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Influence of Compressive Stress in TGO Layer on Impedance Spectroscopy from TBC Coatings

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Abstract Impedance spectroscopy is a non-destructive evaluation (NDE) method first proposed and developed for evaluating TGO layers with compressive stress inside thermally degraded plasma-sprayed thermal barrier coatings (PS TBCs). A bode plot (phase angle (h) vs. frequency (f)) was used to investigate the TGO layer on electrical responses. In our experimental study, the phase angle of Bode plots is sensitive for detecting TGO layers while applying compressive stress on thermal barrier coatings. It is difficult to detect TGO layers in samples isothermally aged for 100 hrs and 200 hrs without compressive stress, and substantial change of phase was observed these samples with compressive stress. Also, the frequency shift of the phase angle and change of the phase angle are observed in samples isothermally aged for more than 400 hrs.

Keywords: Impedance Spectroscopy, Compressive Stress, Thermal Barrier Coating (TBC), TGO Layer

1. Introduction

In the development of next-generation gas turbines and aero engines, increasing the turbine inlet gas temperature is an effective and direct approach for improving efficiency [1]. Thermal barrier coatings (TBCs) have been adopted in gas turbines to increase the operating temperature and provide insulation for hot sections to prolong the life of the components [2-4]. Typical TBC systems are composed of three primary components: an yttria stabilized zirconia (YSZ) topcoat, an aluminum containing bond coat (BC), and a superalloy substrate [5]. A plasma-spray technique is generally used to deposit the top coat and bond coat of TBC systems. During the service of gas turbines, a thermally grown oxide (TGO) layer of alumina oxidation develops between the YSZ and BC coatings under high temperature conditions [6,7].

Recently, the failure mechanisms for plasma-sprayed TBC systems have been widely investigated. These complex processes are controlled by the stresses at the interface that can develop delamination, buckling and cracks [8-10]. During high-temperature exposure of TBCs, a TGO layer forms and grows to induce residual stress in the TGO. In the isothermal treatment, a misfit of the coefficient of thermal expansion between the TGO and constituents of the TBC systems is responsible for out-of-plane stress, which induces local delamination and cracking in the vicinity of TGO, and eventual large-scale spallation of the top coating [11]. Therefore, the stress state in the TGO layer is important to understand the damage mechanism and to develop a life-prediction method. It is necessary to develop evaluating methods to monitor the formation and growth of the TGO as well as the residual stress.

There have been few studies dealing with the residual stress measurement of thermal barrier coatings with NDE methods [12-15]. The ultrasonic velocity is changed by the residual stress in materials introduced by manufacturing

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and post-processing, which corresponds to the change of the residual stress in materials. However, the variation of the ultrasonic velocity is about 0.1% to 0.4% (at the level of 0.1%), and it is very hard to prepare precise equipment to measure these small changes [14]. Thermal barrier coatings have 4 layers, including TGO. So, understanding of the dispersion of ultrasound in such materials is needed to describe the complex behavior of wave propagation in thermal barrier coatings. The eddy current method is usually applicable to TBCs to determine either the layer thicknesses or conductivities from measured impedance signals. However, it is difficult to evaluate the residual stress of TBC because of small changes in conductivity due to stress [15].

Several non-destructive evaluation (NDE) techniques have been applied to study the thickness and residual stress of the TGO layer in PS TBCs. The Cr^{3+} photo luminescence piezo spectroscopy(PLPS) technique was adopted to measure the compressive stress of the TGO [16-19]. However, different compressive stress trends have been reported. Tolpygo et al. [18] observed a small decrease, while Sohn et al. [19] observed a gradual increase for the first 200 cycles, followed by a constant value until a sharp decrease associated with spallation. Both groups worked on EBPVD TBCs, although the materials had different BC layers. In addition, residual stress analysis, phase transitions, decomposition and chemical reactions were studied by Raman microprobe spectroscopy (RMS) technique, which is similar to PLPS [20].

Recently, Diaz et al. set up an in-situ synchrotron X-ray diffraction measurement under thermo-mechanical conditions to measure the stress state with measured lattice constants [21]. They observed positive σ_{22}(+36.4 MPa) on oxide grown under mechanical loads in ramp-up thermal cycle conditions from X-ray diffraction. σ_{22} on grown oxide is usually about -2.59~-4 GPa [21], and a σ_{22} of 36.4 MPa indicates a very severe change on the TGO layer. In gas turbine operation, mechanical loading during ramp up in thermal cycles plays a very important role in changing the residual stress in thermal barrier coatings.

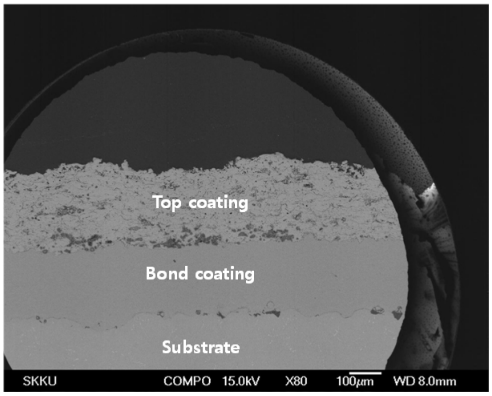
Impedance spectroscopy has also been applied as an NDE technique for TBC systems of actual gas turbine components [1,5,22,23]. The equivalent circuits of TBC systems have been dealt with in other papers [1,22]. Oxide and alumina layers are considered as a combination of resistance and capacitance. These values of resistance and capacitance are changed with thermal exposure time [1]. However, due to the stress in the TGO layer being affected by many factors, it is difficult to understand the influence of residual stress on the impedance spectrum. So, no one has tried to explain the variations of the measured impedance spectrum with respect to change in the TGO layers with different stress states.

The aim of this study is to investigate the influence of stress inside the TGO layer using impedance spectroscopy (IS) method with different applied stresses. The influence of different applied compressive stresses on the impedance spectrum of PS TBCs was investigated. The IS experiment results show that the compressive stress has a great effect on the impedance spectrum of the TGO layer. The phase angle of the Bode plot gives a clear description of different compressive stress effects on the TGO layer. A discussion of the TGO layer with stress applied is summarized, and a conclusion is given in the last section.

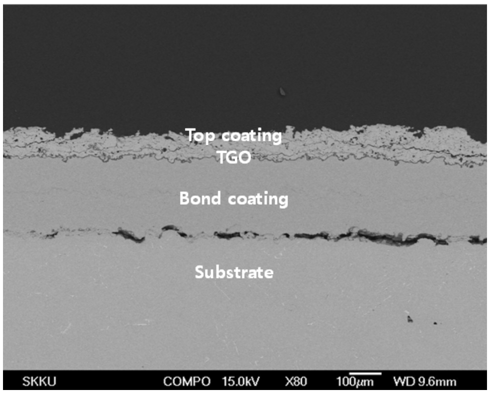
2. Experiments

2.1 Specimens and Preparation

Schematic illustrations of a PS TBC specimen without thermal process used in this



(a)



(b)

Fig. 1 Schematic illustrations of PS TBC specimen, (a) without isothermal process, (b) with isothermal process

study are shown in Fig. 1(a). A plate of INCONEL 738LC Ni-based alloy with a 3-mm thickness was used as a substrate. Commercially available 300-μm yttria stabilized zirconia (6-8 wt.% YSZ) and 250 μm CoNiCrAlY alloy (M/Ni and/or Co) were employed as top coating and bond coating materials, respectively. Normally, as the thermally grown oxide (TGO) grows at the YSZ/bond coat interface due to oxidation of the bond coat, internal stresses are built up at the top coat/bond coat interface since the oxidation is accompanied by volumetric expansion shown in Fig. 1(b) [13]. Prior to impedance measurements, all specimens were isothermally heat-treated at 900°C for 50, 100, 200, 400, 800, and 1600 hrs to grow a continuous TGO layer with a certain thickness.

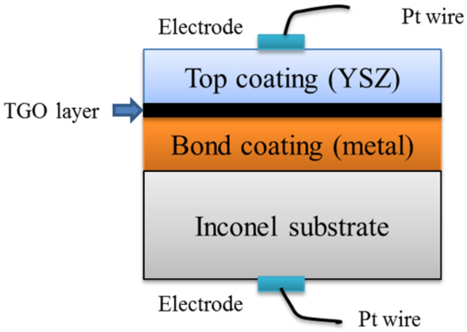


Fig. 2 Specimen preparation

Impedance measurements were carried out using an Ag electrode with a diameter of 5 mm, on the YSZ surface, and with the substrate. The following steps for preparing the specimen in Fig. 2 are as follows.

1) The specimens are cleaned chemically and ultrasonically before the impedance measurements.
2) The impedance is measured by a two-contact-electrode system. One of the Pt electrodes is soldered to the Ni-based superalloy, and the TBC side was coated with a silver paint, which served as the other electrode.
3) In order to consolidate the silver paint and enhance its adhesion to the specimen surface, the paint was solidified at 500°C for 10 minutes, and then the temperature was increased to 900°C for 2-3 minutes.

2.2 Experimental Setup

The impedance properties of all of the specimens are evaluated by the IS technique for the different aging conditions. A schematic illustration of the impedance measurement apparatus used is shown in Fig. 3.

Impedance measurements were carried out using an impedance analyzer 4294A coupled with a computer-controlled tensile machine, with a furnace at 200°C, as shown in Fig. 4.

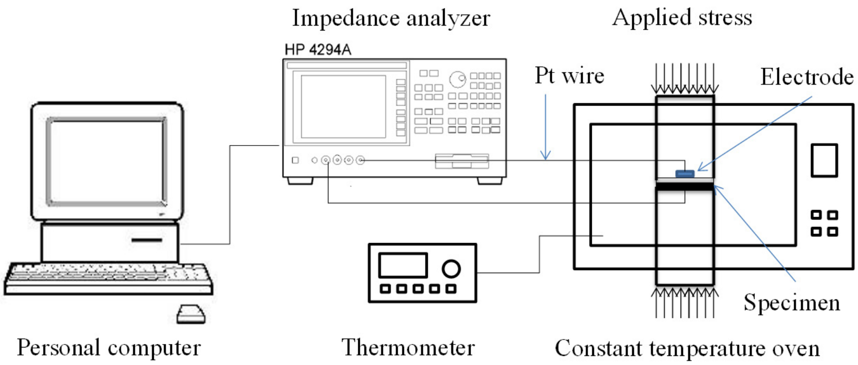


Fig. 3 Impedance measurement setup with applied stress

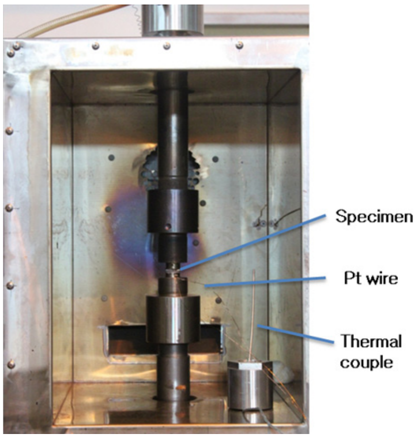


Fig. 4 The detail of experimental setup

2.3 Impedance Measurements

Variation of the temperature during measurements causes a significant variation in the measured values, so the temperature is carefully kept constant at 200°C during the impedance measurements, aiming at decomposition of the moisture on the specimen surface. These treatments of the specimen result in diminishing the leakage surface impedance measurement currents. At a high temperature of 200°C, YSZ is a typical oxygen ion conductor, and the bond coating and substrate are the conductors, while TGO acts as an oxide semiconductor [14]. So, only the properties of the TGO layer are investigated in impedance measurements.

The impedance is measured by a two-contact -electrode system. An alternating current (AC) amplitude of 500 mV and a swept frequency range from 40 Hz to 100 MHz are applied using an impedance analyzer. The measured results were represented by the mean values of data points obtained together with the standard deviation. After the impedance measurements, the cross-sectional microstructure and composition of the TBC specimens were examined by scanning electron microscopy (SEM).

3. Results and Discussion

3.1 SEM Observation

In our study, impedance spectra were first applied to characterize a range of isothermally heat-treated AS TBCs with compressive stress applied.

SEM was used to investigate the thickness and microstructure of the TGO layer. Samples were prepared using a precise diamond wheel saw at low speed to minimize the mechanical damage to the coating [15]. Cut sections were placed in epoxy resin, before grinding and polishing using successively finer SiC paper. Polished samples were carbon coated.

As shown in Fig. 5, TGO layers are grown as the isothermal heat treatment time is increased. These changes of the TGO layers influence the impedance spectra of the TBC coatings.

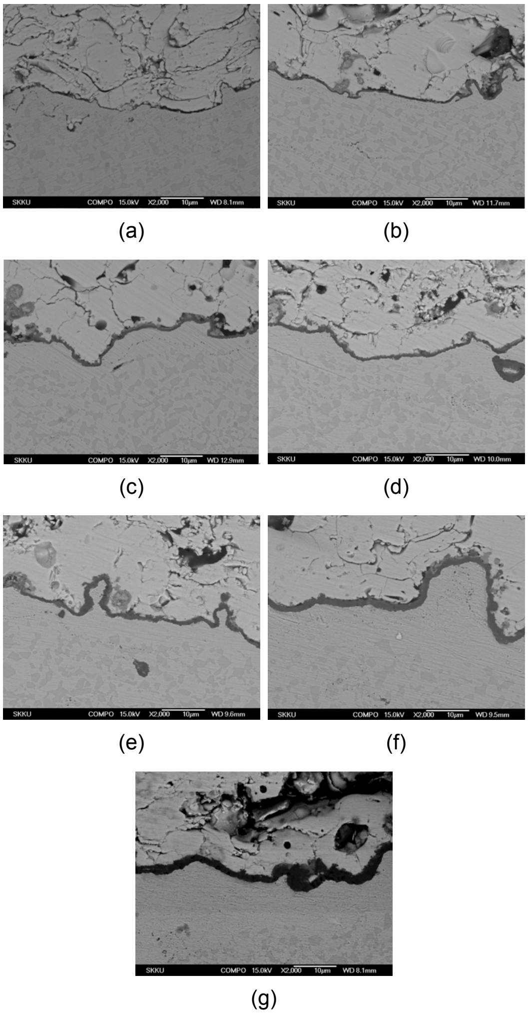


Fig. 5 SEM images of the TBC coating specimens thermally aged at 900°C (a) 0 hr, (b) 50 hrs, (c) 100 hrs, (d) 200 hrs, (e) 400 hrs, (f) 800 hrs, (g) 1600 hrs

3.2 Variation of Impedance Spectrum with Compressive Stress

Impedance spectroscopy was carried out using an impedance analyzer 4294A coupled with a computer-controlled tensile machine to investigate the influence of compressive stress.

As shown in Figs. 6(a) and (b), there were no significant changes in the impedance observation with different stress levels. These results agreed well in the SEM images in Figs. 5(a) and (b). This was because the TGO layers were very small compared to the specimens aged for more than 100 hrs. Also, in this experiment, abnormal change of the phase angle was observed at 240 MPa of compressive stress, as shown in Fig. 6(b). This phenomenon is due to the impedance being very sensitive to variation of the experimental environment, for example odd artifact is included in 240 MPa setup. It is worthwhile to address this abnormal impedance spectrum to emphasize the significance of precise measurement. Phase angle is minimum at 100 kHz~250 kHz in thermally aged TBC specimen without compressive stress as shown in Fig. 8. This phenomenon was already indicated in other papers [11,22]. This is due to relaxation processes in the specimen. As shown in Fig. 6(c), a sharp decrease of the phase angle is observed at 300 MPa of stress level. Until 240 MPa, it is hard to clearly characterize the effect of TGO layers in TBC coatings. But, a sharp decrease is presented at 300 MPa, as shown in Fig. 6(c). Even though thin TGO layers appeared in the 100hrs specimen, as shown in Fig. 6(c), it is hard to detect thin TGO layers without applied stress on the impedance spectrum. However, the impedance spectrum changed enough to detect thin TGO layers with 300 MPa applied stress. A sharp decrease is observed as shown in Fig. 6(d), similar to Fig. 6(c). As stress is increased, the magnitude of the phase angle is increased. Also, the frequency is shifted to low frequency. For aging over 400 hrs, the frequency shift is the dominant phenomenon, as shown in Figs. 6(e)~(g). More research is needed to investigate the frequency shift of phase angle due to applied compressive stress showing firstly observed in this paper.

With the increase of the thickness of TGO, which is due to the longer time of treatment, the effect of stress becomes more and more distinct. From 100 hrs on, the effect of different

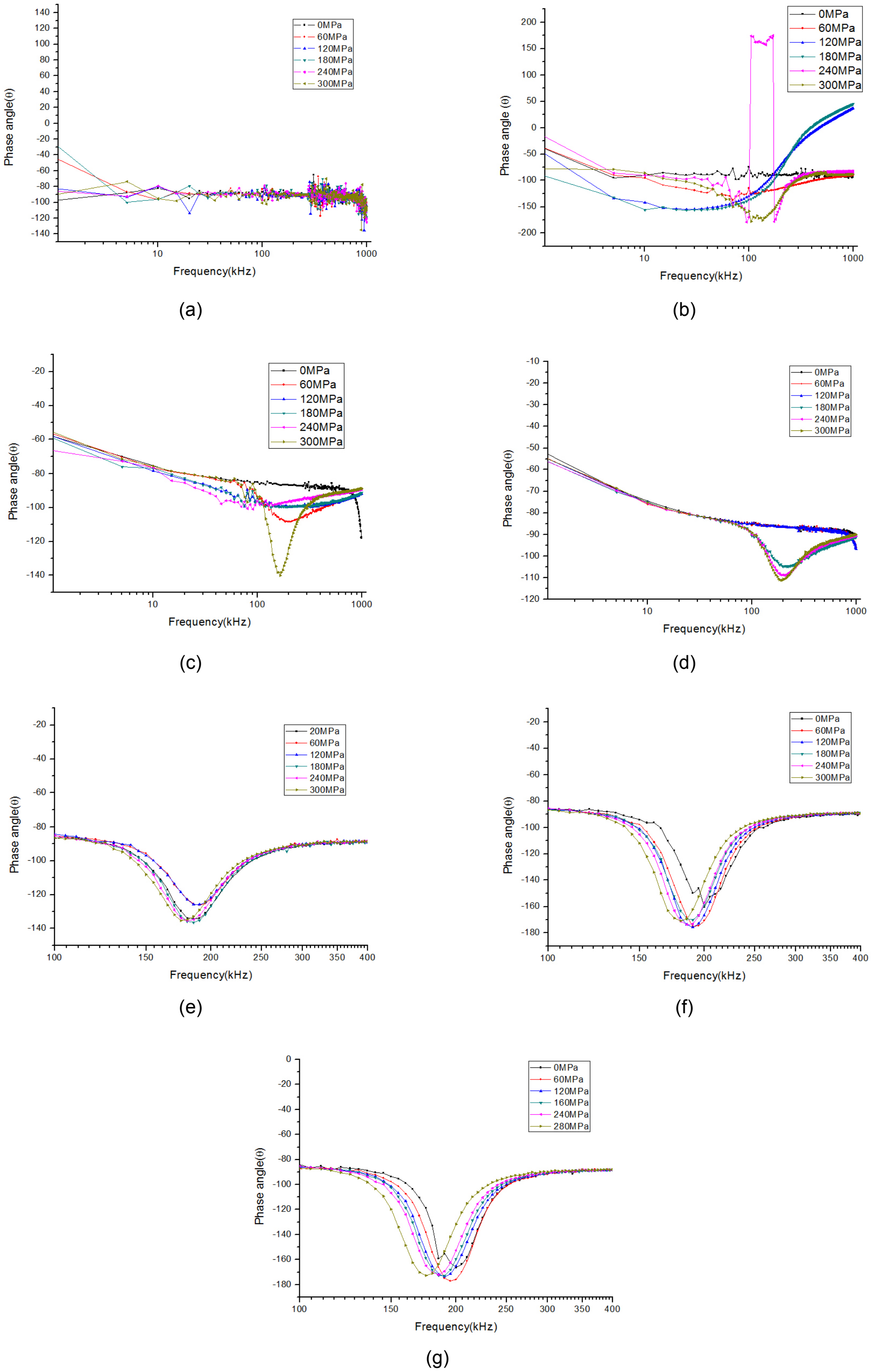


Fig. 6 Experimental results for change of compressive stress in the TBC coating specimens thermally aged for (a) 0 hr (b) 50 hrs (c) 100 hrs (d) 200 hrs (e) 400 hrs (f) 800 hrs (g) 1600 hrs

stress can be shown in the impedance spectrum. At 100 hrs, the applied stress of 300 MPa shows a sharp decrease; at 200 hrs, from the magnitude of 180 MPa on, a sharp decrease appears. From 400 hrs on, all the magnitudes show such phenomena. The higher the applied stress, the sharper the valleys and phase-shift to the left.

4. Conclusions

Impedance spectra have been applied for the first time to characterize a range of isothermally heat-treated AS TBCs with applied compressive stress. In this study, the thermally grown oxide (TGO) layer of the thermal barrier coating after isothermal treatment was measured with compressive stress applied by impedance spectra. The conclusions are as follows:

1) A thin TGO layer of TBC samples isothermally aged for less than 200 hrs can be observed in impedance spectroscopy with applied stress.
2) Low-frequency phase shift is observed in specimens aged for more than 100 hrs.
3) The overall decrease of phase angle is observed in degraded specimens. But, sharp decrease of phase angle in the 200 hrs sample is observed explicitly.

The detailed relationship between the mechanical properties and the change in the impedance spectra needs to be studied in the future.

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