In situ monitoring of microfracture during plasma spray coating by laser AE technique

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Abstract

Atmosphere plasma spray coating materials include many pores and lamellar boundaries formed by flattened particles during spraying process although high reliability are required in ceramic coatings for turbines. These boundaries become an origin of the microcracks and following crack growth. As it is known that spraying parameters strongly affect the microstructure and strength of coating, it is expected to establish in situ monitoring technique for coating process. However, there is a limit to apply the existing non-destructive evaluation techniques to real-time monitoring at elevated temperature. We have investigated a non-contact measuring system to detect acoustic emission (AE) signals due to microfractures using a laser interferometer, and applied this technique for understanding microfracture process of ceramic coating at elevated temperature. In this paper, we evaluated the effect of several spraying parameters on the initiation and growth process of microcrack by detecting AE signals during coating process using a non-contact laser AE technique.

Keywords: Acoustic emission; Laser interferometer; Plasma spraying; Ceramic coating; Microfracture

1. Introduction

As the operation temperature of gas turbines has been rapidly increased to achieve high efficiency in recent years, materials are required improve their stability and mechanical properties at elevated temperature above 1773 K. Thermal barrier coating (TBC) with ceramic coating layer has been developed to shield heat from the outside and increase thermal stability of the surface [1]. As these coatings are subjected to both external stress and internal thermal stress constantly or periodically, the evaluation of mechanical properties and failure process of TBC at elevated temperature is desired to ensure the integrity. A health monitoring of structural components in service is important to assess the integrity of TBC, and a process control is also very important to provide uniform coating thickness and maximize the coating properties, as well as an estimation of the mechanical properties of coatings such as thermal shock resistance, thermal fatigue resistance and creep resistance.

In general, TBC consists of three layers: ceramic layer (top coat, TC) such as yttria partially stabilized zirconia (YSZ) or alumina, intermediate metal layer (bond coat, BC) such as MCrAlY (M: Co, Ni, etc.), and metal substrate such as nickel base superalloys. Failures such as delamination of interfaces or cracking in top coat become a serious problem for practical use [2]. These failures are mainly due to a misfit of thermal expansion between two layers, especially TC/BC interface. Therefore, it is very important to assess the reliability for ceramic coatings, which include many lamellar boundaries introduced during spraying process. These boundaries become an origin of the microcracks and following crack growth [3]. Atmosphere thermal spray (APS) forms a coating layer by the impact and rapid solidification of melted particles from sprayed powders. Many pores which could initiate a crack are generated due to the air inclusion and separation of impact particles with some spraying conditions during APS coating process [4]. Microstructure of coating layer affects a lifetime of structural components as well as an adhesion of the interfaces. As it is important to investigate the influence of spraying parameters on the initial defects in ceramic coating layer, studies for the effect of spraying parameters...
on microstructures [5,6] or mechanical properties of thermal spray coating have been carried out [7,8]. Also it is desired to detect these defects with high sensitivity because they lead to reduce a thermal cycle resistance.

Several non-destructive evaluation techniques, such as infrared thermography and ultrasonic testing, have been applied to detect defects in ceramic coatings [9–11]. However, these techniques have the limits in detection of microcracks. Regarding the detectability in defects, ultrasonic testing has enough sensitive for large-scale delamination at interfaces, but insensitive for vertical cracks in ceramic layer or small-size microcracks. Infrared thermography is not very suitable for the quantitative evaluation of defects because of its low spatial resolution. Also these methods have a difficulty in in-process monitoring. In these techniques some stimulation signals, ultrasonic pulse or temperature difference, are transmitted to a sample, and then the response from a system is detected to identify the defects in a sample. As ultrasonic testing usually requires some couplant, such as water, to transmit an ultrasonic sound from the transducer to the sample, it cannot be applied to the coating process. Infrared thermography also requires to uniform heating of a sample, because temperature distribution in a sample affects the measurement of temperature change by thermography. A sample during coating process is subjected to a complicated temperature distribution, which disturbs the measurement of defects. It is also very difficult to monitor in situ damage in a sample during coating process by these techniques because it takes some time to acquire the response corresponding to defects.

Acoustic emission (AE) technique is a promising tool for reliability assessment of coatings because it can monitor the generation and growth of microcrack in real time. However, conventional contact AE technique has a limit in application at elevated temperature, because a conventional piezoelectric transducer cannot be used above about 800 K. We have investigated the non-contact AE measurement technique using laser interferometer as a AE sensor [12–14]. This laser AE technique has several advantages such as non-contact measurement, absolute velocity measurement of AE signals, and applicability for severe environment. The purpose of this study is to investigate the influence of spraying parameters on the generation and growth process of defects during coating process by means of an in situ monitoring system based on laser AE technique.

2. Experimental

2.1. Plasma spray coatings

Stainless steel SUS304 was used as a substrate and a size of samples was 30 × 30 × 5 mm³. Spraying surface of the substrate was blast-finished to improve an adhesion between the coating/substrate interfaces. Rear surface of the substrate was also mirror-finished to measure a vibration of the surface and obtain a sufficient reflection of laser beam. The 45 kW class atmospheric plasma spraying equipment was used for both top coat and bond coat. METCO Type 9 MB plasma spraying gun was also installed. Firstly, the bond coat of NiCrAlY was sprayed on some samples and then cooled with forced air. A white alumina powder of 10–45 μm grain size (Showa Electric Co., K-16T) was sprayed on the bond coat as top coat and then samples were natural-air-cooled. Parameters of spraying apparatus are listed in Table 1. Working and auxiliary gas were Ar and He, respectively. In this study, the influences of some spray parameters on AE behavior were investigated, such as thickness of the bond coat, \( d_{BC} \), preheating temperature of the substrate, \( T_p \), thickness of the top coat, \( d_{TC} \), and traverse speed of spraying gun, \( v \). All the values of parameters are listed in Table 2.

2.2. In situ monitoring by laser AE technique

Experimental setup of in-process monitoring system is shown in Fig. 1. A sample was fixed to the stage equipped on a turntable in the direction perpendicular to the spraying direction. Temperature at the rear surface of the substrate during coating process was measured using a K type thermocouple welded on the center of sample. Schematic temperature history of the substrate during coating process is shown in Fig. 2. This coating process consists of three stages: first is a preheating of the substrate (stage-1), second is a spraying of top coat (stage-2), and the last stage is an air cooling of the coated specimen (stage-3). After preheating of the surface by gun stroking without powder feeding, a feeding-rate control valve was opened to start the spraying of the top coat. When the thickness of top coat reached a desired value, the spraying apparatus was switched off and then the specimens were air-cooled until room temperature. After spraying, the sample stage was turned and the rear surface was faced to the laser beam of interferometer.

| Parameter | Values |
|-----------|--------|
| \( d_{BC} \) (mm) | 0, 0.15 |
| \( T_p \) (°C) | 0, 300, 350, 450, 500 |
| \( d_{TC} \) (mm) | 0.8, 1.0, 1.2 |
| \( v \) (m/s) | 0.1, 0.2 |

Table 1

| Spraying parameter | Value |
|-------------------|-------|
| Spraying distance | 75 mm |
| Pitch | 5 mm |
| Feeding rate of powders | 20 g/min |
| Current | 500 A |
| Voltage | 70 V |

Table 2

Values for all the spraying parameters used
AE signals during cooling period were detected using a heterodyne type interferometer (AT-0022, Graphtec Corp.). Acquisition of AE signals was started after the power source of the spraying robot, exhaust duct and compressors were switched-off to avoid the influence of mechanical or electromagnetic noise on AE signals. Some dead time for AE measurement was required to adjust the focus of laser beam on the measuring surface. Total dead time before the start of an acquisition was about 1 min. Low noise type demodulator (AT-3600S, Graphtec Corp.) was used to measure an out-of-plane surface velocity on a sample with range of 1 mm/s/V. In order to reduce noise level, output signals were filtered with high pass filter of 50 Hz and low pass filter of 200 kHz. Detected AE waveforms were recorded by AE analyzer (DCM-140, JT-Toshi Corp.).

3. Results

3.1. Microstructures

Fig. 3 shows SEM micrographs of coated Al₂O₃ layer without preheating of the substrate. The top coat consisted of flattened, splat and non-melted particles, as shown in Fig. 3(a). Many pores were also frequently observed in the top coat (Fig. 3(b)). Clear lamellar structure was also found which seems to correspond to the coating layer formed by one stroke of moving spraying gun. Thickness of each lamellar structure was about 85 μm.

3.2. Fracture behaviors

Fig. 4 shows the cross-sections of fractured specimens after cooling, where delamination propagated during cooling along the TC/BC interface or in the top coat. In the sample without bond coat, delamination propagated through the substrate/TC interface or at the 1st boundary between ceramic lamellar layers, as shown in Fig. 4(a). In the sample of d_BC of 70 μm, delamination frequently propagated at the 2nd or 3rd lamellar boundary, as shown in Fig. 4(b). Fig. 4(c) also shows the cross-section of the top coat connected with the substrate in the sample of d_BC of 70 μm. Cracks parallel to the TC/BC interface propagated at lamellar boundaries and vertical cracks were also generated. Several lamellar boundaries in the delaminated top coat were clearly observed, shown in Fig. 5(a). Fig. 5(b) shows a magnified photo around the step between two lamellar boundaries in Fig. 5(a), and well-flattened particles were observed at the surfaces lamellar boundaries.

3.3. AE behaviors

3.3.1. Effect of bond coat

Fig. 6 shows the temperature history and AE behavior for the different thickness of bond coat, where the other processing parameters were common, that is, T_p was 773 K, d_TC was 1 mm, and v was 0.1 m/s. In the sample without
bond coat, only two AE with large amplitude were detected and immediately ceramic layer was delaminated, as shown in Fig. 6(a). On the other hand, a large number of AE events with various amplitudes in the sample of $d_{BC}$ of 70 μm were detected in the temperature range of 200 K before the delamination of TC (Fig. 6(b)).

### 3.3.2. Effect of preheating

Fig. 7 shows the relationship between the amplitude and generation temperature of AE for the different preheating temperature, where the other processing parameters were common, that is, $d_{BC}$ was 0 μm, $d_{TC}$ was 1 mm and $v$ was 0.1 m/s. The temperature where AE events start to generate, $T_{AE}$, was depended on the preheating temperature. However, the temperature difference, $\Delta T = T_p - T_{AE}$, seemed to be almost same as shown in Fig. 7(b). No AE signal was detected in the sample without preheating, and also no visible failure was observed during cooling process down to room temperature.

Fig. 8 also shows the relationship between the amplitude and generation temperature of AE for different preheating temperature, where $d_{BC}$ was 70 μm, $d_{TC}$ was 1 mm and $v$ was 0.1 m/s. $T_{AE}$ also increased with the increase of preheating temperature, and the total number of AE events decreased with the decrease of preheating temperature. The temperature difference $\Delta T$ was smaller than that in the samples without BC.
3.3.3. Effect of thickness of top coat

Fig. 9 shows the relationship between the amplitude and generation temperature of AE for the different thickness of top coat, where $d_{BC}$ was 70 $\mu$m, $T_p$ was 773 K and $v$ was 0.1 m/s. $T_{AE}$ also increased with the increase of thickness of top coat. Many AE events were generated in the sample of $d_{TC}$ with 1.0 mm, on the other hand a few AE events were detected in the sample of $d_{TC}$ with 1.2 mm.

3.3.4. Effect of traverse speed of spraying gun

Fig. 10 shows the relationship between the amplitude and generation temperature of AE for different traverse speed of gun, where $d_{BC}$ was 70 $\mu$m, $T_p$ was 773 K and $d_{TC}$ was 1 mm. $T_{AE}$ in the sample of 0.1 m/s was higher compared with that in the sample of 0.2 m/s. The number of AE events in the case of 0.2 m/s was larger than that of 0.1 m/s, and the temperature range during AE generation of 0.2 m/s sample was wider than that of 0.1 m/s.

3.4. AE waveforms

AE signals detected during cooling process could be classified into two types by the peak frequency, that is, Type-A and Type-B, respectively. Type-A signals have a peak around 75 kHz, on the other hand Type-B signals have several characteristic peaks in 100–200 kHz. Typical examples for each type waveform are shown in Fig. 11(a) and (b), respectively, and fast Fourier transform (FFT) results of them are shown in Fig. 12(a) and (b), respectively. Type-A signals were especially detected before the final delamination of specimens, and Type-B signals were broadly observed during cooling process.

FFT analysis for all the AE signals plotted in Fig. 10 was performed and types for each event were identified. The amplitude distributions of each type signal for both speed of spraying gun are shown in Fig. 13. Many Type-A with low frequency AE events with large amplitude were generated in the sample of $v = 1.0$ m/s, and only few Type-B events with...
high frequency were detected (Fig. 13(a)). On the other hand, there is no significant difference in amplitude distribution between Type-A and Type-B in the sample of $v = 2.0$ m/s (Fig. 13(b)). The relationship between the amplitude and generation temperature of each type AE for different traverse speed of gun is re-plotted with frequency type in Fig. 14. Type-A signals were firstly detected and the following Type-B signals were generated with cooling. Only Type-A signals occurred just before final delamination of TC.

4. Discussion

4.1. Change of microstructures

In general, porosity and distribution of pore radius in alumina coating are strongly affected by the spraying distance. Intergranular vertical cracks are frequently observed in the microstructure of top coat fabricated with relative short spraying distance, and transgranular cracks or delamination of lamellar boundaries are dominant in the case of long spraying distance. The spraying distance of 75 mm, common spraying condition in this study, induced a formation of microstructure with both microfracture types (Fig. 3). Therefore, large cracks will be formed by the coalescence of initial cracks or pores due to thermal stress, and these will be AE sources during cooling process. As shown in Fig. 3(b), pores were frequently observed on the boundary between coating layers at equal spaces formed by one stroke cycle of spraying gun. Cracks will be easily generated at these relative weak boundaries rather than other lamellar boundaries inside of each ceramic layer formed by one stroke cycle. Delamination propagated at these boundaries of lamellar layers as shown in Fig. 4(a) and (b), and intergranular cracks at these boundaries were also observed even in the delaminated top coat as shown in Fig. 4(c). This demonstrates that many intergranular cracks at these weak boundaries occurred apart from the final delamination location. Fig. 5(a) also shows the other
evidence that multiple intergranular cracks at the boundaries of lamellar layers propagated at various site in the top coat. As well-flattened particles were observed at the fracture surfaces of lamellar boundaries in the delaminated top coat layer, as shown in Fig. 5(b), it is understood that crack easily propagate at these boundaries.

As shown in Fig. 10, $T_{AE}$ decreased with the increase of traverse speed and the temperature range from AE generation to final delamination also became large with the increase of traverse speed. This suggests that traverse speed of spraying gun strongly affects the strength of formed coating layer. As the number of spraying cycles must be increased to obtain a desired thickness of the top coat using high transverse speed, the number of boundary of coating layer per unit thickness increases with the increase of traverse speed and the thickness of each layer decreases. Thus a high traverse speed induces many lamellar boundaries in the top coat, which become a possible initiation site of microcrack. In addition, a thermal sprayed ceramic coating has a low density and the thermal conductivity is also low due to many pores and initial microcracks related with the lamellar structure. Thermal conductivity of the top coat can be affected by the changes in thickness and number of lamellar layer due to processing conditions.

4.2. Effect of thermal stress on microfracture

Crack paths were clearly changed by existence of the bond coat. In the sample without bond coat, delamination propagated at the interface of substrate/TC and 1st lamellar boundary as shown in Fig. 4(a). On the other hand, delamination was observed at various lamellar boundaries of top coat in the sample with bond coat as shown in Fig. 5(a). A generation behavior of AE was also strongly influenced by the existence of bond coat, and also it was related to the size and velocity of microcracks. Large delamination propagated rapidly in the case without bond coat, on the other hand microcracks with various sizes generated at various sites of top coat in the case with bond coat. The substrate is usually preheated before spraying and then air cooled. Thermal stress is applied to the top coat during cooling process because coefficient of thermal expansion of alumina is smaller than that of the substrate.
If the top coat and the substrate are directly connected, it is reasonable that crack propagates along the interface where is mostly strained. Bond coat plays important role in relaxation of misfit strain between the top coat and the substrate. Therefore, cracks were formed by the coalescence between initial cracks or pores in the top coat. Cracks propagated in the top coat near the edge of the specimen regardless of existence of the bond coat. An edge should be stress concentrated site so that cracks initiate at the edge of specimens.

As shown in Fig. 8(b), the difference between preheating temperature and AE starting one in the case without bond coat was almost constant regardless of the difference in preheating temperature. This suggests that a criterion of crack propagation is determined by this temperature difference. Thermal stress applied to the top coat when a specimen is cooled down 200 K from the sprayed temperature can be estimated from the following equation

$$\sigma_{TC} = \frac{E_{TC}}{E_m} (\alpha_m - \alpha_{TC}) \Delta T (E_m d_m + E_{TC} d_{TC})^{-1},$$

(1)

where $E$, $\alpha$ and $d$ are Young’s modulus, coefficient of thermal expansion and thickness, respectively. Subscripts of ‘TC’ and ‘m’ represent the properties for the top coat and the substrate, respectively. Substituting $E_{TC}$ of 30 GPa, $E_m$ of 197 GPa, $\alpha_{TC}$ of $6.5 \times 10^{-6}$/K, $\alpha_m$ of $17.3 \times 10^{-6}$/K, $d_{TC}$ of 1 mm and $d_m$ of 5 mm, respectively, $\sigma_{TC}$ was estimated as 63 MPa.

If the substrate is not preheated or forced-cooled, AE start temperature corresponding to delamination temperature decreases. Therefore, it is possible to prevent delamination of the top coat by temperature control of the substrate during spraying process. However, it has been reported that splat pattern and adhesion of melted particles during spraying process is affected by preheating temperature [15,16]. Spraying condition must be determined from correlations of each parameter.

As shown in Fig. 9, AE start temperature in the case with bond coat increased with the increase of preheating temperature and the number of AE events decreased with the decrease of preheating temperature. This behavior was similar to the case without bond coat, but the difference between $T_{AE}$ and $T_p$ was smaller than that in case without bond coat. Bond coat plays important role in relaxation of misfit strain between the top coat and the substrate as already described. This of change of $T_{AE}$ for different $T_p$ can be explained by the relaxation of misfit strain due to existence of the bond coat.

As shown in Fig. 10, AE start temperature increased with the increase of thickness of top coat, and the difference of temperature between AE onset and final delamination also became large with the increase of thickness of top coat. This result can be described by the fact that thermal stress applied to coating layer becomes large with the increase of thickness. Therefore, the temperature distribution in the thickness direction during spraying and cooling process can be a significant factor governing cracking behavior in the top coat.

4.3. Analysis of microfracture by AE waveforms

We have already reported the failure process under thermal cycle of the same specimens in coating system and geometry with this study [17]. As the result of two-dimensional location and simulation of wave propagation, it was demonstrated that a crack propagated toward the center from edges and segmentations of delaminated top coat by vertical cracks were occurred. The difference between previous study and this study is temperature history of the specimen. In the previous case, cracks gradually propagated under cyclic heating/cooling. In this study, cracks propagated during only one cooling process. As shown in Fig. 4, delamination propagated at the interface or lamellar boundaries between ceramic layers. Fractured top coat showed a ragged feature consisting of the lamellar boundaries between coating ceramics layers divided by vertical cracks as shown in Fig. 5(a). The thickness of residual top coat was the maximum around edges and minimum around the center. If AE waveforms correspond to different failure mode, it can be possible to correlate AE behavior with failure process. It has been reported that failure modes in composite materials correspond can be revealed by the frequency analysis of detected signals of each AE event [18]. In this study, first Fourier transfer (FFT) results of each AE signal were discussed to understand the fracture sources.

AE signals detected during cooling process could be classified into two types, as shown in Fig. 12(a) and (b), by the peak frequency. Type-A signals had relative large amplitude and were detected especially before the final delamination of top coat. Type-B signals had rather small amplitude and were broadly found during cooling process. Type-A signals also includes many larger amplitude signals compared with Type-B as shown in Fig. 13. Fig. 14 shows the relationship between the amplitude and generation temperature of each type AE for the different traverse speed of gun. Only Type-B signals were detected at AE start temperature, while only Type-A signals were detected just before the final delamination of top coat. AE behavior during cooling clearly demonstrated the iteration of Type-A and Type-B signals. Firstly, high frequency type signals generated, and then relative low frequency type signals generated. In the case without bond coat, delamination occurred immediately after the generation of only a few Type-A signals. This suggests that Type-A signal can be corresponding to the propagation of delamination. Type-B signals seemed to be corresponding to cracking in top coat such as vertical cracks, because this type signal was detected only when fractured top coat showed a ragged pattern. If multi-channels AE measurement system for in-process monitoring is successfully applied, relationship between crack type and AE source can be analyzed more quantitatively.
5. Conclusions

We developed a non-contact in-process monitoring system for coatings with laser AE technique, and applied this system to the detection of microfracture during cooling process of thermal spraying. Conclusions of this study are as follows:

1. Using the developed in-process monitoring system with laser AE technique, AE generated in cooling process of plasma spraying was successfully detected.
2. In the case without bond coat, few large-amplitude AE signals were detected and immediately delamination occurred. However, many AE signals with different amplitude were detected before delamination in temperature range of 200 K in the case with bond coat.
3. In the case with bond coat, AE start temperature decreased with the increase of traverse speed, preheating temperature or thickness of the top coat, and temperature difference between AE start and final delamination also became large with the increase of traverse speed.
4. AE signals detected during cooling process could be classified into two types by the peak frequency characteristics. AE signals with relative large amplitude and low frequency can correspond to the propagation of delamination. On the other hand, AE signal with rather small amplitude and high frequency seems to be the cracking in top coat such as vertical cracks.

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