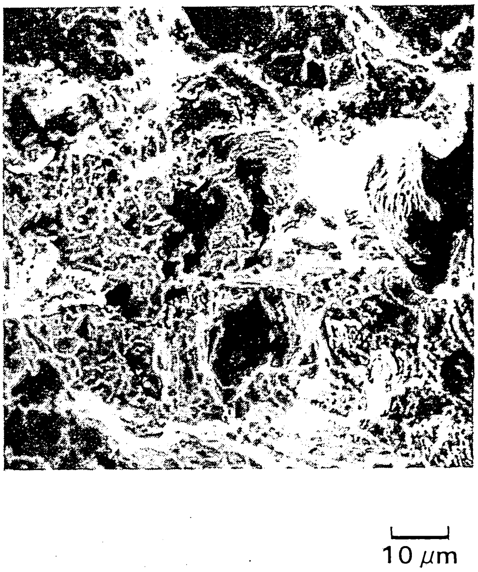


Fig. 6. Results of SSR tests of A27 cast steel in brine at 150°C with varying oxygen content; energy absorbed versus relative oxygen content. Strain rate = 2×10^{-7}/s.

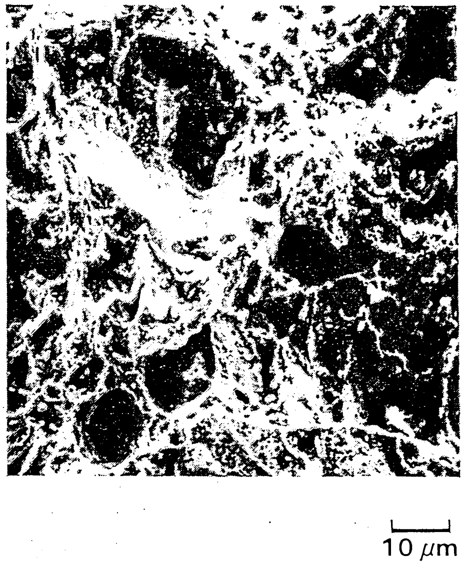
Specimens with relatively high and relatively low ductility were fractographically examined to see if a change in fracture mode occurred. A change, for example, from microvoid coalescence (ductile fracture) to inter- or transgranular cleavage would help identify the mechanism responsible for the observed decreases in ductility.

The fracture mode, as shown in Fig. 7, was the same for low-ductility specimens as for those with high ductility, thus ruling out stress corrosion cracking as the dominant failure mode. Other possible failure modes include hydrogen embrittlement (hydrogen ingress from the corrosion reaction) and dynamic strain aging (deformation restricted by interstitial solute atoms on an atomic scale).

The results of SSR tests at 30°C and 90°C in irradiated brine and in air are given in Table II. The decrease in ductility and ultimate tensile strength observed in irradiated brine is significant because environmentally assisted cracking is unusual at these low temperatures and because ultimate strength was not affected by testing in unirradiated brine at 150°C.



(a) Strain rate = 1×10^{-4}/s



(b) Strain rate = 2×10^{-7}/s

Fig. 7. Fracture surfaces of cast mild steel strained to failure in brine. Both fractures were by microvoid coalescence, with no evidence of intergranular or transgranular fracture.

TABLE II

Results of SSR Tests at 30°C and 90°C

Irradiation Intensity, rad/h	Temperature, °C	Reduction of Area, %	Elongation, %	Ultimate Tensile Strength, MPa
0	30	24.9	20.0	503.0
0	90	38.1	21.5	464.4
0	90	40.0	21.5	481.6
3×10^5	30	12.8	14.0	425.8
3×10^5	90[a]	15.0	[b]	354.8
3×10^5	90	12.9	14.4	443.7

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[a] Moderate overheating occurred during this test, and some salt deposition (drying) occurred.

[b] Elongation was not determined.

CONCLUSIONS

SSR tests were used to characterize the resistance of wrought and cast mild steels to environmentally assisted fracture in a brine solution simulating that anticipated for water intrusion into bedded salt. The following conclusions can be drawn from the test results available to date:

- Both wrought mild steel (AISI 1025) and cast mild steel (ASTM A27) exhibited strain-rate-dependent degradation of ductility in unirradiated SSR tests at 150°C.
- Analysis of the load/deformation curves showed that deformation of the cast steel was most significantly affected in the post-maximum-load region, indicating that fracture occurred easily after cracking was initiated.
- There is no fractographic evidence to indicate that stress corrosion cracking is responsible for the reduced ductility.
- The ductility of cast mild steel (ASTM A216) was significantly affected in tests at 30°C and 90°C in brine irradiated at ~3×10^5 rad/h.
- Ultimate tensile strength was lower in tests done in irradiated brine at 30°C and 90°C than in tests done in air. This is in contrast to the results of unirradiated tests, where ductility was reduced but ultimate strength was not affected.