The Effects of Cathodic Charging on the Acoustic Emission Generated by Intergranular Cracking in Sensitized 304 Stainless Steel

STEVE H. CARPENTER and DANIEL R. SMITH, Jr.

It has been reported that the mode or type of crack propagation (transgranular, intergranular, or microvoid coalescence) in 304 stainless steel could be identified by analysis of the acoustic emissions generated during crack propagation. Different crack propagation modes were produced in double cantilever beam specimens of 304 stainless steel using combinations of heat treatments and cathodic charging. The acoustic emission generated was measured, analyzed, and correlated with metallographic results. Intergranular cracking or separation was observed in three experimental conditions: (1) sensitized samples tested in air at room temperature, (2) sensitized samples which were cathodically charged prior to testing in air at room temperature, and (3) sensitized samples which were simultaneously and continuously cathodically charged while being tested at room temperature. The particular acoustic emissions generated by intergranular separations were identified by careful analysis of the acoustic emission waveforms. The amount of intergranular cracking and the acoustic emissions detected were found to be strongly dependent on the experimental test conditions. The amplitude, duration, and, hence, the energy carried in the waveforms of the emissions from intergranular separations were found to decrease dramatically when there was a constant supply of hydrogen, *i.e.*, during continuous cathodic charging. The results are consistent with the lowering of the cohesive energy along the grain boundary; however, other mechanisms are also plausible.

I. INTRODUCTION

THE purpose of this paper is to report on the acoustic emissions generated from intergranular crack separations in sensitized and cathodically charged 304 stainless steel. Acoustic emissions are the transient elastic waves generated by the rapid release of energy within a material. In many materials, the advance of a crack is an excellent source of acoustic emission. Acoustic emission technology and techniques have been used for many years as a tool to study and investigate the fundamental features of crack initiation and growth as well as a practical tool to locate and characterize cracks and flaws in structures under test.[1,2,3] Recently, it was reported that the mode or type of crack propagation in double cantilever beam (DCB) specimens of 304 stainless steel could be determined by analysis of the acoustic emissions produced during crack growth.[4] This paper will present data on the effects of cathodic charging on the acoustic emission generated from intergranular separations in sensitized 304 stainless steel. The acoustic emissions generated from intergranular separations in samples exposed to hydrogen as well as from samples which were not exposed to hydrogen have been detected, characterized, and analyzed. The presence of hydrogen was found to have a pronounced effect on the amount and magnitude of acoustic emission generated per intergranular separation. A decrease of over two orders of magnitude in the acoustic energy released per grain separation was measured when there was a continuous supply of hydrogen. The results obtained are easily explained by the presence of hydrogen causing a lowering of the cohesive strength at the grain boundary. However, it is also possible to explain the results as a buildup of internal stress due to the presence of hydrogen. Hence, in this investigation it is impossible to unambiguously identify the mechanisms responsible for the observed changes in acoustic emission due to cathodic charging.

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II. EXPERIMENTAL PROCEDURE

The acoustic emissions generated during crack propagation were detected and characterized using standard, commercially available equipment. A schematic of the equipment used is shown in Figure 1. A Dunegan S9204 piezoelectric resonant transducer (resonant frequency approximately 140 kHz) was used in conjunction with a broadband (0.02 to 2 MHz) preamplifier at 40 dB gain. A Physical Acoustics Corporation (PAC) model 3104 acoustic emission system was used to acquire common time domain parameters of the measured acoustic emissions. The PAC system was also used to record the parametric inputs of applied load and clip gauge displacement. Signals from the preamplifier were digitized using a LeCroy model TR8837F transient recorder and stored on magnetic disks for later analysis. Both the PAC and the LeCroy data were analyzed using software programs developed in the ASYST programming environment. Data from individual waveforms, such as amplitude, are referenced to the transducer and have been corrected for noise and gain.

Crack propagation was achieved using DCB specimens fabricated from a rolled sheet of commercial grade

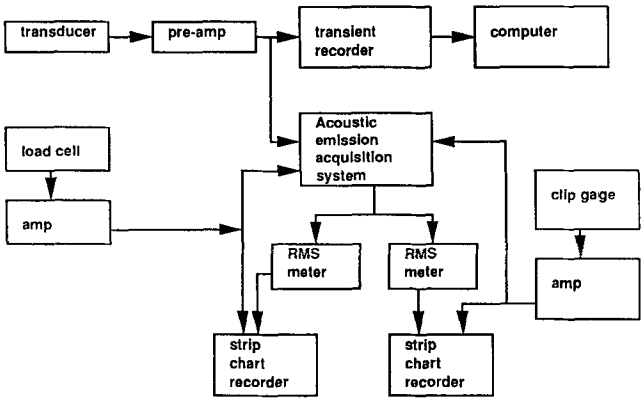

Fig. 1—Schematic of equipment used.

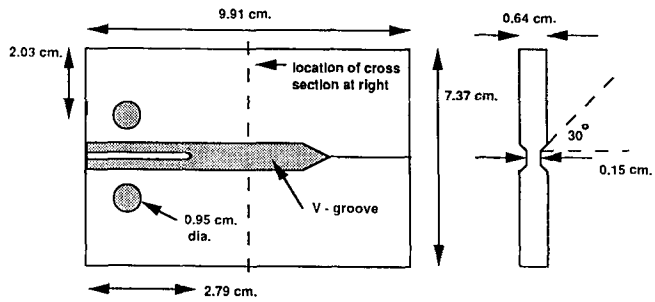

Fig. 2—Schematic of DCB samples used.

304 stainless steel. The chemical composition of the sheet used is given in Table I. Rectangular blanks were cut from the rolled sheet and were then given a sensitizing heat treatment. The blanks were heat-treated at 1373 K for 1 hour, water quenched at 923 K for 24 hours, and furnace cooled.[5] The heat treatment produced an equiaxed grain structure having an average grain diameter of approximately 64 μm (ASTM No. 5). Following heat treatment, the blanks were machined into DCB specimens shown schematically in Figure 2.

In the present investigation, three basic test conditions were used to produce intergranular separations: (1) sensitized samples tested at room temperature in an air environment, (2) sensitized samples which had been cathodically charged prior to testing in air at room temperature, and (3) sensitized samples which were cathodically charged while being tested at room temperature. Details of the different crack modes observed in the three experiments are given in Table II.

All tests were carried out at a constant crosshead displacement rate of 0.025 mm/min. The charging solution used was 1 N H_2SO_4 with traces of As_2O_3 and CS_2 to prevent hydrogen recombination.[6] A variety of charging times and current densities was used; however, the acoustic emission produced did not seem to be strongly dependent on these parameters. Charging times ranged from 2 to 72 hours with current densities ranging from 50 mA/cm^2 to 4 A/cm^2. The threshold of the PAC acoustic emission system was set just above the noise level produced by hydrogen gas evolution and was held constant for all tests. The transducer was acoustically coupled to the sample using VASELINE* petroleum jelly.

*VASELINE is a trademark of Chesebrough-Ponds, Inc., Greenwich, CT.

Preliminary tests showed no evidence of degradation of the coupling due to the presence of the charging solution. The breaking of a 0.3-mm-diameter, 2H Pentil pencil lead was employed to calibrate the system before each test.[7] A complete detailed description of the experimental parameters and procedures is given in Reference 8.

III. RESULTS

Intergranular separations were produced in the three experimental conditions discussed above. Although the measured acoustic emissions included emissions from other sources, it was possible to determine the particular emissions coming directly from intergranular separations.[4] The primary techniques used to identify the emissions generated from the intergranular separations were relative values of the PAC acoustic energy (see discussion concerning this parameter below) and the power spectrum of the emission waveforms. The unique features of the power spectra of the emissions from intergranular separations were unchanged in all three experimental conditions. The PAC acoustic energy (a name provided by the equipment manufacturer) is not a true measure of the energy in the acoustic emission waveform. It is a digital measure used by the PAC equipment that approximates the real energy. Data and results from each of the three experimental test conditions are given below.

A. *Test Condition 1*

Figure 3 shows a plot of the acoustic emission generated and the applied load as a function of clip gage displacement for a sensitized sample tested in air at room temperature with no cathodic charging. The acoustic emission is reported in terms of the PAC acoustic energy. Examination of the fracture face revealed that the crack growth proceeded by microvoid coalescence and

Table I. Chemical Composition of the Commercial 304 Stainless Steel Used in This Investigation

S	C	Si	Mn	Cr	Ni	P	Mo	Fe
0.021	0.073	0.47	1.8	19.7	8.5	0.037	0.20	remainder

Note: Amounts are given in weight percent.

Table II. Details of Testing Conditions and the Crack Modes Produced

Test Condition	Cathodic Charging	Testing Conditions	Crack Modes Observed
1	no charging	air, room temperature	microvoid coalescence and intergranular separations
2	charged prior to testing	air, room temperature	microvoid coalescence and intergranular separations
3	charged while testing	air, room temperature, cathodic charging	intergranular separations

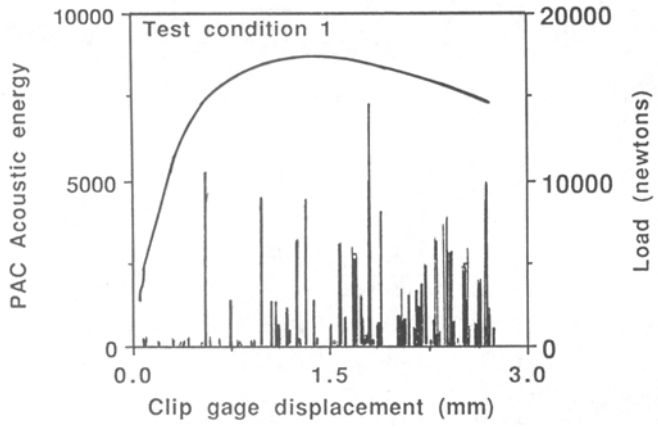

Fig. 3—PAC acoustic energy and load *vs* the clip gage displacement. Test condition 1.

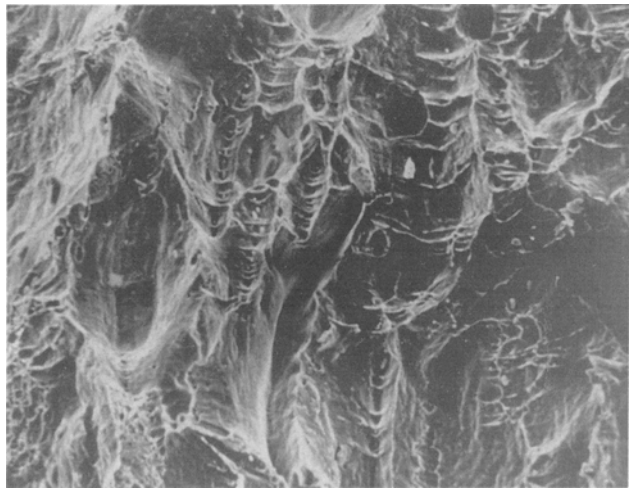

Fig. 4—SEM micrograph of the crack surface in a condition 1 test. Magnification 544 times.

with a small number of intergranular separations perpendicular to the crack interface. Figure 4 is a scanning electron microscopy (SEM) micrograph of a typical fracture face. Figure 5 shows the intergranular separations normal to the interface shown in Figure 4. Two sources of acoustic emission were identified, the fracture of inclusions on or near the crack interface and intergranular separations. The number of acoustic emission bursts believed to be due to intergranular separations correlated well with the number of intergranular separations measured by careful metallographic examination both along and perpendicular to the fracture face. For the particular test shown in Figure 3, 44 bursts of acoustic emission were believed to come from intergranular separations, and 52 intergranular separations were measured. In all of the condition 1 tests, the correlation was similar, with the number of intergranular separations measured slightly exceeding the number of acoustic emission bursts believed to be due to intergranular separations. Analysis of the waveforms associated with intergranular separations revealed an average amplitude of 5.43 ± 1.09 mV and an average duration of 847 ± 25 μs. These data give the values measured at the transducer, having been corrected for both gain and noise.

B. *Test Condition 2*

Figure 6 shows typical acoustic emission and applied load data for a sensitized sample that was cathodically charged at a current density of 1 A/cm^2 for 6 hours immediately prior to testing. The major difference from the data shown in Figure 3 is that there is considerably more acoustic emission. Figure 7 is an SEM micrograph of a typical fracture face showing that the crack propagated by a combination of microvoid coalescence and intergranular separations. Again, two sources of acoustic

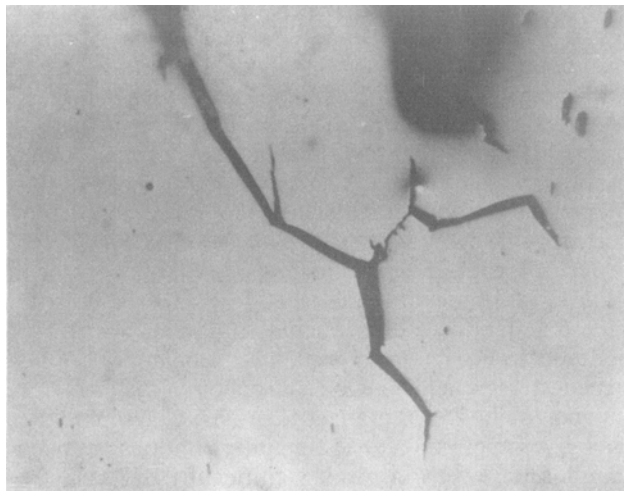

Fig. 5—Optical micrograph showing intergranular separations just below and perpendicular to the main crack surface. Magnification 213 times.

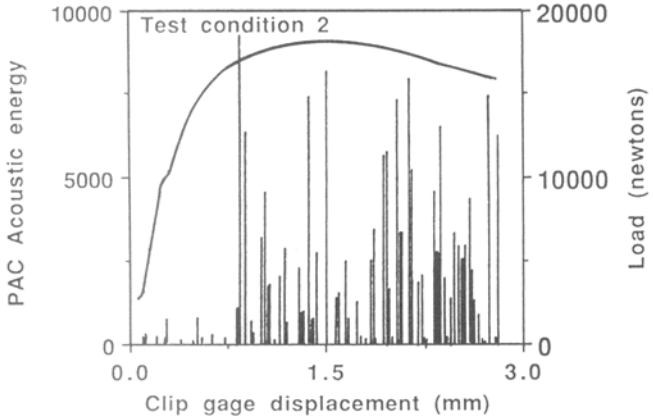

Fig. 6—PAC acoustic energy and load *vs* the clip gage displacement. Test condition 2.

emission were identified, fractured inclusions on and near the crack interface and intergranular separations. The number of fractured inclusions was consistent with that observed in the noncharged samples; however, the number of intergranular separations was found to increase as a result of the prior cathodic charging. For the test shown in Figure 6, 132 intergranular separations were found and 115 bursts of acoustic emission were identified as coming from intergranular separations. The ratio of emissions associated with intergranular separations to the number of intergranular separations measured by metallography was nearly identical to that measured for samples which had not been charged. Neither the value of the current density (50 mA/cm^2 to 4 A/cm^2) nor the time of charging (2 to 72 hours) had a significant effect on the detected acoustic emission. The average amplitude of the waveforms for emissions due to intergranular separations was 7.13 ± 0.99 mV. The average duration of the waveforms was found to be 873 ± 15 μs.

C. *Test Condition 3*

Figure 8 provides typical acoustic emission and applied load data for a sensitized sample that was cathodically charged at a current density of 100 mA/cm^2 during testing. There appears to be less total acoustic emission than observed in the other conditions. Figure 9 is an SEM micrograph of the crack face for the test shown in Figure 8. No fractured inclusions could be found on or near the crack interface; however, small amounts of transgranular cleavage were observed. Clearly the major mode of crack propagation is intergranular cleavage. The applied load necessary to maintain crack propagation is considerably lower, as would be expected. A careful analysis of the acoustic emission revealed that for the data shown in Figure 8, there were 458 actual emissions which were believed to be due to intergranular separations. For the same test, a metallographic examination of the crack faces found a total of 5670 grain separations. Combining these data gives approximately 12 grain separations per acoustic emission event, a result significantly different from that observed in the other experimental test conditions. The acoustic emission waveforms were found to have a significantly lower amplitude and duration, the average

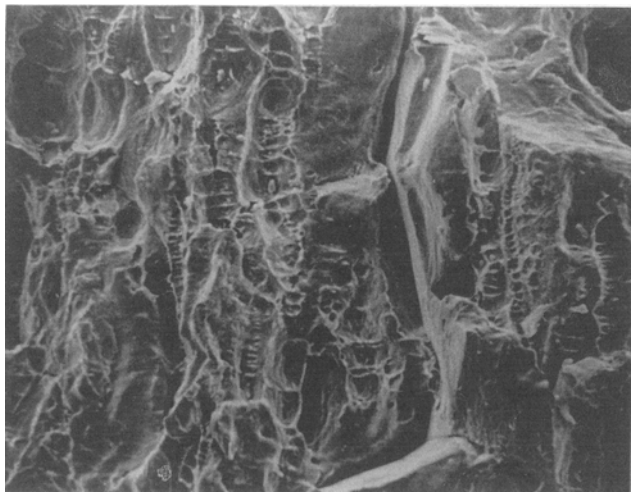

Fig. 7—SEM micrograph of the crack surface in a condition 2 test. Magnification 544 times.

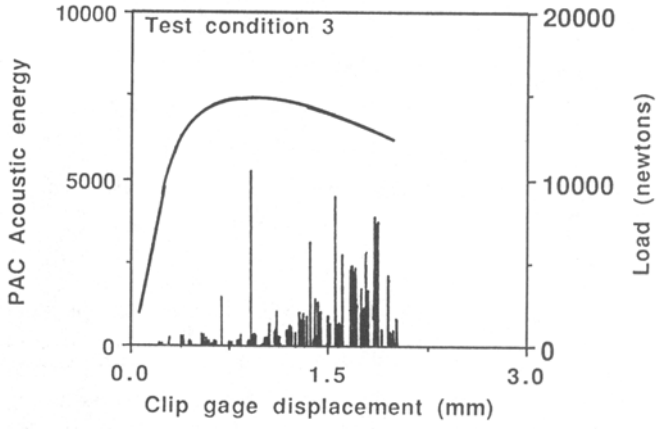

Fig. 8—PAC acoustic energy and load *vs* the clip gage displacement. Test condition 3.

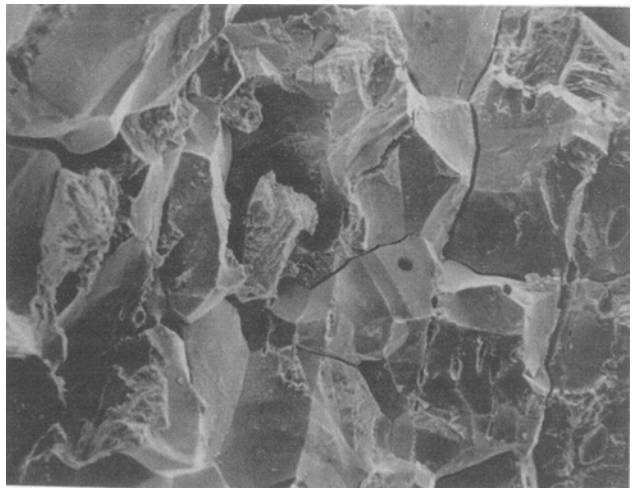

Fig. 9—SEM micrograph of the crack surface in a condition 3 test. Magnification 272 times.

amplitude being 0.74 ± 0.10 mV and the average duration being 287 ± 8 μs.

IV. DISCUSSION

The continuous cathodic charging, which supplies a continuous source of hydrogen, caused a significant reduction of the mechanical properties, a change in the major mode of crack propagation, and a reduction in the acoustic emission from intergranular crack separations in 304 stainless steel. Cathodic charging prior to testing had only a slight effect on the detected acoustic emission. Since the energy carried in the acoustic emission waveform depends directly on both the amplitude and duration of the waveform, an examination of the energy provides an excellent way of determining the effects of cathodic charging. Unfortunately, with our equipment, it was impossible to obtain an exact measure of the energy carried in each of the waveforms, *i.e.*, the waveform squared and integrated over time. However, a fairly close approximation to the energy can be obtained by the following equation:

$$E = \frac{[\text{peak amplitude}]^2}{3} \times [\text{duration}] \qquad [1]$$

Using the acoustic emission data for amplitude and duration of individual waveforms for tests shown in Figures 3, 6, and 8, it is possible to calculate both the total and average energy per acoustic emission (AE) burst according to Eq. [1] for each of the three experimental conditions. The total energy divided by the number of intergranular separations determined by metallographic examination gives an "energy per grain separation" for the three test conditions as well. The experimental data are shown in Table III.

The results clearly show the effects of continuous cathodic charging. Samples which were continuously cathodically charged have an energy per intergranular separation which is two orders of magnitude less than that observed for the other test conditions. Figure 10 graphically demonstrates the differences in the energy per intergranular separation for the three test conditions. To determine if a significant amount of attenuation was due to the charging solution itself, lead breaks (0.3-mm-diameter, 2H Pentil pencil lead) were made at various locations on the test specimen both in and out of the charging solution. Only minor differences were observed as a result of the presence of the charging solution.[8] Recently published work where a similar transducer was used has reported the energy released from a number of different acoustic emission sources.[9,10,11] A comparison

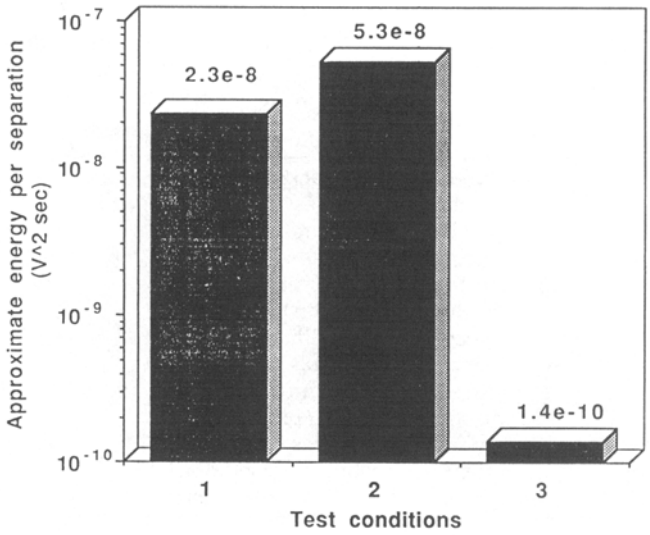

Fig. 10—Approximate acoustic energy per intergranular separation for different test conditions.

of the energy released per acoustic emission event is instructive and allows for an evaluation and comparison of the energy released by intergranular separations. Energies per acoustic emission signal for a variety of sources are provided in Table IV.

The largest values of energy per acoustic emission signal occur for the intergranular separations in conditions 1 and 2. For conditions 1 and 2, each acoustic emission signal correlates with one grain separation. It is reasonable to expect that a grain separation would release more energy than any of the other sources in Table IV. However, an acoustic emission signal in condition 3 correlates with more than 12 grain separations and yet gives an energy comparable with the fracture of a 300-μm boron particle.

Nozue and Kishi[12] have shown that the energy released by acoustic emissions from intergranular crack propagation in 4340 steel was directly proportional to the surface area of crack motion measured for each acoustic emission burst. The data for samples subjected to continuous charging show the opposite behavior. The data indicate a considerable increase in the number of separations per acoustic emission burst (from 1 to more than 12), yet there is an enormous decrease in the average energy per grain separation. A possible and consistent explanation for the observed data could be the lowering of the cohesive strength along the grain boundaries by the presence of hydrogen. The lower cohesive strength would require less stress to propagate the crack, and less

Table III. Listing of Acoustic Emission Data and Approximate Energy Data for Tests Shown in Figures 3, 6, and 8

Test Condition	Maximum Applied Load (N)	Average Amplitude per AE Burst (mV)	Average Duration per AE Burst (μs)	Average Energy per AE Burst (V^2 s)	Number of Separations per AE Burst	Energy per Separation (V^2 s)
1	19,075	5.43 ± 1.09	847 ± 25	$2.8 \pm 1.6 \times 10^{-8}$	1.18	$2.3 \pm 1.4 \times 10^{-8}$
2	18,988	7.13 ± 0.99	872 ± 15	$6.1 \pm 2.3 \times 10^{-8}$	1.15	$5.3 \pm 2.0 \times 10^{-8}$
3	17,853	0.74 ± 0.10	287 ± 8	$1.7 \pm 1.1 \times 10^{-9}$	12.50	$1.4 \pm 0.1 \times 10^{-10}$

Table IV. Listing of Measured Average Energy per Acoustic Emission Event for a Number of Different Acoustic Emission Sources

Acoustic Emission Source	Average Energy per Acoustic Emission Signal (V^2 s)	Reference
Fracture of boron particles in a 2219 aluminum matrix	1×10^{-9}	9
Reverse thermoelastic martensitic phase transformation in Au-47.5Cd	4.3×10^{-9}	9, 10
Forward transformation in Au-47.5Cd	2.5×10^{-11} (largest)	9, 10
Avalanche motion of dislocations in powder source beryllium	2×10^{-11}	9, 11
Avalanche motion of dislocations in ingot source beryllium	2×10^{-12}	9, 11
Twin formation in uranium	2.6×10^{-9}	9
Intergranular separations (condition 1)	2.8×10^{-8}	this study
Intergranular separations (condition 2)	6.1×10^{-8}	this study
Intergranular separations (condition 3)	1.7×10^{-9}	this study

energy would be released when the grain separated. The continuous supply of hydrogen would allow for the process to repeat as the crack advanced. Such models have been postulated in the literature.[13] However, other models have been postulated which propose that the continuous supply of hydrogen causes a buildup of internal pressure sufficient to produce voids and microcracks.[13] The data can also be explained by this type of a model. In this case, the initial advancement of the crack front would cause only a fraction of a grain separation. Since only a fraction of the grain separated, only a fraction of the energy would be released. In a similar manner, the buildup of internal stresses from the supply of hydrogen could cause the crack to open and grow slowly (producing no acoustic emission) until critical conditions had been met for the crack to propagate. This type of mechanism would also give a lower energy release.

An examination of the data in Table III for test conditions 1 and 2 shows that the acoustic emission parameters (amplitude, duration, and energy) are larger for condition 2 than for condition 1. The differences in magnitude are not great, and the data overlap when the error bars are included. At present, it is not known if the differences are meaningful or simply the result of too small a data base.

V. CONCLUSIONS

The acoustic emissions due to intergranular separations in sensitized and cathodically charged commercial grade 304 stainless steel have been measured and characterized. Testing of DCB specimens in three separate test conditions was carried out. In each of the three test conditions, intergranular separations occurred and it was possible to identify the emissions produced from the intergranular separations. Excellent correlation between the acoustic emission data and metallographic analysis was obtained. Continuous cathodic charging of a sensitized 304 stainless steel sample had a pronounced effect on the acoustic emissions generated by intergranular separations. Analysis of the acoustic emission waveforms indicated a significant decrease in both the amplitude and duration when a continuous supply of hydrogen was present. Using the waveform data, an approximate calculation of the energy carried in the waveform was possible. A comparison of energies per acoustic emission event from other sources demonstrates that the energy calculations given for the 304 samples are consistent and of the right order of magnitude. Combining the energy measurements with metallographic data, a calculation of the energy released per grain separation could be made. Data for the three test conditions showed that the energy released per grain separation is at least two orders of magnitude less when there is a continuous supply of hydrogen (continuous cathodic charging during testing) than when there is no cathodic charging or when charging is carried out before testing. The acoustic emission can be explained equally well by the presence of hydrogen causing either a lowering of the cohesive strength along the grain boundary or a buildup of internal stresses within the material.

ACKNOWLEDGMENT

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