

A Phenomenological Model for Stress Corrosion
Cracking of Types 316NG and 316 Stainless Steel*

CONF 850902--3

by

CONF-850902--3

TI85 014974

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May 1985

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To be presented at the Second International Symposium on Environmental
Degradation of Materials in Nuclear Power Systems -- Water Reactors,
September 9-12, 1985, Monterey, CA.

MASTER

*Work supported by the Office of Nuclear Regulatory Research, U.S. Nuclear
Regulatory Commission, Washington, D. C. 20555.

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ABSTRACT

A phenomenological model for both intergranular and transgranular stress corrosion cracking (SCC) in constant extension rate (CERT) tests is discussed. The model is based on an estimate of crack-tip strain rate obtained by use of a J-integral approach, a slip dissolution model for SCC, and a J-integral fracture criterion. The model is used to derive correlations between the SCC parameters (such as time to failure and average crack growth rate) and applied (nominal) strain rate. The model is extended to determine similar relationships between the SCC parameters and average near-crack-tip strain rate. The model predictions are compared with CERT test results obtained for Type 316NG stainless steel (SS) in water containing dissolved oxygen and impurities (viz., Cl^- and SO_4^{2-}) at 289°C over a range of strain rates between 10^{-5} and 10^{-7} /s. Good agreement is obtained between the theoretical description and experimental results. The SCC behavior of Type 316NG SS is also compared with that of Type 316 SS to determine the relative SCC susceptibility of the two materials.

*Work supported by the Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, D. C. 20555.

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I. INTRODUCTION

It is well known¹ that the intergranular stress corrosion cracking (IGSCC) susceptibility of conventional Types 304 and 316 stainless steel (SS) in the high-temperature water environment of boiling water reactors (BWRs) occurs because of the existence of a sensitized microstructure.² Sensitization refers to a local change in the chromium content at the grain boundaries, which results from the precipitation of Cr-rich carbides when the steel undergoes heat treatment through a certain temperature regime. Nuclear grade materials such as Types 316NG and 304NG SS are being considered for use in BWRs because their carbon levels (<0.02 wt.%) are low enough to suppress the grain boundary precipitation of carbides, and their nitrogen levels ($0.06 < N < 0.1$ wt.%) are high enough to impart strength properties equivalent to those of conventional materials. Recent constant extension rate (CERT) tests have shown^{3,4} that although Type 316NG SS is extremely resistant to IGSCC even in the presence of dissolved oxygen and impurities (viz., SO_4^{2-} , Cl^-), it is not immune to transgranular (TG) SCC when impurities are present along with a dissolved oxygen concentration of 0.2-8 ppm.⁴ No TGSCC occurs^{3,4} in high-purity water in slow-strain-rate tests over a range of strain rates from 10^{-5} to 10^{-7} /s.

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Recently, we have discussed⁵ the development of a model for IGSCC susceptibility based on elastic-plastic fracture mechanics^{6,7} and the slip-dissolution model of Ford.⁸ The model predictions for IGSCC are in good agreement with CERT results obtained for sensitized Type 316 SS. The purpose of this paper is to discuss this model further and apply the theoretical description to the TGSCC data obtained in two different environments, namely, oxygenated water (0.2 ppm O₂) with 0.1 ppm SO₄²⁻ added as H₂SO₄ and oxygenated water (8 ppm O₂) with 0.5 ppm Cl⁻ added as HCl. The results are also compared with the IGSCC behavior of Type 316 SS to demonstrate the applicability of the model to both IGSCC and TGSCC and to determine the relative SCC susceptibility over a range of strain rates. Finally, the model is extended to derive correlations between the SCC susceptibility parameters and average crack-tip strain rate, which can be expressed in terms of measurable CERT parameters; thus, the consistency of CERT results with a slip-dissolution mechanism can be examined.

II. MODEL DESCRIPTION

A. (Nominal) Strain Rate Effects on SCC

A phenomenological model for SCC has been developed⁵ which is based on (1) an estimate of the crack-tip strain rate, obtained by use of a J-integral approach;^{6,7} (2) the slip-dissolution model of Ford;⁸ and (3) a J-integral fracture criterion.¹⁰ The model is used to derive correlations between the applied strain rate and such conventional SCC parameters as time to failure, t_f , strain to failure, ϵ_f , and average crack growth rate, \dot{a}_{av} , in the critical-strain-rate regime where the SCC susceptibility increases with a decrease in $\dot{\epsilon}$. For a fully plastic material containing a crack in a uniform strain field, the strain ϵ is greater than the yield strain ϵ_y and J is approximated by⁵⁻⁷

$$J = \alpha \epsilon a = \beta \sigma_f \delta, \quad (1)$$

where a is the crack length, δ is the crack-tip-opening displacement, σ_f is the flow stress, and α and β are constants. One can define a near-crack-tip strain rate $\dot{\epsilon}_T$ in terms of δ and the crack-tip-opening displacement rate $\dot{\delta}$:

$$\dot{\epsilon}_T = \frac{\dot{\delta}}{\delta} = \frac{\dot{J}}{J} = \frac{\alpha \dot{\epsilon} a + \alpha \epsilon \dot{a}}{\alpha \epsilon a} = \frac{\dot{\epsilon}}{\epsilon} + \frac{\dot{a}}{a}. \quad (2)$$

Thus, $\dot{\epsilon}_T$ is insensitive to the values of α and β and the definition of flow stress. In his slip-dissolution model for SCC, Ford⁸ has proposed that

$$\dot{a} = A_T (\dot{\epsilon}_T)^{0.5}, \quad (3)$$

where \dot{a} is the crack growth rate and A_T is a constant that is dependent on parameters which control passivation rate, such as the chemical composition of the steel or the degree of sensitization and oxygen and impurity contents of the aqueous environments. Combining Eqs. (2) and (3), one obtains the following relationship between the crack growth rate and nominal strain rate:

$$\dot{a} = A_T \left(\frac{\dot{a}}{a} + \frac{\dot{\epsilon}}{\epsilon} \right)^{0.5}, \quad (4)$$

where a is the crack length at time t . For CERT tests,

$$\frac{\dot{\epsilon}}{\epsilon} = \frac{1}{t}; \quad (5)$$

therefore,

$$\dot{a} = A_T \left(\frac{\dot{a}}{a} + \frac{1}{t} \right)^{0.5}. \quad (6)$$

The exact analytical solution of Eq. (6) is

$$a = A_T \sqrt{6t}, \quad (7)$$

which is consistent with a diffusion-controlled crack propagation process. If we define $a = a_f$ and $t = t_f$ at failure, Eq. (7) becomes

$$a_f = A_T \sqrt{6t_f} = A t_f^{0.5}, \quad (8)$$

where $A = \sqrt{6} A_T$. Thus, the validity of the above relation is experimentally verifiable in terms of measurable CERT parameters.

Equation (8) is a useful relation, but is not in itself sufficient to determine t_f ; an additional fracture criterion is required. A possible fracture criterion is provided by the J-integral (see Ref. 6), which is given by

$$J = C_0 \epsilon^{n+1} f(n) a \approx C \epsilon^{n+1} a. \quad (9)$$

At failure, i.e., when $a = a_f$ and $\epsilon = \epsilon_f$, the J-integral attains a critical value, J_c :

$$J_c = C_0 \epsilon_f^{n+1} f(n) a_f \quad (10a)$$

$$\approx C \epsilon_f^{n+1} a_f, \quad (10b)$$

$$\approx C \epsilon_f a_f,$$

where J_c depends on the material and geometry; C_0 is a material parameter related to plastic modulus; n is the strain-hardening exponent near fracture at large strains; $f(n)$ = function of n ; ϵ_f = strain at failure; and $C \approx C_0 f(n)$. It is important to point out that in the application of the J-integral fracture criterion to CERT tests, our model makes a clear distinction between SCC and mechanical cracking (viz., J_c refers to the value of J at which the occurrence of rapid mechanical crack initiation and growth leads to failure; this process is preceded by SCC). In other studies,^{9,10} this distinction is not made and environmentally assisted cracking is implicitly included in the J-R curve, giving rise to an apparent dependence on environment and strain rate. When the environmentally assisted crack growth is explicitly accounted for, the fracture-characterizing parameter (more specifically, J_c/C) is expected to be almost independent of both environment and strain rate. CERT test results

permit examination of the validity of this fracture criterion. Since n has a relatively small value compared to unity, it is ignored for simplicity in Eq. (10b). It should be pointed out that higher values of the power law exponent are usually quoted for stainless steel. However, they are based on a description of the stress-strain behavior over a wide range of strains. Equations (8) and (10b), along with $\epsilon_f = \dot{\epsilon} t_f$ and $\dot{a}_{av} \simeq a_f/t_f$ (assuming that crack initiation in CERT tests occurs early in life), can be combined to derive correlations between the SCC susceptibility parameters and the strain rate as follows:

$$a_f = A(J_c/AC)^{1/3} \dot{\epsilon}^{-1/3}, \quad (11)$$

$$t_f = (J_c/AC)^{2/3} \dot{\epsilon}^{-2/3}, \quad (12)$$

$$\dot{a}_{av} = A(AC/J_c)^{1/3} \dot{\epsilon}^{1/3}, \text{ and} \quad (13)$$

$$\epsilon_f = (J_c/AC)^{2/3} \dot{\epsilon}^{1/3}. \quad (14)$$

It should be noted that Eqs. (11)-(14) are valid only in the critical-strain-rate regime where the severity of cracking increases with a decrease in strain rate. Examination of Eqs. (11)-(14) shows that the fracture-characterizing parameter appears as J_c/C . Hence, from one CERT test at the appropriate $\dot{\epsilon}$ (that at which SCC susceptibility occurs), both the strain-rate-independent parameters (for a chosen environment and material condition and geometry) A and J_c/C can be determined [see Eqs. (8) and (10b)] provided a_f is determined from the fracture surface (t_f and hence $\epsilon_f \simeq \dot{\epsilon} t_f$, of course, simply follow from the test). The quantity a_f is usually interpreted as the maximum crack length that can be measured on the fracture surface by means of scanning electron microscopy. A knowledge of the parameters J_c/C and A can then be used to predict the SCC susceptibility at other strain rates [by means

of Eqs. (11)-(14)], provided the value of $\dot{\epsilon}$ lies in the critical-strain-rate regime for the environment and material under investigation.

B. Crack-Tip Strain Rate Considerations

We have used a J-integral approach to estimate the crack-tip strain rate $\dot{\epsilon}_T$ [see Eq. (4)] and Ford's slip-dissolution model to relate $\dot{\epsilon}_T$ to crack growth rate \dot{a} . In Eq. (11), A_T is a constant independent of $\dot{\epsilon}$, but it depends on parameters that control passivation rate, such as degree of sensitization, alloy chemical composition, or dissolved oxygen and impurities in the environment, as discussed by Ford. Apart from the fundamental nature of the crack-tip strain rate and related parameters, estimates of crack-tip strain rates $\dot{\epsilon}_T$ represent a significant step as they potentially allow results obtained with different testing techniques to be correlated by representation on an \dot{a} vs $\dot{\epsilon}_T$ diagram. The relationships between average near-crack-tip strain rate (which can be estimated from CERT parameters) and the strain rate and other SCC susceptibility parameters of CERT tests can be derived as follows:

An average crack-tip strain rate can be defined by

$$\bar{\epsilon}_T = \frac{1}{t_f - t_o} \int_{t_o}^{t_f} \dot{\epsilon}_T dt . \quad (15)$$

Substituting for $\dot{\epsilon}_T$ as defined by Eq. (2) and performing the integration, we obtain¹³

$$\bar{\epsilon}_T = \frac{1}{t_f - t_o} \left(\ln \frac{\epsilon_f}{\epsilon_o} + \ln \frac{a_f}{a_o} \right) , \quad (16)$$

where t_o , ϵ_o , and a_o are the time, strain, and crack length, respectively, at the initiation of the crack and the subscript f refers to the corresponding

values at failure. Assuming that $a_o \ll a_f$ and $t_o \ll t_f$ and substituting Eq. (10) in Eq. (16), we obtain

$$\bar{\epsilon}_T = \frac{1}{t_f - t_o} \ln \frac{J_c}{C \epsilon_o a_o} . \quad (17)$$

By defining

$$K = \ln \frac{J_c}{C \epsilon_o a_o} , \quad (18)$$

Eq. (17) may be simplified to

$$t_f - t_o = K(\dot{\epsilon}_T)^{-1} . \quad (19)$$

Since K depends only on C (a geometric variable) and J_c (and also on ϵ_o and a_o), it is assumed to be independent of failure mode (i.e., IGSCC or TGSCC), and it is also assumed to be fairly insensitive to environmental variables. Combining Eqs. (12) (in which t_f is corrected for crack initiation time) and (19), we obtain the following relationship between $\dot{\epsilon}$ and $\bar{\epsilon}_T$:

$$\dot{\epsilon} = \frac{(J_c/AC)}{K^{3/2}} (\bar{\epsilon}_T)^{3/2} . \quad (20)$$

Since A is strongly dependent on the environment, the relation between $\dot{\epsilon}$ and $\bar{\epsilon}_T$ is not solely dependent on geometry and mechanics. Combining Eqs. (13) and (20) gives the following relationship between \dot{a}_{av} and $\bar{\epsilon}_T$:

$$\dot{a}_{av} = \frac{A}{K^{0.5}} \bar{\epsilon}_T^{0.5} . \quad (21)$$

Equation (21) is identical in form with the power-law relationship proposed by Ford in his slip-dissolution model,⁸ but is expressed in a form that can be

verified in CERT tests. Thus the consistency of CERT test results with the slip-dissolution model of Ford can be examined.

III. COMPARISON BETWEEN MODEL PREDICTIONS AND EXPERIMENTAL RESULTS

CERT tests have been performed on Types 316NG and 316 SS in oxygenated water (0.2 and 8 ppm O_2) containing 0.1 ppm sulfate and 0.5 ppm chloride ion impurities (added as acids), respectively, over a range of strain rates between 10^{-5} and 10^{-8} /s. The conductivity of the water at room temperature is 0.9 $\mu S/cm$ with 0.1 ppm sulfate and 5-7 $\mu S/cm$ with 0.5 ppm Cl^- . Both materials were given the same heat treatment (1050°C/0.5 h + 650°C/24 h). This treatment produced no measurable sensitization in Type 316NG SS, as assessed by the electrochemical potentiokinetic reactivation (EPR) method,¹¹ but caused sensitization of Type 316 SS to $EPR \approx 17 C/cm^2$. The specimens were enclosed in an autoclave with a once-through water system containing dissolved oxygen and impurities, and tested at 289°C, a pressure of 9 MPa, and a flow rate of 10 cm^3/min . The chemical compositions of the two steels are shown in Table I; details of the specimen preparation and experimental procedure for CERT tests have been described elsewhere.^{4,5}

After the tests were completed, the fracture surfaces were examined by scanning electron microscopy to determine the maximum IG or TG crack length. The average crack growth rates \dot{a}_{av} were estimated with the assumption that the crack length and strain at crack initiation (a_0 and ϵ_0 , respectively) are small compared to the corresponding values at failure. Specifically, the estimation of \dot{a}_{av} is based on $a_0 = 1 \mu m$ and $\epsilon_0 = 1\%$. The assumption of a small crack at initiation early in life has been supported by some interrupted tests performed on Types 316NG and 316 SS. Although a range of crack initiation parameters can be obtained depending on the material/environment system and experimental

variables,^{12,13} a sensitivity analysis shows that the results discussed in the present study are only weakly dependent on and not significantly affected by the choice of crack initiation parameters over a wide range of values ($a_0 = 1\text{--}100\ \mu\text{m}$ and $\epsilon_0 = 0.3\text{--}1.0\%$).

The experimentally observed power-law relationships between \dot{a}_{av} and $\dot{\epsilon}$ for Type 316NG SS (TGSCC) and Type 316 SS (IGSCC) tested in impurity environments show good agreement with the model predictions [Eq. (13)], as can be seen from the plots of \dot{a}_{av} vs $\dot{\epsilon}$ in Fig. 1. Similar agreement is obtained when other SCC parameters are considered. In the relatively more aggressive chloride environment, the average TG crack growth rates for Type 316NG SS are an order of magnitude lower than the average IG crack growth rates for Type 316 SS. In the less aggressive sulfate environment, the rates for Type 316NG are approximately a factor of 3 lower than for Type 316. Furthermore, in going from the aggressive chloride environment to an environment with impurity levels within the current allowable BWR water-chemistry limits, the average IG crack growth rate decreases by a factor of ~ 4 , but the average TG crack growth rate decreases by a factor of less than two. Hence the relative crack growth rate for the conventional and the NG materials is dependent on the testing environment.

The average near-crack-tip strain rate $\bar{\epsilon}_T$ estimated from CERT test results and Eq. (16) is plotted vs \dot{a}_{av} for Types 316NG and 316 SS in Figs. 2 and 3. The results obtained for the two impurity environments show that \dot{a}_{av} varies as the square root of $\bar{\epsilon}_T$, as predicted by the model [Eq. (21)]. These results are consistent with Ford's model,⁸ which relates the two in terms of instantaneous quantities rather than in terms of average parameters as has been done here. Formulation of Ford's slip-dissolution model for CERT tests in terms of average quantities makes it amenable to experimental verification. The estimation of

crack-tip strain rates in CERT tests provides a potential link for correlating the results with the crack growth data obtained under different loading conditions, although currently difficulties still exist for estimating crack-tip strain rates under different loading histories.

It is important to observe that K in Eq. (19) depends only on C (a geometric variable), J_c , ϵ_o , and a_o ; it is independent of whether failure occurs by IGSCC or TGSCC and is fairly insensitive to environmental variables such as oxygen and impurity level. Hence, if K is not significantly influenced by the secondary cracks that may form late in life, Eq. (19) is expected to be valid for CERT test results obtained for Types 316NG and 316 SS in several environments. Figure 4 shows a comparison of Eq. (19) with data obtained in oxygenated water (0.2 and 8 ppm O_2) with sulfate and chloride impurities, respectively. The excellent agreement between Eq. (19) and the data lends support to the model and validates some of the assumptions in the analysis, such as constancy of J_c in different environments (as discussed earlier) and the insensitivity of the results to crack initiation parameters over the range considered in this study.

Finally, it should be pointed out that a number of approximations have been made in order to develop a simplified model for strain rate effects on SCC in the critical-strain-rate regime discussed in this study. For example, secondary cracking has not been considered in the model. However, the overall consistency between the measured failure strains and the strains predicted on the basis of our initial correlation between strain and displacement, as well as the agreement of the CERT test results with Eq. (19), suggests that the effect is of second order. Furthermore, a J-based approach is used to characterize the crack growth behavior as well as fracture in the fully plastic material. Some of the simplifications we have introduced in applying the J-integral solutions to CERT

test results are difficult to justify a priori through a rigorous use of elastic-plastic fracture mechanics. However, the good agreement between model predictions and experimental results over a wide range of strain rates and environmental variables lends support to the adequacy of the approximations. The model can also account for the effects of environmental and material variables on SCC in an explicit manner^{14,15} and thus provides a potentially useful approach to the analysis and interpretation of CERT test results.

IV. SUMMARY

A model for quantitatively describing the effects of strain rate (as well as environment and alloy microstructure) on SCC susceptibility in CERT tests is discussed. The theoretical description makes use of a J-integral approach to estimate the crack-tip strain rate; this is used in conjunction with the slip-dissolution model of Ford to derive the crack growth behavior. These results, along with a J-integral fracture criterion, are used to derive the power-law relationships between the SCC susceptibility parameters and strain rate. The theoretical predictions are shown to be in good agreement with the experimental results on TGSCC for Type 316NG SS and IGSCC for Type 316 SS, observed in two different environments. The improved performance of the nuclear grade material over the conventional material is discussed in a quantitative manner. An extension of the model is used to derive a relationship between average crack growth rate and average crack-tip strain rate; this relation is also verified by experimental results for TGSCC and IGSCC in oxygenated water containing chloride and sulfate ion impurities and is consistent with a slip-dissolution mechanism for SCC.

ACKNOWLEDGMENTS

The author wishes to thank W. J. Shack and F. A. Nichols for helpful discussions.

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Table I. Chemical Composition (wt %) of Stainless Steels Used in the Present Study

| Material | Heat No. | C | Mn | P | S | Si | Ni | Cr | Mo | Co | Cu | N | O | B |
|----------|----------|-------|------|-------|-------|------|-------|-------|------|------|------|--------|--------|--------|
| 316 | 0590019 | 0.05 | 1.68 | 0.030 | 0.008 | 0.71 | 10.83 | 17.34 | 2.05 | 0.34 | 0.20 | 0.0145 | 0.0011 | 0.0005 |
| 316NG | P91576 | 0.015 | 1.63 | 0.020 | 0.010 | 0.42 | 10.95 | 16.42 | 2.14 | - | - | 0.068 | - | 0.002 |

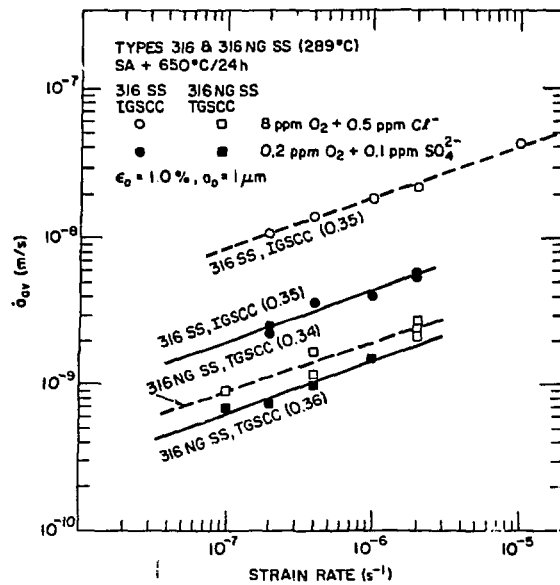


Fig. 1. Correlation between Average IGSCC or TGSCC Growth Rate and Strain Rate for Types 316 and 316NG SS.

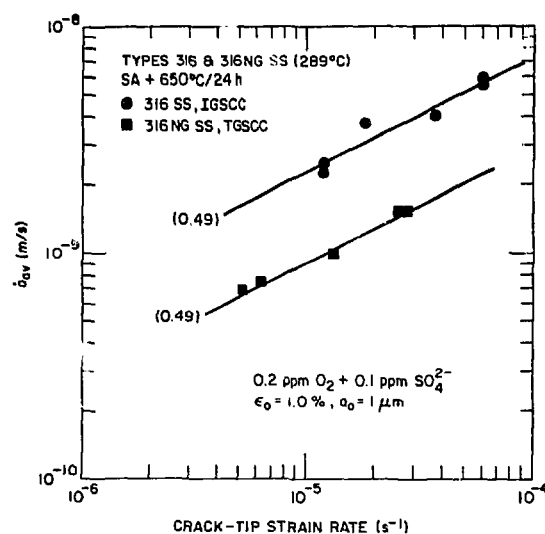


Fig. 2. Average IGSCC or TGSCC Growth Rate as a Function of Average Crack-Tip Strain Rate for Types 316 and 316NG SS in a Sulfate Environment.

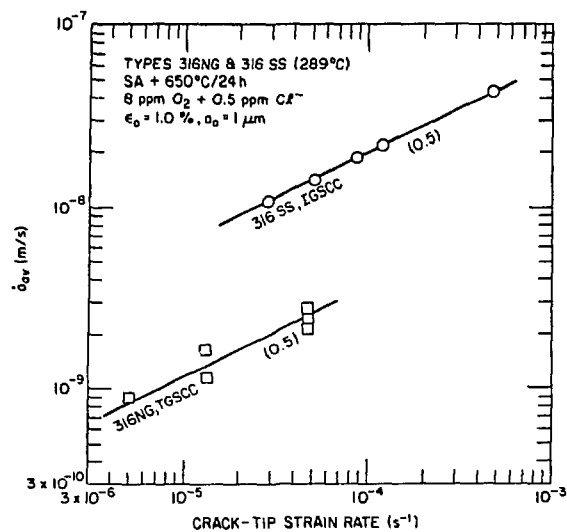


Fig. 3. Correlation between Average IGSCC or TGSCC Growth Rate and Average Crack-Tip Strain Rate.

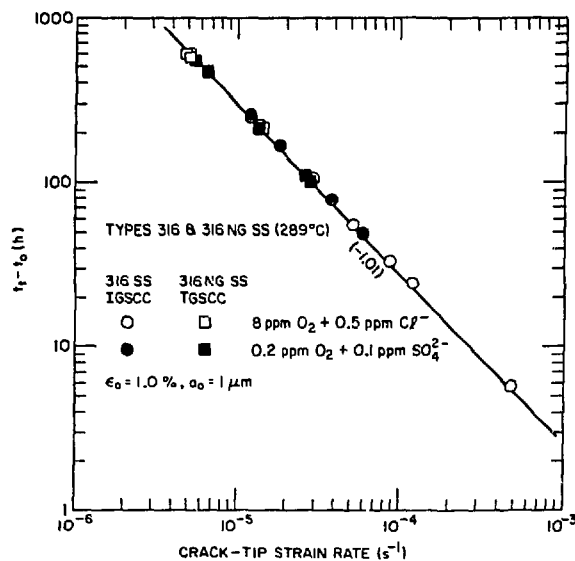


Fig. 4. Relationship between Time to Failure and Average Crack-Tip Strain Rate.