Evaluation of Damage Process of a Coating by Using Nonlinear Ultrasonic Method

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Abstract: During the service or external loading of the surface coating, the damage accumulation may develop in the coating or at the interface between the substrate and the coating, but it is difficult to measure directly in the early stage, so the acoustic nonlinear parameters are used as the early damage index of the coating. In this paper, the nonlinear wave motion equation is solved by the perturbation method and the new relationship between the relative ratio of second-order parameter and third-order parameter was derived. The nonlinear ultrasonic testing system is used to detect received signals during tensile testing of for the specimen with Al₂O₃ coatings. It is found that when the stress is less than 260 MPa, the appearance of the coating has no obvious change, but the nonlinear coefficients measured by the experiment increase with the increase of the tensile stress. By comparing the curves of nonlinear coefficients and stress respectively, the fluctuation of curves the second-order nonlinear coefficient $A_2$ and the relative nonlinear coefficient $\beta'$ to stress is relatively small, and close to the linear relationship with the tensile stress, which indicates that the two parameters of the specimen with Al₂O₃ coatings are more sensitive to the bonding conditions, and can be used as an evaluation method to track the coating damage.

Keywords: acoustic nonlinearity; material degradation; simplified nonlinear parameter; second-order and third-order nonlinear coefficient

1. Introduction

Surface coating technology is increasingly used in defense, aviation, aerospace and other fields, such as solid rocket engine sealing layers and heat-proof coating. However, because the coating is different from the substrate material, it is inevitable that the local separation and exfoliation between the coating and the substrate material will affect the performance of the coating. And the properties of the coating after storage still meet the requirements of use. For the mechanical bonding defects and weak bonding defects existing in the interface, ultrasonic technologies are considered to be valid nondestructive methods and have become a hot research issue [1].

Traditional linear ultrasonic detection technology uses acoustic wave propagation and reflection and scattering of waves and absorption of ultrasonic energy when encountering a defect in the process. Linear characteristics of the defect detection and evaluation of the medium are to be detected. For example, Lescribaa et al. [2] made a preliminary exploration of the ultrasound quality evaluation of the bonding interface, through an analysis of the reflection echo coefficients of various interfaces of the thermal barrier coating. Wu et al. [3] adopted an ultrasound microscope system to test the inner quality of coating and used a 20 MHz probe to conduct an ultrasound C-scan testing of the simulated debonding defects of 3 and 5 mm in the wave absorbing coating. Ingo et al. [4] tested the surface defects, bubbles, debonding, and delaminating defects of polymer coating with an ultrasonic microscope system (SAM). However, the above methods cannot effectively detect micro-cracks or microscopic degradation. The detection of micro-cracks and the decrease of
material strength at the early stage of fracture is a problem, so it is urgent to develop a new method that can solve this problem.

Studies in the fields of mechanics, acoustics and materials show that the reflection and transmission signals of the interface when the ultrasonic wave propagates in the material with degradation (mechanical bonding, weak bonding, plastic deformation, etc.) are accompanied by obvious nonlinear behavior [5]. In the application of nonlinear theory, scholars have mostly adopted the second-order classic nonlinear coefficient to study the nonlinear phenomenon of sound waves [6,7], which has been supposed to be valid for testing material properties such as degradation, including fatigue [8–10], creep [11], thermal aging [12,13] and micro-cracks [14–16]. Meanwhile, the second-order nonlinear technology has also been used to evaluate the early damage of the coating. For instance, Shui et al. [17] employed a second-order nonlinear coefficient to evaluate the early damage of the coating. In recent years, the third-order nonlinear parameter has become a significant index in evaluating material degradations [18,19]. Therefore, the current paper attempts to simultaneously use second-order and third-order nonlinear coefficients to evaluate the bonding condition of the coating.

In this study, an ultrasonic method for the evaluation of coating damage processes based on the second-order and third-order nonlinear coefficients is proposed by analyzing the relative acoustic nonlinearity parameters of coated materials under tensile loading. The specimen with Al₂O₃ coatings is loaded to different stresses, and this will result in cumulative damage at different levels corresponding to the corresponding stress. The relationship between nonlinear ultrasonic parameters and tensile stress during tensile testing is studied, and the nonlinear ultrasonic testing system is used to detect the received signal. The fundamental and harmonic amplitudes are obtained, and the parameters are calculated. The relationship curve of the nonlinear parameters as the functions of tensile stress is obtained to detect or predict the performance of stress plasma-sprayed thermal barrier coating. The nondestructive evaluation method using the higher order harmonic generation technique has the potential to be an effective tracker and predictor of the coating damage.

2. Theory of Higher-Order Acoustic Nonlinearity

When a 1D longitudinal wave propagates in a nonlinear medium, the wave motion equation is given by [20]

\[ \rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial \sigma}{\partial x} \]  

(1)

where \( u \) is the displacement, \( \rho \) is the density, \( t \) is the time, and \( \sigma \) is the Piola–Kirchhoff stress tensor. This paper tries to use the perturbation method to solve the second- and third-order nonlinear coefficients.

In the case of a small strain, the positive strain is defined as

\[ \varepsilon = \frac{\partial u}{\partial x} \]  

(2)

Suppose the nonlinear constitutive relation (i.e., the stress-strain relation) of a solid dielectric is given by

\[ \sigma = E \cdot f(\varepsilon) \]  

(3)

where \( E \) is elastic modulus. Given that the sound velocity \( c \), elastic modulus \( E \) and density \( \rho \) are in the relation \( c = \sqrt{E/\rho} \), Equations (2) and (3) can be substituted into Equation (1), thus obtaining

\[ \frac{\partial^2 u}{\partial t^2} = c^2 f'(\varepsilon) \frac{\partial^2 u}{\partial x^2} \]  

(4)
where $f'(\varepsilon)$ is the derivative of $f(\varepsilon)$ with respect to $\varepsilon$, and this can be dealt with by Taylor series expansion. Thus, $f(\varepsilon)$ can be expressed as

$$f(\varepsilon) = \varepsilon + \frac{1}{2} \beta \varepsilon^2 + \frac{1}{6} \delta \varepsilon^3 + \Delta(\varepsilon^4)$$

(5)

where $\beta$ is the classic second-order nonlinear coefficient, $\delta$ is the classic third-order nonlinear coefficient, and $(\Delta \varepsilon^4)$ is the high-order infinitesimal minterm of $\varepsilon$. By substituting Equation (5) into Equation (4), the high-order minterm is omitted and $\frac{\partial^2 u}{\partial \varepsilon^2}$ can be derived as:

$$\frac{\partial^2 u}{\partial t^2} = c^2 \cdot \frac{\partial^2 u}{\partial x^2} \left[ 1 + \beta \frac{\partial u}{\partial x} + \frac{\delta}{2} \left( \frac{\partial u}{\partial x} \right)^2 \right]$$

(6)

Generally, Equation (6) has no analytical solution, and the perturbation method is mostly adopted to obtain the approximate solution. Hence, $u(x,t)$ is given by

$$u(x,t) = u_0(x,t) + u_1(x,t) + \cdots + u_n(x,t)$$

(7)

When $n = 0$:

$$u_0(x,t) = A_1 \sin(kx - \omega t)$$

(8)

Only considering the contribution of the second-order nonlinear coefficient in Equation (6), then that equation can be written as

$$\frac{\partial^2 u}{\partial t^2} - c^2 \cdot \frac{\partial^2 u}{\partial x^2} = \frac{\partial^2 A_1^2}{\partial x^2} \sin 2(kx - \omega t)$$

(9)

By substituting Equation (8) into the right side of Equation (9), then Equation (10) can be obtained.

$$\frac{\partial^2 u}{\partial t^2} - c^2 \cdot \frac{\partial^2 u}{\partial x^2} = -\frac{c^2 A_1^2}{2} \beta k^3 \sin 2(kx - \omega t)$$

(10)

Assume that the general solution of Equation (10) is

$$u_1(x,t) = f(x) \sin 2(kx - \omega t) + g(x) \cos 2(kx - \omega t)$$

(11)

By substituting Equation (11) into the left side of Equation (10),

$$\left( -4k \frac{df}{dx} + \frac{df}{dx} \right) \sin 2(kx - \omega t) + \left( \frac{d^2 g}{dx^2} + 4k \frac{dg}{dx} \right) \cos 2(kx - \omega t) = -\frac{A_1^2}{2} \beta k^3 \sin 2(kx - \omega t)$$

(12)

Moreover, From Equation (12), we can also obtain the following:

$$-4k \frac{dg}{dx} + \frac{d^2 f}{dx^2} = -\frac{\beta}{2} A_1^2 k^3$$

(13)

$$\frac{d^2 g}{dx^2} + 4k \frac{dg}{dx} = 0$$

(14)

By assuming $\frac{d^2 g}{dx^2} = 0, \frac{df}{dx} = 0$, then it can be obtained from Equation (13) that

$$u_1(x,t) = \frac{A_1^2 k^2}{8} - \beta x \cos 2(kx - \omega t)$$

(15)

In solving the third-order nonlinear coefficient with the above method, the influence of the third-order non-linear coefficient on wave equation is considered, which is given by

$$\frac{\partial^2 u}{\partial t^2} - c^2 \cdot \frac{\partial^2 u}{\partial x^2} = \frac{\partial^2 A_1^2}{\partial x^2} + \frac{\partial^2}{\partial x^2} \left( \frac{\partial u}{\partial x} \right)^2 \frac{\partial^2 u}{\partial x^2}$$

(16)
Then, according to the above formula derivation method and the following expressions of nonlinear parameters are given [21].

\[
\beta = \frac{8A_2}{k^2x A_1^2}
\]  
(17)

\[
\delta = \frac{32A_3}{k^3 x^2 A_1^3}
\]  
(18)

As can be seen in Equations (17) and (18), when the transducer frequency and propagation distance are fixed, the nonlinear coefficients, \(\beta\) and \(\delta\), are in direct proportion to \(A^2/A_1^2\) and \(A^3/A_1^3\), respectively. During the detection process, the relative nonlinear coefficient is defined as [22]

\[
\beta' = \frac{A_2}{A_1} = \frac{k^2}{8} \beta x
\]  
(19)

\[
\delta' = \frac{A_3}{A_1} = \frac{k^3}{32} \delta x^2
\]  
(20)

The relative nonlinear coefficients \(\beta'\) and \(\delta'\), as shown in Equations (19) and (20), respectively, are the functions of the ultrasound propagation distance \(x\). The relationships between the second-order relative nonlinear coefficient \(\beta'\) and the third-order relative nonlinear coefficient \(\delta'\) in the far-field region are a good linearity for the propagation distance.

3. Materials and Methods

3.1. Materials

The coating specimen for the experimental study of the relationship between ultrasound characteristic parameter and stretching stress is shown in Figure 1. The high-temperature alloy steel with a thickness of 2 mm is selected as the substrate material, the hardness is 81HRB, and the yield limit is 365 MPa. The dimensions of specimens for testing are shown in the Figure 1. The \(\text{Al}_2\text{O}_3\) coatings with 100 mm × 25 mm × 0.3 mm are prepared for the plasma spraying of 60-mesh sand blast by following the substrate pretreatment process.

![Figure 1. Dimensions of specimens for testing.](image)

3.2. Nonlinear Experimental System

The ultrasonic nonlinear testing system is made up of the RITEC advance measurement system, a Tektronix DPO 4104B digital phosphor oscilloscope, the exciting and received transducers, and a computer as shown in Figure 2. The center frequency of exciting transducer is 20 MHz and that of the received transducer is 20 MHz. The RITEC advance measurement system produces the exciting wave (shown in Figure 3), which is a sinusoidal tone burst of a 20 MHz single frequency. The interactions between the ultrasonic
wave and the specimen result in distorted waves and nonlinearity. The received wave is shown in Figure 4.

Figure 2. Block diagram of the ultrasonic nonlinear testing system.

Figure 3. Time domain and amplitude frequency diagrams of exciting signal. (a) Time domain diagram; (b) Amplitude frequency diagram.

Figure 4. Time domain and amplitude frequency diagrams of received signal. (a) Time domain diagram; (b) Amplitude frequency diagram.
3.3. Relative Measurements of Nonlinear Parameters

The WDW-E200 universal tensile tester is used to load the specimens. Measurements consist of 21 individual measurements at stress levels ranging from 0 MPa to 400 MPa in increments of 20 MPa. After each stress loading, the specimen is unloaded, the nonlinear ultrasound system is used to make measurements at five points successively, and the average value is taken. The aluminum oxide coating/high-temperature alloy steel belongs to the brittle coating/toughness matrix system. Hence, given that the elasticity moduli of the two are different, the bond interface can produce shear stress, and the bonding condition of the interface is reduced along with the increase of stretching stress. Therefore, this experiment represents the changes of coating bonding condition with the changes of stretching stress. The study also investigates the relationship between the ultrasound characteristic parameter and stretching stress, on the basis of which the parameters that are sensitive to bonding conditions are chosen.

4. Results and Discussion

4.1. The Indicator of Material Nonlinearity

It is necessary to verify that the second harmonics measured by the experimental instrument are caused by material nonlinearity rather than false nonlinearity. With the same sample and the propagation distance, the basic harmonic and the second harmonic are read by adjusting the input voltage excited by the sensor. By increasing the driving voltage, observe the relationship between the second harmonic amplitude \( A_2 \) and the square value of the basic amplitude \( A_1^2 \), as shown in Figure 5. Through linear fitting, \( A_2 / A_1^2 \) remains constant for different voltages at the same propagation distance, indicating a linear relationship. This verifies that the high harmonics measured by the ultrasonic nonlinear testing system are an indicator of the material nonlinearity.

![Figure 5. Relationship between the second harmonic and the square of the fundamental amplitude for different voltage levels.](image)

4.2. The Specimens for Tensile Stretch Testing

Tensile experiments are carried out on the specimen. According to the generation of interface cracks, the whole tensile process is divided into four nodes, as shown in Figure 6. Figure 6a shows the complete interface of the specimen coating. When the stretching stress is loaded to 260 MPa, the margins of the coating interface first produce the transversal cracks pointed by the red arrows in Figure 6b. With the increase of stress, the transversal cracks are produced continuously. When the stretching stress reaches 320 MPa, as shown in Figure 6c, the transversal cracks form larger macro cracks towards the center, the local
delaminating phenomenon appears in the center. Finally, when the stretching stress is larger than 320 MPa, as shown in Figure 6d, the coating falls off in large areas.

![Figure 6. Stretching Process of Specimen. (a) Complete coating interface; (b) Stress: 260 MPa; (c) Stress: 320 MPa; (d) Coating falls off in large areas.](image)

4.3. Results for Relative Measurements of Nonlinear Parameters

Based on the fundamental current amplitude $A_1$, second harmonics amplitude $A_2$, and third harmonics amplitude $A_3$, the second-order relative nonlinear coefficient $\beta'$ and the third-order relative nonlinear coefficient $\delta'$ can be calculated. Then, the values of $A_2$, $A_3$, $\beta'$, $\delta'$ of each measuring point are normalized, and then their relationships with the stretching stress are drawn into the curve. Figure 7 shows the curves of the relationships between nonlinear parameter and the stretching stress of the specimen.
Figure 7. Curves of the relationships between nonlinear parameters and stress of stretching specimen. (a) The relationships between second harmonics $A_2$ and stress; (b) The relationships between third harmonics $A_3$ and stress; (c) The relationships between relative nonlinear coefficient $\beta'$ and stress; (d) The relationships between relative nonlinear coefficient $\delta'$ and stress.

Figure 7a,b are the curves of the second and the third harmonics amplitudes $A_2$ and $A_3$, respectively. Meanwhile, Figure 7c,d show the relative nonlinear coefficients $\beta'$ and $\delta'$ against the stress for the stretching specimen. As can be seen, the average values of $A_2$, $A_3$, $\beta'$ and $\delta'$ of the five measuring points generally increase along with the increase of stress; hence, these parameters can reflect the changes of the coating bonding condition. As Figure 7a shows, the curve of $A_2$ against stress is an obvious monotone increase before 260 MPa, which may be due to dislocations and retention in the specimen [9]. Caused by the accumulation of microdefects such as slip bands, the curve of $A_2$ against the stress shows a relatively flat area in the range of 260 MPa to 320 MPa (i.e., corresponding to the period from the time when transverse cracks first appeared in the coating interface of specimen to that when the coating falls off). Whereas the curve of $A_3$ versus stress shows an even wider range (from 180 MPa to 320 MPa) in Figure 7b, this illustrates that $A_3$ is a greater fluctuation than $A_2$ with respect to the stretching stress. After 260 MPa, the nonlinear coefficient of the ultrasound fluctuates greatly. This may be due to the macroscopic crack in the specimen at the late fatigue stage. Thus, the attenuation of the second harmonic is increased [17]. It is observed in Figure 7c that the curve of the second-order relative nonlinear coefficient $\beta'$ versus stress jumps obviously when the coating cracks appear at
260 MPa, and increases monotonically before the coating falls off at 320 MPa. Nevertheless, the curve of the third-order relative nonlinear coefficient $\delta'$ versus stress shows more gradual fluctuations than the curve of third-order linear versus stress in Figure 7d.

In comparison, the fluctuations of curves $A_2$ and $\beta'$ against stress are relatively smaller and approach the linear relationship with stretching stress, indicating that the two parameters of the specimen are more sensitive to the bonding condition. Therefore, from the experiment results and analysis of the specimen, the nonlinear coefficient $A_2$ and the relative nonlinear coefficients $\beta'$ can be selected as the ultrasound characteristic parameters that can be used in evaluating the coating bonding condition.

5. Conclusions

Results show that the relative nonlinear coefficient increases along with the increase of stress. Although the coating at the surface of each specimen does not show obvious changes in appearance in the section where the stress is smaller than 260 MPa, the nonlinear coefficients measured through experiments increase along with the increase in the loading stress. As the coating interface undergoes certain changes with different loading actions, the experimental results of this paper indicate that the nonlinear coefficient changes along with the change of loading stress. Therefore, the evolution condition of the coating injuries can be reflected through a nonlinear coefficient, thus achieving the objective of conducting a nonlinear ultrasound nondestructive evaluation of surface-coating injuries.

It is necessary to further study the nonlinear ultrasonic detection of the performance change of the surface coating under other different loads (such as fatigue loads) and the corresponding working conditions.

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