Quantitative evaluation of adhesion quality of surface coating by using pulse laser-induced ultrasonic waves

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Abstract

This study aims to evaluate delamination characteristics of surface coating by using strong ultrasonic wave induced by pulsed laser irradiation. Strong tensile stress wave is induced by pulse laser irradiation, and delamination of the coating layer/substrate interface (or spallation) is produced by confined silicone grease breakdown (i.e. ablation). Upon various levels of energy of irradiation laser, the coating delamination and displacement are measured in-situ. Parallel computation of elastic wave propagation using FDTD (finite difference time domain) yields the wave propagation in the specimen, and estimates the interfacial strength with stress wave distribution. The delamination area is visualized by laser ultrasonic wave scanning technique after the laser spallation test. The interaction between displacement waveform and delamination is explored. The present technique may shed some light on various coating depositions for assessing the coating adhesion quality.

Keywords: Pulse laser-induced ultrasonic wave; Spallation; Adhesion quality, Delamination detection; Coating

1. Introduction

Thin films and surface coatings are widely used for wear/contact resistance, which plays a protective role against a soft/ductile substrate. In use, delamination or exfoliation is often experienced due to weak bonding, driven by thermal stress and stress concentration at the edge. Bonding quality is always important and such material integrity imposes a challenge during the process of deposition of films and coatings. Quantitative evaluation of bonding quality through effective and rapid mechanical testing is strongly demanded in industry. An overview of several adhesion testing methods including peel, stud-pull, scratch, bulge, and four-point bending was given by Lacombe [1]. In general, peel tests imposes a high tensile stress to the coating, which may rupture the coating prior to interfacial delamination. In a stud-pull test, a rigid metallic stud is bonded to the target coating with epoxy and then pulled at a controlled rate until interfacial failure occurs; however, if the adhesion of coating is strong, stud separation may occur before the desired interfacial delamination. Interfacial fracture toughness may also be extracted from four-point bend [2, 3], blister [4] or superlayer [5-7] tests. The effectiveness and repeatability of the test are strongly dependent on generation and propagation of a pre-crack along the interface of interest. Specimen preparation is often time consuming due to the difficulties in introducing the pre-crack. Blister and superlayer techniques also require significant sample preparation and are not practical for rapid interface characterization.

The laser ultrasonic wave and laser spallation technique [8-27] appears to be a promising way of fast and efficient measurement of interfacial strength. High amplitude laser-induced acoustic stress pulses are employed to dynamically load a film/coating interface. Vossen [22] initially developed the spallation technique to estimate the bond

strength of millimeter thick aluminum (Al) films on substrates using a high energy laser-induced acoustic pulse loading. An absorbing layer was first coated on the back side of the substrate, and a laser pulse impinging the absorbing layer generates a strong acoustic pulse. The advantage of this technique is use of the pulse wave, inducing the coating delamination in a non-contact manner. Gupta et al. [12-14] and Yuan et al. [15-18] extended Vossen's technique to investigate a wide variety of thin film interfaces. They determined the critical laser power at which the spallation initiated, and extracted the interface strength. One issue is that the measured strength at the level of coating spallation is possibly overestimated, since small delamination first occur prior spallation (i.e. complete delamination and is followed by coating fracture). In fact, most previous studies used high laser power, inducing a large coating spallation clearly (thus, this technique is called laser “spallation” technique)*. In addition, the interfacial strength was estimated through a combination of experimental calibration and corresponding one-dimensional (1D) wave propagation simulations. More recently, Ito et al. [28] conducted simulation of elastic wave propagation using FDTD (Finite Difference Time-Domain method) with two-dimension, to improve the estimation of the interfacial stress. In the light of technical development, the evaluation process for interfacial strength by laser spallation technique is still challenging. Specifically, both detection of delamination and calculation of interfacial stress are critical.

In this work, we improve the laser spallation technique to measure the adhesive strength of hard coating on steel substrate. As a representative material, electroplated Ni coating deposited on stainless steel substrate is used in this study. In order to evaluate

* It may be difficult to stop the coating spallation, with increase in the power of irradiated pulse-laser, since it requires detecting the coating delamination before the spallation with a non-destructive way.

the interfacial strength accurately, new detection techniques for delamination and stress computation procedure are explored. The former one is the use of a correlation factor, which captures the variation of elastic waveform (output wave) since the existence of delamination may affect the output wave. In addition, scanning technique with laser ultrasonic wave (called laser scanning) was also employed to visualize the delamination area. The latter (stress computation) is an improved analysis based on the stress wave propagation by FDTD (similar with Ito et al [28]). The main objective of the present paper is to further improve the accuracy of laser spallation technique for measuring interfacial strength of various coating systems.

2. Experimental setup

2.1 Laser spallation technique

The present setup for laser spallation technique is shown in Fig.1. A pulse laser generated by a Q-switched Nd: YAG laser (Tempest 300, New Wave Research) irradiates onto the break down layer, which is formed on the back surface of the substrate. For the YAG laser, the wavelength is 1064 nm. **In addition, the pulse duration of half bandwidth is about 5 ns (3-5 ns) and the rise time is 5 ns.** The laser is focused to a circular spot of 3.0 mm on the break down layer to induce ablation. This layer is molybdenum disulfide (MoS_2) with viscosity grease. The grease layer with thickness of 50-80 μm is confined by fused quartz plate as shown in Fig.1. The laser-induced ablation of the confined absorbing layer results in the generation of a high amplitude acoustic pulse wave. This elastic wave propagates through the substrate thickness to the coating, and reflects at the free surface of the coating. This leads to reflection and interference of elastic wave, impinging on interfacial stress. When the stress reaches the

interfacial strength of coating, delamination and spallation of the coating may occur.

To estimate the interfacial stress accurately, numerical simulation of elastic wave propagation is carried out (Section 4.1). For this simulation, dynamics of input pulse wave induced by laser ablation is required. However, the ablation is a very complicate physical phenomenon [29, 30][†], and is difficult to clarify theoretically in this study. Therefore, the out-of-displacement velocity at the back surface of laser irradiation epicenter is employed to determine the dynamics of ablation (for input wave). In order to measure the out-of-plane displacement velocity, laser ultrasonic interferometer (Tempo-1D, Bossa Nova Technologies) is used. The spot diameter of this laser is set 0.15 mm. Note the alignment of two laser beams is carefully adjusted. Therefore, the present technique involves two laser systems as shown in Fig.1, in which pulse laser ablation induces high amplitude elastic wave, and travels through the specimen thickness. The out-of-plane displacement at the opposite surface (coating free surface) is measured by laser ultrasonic interferometer. Note that a roller pump is used to circulate MoS₂ grease of ablation layer, such that reproducible laser irradiation can be achieved readily.

2.2 Specimen

The electroplated Ni coating is deposited on a stainless steel substrate. This is a widely used coating and quite representative to demonstrate the versatility of the proposed measurement technique. The austenitic stainless steel SUS304 is employed as

[†] In the previous study [29, 30], physical model of laser ablation has been investigated, and very illustrative to determine the input wave (resulting in elastic wave propagation and coating delamination). However, since the present ablation media is MoS₂ grease with adjustment by machine oil, and may be difficult to be described in the physical model. Thus, we use numerical simulation and output waveform to determine input pulse wave (dynamics) of laser ablation. This procedure will be explained later.

substrate. The substrate is plate shape with 50 mm x 75 mm x 3 mm. Mechanical polishing is carried out in order to make the substrate surface of mirror finish. The thickness of Ni coating is 20 μm and 50 μm . Table 1 shows the deposition condition of Ni electroplating. In addition, material properties (elastic property, density, longitudinal wave velocity) are shown in Table 2. The property of Ni coating is referred to the literature [31]. These material properties will be used for the computation of elastic wave propagation and interfacial stress. Note that the residual stress induced by deposition seems to be minor, since the deposition temperature is not so high (below 50°C) and the post-heat treatment is absent.

3. Experimental result

3.1 Effect of break down layer

Before conducting the laser spallation test, the effect of break down layer (grease layer) is investigated. We prepare a simple plate of SUS304 as specimen and set to the experimental fixture as shown in Fig.1. The energy of pulse laser is set to 30 mJ, and the grease layer thickness is set to 50 μm . Figures 2 show the measurement waveform obtained by the laser ultrasonic interferometer, so that we examine the effect of break down layer. Figure 2(a) is out-of-plane displacement and Figure 2(b) is the velocity of out-of-plane displacement. In Fig.2(a), it is found that the existence of grease layer induces large P-wave and S-wave clearly, compared with the case of no grease layer. Consequently, in Fig.2(b), the result of out-of-plane displacement velocity suggests the importance of grease layer, where the grease layer induces large amplitude of displacement velocity. Therefore, the addition of grease layer is an effective way to induce large stress wave, becoming driving force of delamination.

To convert the stress from the measured out-of-plane displacement, one-dimensional wave equation is first employed (2D simulation will be investigated later). The following Eq.(1) has been used in the previous studies [8, 24] for the laser spallation technique, and will be employed in this study as first approximation .

$$\sigma = \frac{1}{2} \rho C_L \frac{\partial u}{\partial t} \quad (1)$$

where σ is normal stress, ρ is materials density, C_L is P-wave (longitudinal wave) velocity, and $\frac{\partial u}{\partial t}$ is the velocity of out-of-plane displacement (where u is specimen thickness direction and out-of-plane direction). Since Fig.2(b) shows the velocity of out-of-plane displacement, their values convert to normal stress from the Eq.(1). It is observed that the compressive stress wave arrives first, followed by a tensile stress wave[‡]. The important issue is how a large tensile stress wave can be induced with the laser spallation technique.

To generate a large tensile stress wave, the effect of irradiated laser energy on the tensile stress is investigated as shown in Fig.3. In this experiment, two different thickness of grease layer are prepared. Note that the tensile stress is computed from the positive peak value of out-of-plane displacement velocity (see Fig.2(b)). This result suggests that the tensile stress becomes larger with the larger laser energy. It is also found that a thicker grease layer induces a higher tensile stress. Thus, by changing the laser energy (as well as grease layer thickness[§]), the tensile stress for driving force can

[‡] In Fig.2(b), negative side of $\partial u / \partial t$ indicates compressive stress component, while positive one is tensile stress component.

[§] This study did not optimize the grease layer thickness to generate a larger stress wave. Larger

be adjusted.

3.2 Laser spallation test

We conduct the laser spallation test for electroplated Ni coating on SUS304 substrate (in Fig.1). We first examine the Ni coating with 20 μm thickness. The break down layer (grease layer) is 50 μm . The energy of pulse YAG laser changed from 10 mJ to 50 mJ with a step of 10 mJ. At the top free surface of coating, the out-of-plane displacement is measured by laser ultrasonic interferometer (in order to estimate the tensile stress, which will be discussed later). In addition, it is expected that the signal of out-of-plane displacement may possibly capture a delamination existence, since the propagation of tensile stress wave is usually influenced by small defect, i.e. the delamination space induces reflection and diffraction of wave motion. In other words, the propagated waveform is expected to be changed by the occurrence of delamination. Therefore, the measured signal of laser ultrasonic interferometer is carefully investigated. In this study, repeated laser irradiation (with a few times, up to 10 times) is conducted at the same laser energy, so that we investigate the change of waveform signal from laser ultrasonic interferometer. Indeed, this variation of waveform is evaluated by correlation factor. Note that, in this experiment, the diameter of irradiated pulse laser is 3.0 mm, while the spot diameter of laser ultrasonic interferometer is 0.1-0.15 mm. The measurement spot is involved in the irradiated area of pulse laser.

Up to the laser energy of 50 mJ, the measured waveforms do not change. However, the laser energy of 50 mJ indicates a sign of change in waveforms. These waveforms are

stress generation may require a certain volume of grease layer (i.e. grease thickness). However, when the grease layer is too thick, it cannot play a role of a confined space. Thus, there are thought to be appropriate thickness of grease layer.

shown in Figure 4, where several representative waveforms are plotted. The figure also indicates the correlation coefficient at representative number of times. The waveform shows the arrival of the first P-wave (P1) and is followed by second wave of P2. The P2 wave is caused by reflection of P1 wave from the coating surface and the back of the substrate surface. It is observed the P2 waveform is significantly changed with the increase in irradiation number of times. Subsequently, decreasing of the correlation coefficient was also found. Figure 5 shows the change in correlation coefficient as a function of irradiation number. For the reference, the data of 20 and 40 mJ energy is also plotted. The result indicates that low laser energy (lower than 40 mJ) shows a constant coefficient, while the larger energy of 50 mJ shows significantly change with respected to the number of irradiation. Therefore, it is expected that the leaser energy of 50 mJ induces delamination of the coating. However, we observe that the coating surface does not change (there is no crack and exfoliation) by using the optical microscope.

Furthermore, the Ni coating with 50 μm thickness is studied by using similar manner. The grease layer is set to 80 μm to increase the driving force of tensile stress. The laser energy was changed from 10 mJ to 30 mJ with a step of 5 mJ. By investigation of the correlation coefficient (similar with Fig.5), it is expected that the critical laser energy in this case is 25 mJ.

In order to visualize the coating delamination, a new detection method is explored here. We attempt to use a scanning technique based on laser ultrasonic wave propagation. In this technique, low energy laser beam is irradiated on the back surface of substrate (after laser spallation test). This irradiation induces ultrasonic wave, propagating through the specimen, and reaches to the coating surface. The out-of-plane

displacement waveform on the coating surface is measured by laser ultrasonic interferometer. As shown in Fig.4, it is found that the displacement waveform is changed whether delamination occurs or not. Therefore, out-of-plane displacement around pulse laser irradiated area is scanned with the in-plane motion. This measurement relies on scanning of laser ultrasonic waveform (called laser ultrasonic scanning method). The measurement data in the scanning area is automatically accumulated in the in-plane of coating surface, and the software can establish 2D and 3D images. It is thus expected that the delamination area can be derived from the image in a nondestructive and non-contact manner.

In this study, the laser energy is about 20 mJ with no grease layer condition^{**}. The scanning area is about 5 mm and 5 mm with a step of 0.15 mm as space interval. The results of the scanning method for Ni coating with 20 μm thick and 50 μm thick are shown in Figs.6 and 7, respectively. These figures show the contour map of out-of-plane displacement, where we compare the results before the laser spallation test (a) and after the spallation test (b). Note that, for Fig. (b), the laser scanning technique is carried out just after the correlation factor is changed (like Fig.5). For both Fig.6 and 7, bulge-shaped circle (in red) was observed in the specimen after the spallation test. Three dimensional view (in Figs.6(c) and 7(c)) indicates clear delamination area. Thus, this scanning technique can effectively detect delamination area.

To roughly estimate the tensile stress for delamination (at least on the first order of magnitude), the peak value of out-of-displacement velocity is measured (see Fig.2(b)). The positive component can convert to normal tensile stress by using the Eq.(1). For the case of Ni 20 μm thickness, the tensile stress is estimated to be 2.6 MPa, while that of

^{**} We confirm that this condition of laser irradiation energy does not encourage delamination area, due to small tensile stress.

Ni 50 μm thickness indicates the critical stress of a quite close value of 2.9 MPa. Note that the computation procedure does not consider the boundary condition of free surface and interface (i.e. wave reflection/transmission), and also relies on only one-dimensional wave equation (see Eq.(1)). Advance numerical simulation is required to evaluate the interfacial stress more accurately.

4. Computation of critical stress

4.1 Numerical simulation

Most previous studies on laser spallation method used Eq.(1) to estimate interfacial stress, which originated from one-dimensional wave equation with transmitted wave (traveling wave) only [8, 24]. However, when the coating thickness is very small compared with the wave length of traveling wave, complicated wave interference may take place due to reflection wave from free surface and/or interface. This suggested that the Eq.(1) is difficult to estimate interfacial stress accurately, and need to be improved by numerical simulation. In this work, the numerical simulation of elastic wave propagation is carried out using FDTD (finite difference time domain). Indeed, FDTD method calculates the wave propagation in the specimen, by taking an effect of reflection / interference from the boundary (i.e. specimen surface and interface) into account [28], so that it accurately estimates stress wave distribution.

To simulate wave propagation due to a pulse laser ablation, we need to quantitatively characterize the source dynamics of laser ablation, which is used for input source in FDTD simulation. However, physical phenomenon of laser ablation is very complex, and is difficult to be elucidated directly in experiment. Thus, an inverse analysis is carried out by using a transfer function of the specimen, and the out-of-plane

displacement velocity is deduced as the output waveform (experimentally measured by laser ultrasonic interferometer). We first determine the transfer function of the specimen by using FDTD. Subsequently, by using the transfer function and the experimentally obtained out-of-plane displacement velocity, the source dynamics of laser ablation may be identified. Such an input wave (due to a laser ablation) is employed to FDTD simulation, computing the elastic wave propagation in the specimen and determining the interfacial stress.

Figure 8 illustrates the two dimensional model of FDTD. The z -axis indicates the wave propagation direction (along specimen thickness) and x -axis is the direction of specimen width. This is the two layer model, whose mechanical property for FDTD simulation is shown in Table 2. The interfacial condition is employed with the average of two material properties (the coating and substrate). The specimen size and measurement condition are the same with the experimental condition, in which the substrate thickness is 3 mm, and the thickness of Ni coating are 20 and 50 μm . The width is set to 7.5 mm, and both edges are imposed as absorbing boundary condition [32] to mimic semi-infinite plate. Whereas, the top and bottom surfaces (in Fig.8) are imposed with free boundary condition. For the input pulse wave, the spatial distribution of amplitude is assumed to Gaussian distribution, whose diameter is set to 3.0 mm (same with experimental condition). As shown in Fig.8, the measurement point of displacement velocity is on the free surface of coating (in which the laser ultrasonic interferometer is used in the present experiment in Fig.1).

With the above FDTD model, wave propagation simulation for several input waves is carried out, and the transfer function of the specimen $G(w)$ is determined. In other words, the convolution inverse of detected displacement velocity (output

waveform) and the assumed input wave provides the transfer function $G(w)$ for the present specimen. In the simulation, space interval for $\Delta x = \Delta z$ is set to 5 μm and time interval Δt is 0.5 ns. Figures 9 show the input waveform (a), output waveform (b) (displacement velocity) and determined transfer function $G(w)$ (c). These are expressed in the frequency domain, since de-convolution is conducted in the frequency domain. Indeed, this transfer function was investigated for several different input waveforms (by changing the dynamics and shape of wave function), and we confirm that the determined transfer function is universal. Fig.9(c) is thus the transfer function for the present specimen, which will be used to identify experimental input wave (due to ablation) next.

To identify the source dynamics of laser ablation in the present experiment, de-convolution is carried out using the transfer function (see Fig.9(c)) and measurement of displacement velocity (output waveform) as shown in Fig.10(a). In this experimental condition, the thickness of grease layer is 50 μm , energy of pulse laser is 50 mJ and the Ni coating thickness is 20 μm . Here, we refer the output waveform of 1st laser irradiation at critical laser energy (50 mJ). The identified source wave due to laser ablation is shown in Figs.11, indicating frequency domain (a) and time domain in (b). It is found that the present laser ablation induces very fast state variation, i.e. rapid expansion and contraction. Such a strong impact pressure induces large elastic wave propagation, and cases the delamination.

Finally, by using the input wave of ablation (Figs.11) and transfer function (Fig.9(c)), convolution is conducted to obtain the displacement velocity (output wave) as shown Fig.10(b). Subsequently, we compared the output waveform with the experimental one measured by laser ultrasonic interferometer as shown in Fig.10(a).

Note that the experimental waveform is obtained in the test at laser energy of 50 mJ and the 1st irradiation. In Figs.10, this result indicates very good agreement, suggesting that our numerical simulation (including input waveform and transfer function) is acceptable.

4.2 Stress wave propagation and its distribution

By using the above numerical simulation, stress distribution in the specimen is evaluated. The present two dimensional FDTD analysis provides three stress components σ_x , σ_z and $\tau_{xz} = \tau_{zx}$ (see the coordinate axis of Fig.8). Although the driving force for delamination is σ_z dominantly in the laser spallation, other stress components may affect the interfacial fracture due to wave propagation. Figure 12 shows maximum equivalent stress σ_{eq} distribution with respect to wave propagation direction (i.e. along the straight line from the irradiation point of input wave to detected point for output wave, z -axis). In other words, the contour of the maximum σ_{eq} is extracted, and plotted in Fig.12. Since the coating surface is free surface, σ_z component becomes zero. This figure suggested that the maximum σ_{eq} increases up to 100 μm depth. At the interface of 20 μm depth, the σ_{eq} shows 6.6 MPa, corresponding to the critical stress for interfacial fracture. As mentioned above, estimation from Eq.(1) (the one dimensional wave equation model) is 2.6 MPa, which is different from the numerical simulation. This may be due to the fact that during the wave propagation, the reflection from the free surface takes place and interference occurs, leading the larger stress at interface. The one dimensional model Eq.(1) is insufficient to identify the critical interfacial stress, but the present simulation based on FDTD is more reliable to estimate the interfacial stress.

In order to confirm the estimated critical stress, another Ni coating with 50 μm

thick is also investigated. With the similar manner, laser spallation test was carried out and investigate whether delamination occurs or not (see Figs.7(b)(c)). Subsequently, FDTD simulation is carried out to estimate the critical stress. The estimated value is 10.9 MPa, which is roughly the same order of magnitude for 20 μm thickness coating (6.6 MPa). Thus, our method and estimation process are reasonably acceptable for quantitative measurement of coating delamination.

However, it is noted that the present method employs pulse laser-induced ultrasonic wave, which is similar with very high impact loading for coating delamination. This may be high speed interfacial fracture with very high strain rate. Thus, our estimation may not directly correspond to the general interfacial strength under static loading, but quite useful for assessing the quantitative adhesion quality. The effect of strain rate will be addressed in future.

5. Conclusion

We propose an improved framework of employing laser spallation to evaluate the interfacial/adhesion quality for a coating material. Ni electroplating film is employed as a representative coating. In this measurement technique, pulse laser irradiates to the back surface of the specimen. Strong tensile stress wave, impinging on the coating layer/substrate interface, is produced by confined silicone grease breakdown (i.e. ablation) in order to induce the coating spallation. In this study, the spallation test is carried out multiple times at the same level of laser energy. During the test, the out-of-plane displacement (output waveform) on the coating surface is obtained at the irradiation epicenter using laser ultrasonic interferometer. It is found that the coating delamination can be detected by a variation of correlation coefficient, which is

calculated with a series of the output waveform measured by a laser ultrasonic interferometer. In other words, the existence of delamination affects the elastic wave propagation, indicating that the correlation coefficient of series of output waveforms readily identifies the critical laser energy.

The delamination area is visualized by laser ultrasonic wave scanning technique (i.e. called laser scanning) after the laser spallation test. In this technique, the low energy laser beam is irradiated on the back surface of substrate. This irradiation induces ultrasonic wave, propagating through the specimen, and reaches to the coating surface. The out-of-plane displacement waveform on the coating surface is measured by laser ultrasonic interferometer. It is found that the displacement waveform is changed whether delamination occurs or not. In this system, the measured data of displacement waveform are accumulated in the in-plane coating area, and it can establish 2D and 3D images. This image clearly provides the delamination area beneath the coating with nondestructive and noncontact manner. Thus, by using the above correlation coefficient and laser scanning, the critical laser energy can be determined accurately.

In parallel, the computation of elastic wave propagation is carried out using FDTD (finite difference time domain), taking into account the effect of reflection from the boundary (i.e. specimen edge and interface), so that it effectively estimates the interfacial strength based on stress wave propagation. By using the FDTD method, the transfer function of the specimen is first determined, and the source dynamics of input wave due to a laser ablation is estimated, and hence the interfacial stress is deduced quantitatively. In the experiment, the critical stresses of two Ni coatings with different thickness (20 and 50 μm) are obtained. These critical stresses showed roughly the same order of magnitude. Thus, it is concluded that our estimation process may be useful for

assessing the coating adhesion quality.

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Table1 Ni electroplating condition

Composition	g/l	Plating	
$\text{Ni}_2\text{SO}_4 \cdot 6\text{H}_2\text{O}$	250	Temperature	50°C
$\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$	40	pH	4.0~4.6
H_3BO_3	40	Current density	30mA/cm ²

Table2 Mechanical properties of SUS304 substrate and Ni coating

Material	Young's modulus E GPa	Density ρ kg/m ³	Longitudinal wave velocity C_L m/s	Shear wave velocity C_s m/s	Poisson's ratio ν
SUS304 substrate	197	8,000	4,962	3,078	0.3
Ni coating	174*	8,908	4,420	2,740	0.3

*J.-L.Bucaille et al, Thin Solid films 447-448 (2004)