

DEVELOPMENT OF NONDESTRUCTIVE EVALUATION METHODS FOR CERAMIC COATINGS

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ABSTRACT

Nondestructive evaluation (NDE) methods are being developed for ceramic thermal barrier coatings (TBCs) applied to components in the hot-gas path of advanced high-efficiency and low-emission gas turbines, including syn-gas fired turbines. The objectives of the NDE development are to assess TBC condition for quality control as well as to monitor TBC degradation and predict TBC life during service. For these applications, NDE development at Argonne National Laboratory (ANL) is focused on quantitative methods including laser backscatter, mid-infrared reflectance (MIRR), and thermal imaging based on a multilayer analysis method. Both laser backscatter and MIRR are optical imaging methods and have been investigated for TBC health monitoring. The thermal imaging method developed recently at ANL can measure TBC's thermal properties with high accuracy. Because TBC properties change with life, measured TBC properties may be used for TBC health monitoring based on a model proposed in 2009 under this project. This model accounts for the TBC conductivity increase due to material sintering as well as the conductivity decrease due to interface cracking. These optical and thermal imaging NDE methods were evaluated based on preliminary experimental results for TBC samples that were thermal cycled to various lifetimes. This paper presents these recent developments and experimental results related to TBC health monitoring.

INTRODUCTION

Ceramic thermal barrier coatings (TBCs) are extensively used on hot gas-path components in advanced high-efficiency and low-emission gas turbines, including syn-gas fired turbines.¹⁻² In this application, a thermally insulating ceramic topcoat (the TBC) is bonded to a thin oxidation-resistant metal coating (the bond coat) on a metal substrate. TBC coated components can therefore be operated at higher temperatures, with improved performance and extended lifetime. TBCs are usually applied by electron beam-physical vapor deposition (EB-PVD) and air plasma spraying (APS). As TBCs become "prime reliant," it becomes important to know their conditions nondestructively to assure the reliability of these components. NDE methods can be used to assess the quality of new coatings, identify defective components that could cause unscheduled outages, monitor degradation rates during engine service, and provide data for reaching rational decisions on replace/repair/re-use of components.

Work at Argonne National Laboratory (ANL) is underway to develop advanced NDE methods for TBCs. TBC failure normally starts from initiation of small cracks at the TBC/bond coat interface. These cracks then grow and link together to form delaminations which eventually cause TBC spallation (i.e., failure). Although qualitative NDE methods can detect large-scale flaws/damages, quantitative methods are necessary to determine material property degradation and crack progression during the entire TBC life cycle. Therefore, for TBC health monitoring,

NDE development at ANL is focused on quantitative methods including laser backscatter, mid-IR (infrared) reflectance (MIRR), and thermal imaging based on a multilayer analysis method. Laser backscatter is an optical scanning method developed at ANL for detection of subsurface flaws in ceramic materials.³⁻⁴ MIRR is an optical imaging method developed at NASA for TBC health monitoring.⁵ Both optical methods detect the increased light scattering/reflection from the progressive crack development during the TBC life. The thermal multilayer analysis method developed at ANL measures TBC's thermal properties including thermal conductivity and heat capacity. These properties, especially the thermal conductivity, change with TBC life.⁶⁻⁷ The characteristic change of TBC's thermal conductivity with life has been modeled under this project.⁸

In this paper, the principles of the optical methods and the thermal imaging method and model for TBC degradation monitoring are described first. Preliminary experimental results for TBC samples that were thermal cycled to various lifetimes are then presented and analyzed.

OPTICAL AND THERMAL NDE METHODS FOR TBC HEALTH MONITORING

Laser Backscatter Method

The laser backscatter method is based on the cross-polarization detection of the scattered light from the subsurface of a translucent material.³ When a polarized laser beam is incident on a translucent material such as a TBC, the total backscattered light consists of surface reflection and subsurface backscatter. However, the surface reflection typically has no change in its polarization state while the subsurface scatter has a significant change. This method selectively measures only the subsurface backscatter from cracks and interfaces, while filtering out the surface reflection. This method has been investigated for NDE of TBCs, specifically for health monitoring during isothermal heat-treatment testing.⁴ The current ANL system uses a HeNe laser at 633nm wavelength and performs scanning for image construction; however, direct imaging at other wavelengths is possible.

Mid-IR Reflectance (MIRR) Method

MIRR is an optical imaging method developed by Dr. Eldridge of NASA for TBC health monitoring.⁵ Because optical penetration for TBCs is at maximum in mid-infrared wavelengths (3-5 μm), MIRR has higher sensitivity to detect TBC cracking near the TBC/substrate interface. Therefore, changes in MIRR may be used to monitor the progression of TBC cracking and delamination.

In MIRR, a steady-state infrared light source is used to illuminate the TBC surface and the total reflection, including those from the TBC surface, TBC volume, and cracks near TBC/substrate interface, is imaged by an infrared camera. In the NASA system, a SiC IR emitter coupled with an off-axis parabolic mirror is used to provide collimated illumination to the TBC samples, and imaging is performed at the wavelength of 4 μm with a narrow-band filter.⁵ The ANL system utilizes a standard IR heating lamp coupled with a sanded-glass plate to provide smooth illumination, and imaging is taken in the entire wavelength band (3-5 μm) of an mid-wavelength IR camera. The wide-band imaging allows a weak IR illumination to be used, which reduces sample heating and improves detection sensitivity. The effect of OH absorption band near 3 μm ⁵ was estimated to be not significant to the detection accuracy.⁹

Thermal Imaging Method

The change of thermal conductivity with TBC life has been studied by many authors. In early exposure times, TBC undergoes a sintering process, resulting in a denser material and an increased conductivity. As exposure time increases, the initiation and growth of microcracks causes a gradual reduction of TBC conductivity. When delamination occurs in the later stage of the TBC life, a significant conductivity decrease can be observed in the delaminated regions. This characteristic change in TBC conductivity may be used to monitor TBC degradation condition and predict TBC life. Such approach for TBC health monitoring apparently has not been attempted before likely because of the lack of a NDE method that can accurately measure TBC conductivity on real components. TBC conductivity can be measured by several methods. The most commonly used method is laser flash, which is normally a destructive method because it uses stand-alone TBC coating samples (sometimes with specially-prepared TBC samples with substrate). Although a large number of TBC thermal conductivity data is available, they are mostly for the intrinsic property of the TBC coating material without the contribution from the interface degradation inside a real TBC system. Therefore, these data are not suitable for TBC life prediction. What is needed is a NDE method that can measure the thermal conductivity of a TBC system (i.e., TBC coating on substrate), so TBC degradation and cracking at the interface is included within the measured data. Such a NDE method has been developed recently: the multilayer analysis method based on one-sided flash thermal imaging.⁸ Preliminary studies showed that it has high measurement accuracy and repeatability.

For TBC health monitoring, it is necessary to establish a model based on a characteristic “damage” parameter. There are two major mechanisms affecting TBC conductivity, as illustrated in Fig. 1: one is the increase of the intrinsic conductivity of the coating material due to continued sintering, and the other is the decrease of the TBC system conductivity due to the progressive cracking at the interface. Because the conductivity decrease due to cracking is the major TBC degradation mechanism, a damage parameter based on the conductivity decrease is an indicator of the TBC degradation status. A model⁸ to determine this parameter is described below.

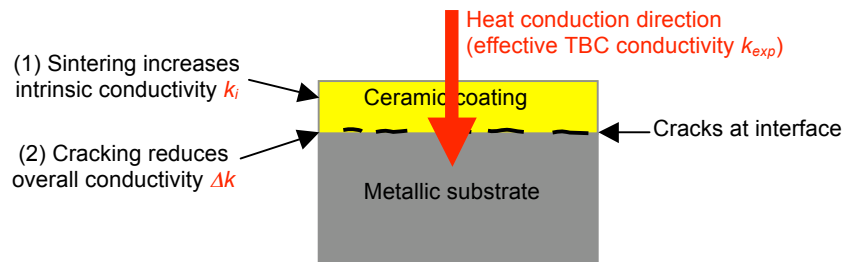


Figure 1. Effects of TBC degradation to the change of its thermal conductivity.

Recent studies⁶⁻⁷ showed that the increase of TBC's intrinsic thermal conductivity k_i due to sintering may be correlated with the Larson-Miller parameter (LMP):

$$\ln(k_i / k_{i0}) = a + bLMP, \quad (1)$$

where LMP is defined as:

$$LMP = T(\ln t + C), \quad (2)$$

k_{i0} is the initial conductivity (at $t=0$ for as-sprayed coating), T is an average exposure temperature, t is time, and a , b , and C are fitting constants. This correlation indicates that if a TBC is not

damaged during its service life its conductivity should increase monotonically with time (or life). Any damage inside the coating or at the interface should reduce the TBC-system conductivity, k_{exp} , which is measured by thermal imaging. The conductivity difference Δk ,

$$\Delta k = k_i - k_{exp}, \quad (3)$$

is therefore a TBC damage parameter. Δk is related through LMP to the thermal exposure temperature and time, both are important parameters affecting TBC lifetime. It is noted that in Eq. (1) the initial conductivity k_{i0} can be combined into the fitting parameter a , because $\ln(k_i/k_{i0}) = \ln(k_i) - \ln(k_{i0}) = \ln(k_i) - \text{constant}$, so k_{i0} may not need to be measured when using this model.

In practical application, it is necessary to establish an approach to determine the intrinsic TBC conductivity k_i , which may be different for coatings with different composition and microstructure as well as under different sintering conditions. Therefore, the correlation Eq. (1) should be determined for each TBC system. To do this, we note that a TBC system is normally not damaged during its early life, so measured conductivity should be equal to the intrinsic conductivity, i.e., $k_i = k_{exp}$ or $\Delta k = 0$. This approach may then establish the relationship Eq. (1)-(3) for each TBC system from experimental data. The remaining issue involves with how to determine a criterion for Δk that corresponds to the level of TBC damage. Although more validations are necessary, development of this model may establish another fundamental approach to understand TBC damage process based on an important and most-studied TBC property: thermal conductivity. Figure 2 illustrates the use of this model for TBC life prediction, as presented for this project in 2009.⁸

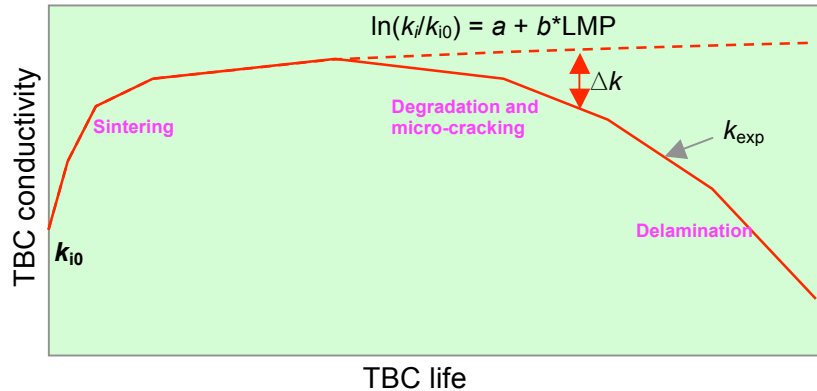


Figure 2. Illustration of thermal conductivity model for TBC degradation and life prediction.

EXPERIMENTAL RESULTS

TBC Samples

Two sets of thermal-cycled APS TBC samples were obtained from Dr. Kulkarni of Siemens Energy Inc. and used to evaluate NDE methods for TBC life prediction. All TBC samples have a dimension of 1-inch in diameter. One set consists of 7 samples exposed at a high temperature T_1 , within which two of the longest exposed coatings have already spalled. The other set consists of 4 samples exposed at lower temperatures: two at T_1 and two at T_2 ($>T_1$). Figure 3 shows photographs of these TBC samples; it is noted that some TBC surface has contaminations of different colors.

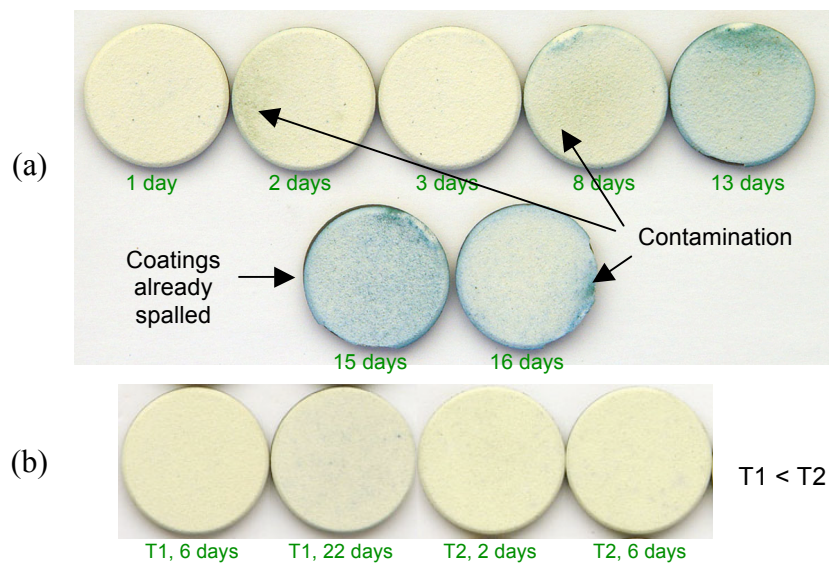


Figure 3. Photographs of APS TBC samples that were thermal cycled at (a) a high temperature T and (b) lower temperatures T_1 and T_2 .

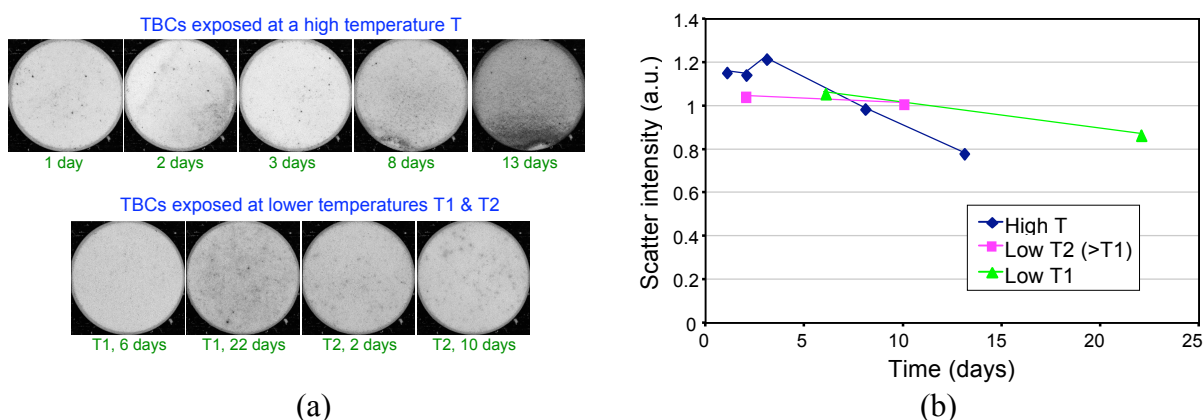


Figure 4. Laser backscatter (a) images and (b) intensity as function of TBC exposure time.

Experimental Results from Laser Backscatter and MIRR Methods

Figure 4a shows the scan images of all TBC samples using the laser backscatter method. The scatter intensity is clearly affected by the surface contaminations as seen in Fig. 3: the intensity is lower in contaminated surface areas. In addition, the intensity is low in many spots areas; the nature of this lower scattering is not clear at present. In Fig. 4b the average scatter intensity is plotted as a function of TBC exposure time for both sets of TBC samples; the intensity data generally show a decreasing trend with exposure time which however is likely due to the effects of surface contamination and low scattering spots. Because of the shallow optical penetration with the current system operated at visible 633nm wavelength, it was suggested that laser backscatter may become more sensitive to TBC degradation when using a longer wavelength that has deeper optical penetration.⁹

Figure 5a shows the MIRR image of all TBC samples. MIRR data are also affected by the surface contaminations. However, by comparing the images in Figs. 3 and 5a, it is evident that the surface reflectance value is not correlated with the visible contamination level. The average

reflectance value generally increases with TBC exposure time, as shown in Fig. 5b. Although the reflectance that corresponds to TBC spallation can be identified to be at ~74% from the samples exposed at the high temperature T, it is unclear if this “spallation” value can be applied to other samples exposed at lower temperatures T1 and T2 because of the contamination problem.

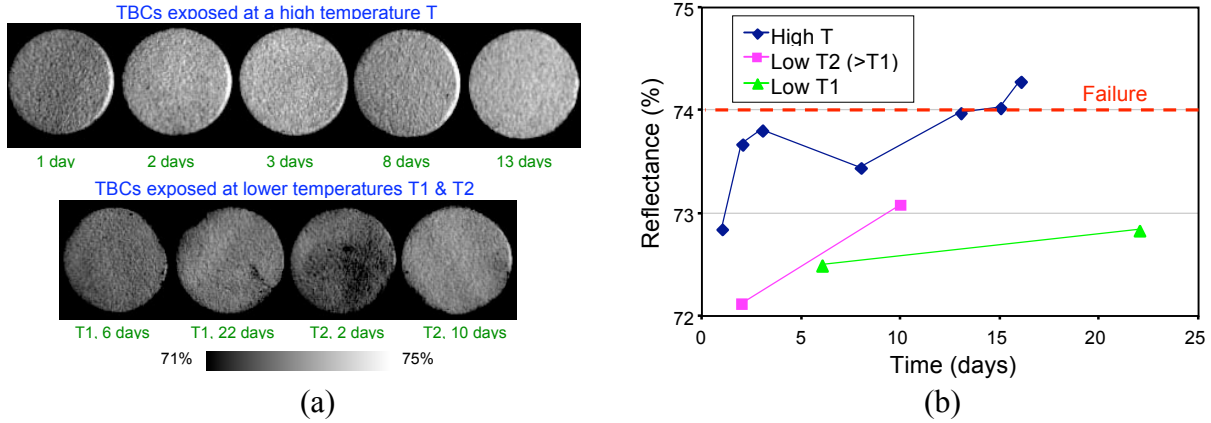


Figure 5. MIRR reflectance (a) image and (b) value as function of TBC exposure time.

Experimental Results from Thermal Imaging Method

Figure 6a shows measured thermal conductivity of all thermal-cycled TBC samples using the thermal-imaging multilayer analysis method.⁸ TBC conductivity is generally uniform on each sample surface, not affected by the surface contaminations. For TBCs exposed at lower temperatures T1 and T2, a faint stripe pattern is visible, which could be due to slight thickness variation that was not considered in experimental data processing. Further, all coatings were assumed to have the same thickness of 0.3 mm; this assumption might have introduced some measurement error as small difference in coating thickness is expected for these samples. In general, TBCs exposed at the high temperature T have much lower conductivity than those exposed at lower temperatures T1 and T2. However, the thermal response of the high-temperature-exposed TBCs was significantly different from that of the lower-temperature-exposed ones (and all other APS TBCs previously tested by ANL) as seen in Fig. 6b. Because all TBC samples were prepared with same surface treatment (i.e., painted black) before testing, the difference seems to suggest that there is a conductivity variation along the coating depth in all high-temperature-exposed TBCs. For the high-temperature-exposed TBCs, thermal conductivity images in Fig. 6a indicate some damage (i.e., low conductivity spots) in coatings with longer exposure times, 8 and 13 days (note that coatings failed after 13 days, see Fig. 3).

The thermal conductivity model described above was attempted for TBC degradation prediction. Because of the thermal response difference as noted above, the correlation for the lower-temperature and high-temperature exposed TBC samples was made separately. Figure 7a shows the conductivity-LMP correlation for the low-temperature exposed TBCs. Because the correlation, Eqs. (1)-(2), has three unknowns, three data points (out of four) were used to establish an exact conductivity-LMP relationship ($b=7 \times 10^{-5}$, $C=58$). The remaining data point, for the sample exposed with higher temperature T2 and longer time 10 days, showed a conductivity difference Δk from the correlation, which indicates some damage within this sample. The conductivity-LMP correlation for the high-temperature-exposed TBCs is displayed in Fig. 7b. The correlation is not good in early times, as the TBC conductivity showed a

monotonic decrease with exposure time. However, the conductivity decrease in later times is a clear indication of damage accumulation with time.

It is interesting to compare the TBC life prediction models based on MIRR and thermal conductivity. In MIRR the reflectance has small increases in later TBC life (see Fig. 5b), while the conductivity shows larger decreases in later TBC life (see Fig. 7b). Therefore, the conductivity model may have a better sensitivity to detect the damage progression near the end of the TBC life.

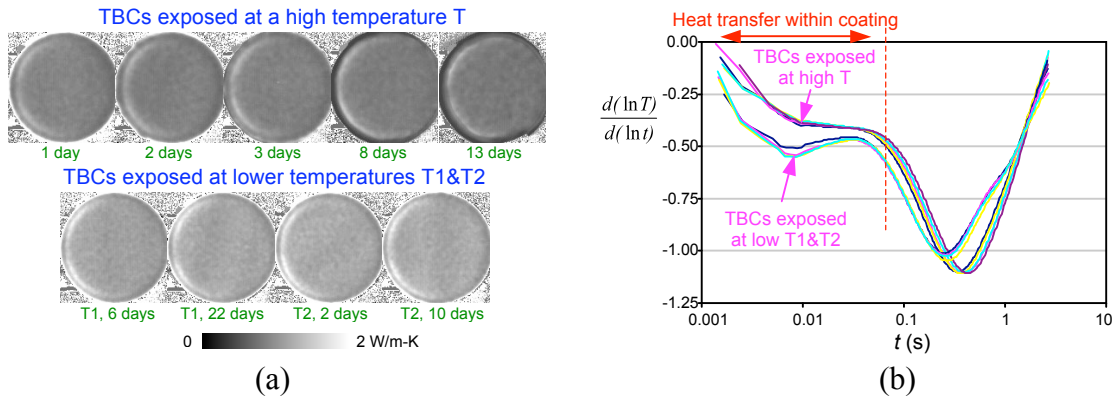


Figure 6. Measured (a) thermal conductivity images and (b) thermal responses of all TBC samples.

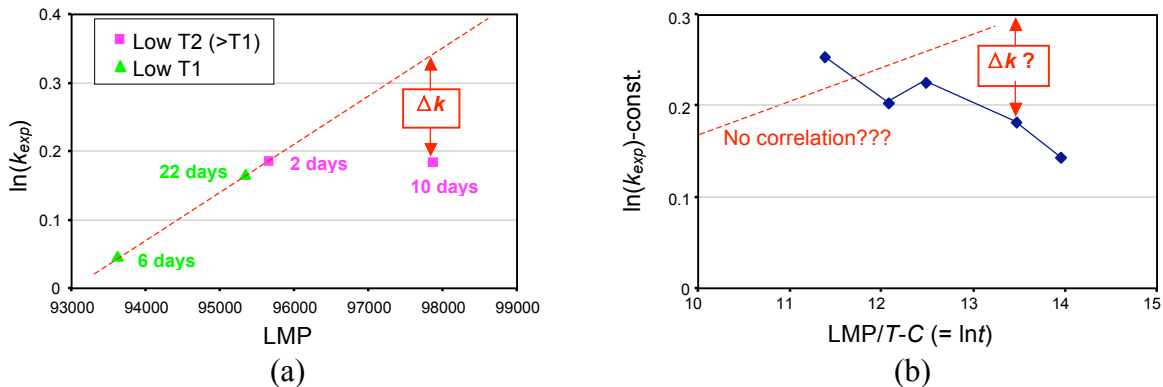


Figure 7. TBC degradation evaluation based on thermal conductivity model for TBC samples exposed at (a) lower temperatures T1 & T2 and (b) high temperature T.

CONCLUSION

Quantitative optical and thermal imaging NDE methods are being developed for TBC health monitoring and life prediction. The optical methods include laser backscatter developed at ANL and mid-IR reflectance (MIRR) developed at NASA. For thermal imaging, a multilayer analysis method developed by ANL was used to measure TBC thermal properties, and a TBC degradation prediction model based on a correlation between TBC conductivity and LMP was established. These NDE methods were evaluated using two sets of thermal-cycled APS TBC samples provided by Dr. Kulkarni of Siemens Energy Inc. Experimental results showed that surface contamination may affect the data of both optical methods. The MIRR data were found generally correlated with the TBC exposure time although the sensitivity becomes small near the

end of TBC life, while the laser backscatter was not very sensitive to the range of TBC exposure conditions investigated in this study (longer wavelength operation should improve sensitivity). On the other hand, the TBC conductivity-LMP correlation was demonstrated to be capable of identifying accumulated damage in thermal-cycled samples with long-time exposures. However, the correlation was not completely satisfied for the high-temperature-exposed samples because of an abnormal thermal response within the coating layer for these samples; the reason for this abnormality will be further investigated.

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