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METALLURGICAL AND PROTECTIVE COATINGS

FAILURE DURING THERMAL CYCLING OF PLASMA-SPRAYED THERMAL BARRIER COATINGS*

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The thermal cycling behavior of plasma-sprayed ZrO_2–12wt.%Y_2O_3 coatings was studied. Coatings were produced with and without bond coats of Ni–Cr–Al–Zr and in some cases the substrates were heated to above the optimum temperature prior to spraying. The coatings (attached to the substrate) were thermal cycled to 1200°C and their cracking behavior was followed by acoustic emission (AE) techniques. It was possible to examine the failure mechanisms by statistical analysis of the AE data and to evaluate the influence of preheating and bond coating. It is shown that the AE spectrum changes when a bond coat is used because of the presence of microcracks which, in turn, dissipate energy and improve the coating integrity. The preheating effect is reflected by a decrease in the peak count rate and an increase in the temperature at which AE activity is initiated.

1. INTRODUCTION

Early studies of plasma-sprayed coatings stressed microstructural features[1,2] and mechanical properties[3,4] in order to characterize coating performance. More recently, the focus of mechanical properties has turned to fracture mechanics[5], statistical analysis[6] and failure mechanisms[6*"].

A major problem in the development of thermal barrier coatings is establishing their material properties and correlating these properties with the fracture behavior. Coating failure is often classified as adhesive or cohesive, there generally being little indication of the specific failure mechanism, *i.e.* the description of fracture morphology does not per *se* indicate the mechanism(s) of failure. Mechanistic studies of coating failure will aid in an understanding of coating integrity and thus are expected to lead to improvements in future thermal barrier coating systems.

Acoustic emission experiments carried out during the thermal cycling of plasma-sprayed coatings are described in this paper. In this way the progressive failure of coatings was ascertained.

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In *situ* evaluation during mechanical and thermal tests was carried out by analyzing acoustic emission (AE) spectra (*i.e.* count rate data as a function of time). The mechanical studies were based on tensile adhesion tests[8] and showed that coatings which fail at high tensile stress also exhibit a high AE count. More detailed analysis[9] was employed to relate the time dependence of count rate distribution to the fracture morphology.

In another AE study carried out during thermal cycling of coatings[10] it was found that the total AE count changed with respect to specimen temperature and that these trends could be correlated with coating integrity. However, in that study edge effects in the planar sample geometry may have masked failure processes for the bulk of the coating. Also, the mild steel substrate was not a typical high temperature alloy and thus the temperature limit of the tests was 650 °C; otherwise oxidation-related effects of the substrate would have generated high AE activity. More importantly, it is considered that the count rate distribution (rather than total counts) is more indicative of failure phenomena, since analysis of these distributions has proven valuable in interpreting AE events resulting from tensile adhesion tests[9,11].

2. EXPERIMENTAL DETAILS

Coatings were prepared in a manner that maximized differences in their AE behavior. For example, substrates which are overheated prior to plasma spraying are known to produce coatings with short lifetimes compared with the "optimally" processed substrate. Also, it is well known that bond coating greatly enhances coating lifetime. In this manner coatings of yttria-stabilized zirconia (YSZ)(ZrO_2–12wt.%Y_2O_3 0.04 cm thick) were deposited onto the grit-blasted substrate with and without a bond coat (Ni–Cr–Al–Zr 0.01 mm thick) such that "good" and "bad" coatings could be produced. The Udimet-700 substrate rod was 35.6 cm long and 1.25 cm in diameter and both ends were paper ground to a smooth finish. Coatings 2.5 cm wide were plasma sprayed 7.5 cm from one end of the bar which was supported in a cantilever fashion from the opposite end. The AE transducer was attached using a suitable high temperature couplant and spring arrangement. A 20 kW tubular furnace was employed together with a programmed temperature controller which allowed a nominal heating and cooling rate of 5.5 °C min^{-1}. The specimen was normally cycled (for up to seven times) to 1200 °C and the trends in the AE count rate and the total AE counts were measured. The AE arrangement incorporated a Dunegan–Endevco modular system (3000 series) to amplify the signal from a 0.1–0.5 MHz transducer for a total system gain of 96 dB. The AE count rate data were subsequently digitized and computer processed. A schematic representation of the experimental arrangement is shown in Fig. 1.

3. EXPERIMENTAL RESULTS

The AE was generated on cooling, the count rate being measured over 1 min intervals with respect to time (and thus temperature). The gain of the AE instrumentation was adjusted so that no counts were generated during the thermal cycling of the uncoated substrate. Thus the AE activity arose from processes which

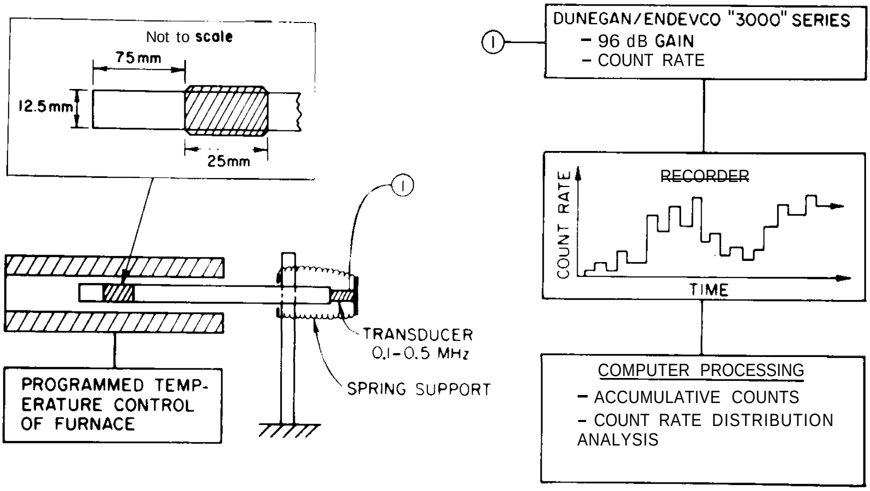

Fig. 1. Experimental arrangement.

depended only on the coating behavior. It should be emphasized that these data were manipulated in several analyses to discern different trends.

The AE spectra of specimens prepared in exactly the same manner showed the same trends; however, the detailed shapes of the response curves were not identical. It should be noted that the specimens showed significant AE activity only on cooling (Fig. 2).

3.1. Cumulative acoustic emission data

3.1.1. Effect of pretreating the substrate

Preheating the substrate resulted in high AE activity during the initial cooling period for all the coatings (Figs. 2(b) and 2(d)). For example, the bond-coated samples exhibited AE activity immediately upon the start of cooling (*i.e.* at 1200 °C) only when the substrates were preheated. However, the "optimally" sprayed bond-coated specimens showed activity only at temperatures below 320–420 °C (Fig. 2(c)).

The samples with no bond coat showed similar behavior, except that AE activity was recorded immediately on the cooling portion of the cycle for both previous heat treatment conditions (Figs. 2(a) and 2(b)). The preheated substrate exhibited more counts in the early part of the cooling cycle (above 440 °C), however, and both samples, as in the case of the preheated sample with a bond coat, showed a distinct rise in activity at approximately 440 °C.

3.1.2. Effect of the bond coat

The incorporation of the bond coat decreased the total number of AE counts, regardless of the previous treatment of the substrate. Bond coating also delayed failure, as measured by AE activity, of the coating which was optimally sprayed (Fig. 2(c)).

3.1.3. Influence of thermal cycling on the acoustic emission spectrum

The shape of the AE spectrum was retained on cycling. Small differences in the displacements of the spectrum were associated with a change in the count rate data; these are best seen in the following analysis.

3.2. Analysis of count rate data

The count rate is a more sensitive measurement than are the cumulative counts (as in Fig. 2), since it can be employed to identify the cracking mechanism. For example, additional work has shown that high count rates are usually associated with cracking around fine topographic features on the surfaces of individual particles, while low count rates can be related to interlamellar failure. Therefore the raw count rate data were arranged in ascending order and classified into count rate intervals of 5 x 10^2. The original time (and therefore temperature) sequence of each count rate datum point could be recalled from the original data file. The distribution of any particular count rate interval was thus evaluated in terms of a mean time value, the standard deviation of time, the skewness parameter (the third moment) and the kurtosis parameter (the fourth moment). The last two parameters give an indication of the distribution shape with respect to a normal distribution.

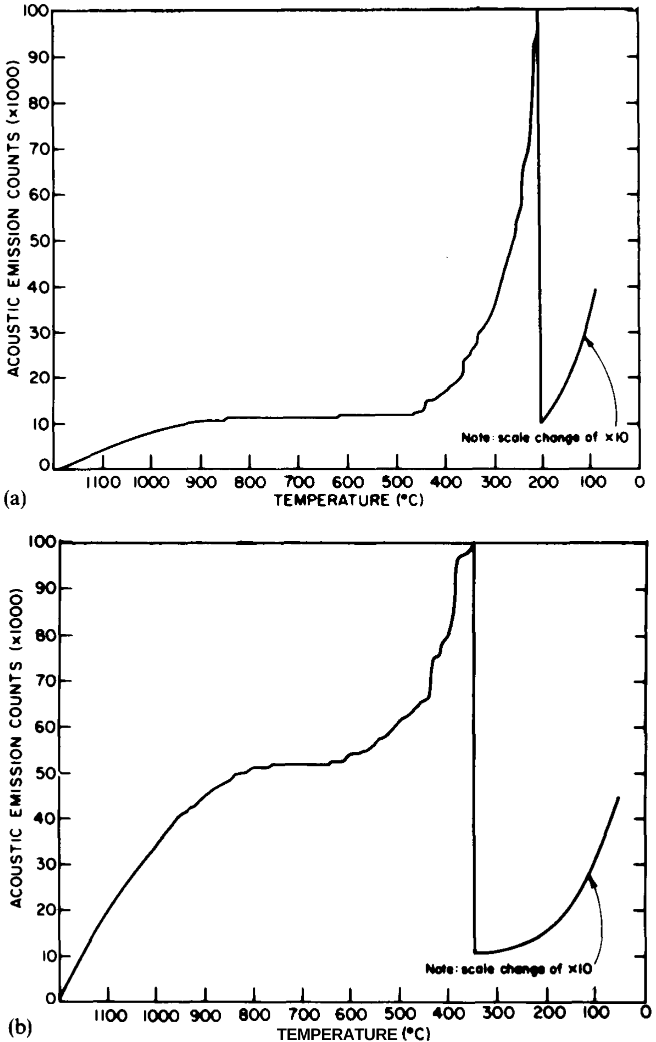

Fig. 2 *(continued).*

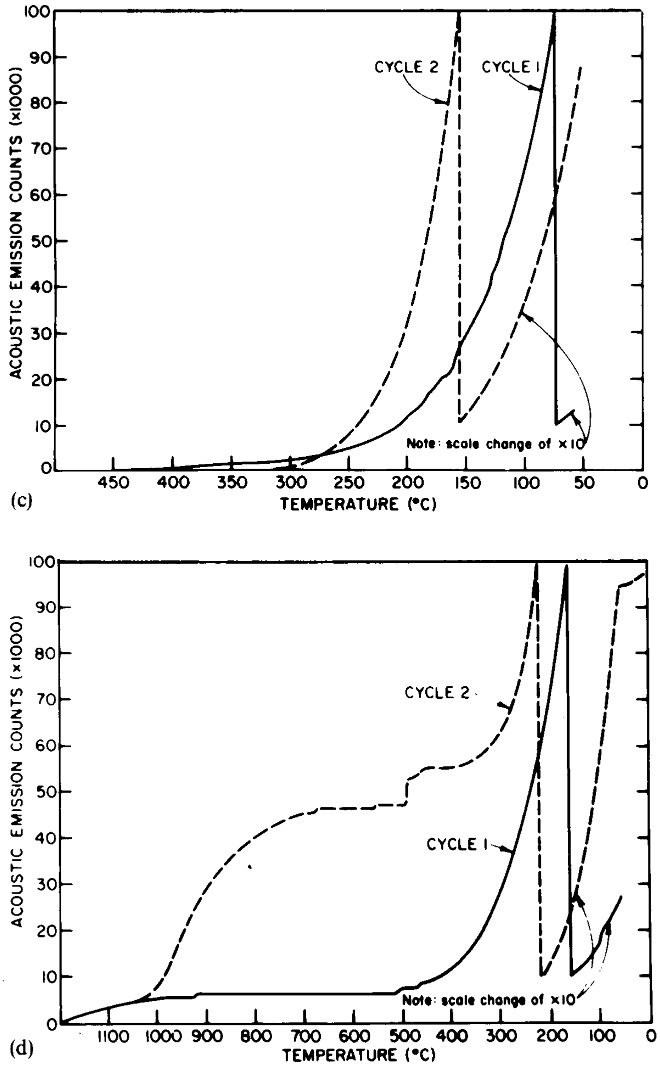

Fig. 2. Cumulative AE data: (a) specimen 21 (YSZ coated); (b) specimen 22 (preheated and YSZ coated); (c) specimen 18 (bond coated and YSZ coated); (d) specimen 20 (preheated, bond coated and YSZ coated).

The results of such an analysis are shown in Fig. 3 and are summarized in Table I. The total number of counts for a specific count rate interval are plotted with respect to the count rate interval. In this way it is possible to ascertain which *specific* count rate makes a significant contribution to the overall cracking process. The graphs also show the average temperature of the count rate interval and Table I includes the coefficient of variance (defined as 100 x standard deviation/mean). A high coefficient of variance indicates that the counts did not occur within a narrow temperature range.

3.2.1. Effect of preheating the substrate

Preheating of the substrate increased the temperature where maximum counts

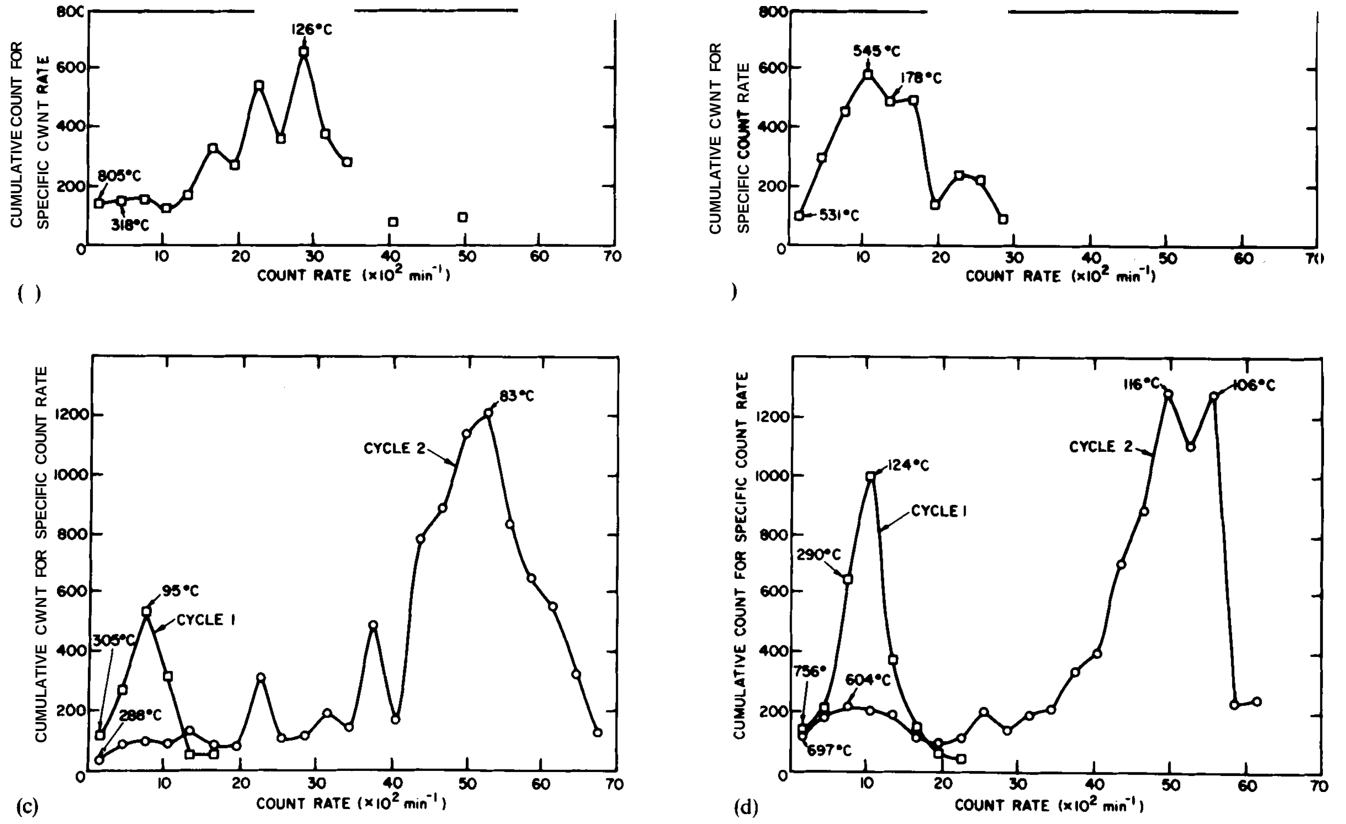

Fig. 3. Count-rate analysis of the AE data: (a) specimen 21 (YSZ coated); (b) specimen 22 (preheated and YSZ coated); (c) specimen 18 (bond coated and YSZ coated); (d) specimen 20 (preheated, bond coated and YSZ coated).

TABLE I

SUMMARY OF ACOUSTIC EMISSION DATA ANALYSIS FOR THERMALLY CYCLED ZrO_2–12wt.%Y_2O_3 COATINGS

Substrate treatment and coating	*Thermal cycles*	*Specimen*[a]	*Initial AE activity*		*Peak counts*				*Maximum count rate*			*Number of counts*	TP/TC[c] (%)
			Temperature (°C)	CV[b] (%)	*Temperature* (°C)	CV[b] (%)	*Number of counts*	*Median count rate*	*Temperature* (°C)	CV[b] (%)	*Median count rate*		
YSZ	2	15(1)	328	8	92	23	1610	37.5	88	26	49.5	7600	21
	1	21(1)	805	39	126	19	660	28.5	98	11	49.5	3920	17
Preheat + YSZ	1	19(1)	708	44	156	26	270	19.5	308	36	61.5	2450	11
	1	22(1)	531	32	545	83	590	10.5	156	28	28.5	4490	13
Bond coat + YSZ	7	16(1)	406	18	139	34	750	10.5	135	28	16.5	2000	38
		16(2)	361	17	74	25	1610	76.5	82	2	94.5	14060	11
	7	18(1)	305	79	95	40	530	7.5	116	30	16.5	1330	40
		18(2)	288	6	83	19	1210	52.5	78	9	67.5	8590	14
Preheat + bond coat + YSZ	7	17(1)	338	22	119	155	660	19.5	120	1	31.5[d]	3850	17
		17(2)	571	42	88	36	1210	52.5	119	25	85.5	10480	12
	2	20(1)	756	45	124	48	1000	10.5	95	1	22.5	2720	37
		20(2)	697	48	116	66	1290	49.5	105	13	61.5	9660	13

[a] The number in parentheses is the run number.

[b] CV, coefficient of variance.

[c] TP, total peak counts; TC, total number of counts.

[d] Isolated value of 79.5 counts min^{-1} at 1153 °C.

occurred. For example, maximum AE activity of the YSZ coating was observed at 126 °C (Fig. 3(a)) as compared with 545 °C for the preheated substrate (Fig. 3(b)). The YSZ/bond coat samples exhibited a maximum activity at 95 °C (Fig. 3(c)) as compared with 124 °C for the preheated substrates (Fig. 3(d)). It is also important to note that the preheated samples displayed a larger coefficient of variance, this being a mitigating factor in generalizing these AE trends for all of the specimens. The data show that the count rate increases during the cooling period and, for most coatings, preheating results in a low count rate which is initiated at a high temperature.

3.2.2. Effect of the bond coat

The incorporation of a bond coat decreases the peak count rate (*i.e.* the interval where most counts occur) as well as the maximum count rate. This is true for both optimally prepared and preheated substrates during the first thermal cycle. For the optimally prepared coatings the temperature range for the peak count rate is approximately the same for coatings with (95–139 °C) and without (92–126 °C) bond coats. However, preheating the substrate increased the temperature range of the peak count rate from 119–124 °C to 156–545 °C for the optimally sprayed YSZ.

3.2.3. Influence of thermal cycling on acoustic emission spectra

The second and subsequent thermal cycles of the bond-coated specimens exhibited (i) a significantly higher peak count rate at a lower temperature and (ii) a greater maximum count rate at a lower temperature. A major distinction between the optimally prepared and the preheated substrates was that the former exhibited a lower temperature for the onset of AE activity on the second thermal cycle. The preheated samples, however, started cracking at a higher temperature on cooling during the second cycle.

4. DISCUSSION

The central aspect of this study was to gain an understanding of how the AE phenomena are related to physical processes within the coating. The "extent of cracking" (or crack distribution and growth) is directly related to coating durability. The "cracking mechanism", however, is a process which identifies whether failure occurs through a lamellar or translamellar cracking mechanism or through interactions between the bond coat and the substrate. The present AE analysis cannot directly be used to identify cracking mechanisms. However, the effects can be studied by correlating the results for different coatings. Thus it is certain that any specific count rate value, in itself, is the total for many different cracking events, each of which may have their own AE generation characteristics. However, since the count rates do not appear in a completely random manner, but are related to some narrow temperature range, it is our view that it is justifiable to associate a particular count rate with a specific cracking mechanism.

The effect of substrate preheating is to increase the number of cracking events immediately upon cooling (Fig. 2). The character of this *extra* activity varies for samples with and without bond coats, as shown in Fig. 3. The specimens with no bond coat which were optimally prepared (Fig. 3(a)) displayed a larger peak count rate than did samples which were preheated (Fig. 3(b)). Thus any conditioning of the substrate causes macrocracking rather than microcracking. In the case of specimens without a bond coat, these microcracks occur at a higher temperature, thus implying pre-existing defects. This phenomenon arises from high residual stresses that are established by depositing the ceramic coating directly onto an overheated substrate. The use of a ceramic coating alone is impractical; however, the preheating effect with this specimen was used to discern a change in the cracking mechanism. It should also be remembered that both of these spraying conditions result in catastrophic coating failure upon the first thermal cycle. Therefore neither spraying procedure (*i.e.* YSZ or YSZ and preheating) should be interpreted as providing less susceptibility to coating failure.

All of the bond-coated specimens (Fig. 3) exhibited cumulative count rate peak positions at almost identical count rates for the same thermal cycle number, the only differences being that the preheated specimens exhibited more cracking on the first cycle (*i.e.* higher count value at the peak count rate) and that this occurred at higher temperatures. The AE spectra of the second cycle were almost identical, as shown in Figs. 3(c) and 3(d), but it should be remembered that these are actually displaced with respect to temperature. If this fact is taken into account, it can then be observed that the optimally prepared coating was far more crack resistant since cracking is activated by thermal stress and thus by temperature. The bond-coated specimens had the same peak count rate value as the preheated YSZ samples for the first thermal cycle. However, the temperature where this maximum AE activity occurs is much less for the bond-coated specimens, indicating more resistance to cracking. It is not straightforward to relate the peak count rates of the duplex and single coating systems because of their different cracking mechanisms.

It is interesting to notice that the peak count rate for the bond-coated specimens is the same as that for the preheated specimens without a bond coat. This leads to one of two inferences with regard to cracking mechanism. If cracks occur only within the ceramic coating, the bond coat reduces the extent of cracking (on the first cycle) since the peak in Fig. 3(c) is narrower than that in Fig. 3(b). However, if this were the only possibility then bond coating and preheating (Fig. 3(d)) have an adverse effect relative to coating integrity. However, experience does not support this conclusion. The more probable mechanism is that cracking occurs within both the bond coat and the ceramic coating. In this case the AE from both types of cracking gives rise to the peak count rate. The contribution from each type of event cannot be deconvoluted, but it is clear that the major event is bond-coat cracking. Thus if ceramic failure gives rise to a broad spectrum (Fig. 3(b)), the bond-coat effect must be significant in order for this spectrum range to decrease. Cracking within the ductile bond coat is more tolerable and preferable than failure within the brittle ceramic and thus coating integrity is improved. The shape of the AE spectrum changes significantly on further cycling because the coating structure has a high density of cracks.

Statistical techniques were used to analyze the AE spectra. Therefore it is important to appreciate the errors and the dependence (if any) of the results on the methods used. For example, count rate intervals of 100, 200, 300 and 500 showed identical results. In all the analyses, the kurtosis and the skewness parameters were approximately from $+2.0$ to $+4.0$ and from -2.0 to $+2.0$ respectively. The kurtosis parameter is more statistically significant since the inference is that the distributions are "highly peaked" compared with the normal distribution. Thus, ± 1 standard deviation (which is used as the error range) would incorporate more than 68% of the observations. A major problem in interpreting AE data is the difficulty in ascertaining whether exactly the same failure phenomenon occurs within duplicate specimens. For example, equivalent specimens display up to a 55% difference in cumulative counts. However, upon normalizing the peak count with respect to the total count it is seen that the duplicates follow the same trends.

5. CONCLUSION

The failure mechanisms of plasma-sprayed thermal barrier coatings are a complex function of imperfections contained within the oxide coating and at the interfacial region, and the means by which these imperfections respond to thermal cycling. Cracking mechanisms, both initiation and propagation, generate significant acoustic energy which, we believe, can be used to probe these mechanisms.

Details on the statistical evaluation of acoustic emission spectra associated with the failure processes of thermal barrier coatings are presented in this paper. It is emphasized that great care must be exercised in interpretations of these results. In fact, it is our ultimate goal in this program to develop rather more straightforward correlations among AE behavior and the wide range of microstructural features which are available for study.

Several plasma spray coating parameters were varied and the changes in cracking response were analyzed. The degree of preheating and the incorporation of a bond coat significantly affects the cracking behavior as determined, by AE response. The AE data can be analyzed in terms of cumulative counts or peak count rate.

It was established that preheating of the substrate gives rise to cracking of the coating during the early stages of cooling. These effects were observed whether or not a bond coat was used. The incorporation of a bond coat had a similar effect in reducing the temperature at which cracking occurred.

The cracking susceptibility, or AE count, is a measure of the coating integrity and is directly related to its performance in thermal barrier applications. Further study is clearly suggested, both of detailing the pertinent frequency spectra of the AE and of correlating such information with microscopy.

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