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Stress-corrosion cracking of reactor pressure-vessel materials in a simulated pressurized water-reactor environment

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SYNOPSIS

The paper gives an assessment of the available data on the linear elastic fracture mechanics involved in stress-corrosion cracking and in the behaviour of reactor pressure-vessel materials exposed to a simulated pressurized water-reactor environment. As a result of the different experimental techniques used in the various studies mentioned, only a few detailed conclusions can be drawn.

The data from constant-displacement wedge-opening loading (WOL) tests, with the exception of one study, exhibited very limited or no crack extension during exposure periods of up to 14 000 hours.

The data from constant-load tests indicated evidence of crack extension, but the nature and extent of the crack extension are still unclear. When crack extension did occur, it was exclusively transgranular in nature.

Certain differences in the available test data are tentatively explained in terms of the experimental procedures used, and the need for a consistent code of experimental techniques is highlighted.

SAMEVATTING

Die referaat gee 'n evaluering van die beskikbare data oor die meganika van lineêre elastiese breuke betrokke by spanningskorrosiekraking en by die gedrag van reaktordrukhouermateriale wat aan 'n gesimuleerde water-reaktoromgewing met drukreëling blootgestel word. As gevolg van die verskillende eksperimentele tegnieke wat gebruik is in die verskillende studies wat genoem word, kan daar net 'n paar uitvoerige gevolgtrekkings gemaak word.

Met die uitsondering van een studie, het die data oor konstanteverplasing-wigopeningbelastingtoetse baie beperkte, of glad geen kraakverlenging nie, tydens blootstellingstydperke van tot 14 000 uur getoon.

Die data oor konstantebelastingtoetse het tekens van kraakverlenging getoon, maar die aard en omvang van die kraakverlenging is nog nie duidelik nie. Waar daar wel kraakverlenging voorgekom het, was dit uitsluitlik transgranulêr van aard.

Sekere verskille in die beskikbare toetsdata word tentatief verklaar in terme van die eksperimentele prosedures wat gebruik is en die behoefte aan 'n konsekwente kode van eksperimentele tegnieke word benadruk.

Introduction

Environmentally assisted cracking (EAC) refers to the cracking of materials under the conjoint actions of an applied stress and an aggressive environment. This phenomenon is known to occur in many engineering alloys over a wide range of environmental conditions, and can have a serious effect on structural reliability. Stress-corrosion cracking (SCC) is the generic term for one particular form of EAC that occurs under constant stress conditions.

The first reported instance (some forty years ago) of SCC was by Hodge and Miller[1], who discussed brittle failures observed in an austenite Cr–Ni steel container pressurized with wet ethyl chloride. Later, however, in the 1950s the incidence of industrial failures of austenitic steels by SCC was at such a level that a great deal of concern was created. Indeed, over subsequent decades there has been a proliferation of alloy–environment systems in which SCC has been found to occur.

One such alloy–environment system that is extremely important in modern terms is that of light water reactors (LWR) within which reactor pressure-vessel (RPV) materials are in contact with purified water in the reactor primary system. Over a decade ago, the potentiality of the purified water in the reactor primary system in causing the SCC of austenitic stainless steel was recognized through failures in the reactors after some period of operation. Indeed, a striking number of instances of the SCC of austenitic stainless steel in boiling-water reactors (BWR) were recognized through failures after some period of operation[2].

The water chemistry specified for a BWR is given in Table I. The first SCC failure was observed in 1966 in the stainless-steel weld overlay of a BWR steel pressure vessel[3]. It was concluded that the SCC of the weld deposit was the result of improper welding. In 1972, SCC of stainless steel piping was found in the heat-affected zone (HAZ) of some weld joints. However, in that case the SCC was confined to sensitized regions in the stainless steel and was intergranular in nature.

Details of the failure of steam turbine No. 5 at the A Nuclear Power Station in Hinkley Point were reported by Kalderon[4] and Gray[5] in 1972. It was established that fast catastrophic failure had propagated from small stress–corrosion cracks in highly stressed disc keyways fabricated from a 3 per cent Cr – 0,5 per cent Mo low-alloy ferritic steel. Subsequent studies[6] found that SCC occurred in this low-alloy steel when it was subjected to a high-purity steam environment at 120°C. The crack extension was mainly intergranular and propagated at a maximum rate of 4×10^{-7} mm/s.

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TABLE I

SPECIFICATIONS FOR THE WATER CHEMISTRY IN AN LWR

	PWR*	BWR
Boric acid, p.p.m.	0 to 13 000	–
Lithium hydroxide, p.p.m.	1,3 to 13	–
pH at 25°C	4,2 to 10,5	6,5 to 7,0
Dissolved oxygen, p.p.b.	$\leqslant$100	50 to 200
Chloride, p.p.b.	$\leqslant$150	$\leqslant$100
Conductivity, μs	1 to 40	$\leqslant$0,1
Hydrogen, cm^3/kg	25 to 35	–

*Pressurized water reactor

Another type of LWR is the pressurized water reactor (PWR). The specification for the water chemistry of a PWR is also listed in Table I. Indeed, over the past few years, numerous studies have been conducted in an effort to establish the corrosion-fatigue behaviour of RPV materials in a PWR environment, and a brief review of these studies was published recently[7]. However, SCC studies in this area have not been as numerous or as widely reported, and the present paper reviews the SCC behaviour of ferritic RPV materials in a PWR environment utilizing precracked test specimens of the linear elastic fracture mechanics (LEFM) type.

Experimental Details

Essentially two types of testing procedures were adopted in the study of SCC in PWR water, and the test specimens are shown in Fig. 1. Fig. 1(a) shows the wedge-opening loading (WOL) specimen, which is loaded to a fixed crack-tip displacement value and, hence, a stress intensity level K. In such tests, the K value decreases with crack extension because the displacement remains constant. After being preloaded to a selected K value, the WOL specimens are inserted into autoclaves specifically used for tests on corrosion fatigue. After a period of exposure, i.e. the duration of the corrosion fatigue test, the WOL specimens are removed and measurements of the crack lengths are recorded. The WOL specimens are then re-inserted in the autoclaves, along with the specimens for the next corrosion-fatigue test, and so on. The second type of test is a constant-load test, Fig. 1(b), in which the K value increases with crack extension. These particular tests are conducted in an autoclave that is devoted to SCC tests, and the specimens can remain exposed without interruption to a PWR environment for significant periods of time. This uninterrupted period, however, depends on the nature of the monitoring of crack growth that is being used.

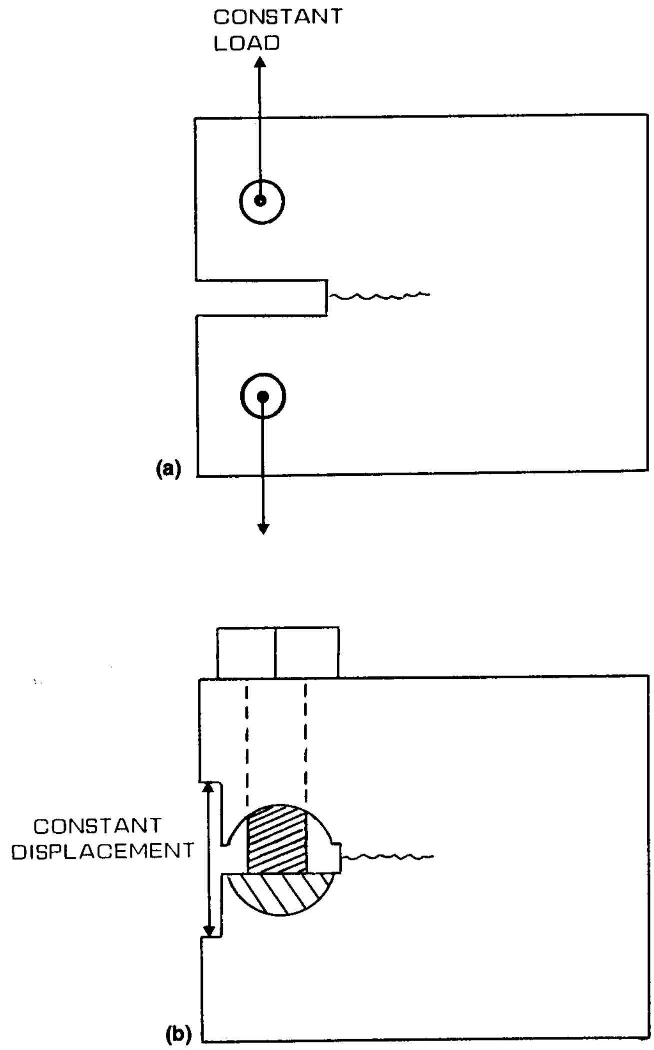

Fig. 1—Configuration of the test specimens used in SCC studies
(a) Compact-tension test
(b) Constant-displacement wedge-opening load (WOL) test

Assessment of Results

Constant Displacement Experiments

The main study conducted in this area is that of the Westinghouse Electric Corporation[8], which has been exposing specimens to LWR environments for some twelve years. Significant crack extensions have been recorded in several HAZ materials, and some selected data are depicted in Fig. 2. The diagram shows that there is an incubation period of about 1000 hours before crack extension occurs. Over the period 1000 to 3000 hours, significant crack extension was recorded (20 to 30 mm), and the average crack velocity, da/dt for the range of HAZ materials was about 4×10^{-6} mm/s. As part of this study, tests were carried out on A533B plate material, and the crack extension versus exposure time is shown in Fig. 3, from which it is evident that significant crack extension occurred after an incubation time of over 37 000 hours. Again, the average velocity of crack growth in the plate materials was approximately 4×10^{-6} mm/s.

Crack extension in the HAZ material was irregular and tended to occur intermittently and, although the initial crack growth tended to be uneven, after a time the lengths of the cracks evened out on each side. No crack extension was recorded in this study in weldments or A508 Cl III materials after nearly 45 000 hours of exposure to a PWR environment.

Atkinson[9] examined a range of A533B plate and A508 Cl III forging materials that had been tested at K values of between 60 and 90 MPa $\sqrt{m}$ over exposure times varying from 3000 to over 12 000 hours. Small amounts of crack extension were recorded at crack velocities of about 1×10^{-9} mm/s.

Of a wide range of materials studied in the U.K., it was reported[10] that no cracking was observed, even in a number of HAZ specimens exposed for some 14 000

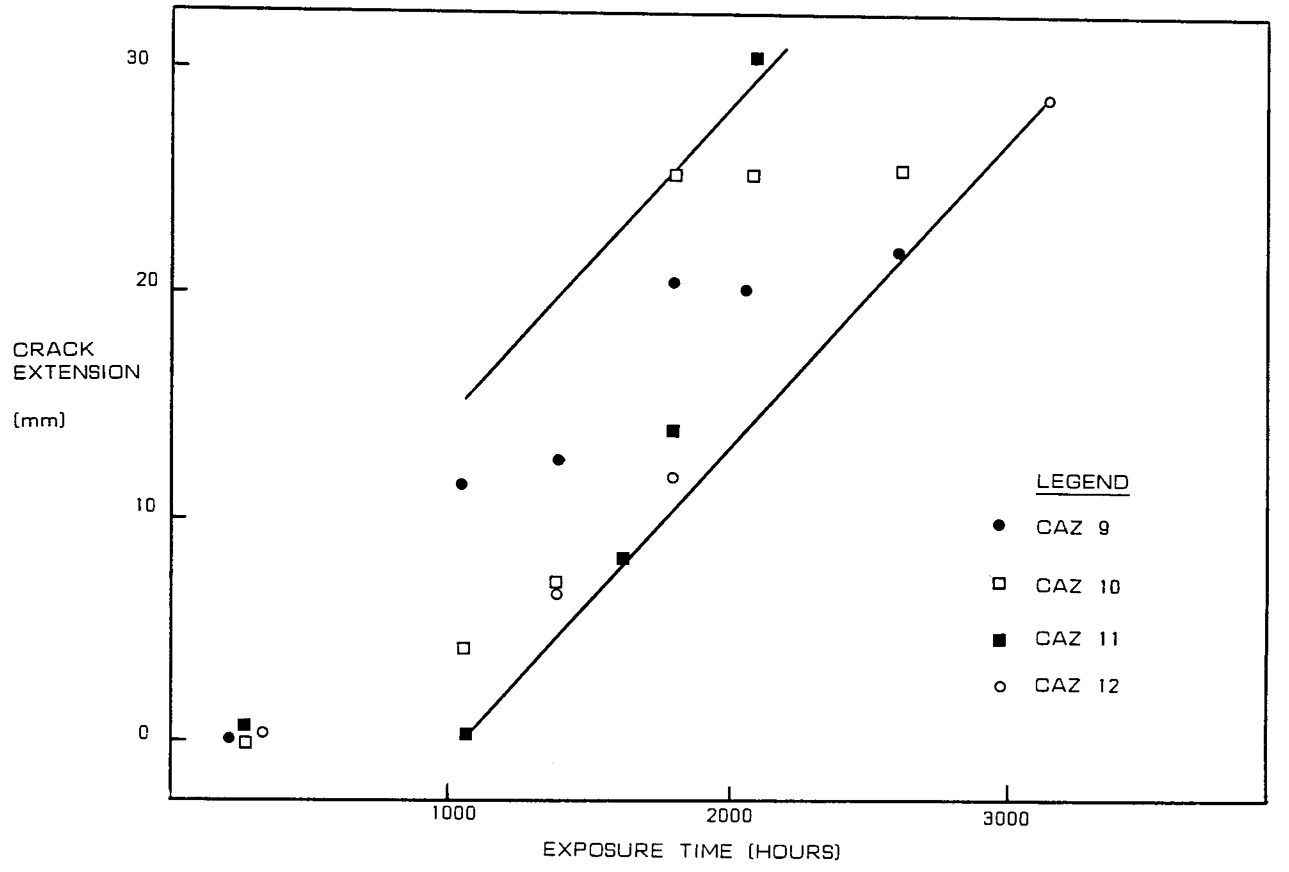

Fig. 2—Results for WOL test specimens of HAZ materials exposed to a PWR environment for which the crack length was monitored by optical measurement of the side cracks (after Gray[5])

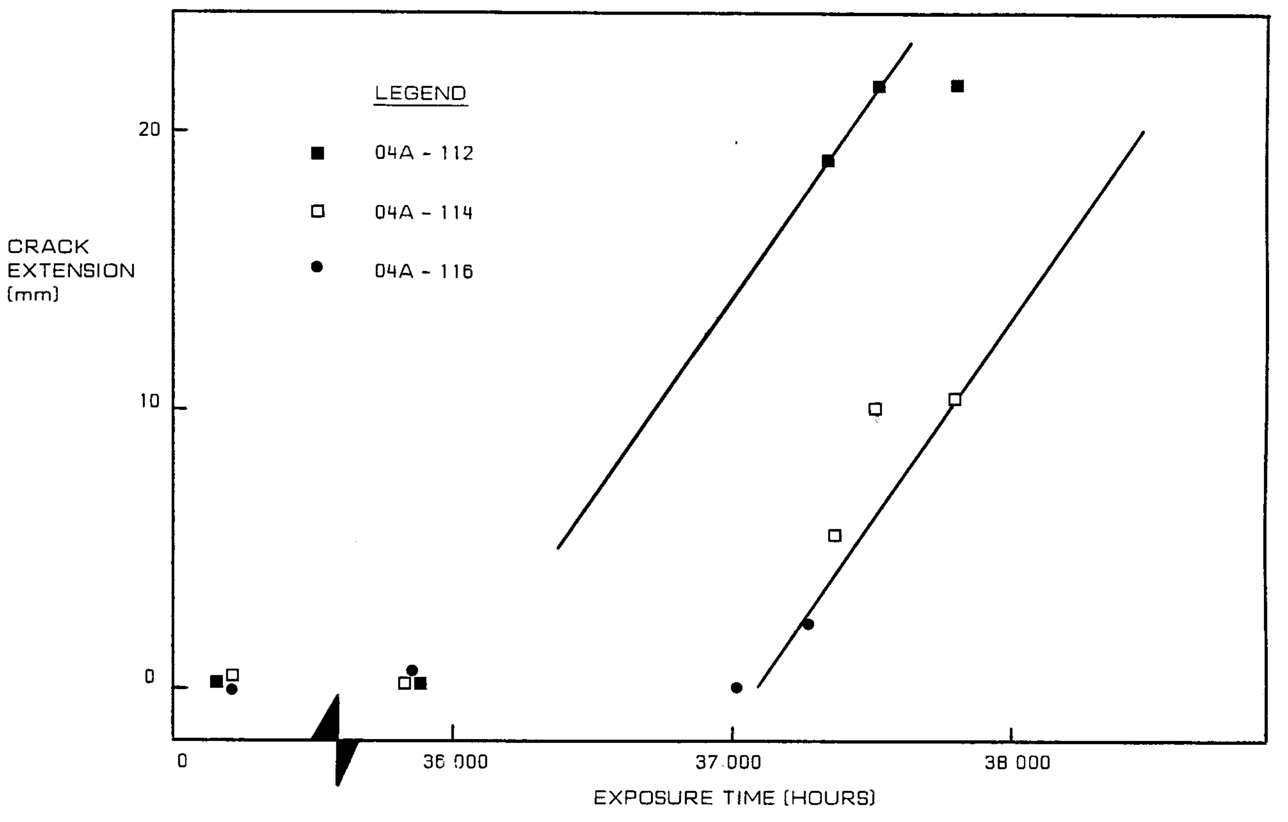

Fig. 3—Results for WOL test specimens of A533B plate materials exposed to a PWR environment for which the crack length was monitored by optical measurement of the side cracks (after Gray[5])

hours, which is considerably in excess of the incubation period observed by Bamford *et al.*[8] for HAZ materials.

Babcock Power[11] reported the results of a series of WOL tests conducted on A533B plate, A508 Cl III forging, A533B HAZ, A508 Cl III HAZ, and a weld metal. The WOL specimens were loaded to an initial K value of 105 MPa $\sqrt{m}$ (to simulate the Westinghouse test conditions) and, after some 7000 hours of exposure to a PWR environment, no crack extension was recorded.

Constant-load Experiments

In a series of constant-load SCC tests, only two studies were reported: one conducted by Bulloch[11] and recently reported by Lloyd[10], and one by Tice[12].

The former study involved a series of compact tension specimens 25 mm thick that had been precracked in air and then dead-loaded into three separate autoclaves in strings of ten specimens (Fig. 4) in a simulated PWR environment. The specimens were A533B and A508 Cl III steel weldments and associated HAZ materials; each was tested at the following three K levels: 40, 60, and 85 MPa $\sqrt{m}$. The chemical analyses of the water, which were carried out consistently throughout the exposure time of 8000 hours, are depicted in Fig. 5. The crack growth was estimated initially by optical monitoring of the crack length in the sides of the specimens after each three-monthly routine visual inspection.

These measurements indicated that crack growth had occurred during the first exposure of some 2000 hours but there was very little, if any, after that (Figs. 6 to 8). These diagrams indicate that the crack extension varied from less than 0,1 mm to a maximum of 0,7 mm. In general, the A533B plate and A508 Cl III forging materials exhibited very small amounts of crack growth, while the HAZ materials and the weld metal showed the greatest amount of crack extension. The crack growth in both cases was insensitive to the K level. The average velocities of crack growth over the initial 2000 hours of exposure for the HAZ materials and weld metals varied over the range 4 to 9 $\times$ 10^{-8} mm/s, while the plate and forging materials exhibited very low velocities, being lower by over an order of magnitude. A subsequent detailed examination by scanning electron microscope (SEM) of five selected specimens[11] that had been broken open to facilitate the measurement of the fracture surfaces showed that the side measurements were larger than the actual crack extensions, which averaged about 0,2 mm with a maximum of 0,5 mm (Fig. 9). These differences between the side measurements and the measurements of the fracture surfaces were the result of the thin side ligaments of the materials that had not cracked during the fatigue precracking exercise; such ligaments could have arisen from residual stresses due to surface machining, and crack extension could have occurred through them by tearing immediately after the static test load was applied.

Tice[12] reported a series of three short-term tests on A533B, A508 Cl III, and A533B HAZ materials during which the crack length was monitored continuously by a d.c. potential drop method. All the test specimens were precracked in a PWR environment at 290°C. No significant crack growth was recorded in the A533B and A508 Cl III materials, while the A533B HAZ test specimen, loaded to an initial K level of 60 MPa $\sqrt{m}$, exhibited about

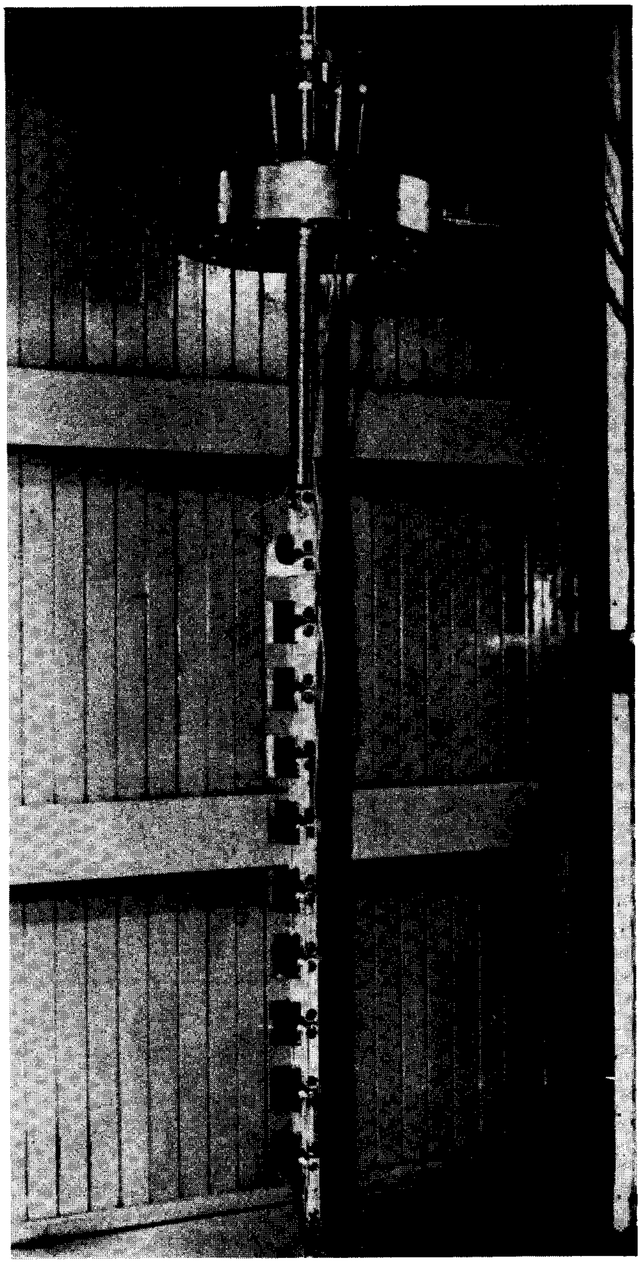

Fig. 4—Details of a string of ten 25-mm thick compact-tension specimens inserted into a test autoclave under constant load (after Bulloch[11])

1 mm of crack extension within about 230 hours of testing (Fig. 10). This crack extension was transient in nature inasmuch as the initial crack velocity was approximately 4 $\times$ 10^{-6} mm/s; this velocity, however, continually diminished with time to about 1 $\times$ 10^{-7} mm/s after some 250 hours of testing. Subsequent detailed studies of the broken fracture surfaces revealed that about 1 mm of crack extension had occurred only over half of the specimen (Fig. 11), where the initial precrack length (hence K level) had been largest. In all instances in which measurable crack growth was recorded, crack extension occurred exclusively in a transgranular manner.

Discussion

Certain trends are evident in the experimental data reported above.

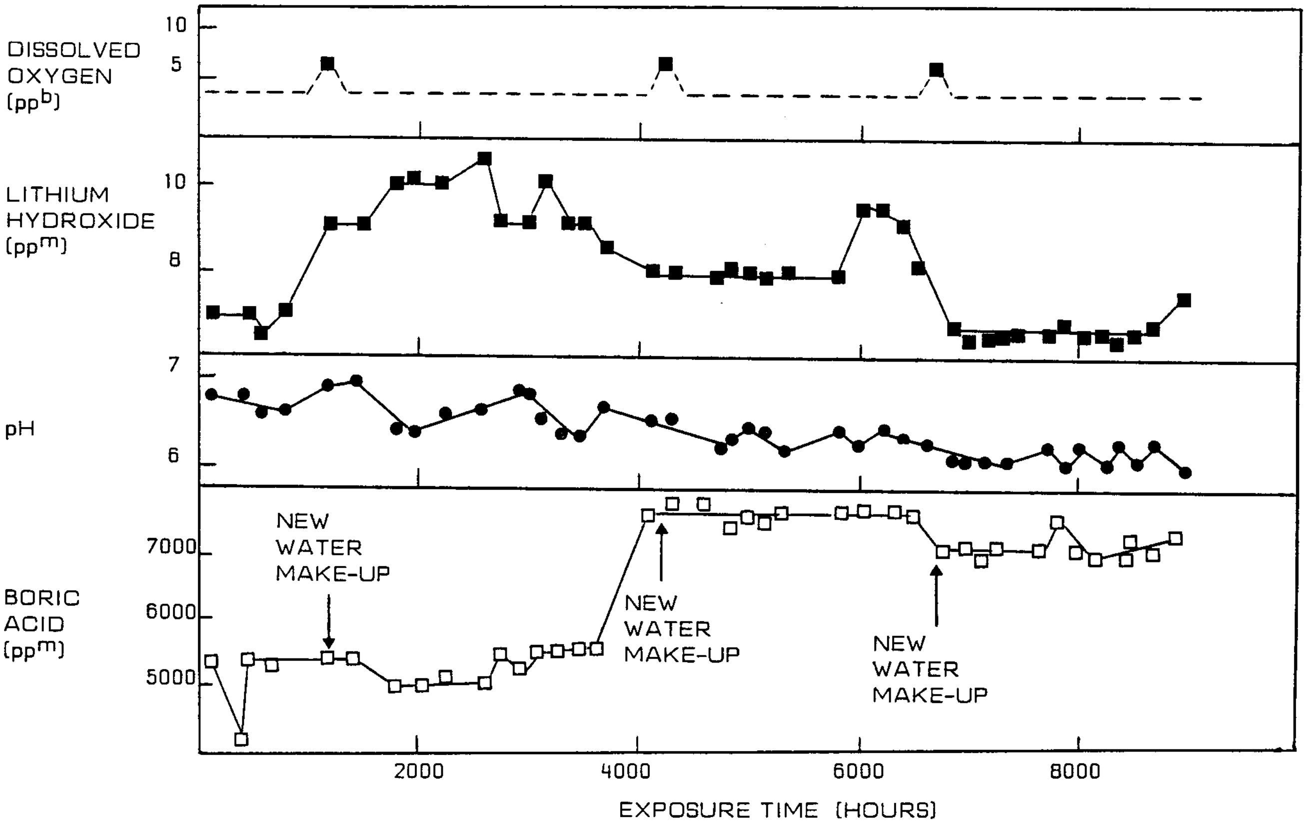

Fig. 5—Details of water chemistry as a function of exposure time (after Bulloch[11])

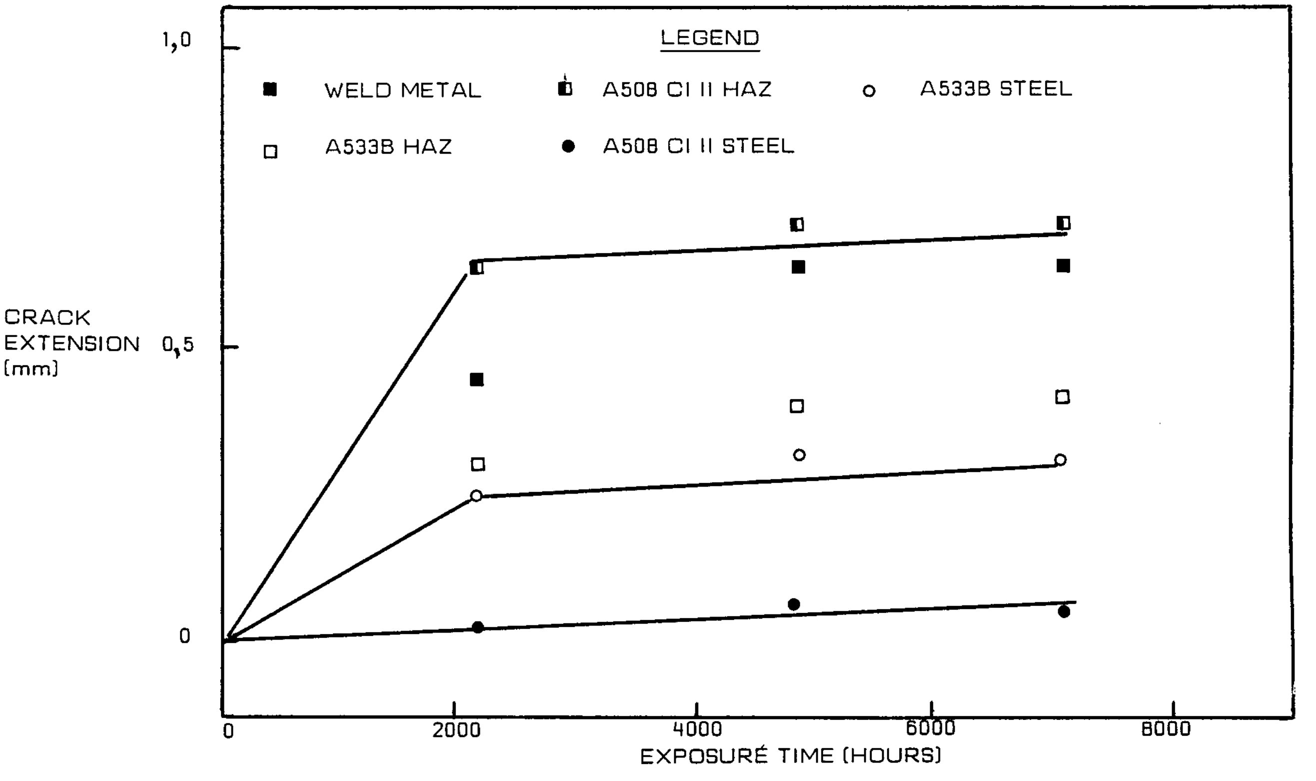

Fig. 6—Results for constant-load specimens of RPV materials exposed to a PWR environment for which the initial loading, *K*, was 40 MPa $\sqrt{m}$ and the crack length was monitored by optical measurement of the side cracks (after Bulloch[11])

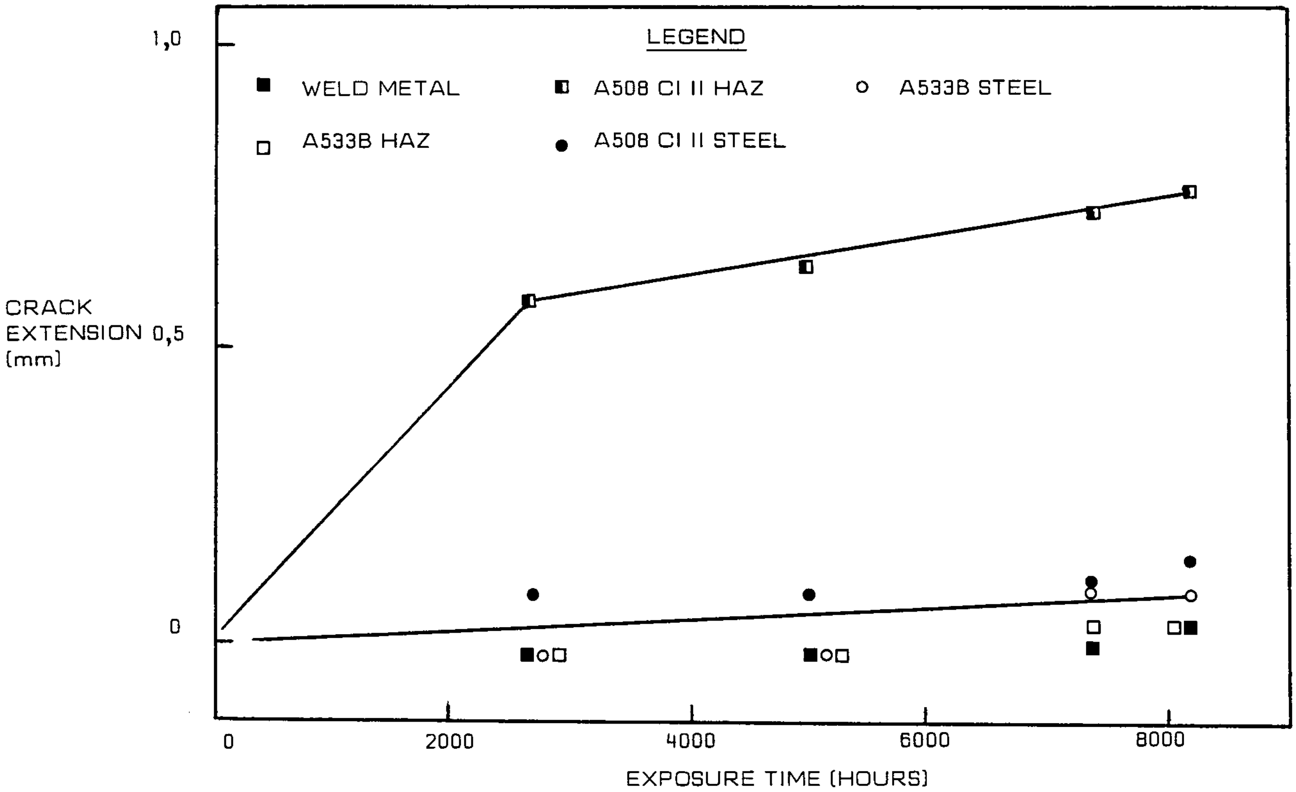

Fig. 7—Results for constant-load specimens of RPV materials exposed to a PWR environment for which the initial loading, *K*, was 60 MPa $\sqrt{m}$ and the crack length was monitored by optical measurement of the side cracks (after Bulloch[11])

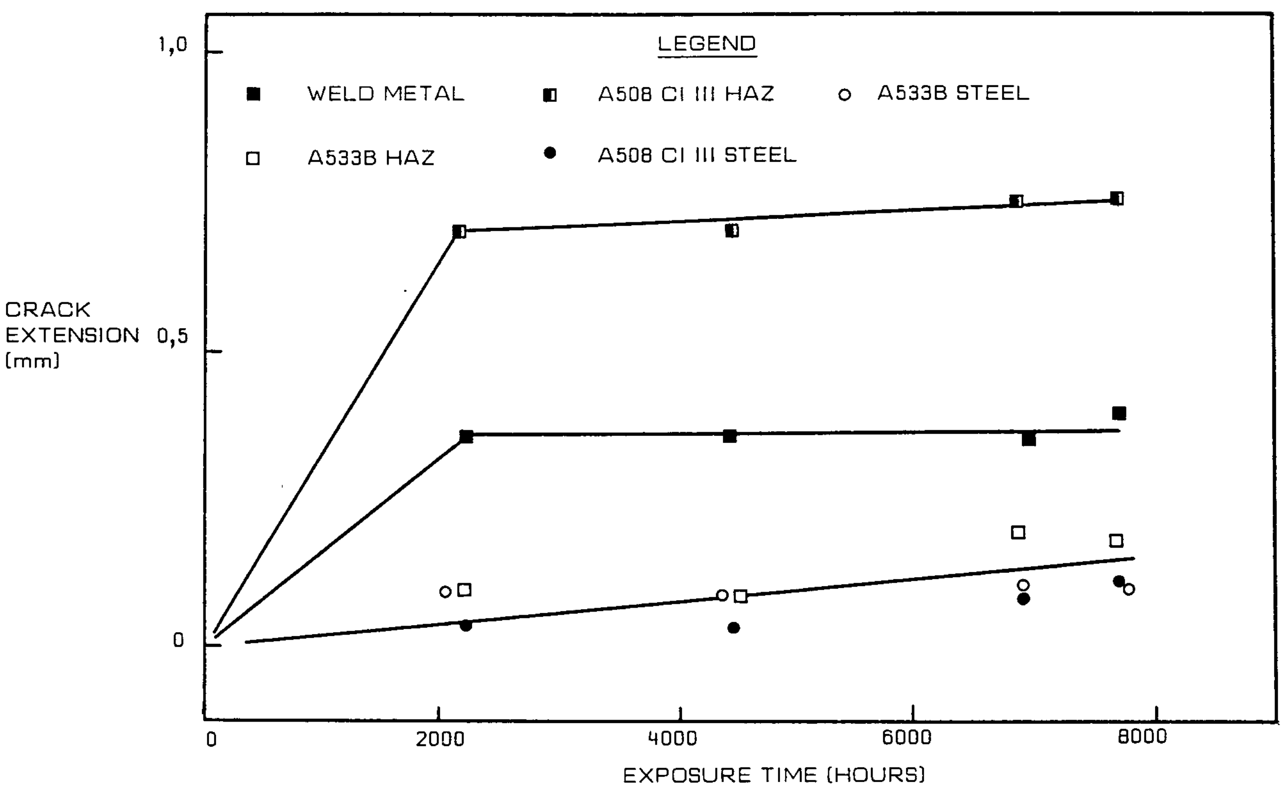

Fig. 8—Results for constant-load specimens of RPV materials exposed to a PWR environment for which the initial loading, *K*, was 85 MPa $\sqrt{m}$ and the crack length was monitored by optical measurement of the side cracks (after Bulloch[11])

(a) With the exception of the data of Bamford *et al.*[8] on plate and HAZ material in a PWR environment, no evidence of significant crack extension was recorded for WOL specimens.
(b) Crack extension was observed in the constant-load SCC tests. However, characteristics of transient crack growth were evident in one instance[12], while in the other it was evident that the side measurements of crack extension were unreliable and over-estimated the amount of crack extension that had occurred during an exposure period.

In constant-displacement SCC tests on WOL, only the work of Bamford *et al.* that was conducted at Westinghouse showed significant crack extension; numerous other studies did not record any crack growth. This discrepancy can be explained in part by the work of Atkinson[9], who examined the fatigue-fracture surfaces of the corrosion-fatigue specimens removed from the Westinghouse rig and found evidence of hematite (Fe_2O_3), which suggests that oxygen contamination could have occurred during testing. As the WOL tests were conducted in the same PWR water as the corrosion-fatigue tests, they too could have been exposed to a dissolved-oxygen level that was higher than that of normal PWR water.

Indeed the effect of dissolved oxygen on the electrochemical potential of ferritic steels is well established, and small quantities (100 p.p.b.) or less can increase the potential by several hundred millivolts so that anodic dissolution is favoured and the stable oxide species is hematite rather than magnetite (Fe_3O_4)[13], which is the usual oxide

Fig. 9—Crack extension of about 0,2 mm in a specimen of weld metal after it had been exposed to a PWR environment with an initial loading, *K*, of 85 MPa $\sqrt{m}$ for 7600 hours

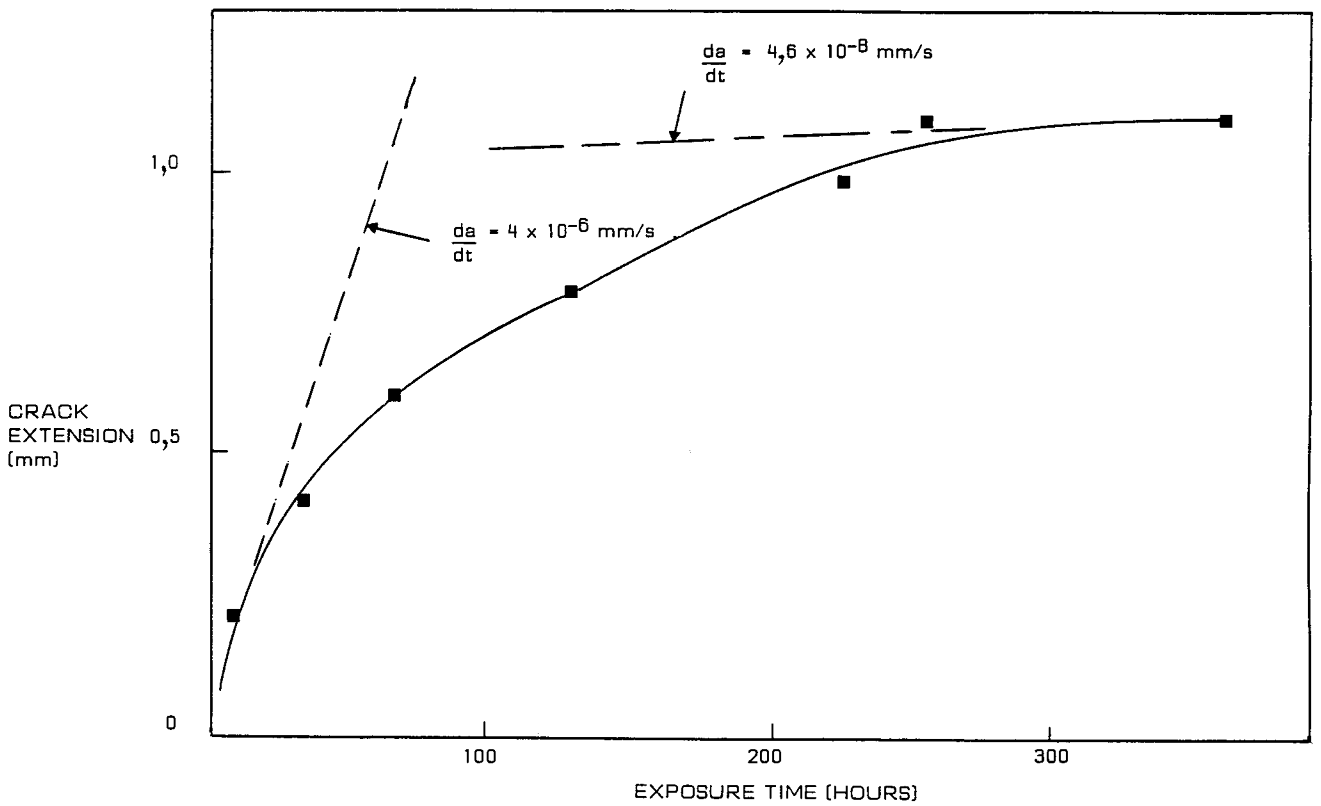

Fig. 10—Results for a constant-load specimen of A533B HAZ material exposed to a PWR environment for which the initial loading, *K*, was 60 MPa $\sqrt{m}$ and the crack length was monitored continuously by a d.c. potential drop method (after Tice[12])

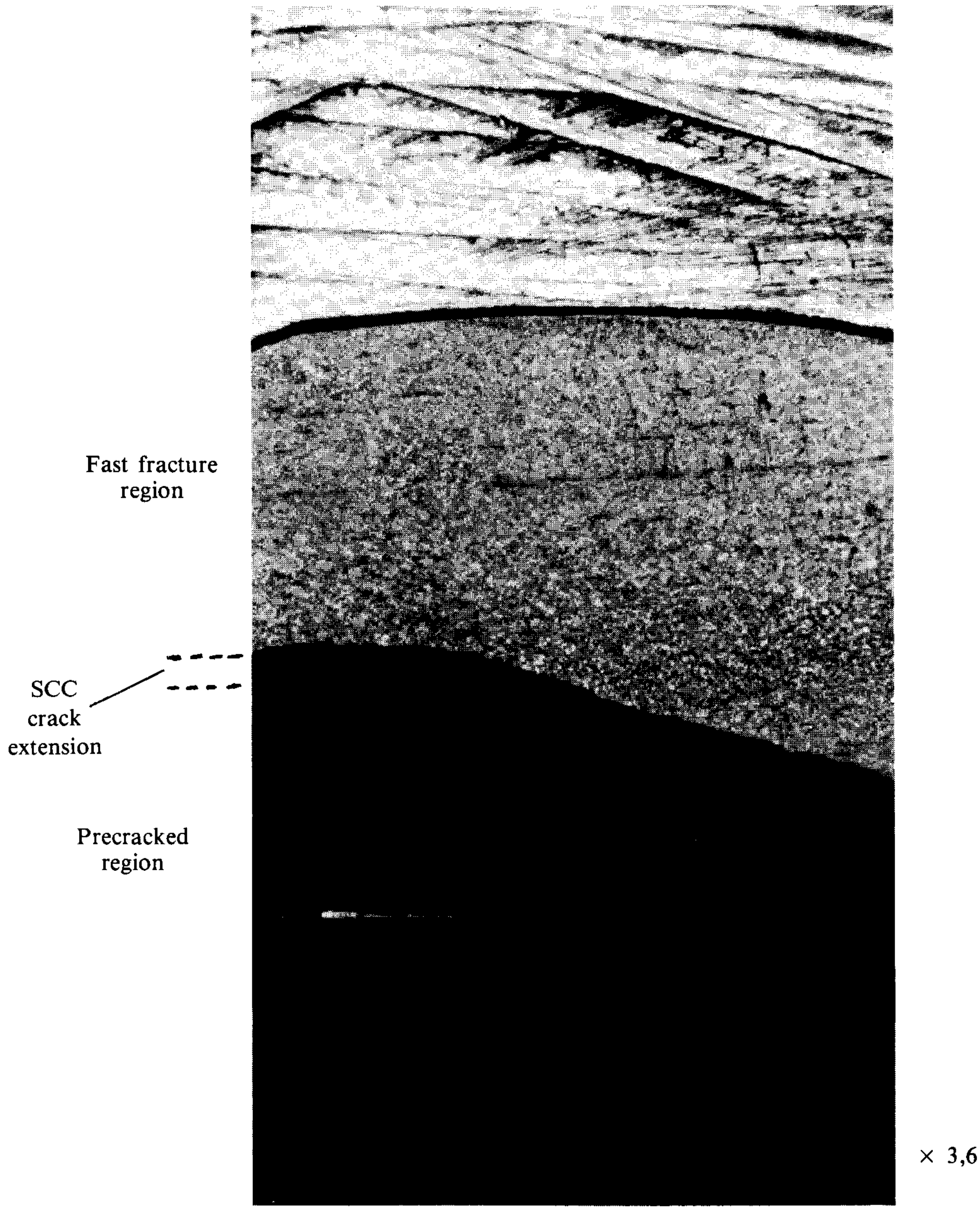

Fig. 11—Details of constant-load crack extension (after Tice[12]), in which 1 mm of growth occurred over the left-hand side of the compact-tension specimen where the precracked length was at a maximum

prevalent in a good low-oxygen PWR-water environment. Under the conditions that result in the formation of hematite, SCC is known to occur in A533B steels[14-16]. As a result, it is possible that the SCC WOL test specimens at Westinghouse could have been exposed at some stage to PWR water that was contaminated with oxygen; this would explain the differences in the test results. The other tests conducted on WOL specimens[10,11] that showed no crack extension were conducted in low-oxygen PWR water. Indeed, in one study[11] the monitoring equipment indicated that the environment contained 10 p.p.b. or less of dissolved oxygen throughout the exposure time of about 7000 hours. This observation was supported by the detection of magnetite on selected test specimens and a short-term measurement of -720 mV (SHE) for the corrosion potential of an A533B test coupon[17].

Generally, with the exception of the Westinghouse results[8], the WOL constant-displacement tests showed no growth while the constant-load tests exhibited some crack extension when exposed to a PWR environment. This difference can be explained in terms of the driving force at the tip of a crack. In a constant-load test, the driving force at the crack tip is continuously re-established during crack growth and the load is always acting at its constant original value. In a constant-displacement test, the load is at its maximum only at the start of the test, and diminishes continuously as the cracks extend. Hence, the crack-tip driving force of a WOL specimen is time-dependent, and is reduced with exposure time in a PWR environment. Such differences in the characteristics of crack tips could have a significant influence in sustaining any cracking.

The results of the constant-load tests, although exhibiting certain differences, show that crack extension occurred in the HAZ materials and the weld metal. The Babcock Power study[11] showed that the side measurements of crack lengths were unreliable, and that the only reliable measurements of crack extension were those from fractographic measurements on broken specimens, i.e. an average of 0,2 mm (0,5 mm maximum) in the HAZ and weld-metal materials, and almost zero in A533B and A508 Cl III steels over an exposure period of some 8000 hours. Hence, the average velocity of crack growth in the HAZ and weld-metal materials was approximately 7×10^{-9} mm/s. These velocity values are based on the assumptions that there was no incubation period, and that growth occurred slowly and continuously over the exposure period of 8000 hours. However, it is possible that there was an incubation period and that crack extension occurred as a result of the unloading–loading sequences.

The experimental data for constant-load specimens reported by Tice[12] (Fig. 11), in which the crack length was monitored continuously, exhibits the significant advantages that this type of approach has over periodic determinations of crack length. The crack-extension behaviour shown in Fig. 10 exhibited an initially high crack velocity of 4×10^{-6} mm/s, which was transient in nature. It should be noted that these data agree well with the crack-velocity results of Bamford *et al.*[8] but did not show any incubation period, whereas the Westinghouse tests required an incubation period of about 1000 hours before crack extension was observed. With increasing crack extension, however, the velocity of growth diminished until, at about 300 hours, the crack velocity compared with those recorded in the Babcock Power study of about 10^{-8} mm/s.

Such transient crack growth is not fully understood at present but could possibly have been due to two factors: the initial level of dissolved oxygen in the vicinity of the crack tip could have been higher than that measured at the water inlet and outlet (5 p.p.b. or less) since the test was started only 22 hours after the autoclave attained the operating temperature of 290°C; and the precracking under PWR conditions could have resulted in creating, during the initial stages of the constant-load test, an aggressive crack-tip environment (possibly through the dissolution of manganese sulphide inclusions[18] encountered during the precracking operation). However, with increasing crack extension during the test, these aggressive factors could either have been flushed out or diluted, causing the velocity of crack growth to diminish. It is probable that the former explanation accounts better for the velocity of transient cracks because the initial crack velocity agrees well with the Westinghouse data, which could have been generated in oxygen-contaminated PWR water.

It is evident from the above discussion that the nature of SCC extension in a PWR environment is not well understood at present and, indeed, cracking could occur in at least four different ways:

(1) initial high velocity of crack growth that continuously diminishes with crack extension, i.e. zero incubation period,
(2) high, sustained velocity of crack growth that occurs after a significant incubation period,
(3) slow, constant velocity of crack growth that is continuous over the whole exposure period, i.e. zero incubation period, and
(4) crack extension that occurs only during the unloading–loading cycle, i.e. zero crack growth during the constant-load test period.

Schematic details of these various characteristics of crack extension are shown in Fig. 12.

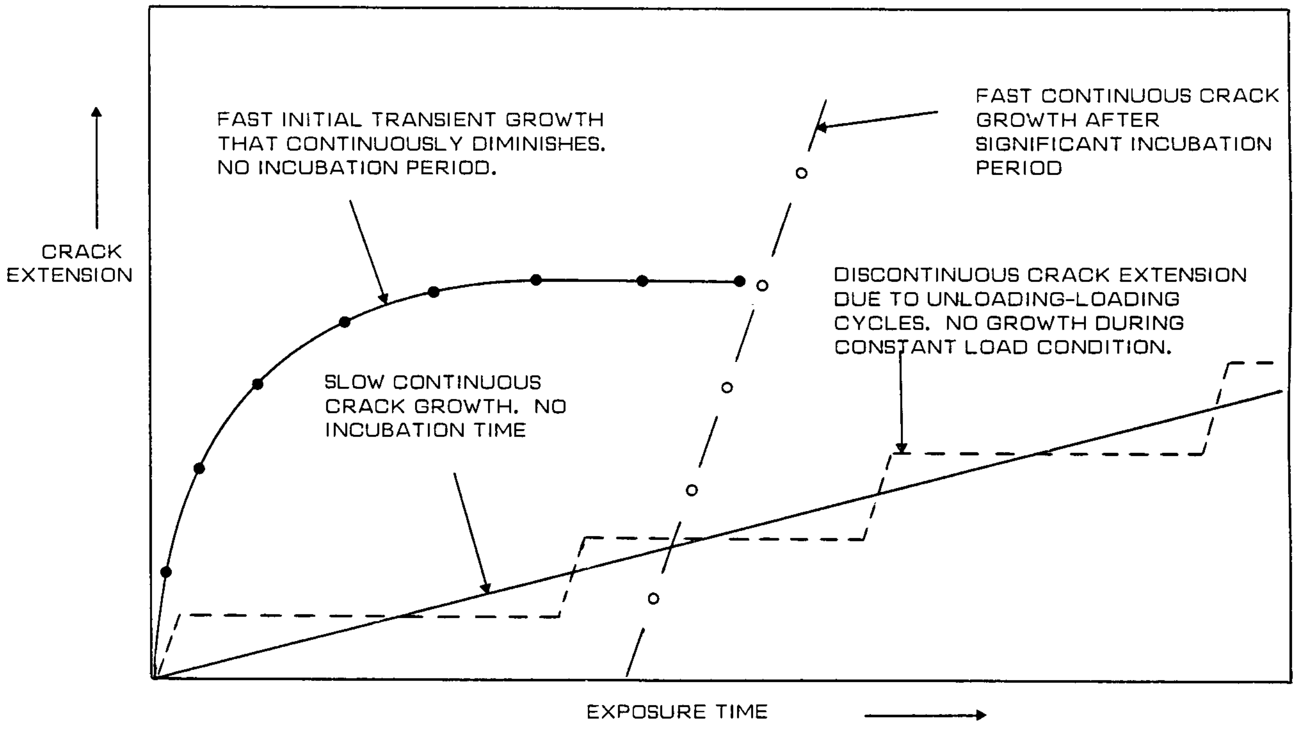

Fig. 12—Schematic illustration of the various characteristics of SCC in RPV materials exposed to a PWR environment

Finally, it should be noted that the experimental data were generated in a number of separate studies, each with its own experimental procedure. Such experimental differences led to some difficulty in the interpretation of the data, and it is clear that there is a need for a consistent code of experimental procedures in the testing of the SCC behaviour of RPV materials in PWR environments.

Summary and Conclusion

From a consideration of the various experimental conditions under which the data were obtained, it is clear that only a few conclusions can be drawn because the different experimental techniques have a significant effect on crack growth.

The results from the constant-displacement WOL tests, with the exception of one study, showed very little or no crack extension during exposure periods of up to 14 000 hours. Although the constant-load tests gave evidence of crack extension, the nature and extent of the crack extension are still unclear. When crack extension did occur, it was exclusively transgranular in nature.

Certain differences in crack-extension results may be due to differences in the crack-tip driving force between test specimens and in the level of dissolved oxygen in the crack-tip enclave.

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Mining machinery

The second International Conference on Mining Machinery will be held in Brisbane, Australia, from 8th to 11th May, 1988. This is a sequel to the extremely successful international conference held in 1979, at which a record of 89 papers were presented and over 600 delegates attended.

The mining industry worldwide operates in a very demanding, cost-competitive environment. With continuing pressures for achieving more economic and more efficient production, the industry must rely increasingly for survival on improvements available through mechanization. This Conference will provide a forum where the manufacturers, designers, and end-users can meet to discuss current problems, likely future requirements, and development trends.

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