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Research on Ultrasonic Testing Methods for Adhesion Quality of Ceramic Coatings

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Abstract. Ceramic coatings are widely used in aerospace field as thermal barrier materials for their high-temperature performance. The adhesion quality of ceramic coatings on metallic substrate is an important index for coating performance. The uniformity of coating thickness is also important since it determines the thermal stresses in the coatings, which means the coating performance is closely related with the uniformity. The reflection characteristic of the coating-substrate interface is an essential factor for evaluating the adhesion quality. Since the acoustic impedance of ceramic coatings is very close to that of substrate, it is not easy to distinguish their reflection wave in the received waves. In this paper, the propagation characteristics of ultrasonic wave in coatings with different thicknesses on the substrate were researched using immersion ultrasonic testing with pulse-echo method. An algorithm based on the amplitude and phase of reflection waves was proposed for the evaluation of adhesive quality. The results show that, pulse-echo method is highly reliable for the measurement of coating thickness and that the algorithm is effective for the evaluation of adhesion quality of ceramic coatings.

1 Introduction

Metallic materials, especially steels and aluminum alloys, are now the most widely-used engineering materials in the society for their excellent comprehensive mechanical properties. However, in certain extreme conditions, such as high temperature, strong corrosion and wear, etc., ceramic materials perform better than traditional metallic materials. In order to enhance the performance of metallic materials in extreme conditions mentioned above, ceramic materials are usually sprayed or bonded on the surface of metallic substrate as coatings [1, 2]. The ceramic coatings are usually very thin, in the order of several hundred micrometers, and are made by two main methods, i.e. atmosphere plasma spraying (ASP) and electron beam physical vapor deposition (EB-PVD) [3,4]. In the paper, thin ceramic layers with thickness from several hundred micrometers to several millimeters were bonded on the surface of metallic substrate as coatings.

Several bonding theories have been proposed in the past few decades, including mechanical interlocking theory, adsorption interaction theory, diffusion mechanisms theory, electrostatic forces theory and acid-base interaction theory [5, 6]. The adhesive between coating and substrate can be

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considered as a viscoelasticity model. There are mainly two models used to describe the adhesion properties, namely Maxwell model and Voigt model. In Maxwell model, the adhesive is considered as a spring and a damper in series and is usually used to describe stress relaxation. While in Voigt model, a spring and a damper in parallel and is usually used to describe creep. Both Maxwell model and Voigt model can be generalized by using a group of such models to better the model [7]. For the model of interface of sample, Tattersall [8] proposed a spring-based model. In his model, the interfacial forces are represented by a density of springs between the two media and the springs are light enough to transmit stress instantaneously across the boundary. L. Singher [9] regarded the interface between adhesive and adherend as a spring-mass structure, and proposed a quasi-static approach, in which a density of springs with constant stiffness were used to describe the shear mechanical behavior of the interfacial adhesive-adherend.

These models provide the theory foundation for nondestructive testing (NDT). Several NDT methods are used for the detection of adhesion quality, for example, acoustic emission testing [10], microwave testing [11], infrared testing [12], ultrasonic testing [13], etc. Ultrasonic testing is widely used for its high sensitivity, strong penetrability, wide detection-range and simple equipment. The spring model mentioned above provides an idea to use ultrasonic wave as a tool to test the adhesion quality of interface, since the ultrasonic wave is one kind of mechanical wave. A. Wegner [14] believes the an-harmonic wave is caused by weak bonds and manifests itself at high dynamic strains exerted by the ultrasonic waves, and proposed a distortion factor as a quantitative index for characterizing the adhesion quality of fusion-welded silicon wafers. L. Czarnecki [15] mainly uses the changes of a mean square value parameter (MS) and the value of variation coefficient (CV) of MS to characterize the propagation of ultrasonic waves through the polymer coating and concrete substrate system. S. I. Rokhlin [16] used high frequency pulsed angle beam ultrasonic waves modulated by low frequency vibrations to test the bonded structure, and used parametric and nonlinear mixing frequencies to characterize the adhesion degradation.

In this paper, an immersion-based pulse-echo ultrasonic testing method is used to measure the thickness of ceramic coatings and to evaluate the adhesion quality of samples.

2 Basic theory

As shown in Fig. 1, there are 4 kinds of media on the propagation route of ultrasonic waves, i.e. water, coating layer, bonding layer or adhesive layer and substrate. The thickness of coating layer is much thinner than that of substrate. And the bonding layer is thin enough comparing with coating layer and substrate. The bonding status of the bonding layer affects the propagation a lot. If an immersion ultrasonic transducer is used for ultrasonic testing, water between the transducer and coating not only serves as couplant, but overcomes the influence of the blind area of ultrasonic testing.

Suppose the complex form of the incident wave can be written as

$$s_i(t) = A_i \exp\,[\mathrm{i}\omega(t - x/c_0)] \tag{1}$$

where A_i, ω, and c_0 are the amplitude, angular frequency and velocity in water of the incident wave, respectively, and $x = 0$ at the water-coating interface.

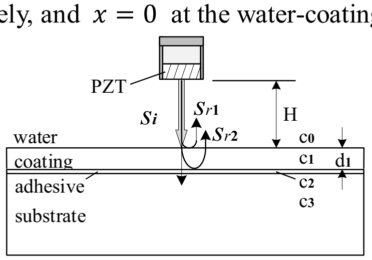


Figure1. Ultrasonic testing for adhesion quality of coating

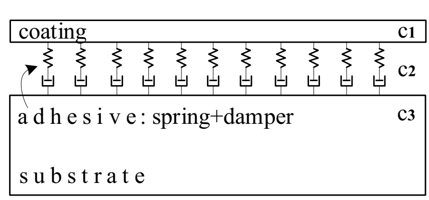


Figure2. Generalized Maxwell model for adhesive layer

Then the complex form of reflection waves from the water-coating interface and coating-substrate interface (adhesive layer ignored) can be written as

$$s_{r1}(t) = A_{r1} \exp\,[\mathrm{i}\omega(t + x/c_0)] \tag{2}$$

and

$$s_{r2}(t) = A_{r2}\exp[\,(\mathrm{i}\omega(t + x/c_0 - 2d_1/c_1)] \tag{3}$$

where d_1 is the thickness of the coating, and other symbols are similar to those in equation (1).

The relationship between A_{r1} and A_i can be determined by reflection coefficient and transmission coefficient of acoustic pressure of the water-coating interface, given by J. L. Rose [17]:

$$r_{01} = (Z_1 - Z_0)/(Z_0 + Z_1) \tag{4}$$

$$t_{01} = 2Z_1/(Z_0 + Z_1) \tag{5}$$

where, Z_0 and Z_1 are the acoustic impedance of water and ceramic coating.

The generalized Maxwell model is used to describe the adhesive layer, which is considered as a series of model of a spring and a damper in series, as shown in Fig. 2.

In this model, the stiffness coefficient K of the spring characterizes the bonding status of the adhesive layer. If K is small, the bonding statue is poor. Vice versa. The viscosity of the damper influences the attenuation coefficient of ultrasonic waves.

According to Q. Zhang [18], there are three types of attenuation in the propagation of ultrasonic waves, i.e. viscous attenuation, heat conduction attenuation and scattering attenuation, and the majority (about 92%) of the attenuation is caused by the last one. The coefficient of scattering attenuation α_s can be described by the equation below:

$$\alpha_s = \frac{8}{3}\pi^4 r^3 f^4/c^4 \tag{6}$$

where, r is the size of the medium particles, c and f are the velocity and frequency of the ultrasonic waves in the medium. From equation (6), we can see that, since the velocity and the particle size are basically constants in certain samples, scattering attenuation is strongly affected by frequency.

Take the scattering attenuation in the coating into consideration, then the amplitude of wave can be written as:

$$A = A_i \mathrm{e}^{-\alpha_s x} \tag{7}$$

where x is the propagation distance in the coating. In the coating, the propagation distance is the thickness of coating d_1, which is quite thin, in the order of 10^{-3}m and the size of the ceramic coating is small, typically 3~5 μm. Then the attenuation factor α_s can then be calculated, which is in the order of $0.1\mathrm{m}^{-1}$. So the factor $\mathrm{e}^{-2\alpha_s d_1}$ is very close to 1, which means the attenuation in the ceramic coating can be ignored.

Since the attenuation in the coating is ignored, the generalized Maxwell model can be simplified to spring model. The adhesion quality can be evaluated by the percentage of energy across the interface. This can be described by adhesion coefficient (CA).

$$CA = \frac{I_{t2}}{I_{t1}} = 1 - \frac{I_{r2}}{I_i(1 - I_{r1}/I_i)^2} = 1 - \frac{\eta_2}{(1-\eta_1)^2} \tag{8}$$

where, $\eta_1 = I_{r1}/I_i$, $\eta_2 = I_{r2}/I_i$.

In equation (8), the symbol r_{12} is directly related to the coating adhesion quality, so are I_{r2} and the adhesion coefficient CA. If the coating is well-bonded, the factor CA is relatively high. If the coating is weakly-bonded, the factor CA is relatively low. Since the parameters η_1 and η_2 can be measured by reflection method of normal incidence, then the adhesion coefficient can be determined.

3 Experimental setup

Two group of samples with different thicknesses were made for the measurement of coating thickness and the evaluation of adhesion quality. The samples consist mainly three layers, namely, coating layer, bonding layer and substrate layer. The coating layer is a relatively thin ceramic layer with thickness ranging from several hundred micrometers to a few millimeter. The bonding layer is an epoxy resin layer much thinner than that of the coating layer. The substrate layer is a stainless steel layer, usually much thicker than that of the coating layer.

The coatings of group I are relatively thick, in the order of a few millimeters, used for characterizing the propagation of ultrasonic waves in the multi-layer samples. The coatings of group II are relatively thin, in the order of several hundred micrometers, and both well-bonded and weakly-bonded samples were made in group II. These samples are used for the thickness measurement and the evaluation of adhesion quality. The mechanical properties of samples are listed in table 1. And the acoustic parameters of each layer of samples are listed in table 2.

Table 1. Coating properties of samples

Sample number	Coating thickness /mm	Bonding status
I-1	2.3	Well
I-2	1.5	Well
I-3	0.67	Well
II-1	0.35	Well
II-2	0.35	Weakly
II-3	0.35	Debonding

Table 2. Acoustic parameters of samples ($\times 10^3$)

Layers	Velocity/ $m \cdot s^{-1}$	Density/ $kg \cdot m^{-3}$	Impedance/ $g \cdot mm^{-2}s^{-1}$
Water	1.50	1.0	1.5
Coating	10.0	3.8	38.0
Adhesion	2.54	1.2	3.0
Substrate	5.72	7.9	45.2

The detection system is shown in Fig. 3. In this system, a normal-incidence pulse-echo mode is used for the measurement of coating thickness and the evaluation of adhesion quality. The immersion piezoelectric ultrasonic probe is excited by an ultrasonic frequency generator that produces pulse square waves. The ultrasonic waves generated by the transducer, which is immersed in water, propagates in the water medium above and around the samples. And the ultrasonic reflection waves received by ultrasonic probe can be transformed into electrical signals, according to the piezoelectric effect. The signals then will be sampled by an oscilloscope, transforming the analog signals into digital signals. The received digital signals then will be analyzed by spectral analysis method and time-frequency analysis method. Finally, the coating thickness and adhesion quality will be evaluated by the characteristics researched in the analyses above.

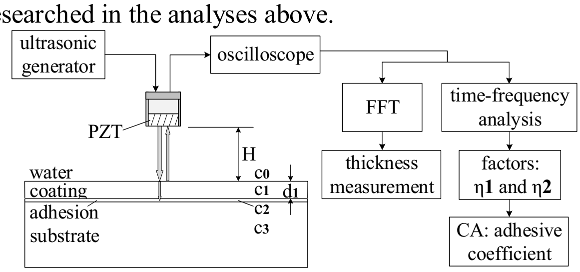
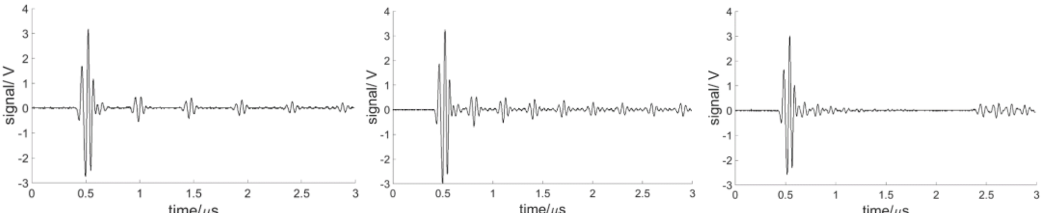


Figure 3. Schematic of detection system

4 Propagation characteristics

When the coating is quite thin, the received waves are a superposition of a series of reflected waves, which is not easy to distinguish. Therefore, thicker samples (group I) are made to research the propagation characteristics of ultrasonic waves. In order to better distinguish the wave series of each pulse excitation, a high-resolution ultrasonic transducer of 15MHz was used.

Fig. 4 shows the ultrasonic waves of samples with different thickness received by an immersion transducer, which also serve as a generator. The received waves consist of a surface echo wave, which is relatively large, and a series of waves that reflects once or more in the coating. The coating thickness of Fig. 4(a) and 4(b) are relatively thick, and every wave received can be clearly distinguished. In Fig. 4(c), the coating thickness is relatively thin, and the received waves are overlapping. It is not easy to distinguish each wave.



a) Coating thickness: 2.3mm b) Coating thickness: 1.5mm c) Coating thickness: 0.67mm

Figure 4. Ultrasonic waves received by transducer

The attenuation of peak-to-peak amplitude of received waves in the ceramic coating can be described by Fig. 5. We can see that the relative amplitude decrease nearly linearly from the second to the sixth received wave, and that the degree of attenuation is not dependent on the coating thickness. Thus, this attenuation is mainly caused by the bonding status.

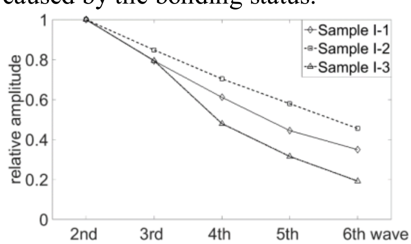


Figure 5. Ultrasonic attenuation in coatings

5 Thickness measurement

The thickness of coating can be measured by time-domain method and frequency-domain method. The time-domain method is commonly used and can be described by the follow equation

$$d = ct/2 \quad (9)$$

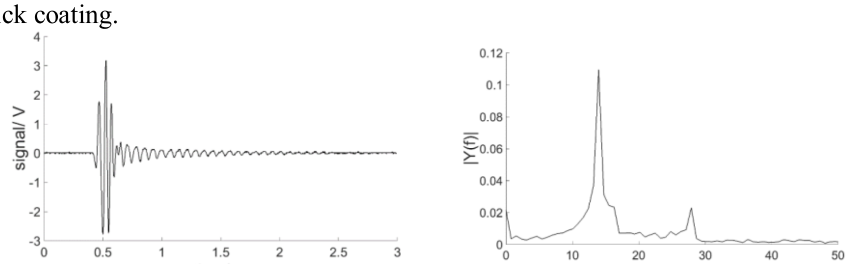
where d is the thickness of coating, c is the velocity of ultrasonic waves and t is the time interval of two peaks in the time-domain waves.

The frequency-domain method is based on the ultrasonic resonance in coating. According to the superposition principle of wave, when the thickness of coating is an integer multiple of half-wavelength, the reflection wave gets an extreme value. Since the ultrasonic probe covers a wide bandwidth, several peaks can be got in the frequency-domain spectrum. This method can be described by the follow equation

$$d = c/2\Delta f \quad (10)$$

where Δf is the frequency interval of two peaks in the frequency-domain waves.

When the thickness is thick, for instance 1 mm or more, both methods are available. When the thickness is very thin, the time-domain method is not available since the waves are superimposed, but the frequency-domain method is still available. The frequency-domain method is mainly based on fast Fourier transform (FFT). The first received wave is ignored in FFT, because it is not associated with the thickness of coating. Fig. 6 shows the time-domain waves and frequency-domain spectrum of 0.35mm thick coating.



a) Waves in time-domain b) Spectrum in frequency-domain

Figure 6. Received ultrasonic waves and spectrum

Apparently, the received waves in Fig. 6(a) are overlapped and it is not easy to calculate the time difference of a round trip in the coating. In the frequency domain, as shown in Fig. 6(b), there are clearly two peaks, the frequency interval of which is 13.96MHz. Then the thickness we get by equation (10) is 0.358mm, which is very close to the nominal value.

6 Adhesion quality evaluation

In order to distinguish the reflection wave of the coating-substrate interface (i.e. the second received wave) from the full received waves, an algorithm based on the amplitude and phase of the received waves is proposed. The received waves, or the synthesized waves mainly consist of the first received wave and the second received wave. According to the synthesis rules of waves, if the amplitudes of the synthesized waves and the first received wave are known and so is the phase difference of the first and the second received waves, the amplitude of the second received wave then can be determined.

Suppose the equations of the full received waves and the first and the second received waves can be written as:

$$s = A\sin(\omega n - \varphi).* \mathrm{Hn}(n - \varphi/\omega) \tag{11}$$

and

$$s_1 = A_1 \sin(\omega n).* \mathrm{Hn}(n) \tag{12}$$

and

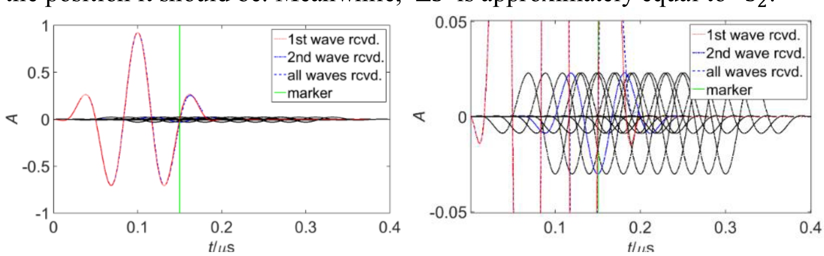
$$s_2 = A_2 \sin(\omega n - \beta).* \mathrm{Hn}(n - \beta/\omega) \tag{13}$$

where A, A_1 and β are known, and A_2 and φ are unknown. As the thickness of the coating has been measured by the means of FFT, then the phase difference of s_1 and s_2, namely β, can be determined. The amplitudes of s and s_1, namely A and A_1, can be determined by pulse-echo method with thick ceramic plates (the same properties as the ceramic coatings) and samples to be detected. The symbol dot with star (.*) denotes the dot product of the vectors. And N is the length of wave series and Hn represents the Hann function:

$$\mathrm{Hn}(n) = \sin^2[\pi n/(N-1)] \tag{14}$$

Since we know the regular form of all the three waves, we can choose a specific feature point as reference point to determine the unknowns. In this paper, the time position of the absolute maximum of wave was chosen as the reference point. The algorithm can be described as follow:

a. Fix the position of s_1 on the *t*-axis, and the time position of the absolute maximum of s_1 is t_{max1}.
b. Mark the time position t_{max2} of the absolute maximum of s_2 on the *t*-axis.
c. Move the wave s along the *t*-axis, and calculate $\Delta s = s - s_1$. Determine the position $t_{max\Delta}$ of the absolute maximum of Δs.
d. If $|t_{max\Delta} - t_{max2}|$ is less than δ (δ is a tiny constant), then the time position of s is exactly the position it should be. Meanwhile, Δs is approximately equal to s_2.



a) A series of Δs from the moving of s b) Details of the series of Δs

Figure 7. Schematic of determination of s_2

In step c, a series of Δs waves were obtained from the moving of the full waves s along the time-axis, as shown in Fig. 7. In these waves, only the one whose time position of the maximum coincides with the marker is exactly the wave s_2 we are looking for. Meanwhile, the amplitude A_2

of wave s_2 is determined. Since A, A_1 and A_2 are known now, we can calculate the adhesion coefficient CA with equation (8).

The absolute maximums of the three samples in group II measured by the algorithm are listed in table 3, as well as the adhesion coefficient CA. The parameter η_1 is about 0.8, according the reflection coefficient of acoustic pressure from water to alumina ceramic coating and the attenuation of ultrasonic waves in propagation. The results obtained by the algorithm above are consistent with expectations: the better the bonding status, the higher the adhesion coefficient CA.

Table 3. Results of evaluation of adhesion quality

Sample number	Absolute maximum	η_1	η_2	Adhesion coefficient
II-1	0.28	0.80	0.014	0.66
II-2	0.32	0.80	0.018	0.55
II-3	0.39	0.80	0.027	0.34

7 Discussion

For the thickness measurement. Generally, an ultrasonic transducer with a higher center frequency has a higher resolution. When the thickness of coating is smaller, transducers with a higher center frequency are needed. In order to obtain a higher amplitude in frequency domain, the center frequency of the transducer should be close to the resonant frequency of the coating, and bandwidth of the transducer should be wide enough for two peaks if possible.

For the evaluation of adhesion quality. If the reflection waves are distinguishable, i.e. the waves are not overlapping, we can use the reflection energy method to evaluation the adhesion quality. However, in many cases, the reflection waves are overlapping (when the coating is very thin), the reflection energy method cannot be used directly. In this paper, the algorithm is proposed to discover the reflection energy hidden in the surface echo wave (i.e. the first received wave).

8 Conclusion

In this paper, the propagation characteristics of ultrasonic wave in coatings with different thicknesses on the substrate were researched using ultrasonic testing with simulation method. An algorithm based on the amplitude and phase of reflection waves was proposed for the evaluation of adhesion quality. From the experimental results, we can conclude that pulse-echo method with FFT is reliable for the measurement of coating thickness and that the algorithm is feasible for the evaluation of adhesion quality of ceramic coatings.

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