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In-Situ Simultaneous Measurement of Thickness, Elastic Moduli and Density of Thermal Sprayed WC-Co Coatings by Laser-Ultrasonics

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A method for simultaneous nondestructive evaluation of WC-Co coating thickness, elastic moduli, and density is presented. The technique, known as laser-ultrasonics, is used to generate and detect surface acoustic waves in a noncontact and nondestructive manner. The surface acoustic wave velocity dependence on frequency is compared to a model and an optimization procedure is used to evaluate the coating properties. The results obtained demonstrate the ability of the technique to simultaneously determine such properties with a single and possibly in situ measurement.

Keywords elastic moduli, laser-ultrasonics, nondestructive testing, WC-12Co

1. Introduction

1.1 NonDestructive Evaluation of Thermal Spray Coatings

Thermal spraying presents a cost-effective solution for depositing coatings to reduce wear and corrosion and also for producing thermal barrier coatings. High-velocity oxy-fuel (HVOF) thermal spray WC-Co coatings are also an excellent replacement for hard-chromium electroplating that is extensively used by aircraft manufacturers to provide wear and corrosion resistance or to restore dimensional tolerance to components, but that utilizes highly toxic and carcinogenic hexavalent chromium. The nondestructive characterization of these types of thermal spray coatings as well as those used in other applications is of particular importance for understanding and modeling the mechanical and thermo-mechanical behavior of parts and then for the development of life prediction models. Because of their special lamellar structure, inhomogeneous composition and presence of discontinuities like pores and cracks, the elastic properties of thermal sprayed coatings are significantly different from those measured in the same bulk material processed by other means and may vary with the spray conditions employed for deposition. If the assessment of coating properties is done in a nondestructive way, it provides a powerful quality-control tool, since these properties should be directly related to the desired technological performance characteristics like wear behavior and fatigue life.

Methods used for measurement of properties of coatings in nondestructive manner include thermal imaging, infrared spectroscopy, x-ray diffraction, eddy current and ultrasound. Previous works on ultrasound for the characterization of thermal sprayed coatings have shown the great potential of the technique to measure various properties like elastic constants (Ref 1-8), thickness (Ref 1, 4) and adhesion (Ref 5, 9, 10). The ultrasonic waves propagate through the thickness of the coating (bulk waves) or along the coating (surface waves). Surface waves have been used largely for characterization of coatings due to their sensitivity to many properties (Ref 11). Surface acoustic waves can be generated and detected by piezoelectric or electromagnetic transducers and by lasers. When lasers are used, the technique is known as laser-ultrasonics (Ref 12, 13) and is by far the most flexible and powerful technique for surface acoustic wave measurements. Among the advantages of the laser-ultrasonic technique compared to conventional ultrasonics are its noncontact character (it can be applied on hot and moving materials), the ease of changing the generation and detection spot geometries according to the type of wave to be measured, the large bandwidth of signals, and the absence of a coupling medium such as water that could give rise to erroneous measurements on porous coatings.

In a previous paper (Ref 3), the elastic property measurements of WC-Co and TiO_2 coatings with the laser-ultrasonic technique and a comparison of the results with measurements obtained with the Knoop indentation technique were presented. The laser-ultrasonic technique was used to generate and detect surface acoustic waves of both Rayleigh and longitudinal types in the high-frequency limit (waves propagate only in the coating with no

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influence of the substrate). In this case, the elastic constants can be determined if density is previously known or fixed. In the present paper, we limit the problem to WC-Co coatings, but in addition to Young's modulus, the thickness, the Poisson's ratio and the density are also obtained with broadband surface acoustic pulses. The coatings are fully characterized solving an inverse problem with an optimization procedure to minimize the difference between the measured and calculated Rayleigh wave dispersion curves. Although this concept has already been used for the characterization of thermal sprayed coatings (Ref 1, 4, 6) and other coatings (Ref 14-17), to the best knowledge of the authors, it is the first time that elastic properties, thickness, and density are determined with a single measurement. This is made possible due to the wide frequency response of the system, including the low frequencies, the simultaneous measurement of the Rayleigh and the longitudinal waves and the high signal-to-noise ratio of the measured signals. The low porosity of the coating that results in a low attenuation also contributes positively to the success of the technique by providing signals with good signal-to-noise ratio at relatively high frequencies. Also, the dense character of the WC-Co coating allows a special signature for density, otherwise not easy to separate from the other parameters.

1.2 Surface Acoustic Wave Propagation in Coatings

Surface waves have been applied for many decades in nondestructive evaluation of materials mainly due to their capability to sense properties near the surface of the material. The most used wave mode is known as Rayleigh wave and was first described by Lord Rayleigh at the end of the nineteenth century. In its simplest form for isotropic materials, the displacement of a point near the surface of the material due to a Rayleigh wave is an elliptic movement and the amplitude of this displacement decreases rapidly with the distance to the surface, as illustrated in Fig. 1a. This decrease of amplitude with depth is strongly dependent on the wavelength (or frequency). Long-wavelengths (low frequency) penetrate deeper in the material while shorter-wavelengths (high frequency) propagate closer to the surface, making this technique very attractive for characterizing materials with depth dependent properties like coatings. Figure 1b illustrates this occurrence for waves of two different frequencies propagating in a coated material. For an impulsive wave, where a broad range of frequencies are present, propagation in a coated material will show a dispersive behavior with the higher-frequencies having the velocity of the coating and the lower-frequencies sensing both the coating and the substrate.

The velocity dispersion of Rayleigh waves propagating in a coated material can be calculated if the thickness, elastic constants and density of both coating and substrate are known. A theoretical description of Rayleigh wave in such a medium and the related 6×6 matrix for which the determinant is numerically vanished can be found in Ref 11. Figure 2 shows a curve calculated with values expected

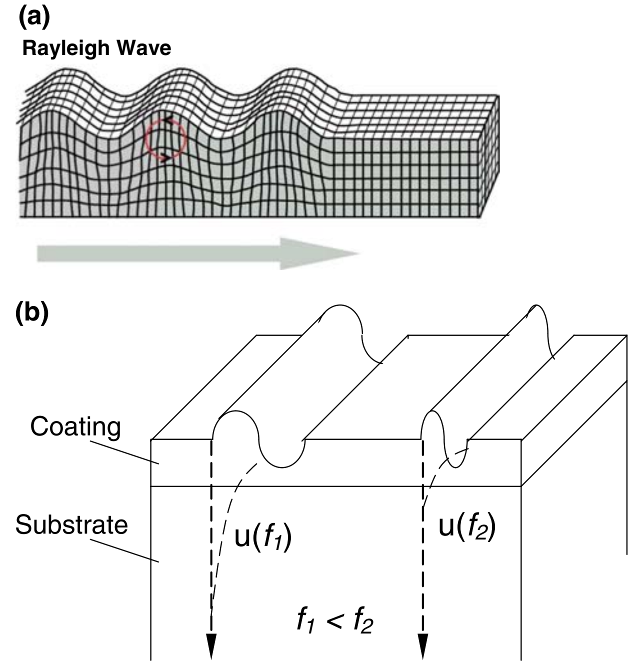

Fig. 1 (a) Surface and sub-surface deformation due to a Rayleigh wave propagation. (b) Rayleigh waves with different wavelengths propagating in a coated material where $u(f)$ illustrates the wave amplitude decreasing with depth

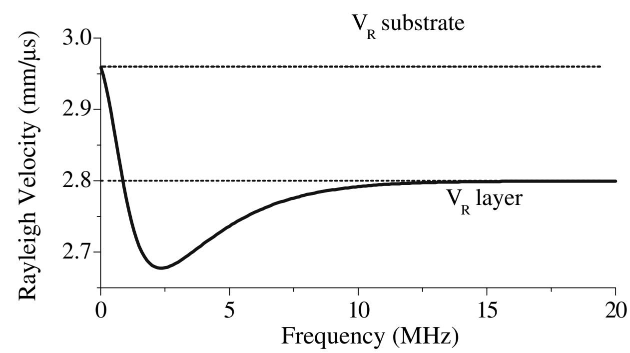

Fig. 2 Rayleigh velocity dependence on frequency as calculated by a model for a WC-Co coating on a steel substrate

for a WC-Co coating on a steel substrate. The WC-Co coating on a steel substrate correspond to the case where the velocity of the coating is lower (less than 10%) than that of the substrate. As expected, the Rayleigh velocity dispersion curve tends to the Rayleigh velocity in steel at low frequency and to the Rayleigh velocity in WC-Co at high frequency. Note that this dispersion curve presents a minimum at intermediate frequencies. This minimum, as shown by the simulations in Fig. 3a, comes from the important difference between the density value of the WC-Co coating and that of the steel substrate. In this figure, all other parameters are kept constant, and changing the density value reveals the special signature associated with this property. Fig. 3b shows the effect of

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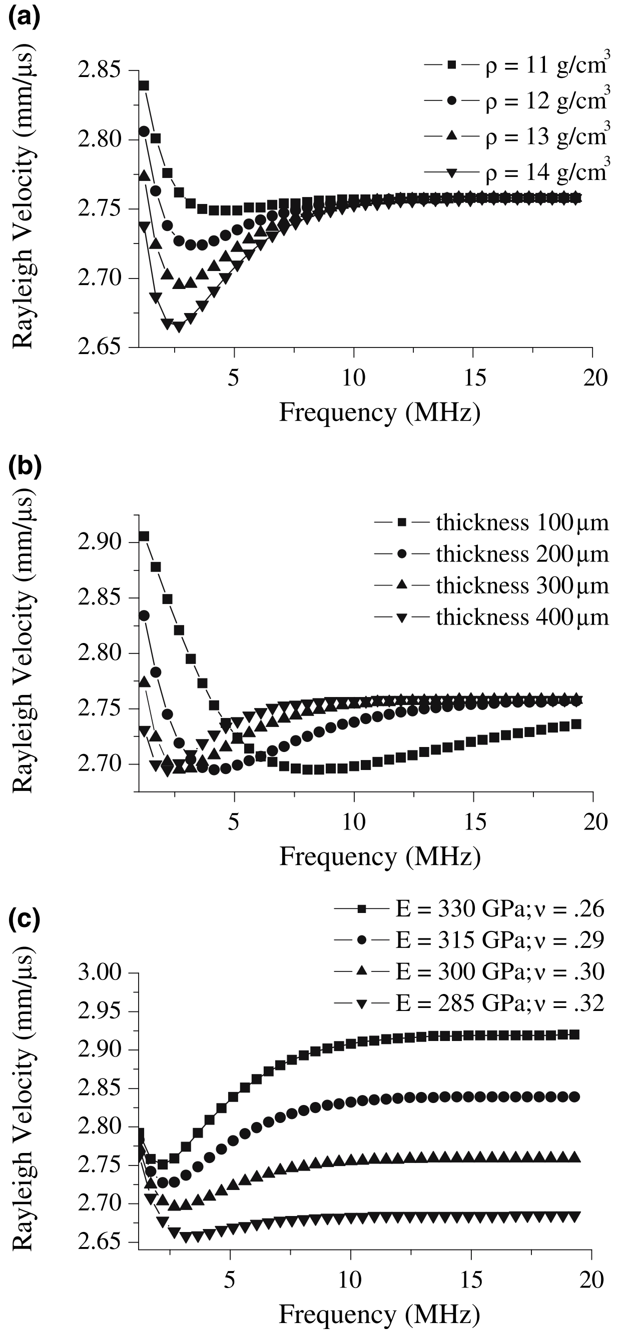

Fig. 3 Simulations illustrating the dependence of the dispersion curve with (a) coating density, (b) coating thickness and (c) coating elastic constants. The values of material properties used in these simulations are close to those of a WC-Co coating on a steel substrate

the thickness that basically scales the horizontal axis. The effect of the elastic constants is more complex, since two independent constants are necessary to characterize an isotropic material. Fig. 3c shows Rayleigh dispersion curves where the Young's modulus and the Poisson's ratio are changed, resulting in changes in the velocity-plateau for higher frequencies.

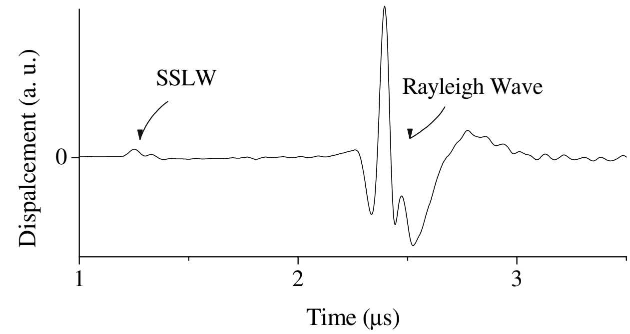

Fig. 4 Typical experimental signal of surface acoustic waves obtained in a WC-Co coating. The Rayleigh wave and the surface skimming longitudinal wave (SSLW) are indicated

However, with only the Rayleigh wave dispersion curve, the measure of two independent elastic constants is not robust. Different combinations of Young's modulus and shear modulus can result in similar dispersion curves. Fortunately, the laser-ultrasonic setup used for measurement of Rayleigh waves also produces a longitudinal mode propagating near the surface as shown in the signal measured in a WC-Co coating depicted in Fig. 4. This wave mode is sometimes called surface skimming longitudinal wave (SSLW) or P-wave. When propagating only or mostly in the coating, it can provide an independent measure that combined with the Rayleigh wave will result in a full characterization of an isotropic material.

2. The Inverse Problem

As shown in Fig. 3, Rayleigh waves are very sensitive to small variations in the properties of the WC-Co coating. But to determine the properties of the coating from an experimental velocity dispersion curve, a numerical inversion problem should be addressed, since no simple analytical expression is available. As already mentioned, the determination of the two independent elastic constants should be greatly improved by an independent measure of the longitudinal wave propagating in the coating. From longitudinal velocity and Rayleigh velocity dispersion data, the optimization process consists in minimizing the function:

$$\underset{h,\rho,V_S}{Min}\left\{\sum_f \left[V_R^{meas}(f) - V_R^{calc}(V_L^{meas}, f)\right]^2\right\} \qquad \text{(Eq 1)}$$

where the unknowns, h, ρ, V_S, to be determined are respectively the thickness, the density and the shear wave velocity, f is the frequency, V_L the longitudinal velocity, V_R the Rayleigh velocity and the superscripts ''meas'' and ''calc'' denote measured and calculated values. The Rayleigh velocity is calculated by vanishing the determinant of a 6×6 matrix as found in Ref 11. This calculation also involves the properties of the substrate, i.e., steel in this work. Density was taken from the literature with a value of 7.8 g/cm^3. The longitudinal and shear velocities were measured on a steel plate without coating in the thickness direction using the same system, with the values of 5.8 mm/μs and 3.2 mm/μs, respectively. Substrate properties have an impact on the Rayleigh velocity of the coating at the lower frequencies. The minimization process is performed on a summation of the measured Rayleigh velocity dispersion data in the available frequency range. The minimization is performed using the Downhill simplex method (Ref 18) and provides optimal values for the three unknowns h, ρ and V_S. With the shear and longitudinal velocities combined with the density, the elastic constants are determined by (Ref 19):

$$E = \frac{\rho V_S^2 (3V_L^2 - 4V_S^2)}{(V_L^2 - V_S^2)} \qquad \text{(Eq 2)}$$

$$G = \rho V_S^2 \qquad \text{(Eq 3)}$$

$$\nu = \frac{(V_L^2 - 2V_S^2)}{2\,(V_L^2 - V_S^2)} \qquad \text{(Eq 4)}$$

where E is the Young's modulus, G the shear modulus and ν the Poisson's ratio. In Fig. 5, an example of the optimization process is illustrated. The experimental data points are shown with crosses, the dotted line is the dispersion-curve calculated with the initial values inserted in the minimization process and the solid line (nearly superimposed to the data) is the dispersion-curve calculated with the values found from the optimization process. Note that the algorithm converges to the optimal values even if the initial values are far from the solution.

3. Experimental Procedure

3.1 Thermal Spraying

The WC-12%Co agglomerated and sintered feedstock was deposited by HVOF on low-carbon steel substrates. The HVOF torch used was JP5000 from Praxair-Tafa, Concord, NH, USA. Eight samples were obtained by variation of the processing parameters, i.e., standoff projection distance, kerosene flow, and oxygen flow. The sample thicknesses were measured by optical microscopy of cross-sectioned samples and are in the range of 190-360 μm. The porosity, measured by image analysis and scanning electron microscopy (SEM), was found to be below 1% for all samples. The samples were also submitted to an abrasion test using standard procedures and equipment (Ref 20).

3.2 Laser-Ultrasonic Experimental Set-Up

The laser-ultrasonic experimental set-up is presented in Fig. 6. A pulsed Nd:YAG laser (3rd harmonic: 355 nm wavelength, 35 ps pulse duration) is employed to generate a Rayleigh wave in the ablation regime. This generation laser, in the ultraviolet wavelength with very short pulse duration is chosen to get good generation efficiency with minimum damage to the sample surface. Although the technique can be considered as nondestructive for practical purposes, it always produces marks of about 10 nm depth on the tested surface due to ablation. For the detection, a long-pulsed Nd:YAG laser (1064 nm wavelength, 50 μs pulse duration) is coupled to an InP:Fe photorefractive interferometer by optical fibers. This photorefractive interferometer provides good sensitivity on unpolished surfaces and good response to lower ultrasonic frequencies.

A line-source and line-detection configuration was chosen in this work to minimize the noise from the coating microstructure. The line dimensions are about 10 mm long per 50 μm wide for both the generation and detection. With this configuration, Rayleigh waves with a 60 MHz bandwidth can be generated and detected in low-attenuating materials. A scanning system with a mirror and a translation table allows the Rayleigh wave to be generated at different locations on the specimen while the line detection is kept fixed. As a result, precise Rayleigh velocity measurements are obtained from different source-to-receiver distances between 1 and 10 mm.

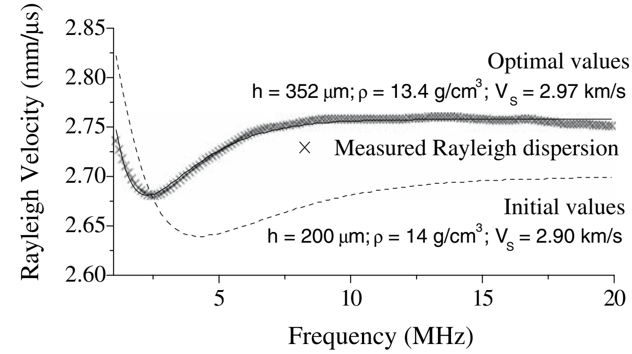

Fig. 5 Optimization process for finding the coating properties by adjusting a theoretical dispersion curve with that obtained experimentally

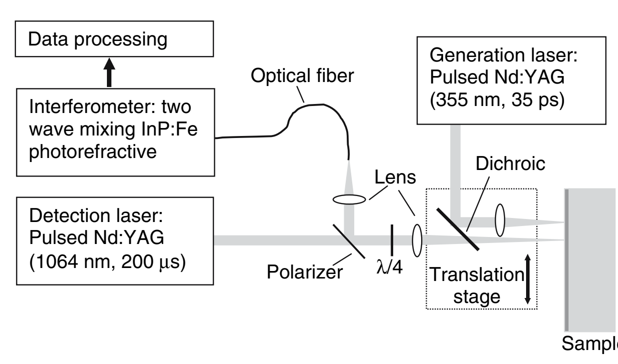

Fig. 6 The laser-ultrasonic setup

4. Results and Discussion

4.1 The Inverse Problem Results

Measurements were performed on the eight samples having WC-Co coatings processed under different conditions. In Fig. 7, the inverse problem results are shown for three of these samples where the squares, circles, and triangles are the experimental data and the solid line are the Rayleigh velocity dispersion curves calculated with the optimal values. The thickness, the density, the Young's modulus and the Poisson's ratio found with the technique are indicated in the figure. The minimization process works very well since the Rayleigh velocity dispersion curves are virtually superimposed to the data. The optimization process was repeated for many propagation distances on each sample and averaged.

4.2 Thickness Measurements via Laser-Ultrasonics

The results obtained with laser-ultrasonic measurements were compared to those from destructive techniques. In Fig. 8, the thickness measured with laser-ultrasonics and with an optical microscope after sectioning the sample are compared for the eight coatings produced in this study. In this figure, the straight line indicates the case of a perfect correlation between the two thickness measurements. It is observed that the data points are well fitted by the straight line, thus indicating an excellent correlation. Note that, for such coatings, the thickness is not perfectly uniform and their variations are indicated by the error bars representing plus or minus one standard deviation. The error bars for the laser-ultrasonic measures are obtained from the variations with different propagation distances. About ten different propagation distances between 1 and 10 mm were used to estimate the accuracy for each sample. They reflect the nonuniformity of the thickness but also the precision of the technique. Considering that the present laser-ultrasonic technique does not use any calibration, the accuracy found in Fig. 8 appears very satisfactory.

4.3 Density and Elastic Modulus Measurements via Laser-Ultrasonics

The coating of one of the eight samples was separated from the substrate for further measurements. Its density, as measured by Archimedes's method, was found to be 13.4 g/cm^3, which is very close to the value of 13.5 g/cm^3 obtained by laser-ultrasonics. The Young's modulus was measured for this sample by a three-point bending test and found to be 318 GPa with an estimated accuracy of ±10%. This is somewhat different than the value of 300 GPa found by laser-ultrasonics, but well within the measurement error of such bending test. The precision on the Young's modulus of the proposed laser-ultrasonic method estimated from different propagation distances is about 2%. This accuracy could not be verified by an independent method due to the intrinsic inaccuracy of available techniques.

The main concern on the application of the model used in the present problem is the assumption of isotropic elastic properties. Previous measurements with Knoop indentation (Ref 3) have shown different elastic behavior when measured on the top surface or in the cross-section of a WC-Co coating. But laser-ultrasonic measurements with longitudinal waves in the plane of the coating (SSLW) and normal to the plane (LW) have shown a minor difference of about 5%, comparable to the accuracy of single LW measurements. The LW velocity measurements were done in the coating detached from the substrate (the same sample used to measure the density and for three-point bending test) and the inaccuracy comes

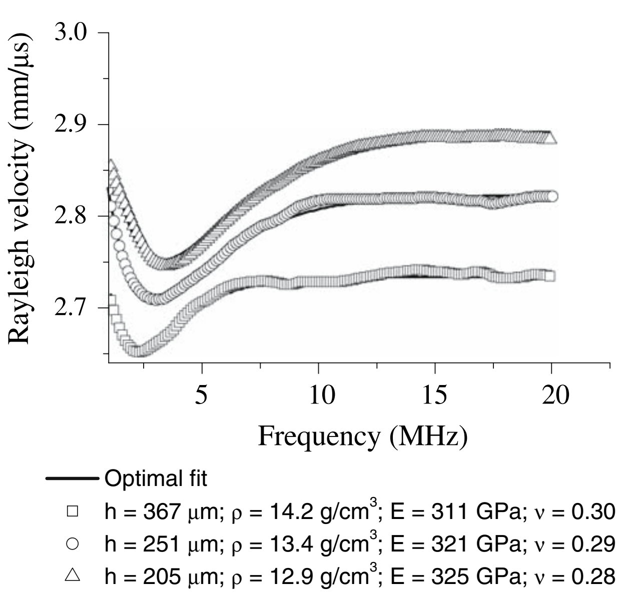

Fig. 7 Examples of optimization results where the symbols represent experimentally obtained velocity dispersion and the solid lines are the optimal fitting curves from the model

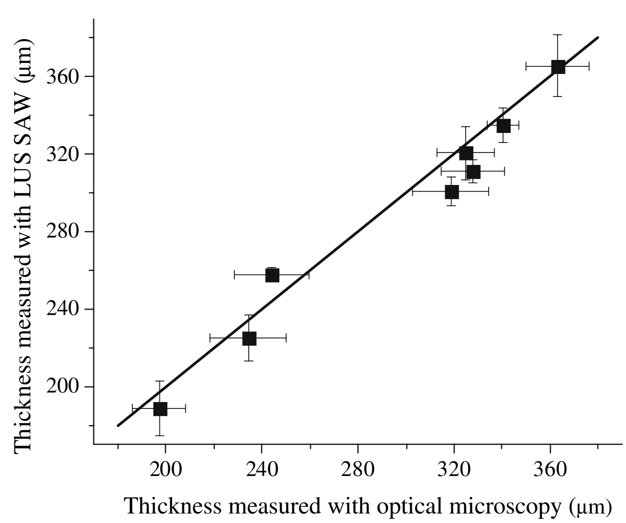

Fig. 8 Thicknesses obtained by laser-ultrasonics compared to that obtained by sectioning the sample and measuring by optical microscopy

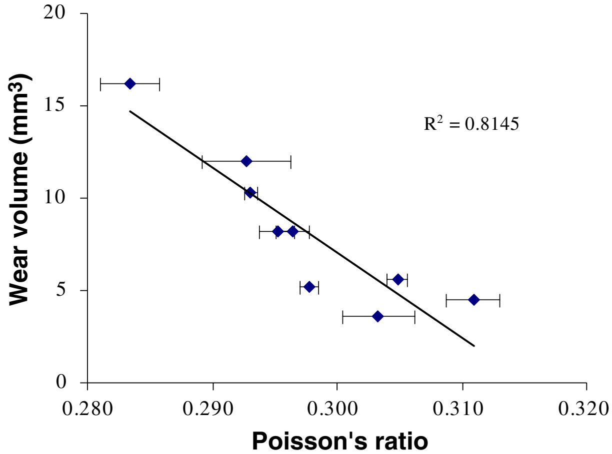

Fig. 9 Correlation obtained between the Poisson's ratio measured by laser-ultrasonics and the wear property

mainly from the separate thickness measurement used to calculate the velocity from the time-of-flight. Also, measurements of surface acoustic waves performed at different angles have shown minor differences of about 1%. This leads to the conclusion that, from the ultrasonic point of view, the tested WC-Co coatings could be considered nearly isotropic. Consequently, the agreement of the thickness and density determined by laser-ultrasonics and those measured by independent methods is not surprising and strongly suggests that the elastic constants determined by laser-ultrasonics also have good accuracy. It should be noticed that considering the coating as an anisotropic material would complicate the inverse problem.

4.4 The Relationship Between the Poisson's Ratio Measured by Laser-Ultrasonics and the Wear Behavior of WC-Co Coatings

The properties measured by laser-ultrasonics were compared with the wear performance on these samples. Figure 9 shows the correlation obtained between the wear volume and the Poisson's ratio measured by laser-ultrasonics. Similar correlations were also found for the other measured parameters like Young's modulus and density, but they are not as good. One reason may be the small range of variations of the elastic properties on the samples measured (less than 10% for the Young's modulus and Poisson's ratio). A set of samples with larger variations of these properties could reveal additional and more clear correlations. Although such kind of correlations can be very valuable, the physical mechanism behind them can be complex and its usefulness limited to a specific type of coating and process.

5. Conclusion

A laser-ultrasonic method is proposed for simultaneous nondestructive evaluation of WC-Co coating thickness, density, Young's modulus and Poisson's ratio. The technique uses lasers for generation and detection of two surface acoustic waves, the Rayleigh wave and the surface skimming longitudinal wave, in a noncontact manner. It also includes a photorefractive interferometer that provides good sensitivity on unpolished surfaces and good response to lower ultrasonic frequencies. The surface acoustic wave velocity dependence over a broad range of frequency is compared to a model with an optimization procedure to determine the coating properties.

Measurements have been performed on WC-Co coatings processed under different conditions. A comparison of thickness measured with laser-ultrasonics and with an optical microscope shows a very good accuracy taking into account the inherent nonuniformity of coating thickness. Also, very good results are obtained for the density by comparison with the method using the Archimedes' principle. For the Young's modulus, the values obtained by laser-ultrasonics are found within the error bar of the three-point bending measurement used for comparison. Indeed, the estimated accuracy of the proposed method on the Young's modulus is about 2%. Regarding the Poisson's ratio, a good correlation was obtained between laser-ultrasonic measurement and wear performance on the tested coatings. It is noted that a set of samples with wider variations of these properties could reveal additional and more clear correlations.

To our knowledge, this is the first time that elastic properties, thickness and density are determined with a single measurement. Both the low porosity of the WC-Co coating that results in a low attenuation and its rather high density producing a particular signature contribute positively to the success of the technique. The results obtained demonstrate the ability of the technique to simultaneously determine such properties with a single and possibly in situ measurement. The application of the technique to other thermal sprayed coatings could also be considered. Although in most cases it will not be possible to measure all parameters simultaneously like in the WC-Co case, the technique should be still a valuable tool to thermal spray coating characterization.

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References

1. A. Abbate, W. Russell, J. Goldman, P. Kotidis, and C.C. Berndt, Nondestructive Determination of Thickness and Elastic Modulus of Plasma Spray Coatings Using Laser Ultrasonics. In D.O. Thompson and D.E. Chimenti (Eds.), *Review of Progress in Quantitative Nondestructive Evaluation*, Vol 18, New York: Plenum, 1999, p 373-380
2. J.A. Slotwinski and G.B. Blessing, Ultrasonic NDE of Sprayed Ceramic Coatings. In D.O. Thompson and D.E. Chimenti (Eds.), *Review of Progress in Quantitative Nondestructive Evaluation*, Vol 15, New York: Plenum, 1996, p 1613-1620

Peer Reviewed

3. R.S. Lima, S.E. Kruger, G. Lamouche, and B.R. Marple, Elastic Modulus Measurements via Laser-ultrasonic and Knoop Indentation Techniques in Thermally Sprayed Coatings, *J. Therm. Spray Technol.*, 2005, **14**, p 52-60
4. D. Schneider, T. Schwarz, H.-P Buchkremer, and D. Stover, Nondestructive Characterization of Plasma-sprayed ZrO_2 Coatings by Ultrasonic Surface Waves, *Thin Solid Films*, 1993, **224**, p 177-183
5. G. Rosa, P. Psyllaki, T. Montesin, C. Coddet, and S. Costil, Laser Ultrasonic Testing for Estimation of Adhesion of Al_2O_3 Plasma Sprayed Coatings, *Surf. Eng.*, 2001, **17**, p 332-338
6. X.Q. Ma, Y. Mizutani, and M. Takemoto, Laser-induced Surface Acoustic Waves for Evaluation of Elastic Stiffness of Plasma Sprayed Materials, *J. Mat. Sci.*, 2001, **36**, p 5633-5641
7. L. Paradis and J.F. Salin, NonDestructive Evaluation of the Mechanical Characteristics of Plasma Sprayed Coatings. In D.O. Thompson and D.E. Chimenti (Eds.), *Review of Progress in Quantitative Nondestructive Evaluation*, Vol 13, New York: Plenum, 1994, p 1887-1894
8. S. Parthasarathi, B.R. Tittmann, K. Sampath, and E.J. Onesto, Ultrasonic Characterization of Elastic Anisotropy in Plasma-sprayed Alumina Coatings, *J. of Therm. Spray Technol.*, 1995, **4**, p 367-373
9. Y. Namba, H. Nakazato, and K. Honma, An Ultrasonic Method to Study the Adhesion of Thermally Sprayed Coatings, *Thermal Spray: International Advances in Coatings Technology*, C.C. Berndt, Ed., 25 May to 5 June 1992 (Orlando, FL), ASM International, 1992, p 241-245
10. H. Hatanaka, I. Kajigaya, and T. Arakawa, Ultrasonic Testing with a Signal Processing Method for Thermal Sprayed Coatings, *Mat. Eval.*, 2004, **62**, p 683-689
11. G.W. Farnell and E.L. Adler, Physical Acoustics, Vol 2, Academic Press, New York, 1972
12. C.B. Scruby and L.E. Drain, Laser Ultrasonics, Techniques and Applications, . , Adam Hilger, Bristol, UK, 1990
13. J.-P. Monchalin, Laser-Ultrasonics: from the Laboratory to Industry, *Review of the Quantitative Nondestructive Evaluation*, D.O. Thompson and D.E. Chimenti, Eds, Vol 23, American Institute of Physics, 2004, p 3-31
14. D. Schneider and T. Schwarz, A Photoacoustic Method for Characterising Thin Films, *Surf. Coat. Technol*, 1997, **91**, p 136-146
15. T.-T. Wu and Y.-H. Liu, Inverse Determinations of Thickness and Elastic Properties of a Bonding Layer Using Laser-Generated Surface Waves, *Ultrasonics*, 1999, **37**, p 23-30
16. B. Knight, J. Braunstein, J.F. Cox, and J. Frankel, Laser-Ultrasonic Characterization of Electrodeposited Chromium Coatings. In D.O. Thompson and D.E. Chimenti (Eds.), *Review of Progress in Quantitative Nondestructive Evaluation*, Vol 18, New York: Plenum, 1999, p 365-372
17. D.C. Hurley, V.K. Tewary, and A.J. Richards, Surface Acoustic Wave Methods to Determine the Anisotropic Elastic Properties of Thin Films, *Meas. Sci. Technol*, 2001, **12**, p 1486-1494
18. W.H. Press, S.A. Teukolsky, W.T. Vetterling, and B.P. Flannery, Numerical Recipes in Fortran, . , Cambridge Univ. Press, NY, 1992
19. D. Royer and E. Dieulesaint, Elastic Waves in Solids I: Free and Guided Propagation, . , Springer-Verlag, NY, 2000
20. Standard Test Method for Measuring Abrasion Using the Dry Sand/Rubber Wheel Apparatus, G 65–91, Annual Book of ASTM Standards, ASTM, 1994, p 246–258