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A practical method for the determination of the Young's modulus and residual stresses of PVD thin films

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Abstract

In a previous article [R.O.E. Vijgen and J.H. Dautzenberg, *Thin Solid Films*, 270 (1995) 264–269], it is shown that the residual stress of thin hard physical vapour deposited (PVD) coatings can be easily measured by the thin foil method. With this method, TiN is deposited on a thin circular stainless steel foil. Sectioning of this foil into narrow strips results in an easily measurable radius of curvature from which the residual stress can be calculated. In this paper, it is shown that the same strip can also be used to determine the Young's modulus of the coating. For this purpose the strip is heated up while its curvature change is observed. To ensure a homogeneous temperature of the strip, oil is used as a heat exchange medium. A calibrated video camera and image processing software are used to measure the curvature of the strip. The results show a linear relationship between the curvature and the temperature of the strip. From this the Young's modulus can be calculated. This method is used to determine the Young's modulus and residual stresses of magnetron sputtered TiN coatings with different deposition parameters. © 1997 Elsevier Science S.A.

Keywords: Young's modulus; Residual stresses; Thin hard physical vapour deposited coatings

1. Introduction

Thin hard physical vapour deposited (PVD) coatings are now widely used in many engineering applications. Depending on the applications different requirements are put on the mechanical properties of the coating, e.g. residual stresses, fracture toughness, hardness, and Young's modulus.

With the magnetron sputtering PVD technique, it is well known that the mechanical properties strongly depend upon the deposition parameters, i.e. bias voltage, reactive gas flow rate, etc. This allows one to tailor those properties according to the value that is desirable for a certain application by adjusting the coating process conditions.

It is generally recognized that performance of a coated system is strongly influenced by the residual stresses in the coating since it directly influences other coatings properties, e.g. strength and adherence. Strictly speaking the residual stresses is in fact the product of the elastic modulus of the coating substrate system and the total residual strain (= sum of the thermal strain and the growth strain). So

residual stresses and Young's modulus are two of the properties of the greatest interest.

There are many ways to measure the residual stresses and the Young's modulus of a coating. The most often used and most practical method is the deflection method [2], where the coating residual stresses are determined by measuring the curvature or deflection of the substrate induced by the residual stresses in the coating. By exploiting the thermal properties of the coating substrate system, the Young's modulus of the coating can also be deduced.

To calculate the residual stresses, the linear relation of Stoney [3] or Brenner and Senderoff [4] is widely used to predict film stress from the measured curvature. However, due to the small strain assumption, those relations are only valid if the measured curvature (deflection) is small. As a rule of thumb, the deflection measured should be at least an order of magnitude less than the substrate thickness. Consequently, high demands are placed on the curvature measuring apparatus regarding the accuracy and also on the substrate in terms of flatness and surface roughness.

Using non-linear relations [5,6] much larger curvature (deflection) is allowed, but the resulting expressions are too complicated to be practical. There are, however particular cases where the linear relations discussed above are

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still valid even for very large curvatures. Vijgen and Dautzenberg [1] carried out large deflection finite element analysis of film stress in thin PVD film and showed that the Brenner and Senderoff formula is still valid if the width-to-length ratio of the strip is small ($w/l < 0.07$). He proposed a simple and accurate method to measure the residual stress of thin PVD coatings. However there still are some drawbacks. First, the given width-to-length ratio (0.07) of the thin strip is of course only valid for his chosen experimental conditions and cannot be considered as a general rule. Second, using the Brenner and Senderoff formula requires the knowledge of the Young's modulus of the coating.

Thus, the purpose of the present work is to extend the method discussed above. Firstly, to provide some general rule regarding the chosen width-to-length ratio of the strip allowing the Brenner and Senderoff formula to be valid, the non-linear relation given by Masters and Salamon [5] is compared to the Brenner and Senderoff formula for the particular cases of very small strips. Secondly, a method is proposed to measure the Young's modulus of the coating. For this purpose the same sample from earlier residual stress measurement is heated while its curvature change is observed. Assuming the validity of the linear relations, the Young's modulus of the coating is evaluated from the residual thermal stressed component using the formula of Chiu [7] or Hsueh and Evans [8]. The finite element calculations were performed to establish the range over which the formula is still valid. Finally, Young's modulus measurements were carried out on different TiN coatings.

2. Theory

2.1. Comparison between linear and non-linear theory

As discussed in Section 1, the Brenner and Senderoff formula yielded excellent agreement with the non-linear finite element calculation for the case of a small coated-thin strip. In this section the same formula is compared to the geometrical non-linear relations of Masters and Salamon [5] to determine whether the same results are predicted as in the first cases. Since it goes beyond the scope of this paper, we have chosen to omit the details regarding the theoretical derivation as well as numerical results of the non-linear theory. Briefly, the theory modelled the film/substrate system as a two layer composite using non-linear plate theory.

Table 1

Physical constants used in calculations which correspond to the realistic value of PVD TiN coatings on a stainless steel substrate

	E (GPa)	$\alpha \times 10^{-6}$ ($^{\circ}\text{C}^{-1}$)	ν
TiN	600	9.4	0.25
AISI 316	192	16.2	0.30

The Young's modulus, E, the Poisson's ratio ν and the linear expansion coefficient α of the TiN coating and the stainless steel substrate.

Fig. 1. Comparison of the calculated curvature obtained by the non-linear model and the Brenner and Senderoff formula as functions of the width-to-length ratio (w/l) for a fixed compressive stress of -5.3 GPa.

The deformations of the film substrate system are approximated by a polynomial with unknown coefficients. The values for these coefficients are found by minimizing the strain energy of the system. This results in highly non-linear equations which describe the equilibrium shape of the plate. Two shapes can be distinguished. In the region of low stress a spherical shape is predicted with equal curvature in the x - and y -direction of the plate. As the stress increases the theory predicts three possible shapes. A spherical shape that is unstable and two ellipsoide shapes that are stable. When other parameters of the plate are considered, e.g. the width-to-length ratio, substrate thickness to the overall plate dimension, etc., a spherical shape and two ellipsoide shapes are predicted by the non-linear theory as a function of the parameters concerned.

In this section, we only considered the results predicted by the non-linear theory for our chosen experimental configuration, namely the relation between the curvature as functions of the width-to-length ratio of the plate. These results are compared to the linear theory of Brenner and Senderoff.

The results presented below are calculated with the second order relations taken from [5] as case 3. The physical constants used in the calculations correspond to the realistic value of PVD TiN coatings on a stainless steel substrate. These constants are depicted in Table 1. Fig. 1 superposes the second order non-linear and linear solutions for the case of TiN coatings on stainless steel substrate. The graph shows x -axis and y -axis curvature of a thin strip as a function of the width-to-length ratio (w/l) at a fixed level of the coating residual stresses, predicted by the non-linear theory. The linear solution is included in Fig. 1 as a dotted line. In the region of small width-to-length ratio ($w/l < 0.03$), the

Fig. 2. Comparison of the calculated curvature between the second order spherical solution and the Brenner and Senderoff formula as functions of the width-to-length ratio for different coating thicknesses. The residual stress is -5.3 GPa.

non-linear theory predicts a spherical shape. In this region, the Senderoff formula yields excellent agreement. Above this value significant differences between the two solutions occur. For $w/l > 0.18$ the solutions of the non-linear theory bifurcated into three solutions in which the spherical shape is unstable and the two ellipsoide shape are stable.

For the purpose of comparison, the linear solution is compared to the spherical non-linear solution for a range of coating thickness; see Fig. 2. The linear solution yields excellent agreement for these cases too. It should also be noted that the limiting width-to-length ratio, i.e. the ratio over which the Brenner and Senderoff solutions is valid, is a variable. This value therefore should be considered from case to case.

2.2. Young's modulus calculation

The relation required to describe the evolution of the curvature of a thin coated strip as function of temperature has been given by Hsueh [8].

$$\frac{1}{R} = \frac{6E_s'E_c't_s t_c (t_s + t_c) (\alpha_c - \alpha_s) \Delta T}{E_s't_s^4 + E_c't_c^4 + 2E_s'E_c't_s t_c (2t_s^2 + 2t_c^2 + 3t_s t_c)} \quad (1)$$

Here E_s' , E_c' the biaxial modulus $E_i' = E_i/(1 - \nu_i)$ of the substrate and coating, respectively, t_s , t_c the substrate and coating's thickness, respectively, α_s , α_c the linear expansion coefficient of the substrate and coating and R the radius of curvature of the strip.

Eq. (1) indicates that the Young's modulus of the coating can be evaluated from the measured change in curvature as a function of temperature if other parameters are known. However, there is a significant difference in the basic

assumption between the proposed experimental technique and the theory. The linear elastic framework above calculates the curvature starting from an initially flat substrate. The Young's modulus measurement start from an initially curved substrate (due to residual stresses).

The validity of the above assumption was checked by the finite element calculations (ABAQUS). In Fig. 3 the change in curvature of the sample from room temperature to 500°C was predicted by the elastic approximation and finite element calculation for different values of initial curvature for the substrate. It is apparent that the FEM predict a lower curvature than the elastic approximation and the differences increased with the increase in initial substrate curvature. However, the difference is small between the two predictions in the temperature range from 20°C to 200°C for all initial curvatures. The experiment therefore will be limited to this temperature range.

3. Experiment

3.1. Film preparation

TiN films were produced with a Teer UDP 350-4-RF system [9] using four unbalanced magnetrons (Type II), a closed magnetic field configuration, an optical gas control and a radio-frequency bias supply. A standard coating cycle consists of a cleaning procedure, a deposition stage and a cooling down period. Details of the apparatus and the film deposition process are given elsewhere [10]. The nitrogen partial pressure is controlled dynamically by monitoring the emission signal of the titanium atoms in the plasma. Nitrogen is admitted through a piezoelectric valve under feedback controlled using the intensity of the Ti plasma

Fig. 3. Comparison of the curvature change versus temperature obtained with the finite element calculation and Eq. (1).

Fig. 4. Experimental set-up for the Young's modulus measurement.

emission ($\lambda = 502$ nm). The set point I is normalized to the maximum I_0 encountered during sputtering of Ti in pure argon. The PEM controller allows the use of a set point anywhere between the range of 20–100%. In this work, the Ti line set point was used as a parameter.

Fig. 5. (a) An example of an original image showing three coated strips. The image was digitized with the frame-grabber card and contains 768×512 pixels. (b) The image of (a) after a sequence of image processing steps. This image shows the neutral line of the three strips. In each strip a different gray level is allocated. The background contains only one grey level.

Fig. 6. The curvature change as a function of temperature for three strips from the same batch.

3.2. Samples preparation

A thin circular foil (AISI 316, $75 \mu\text{m}$) was mounted onto the substrate holder. The substrate was partially covered during the deposition process leaving a circular area open on which the coating was deposited. During the deposition process the substrate was held flat. After the deposition process, a square sample (30×30 mm) was punched out of the center of the foil. Thin strips (30×0.2 mm) were sectioned from the square sample using a shearing machine. The coating thickness was measured with the ball cratering method performed on an additional coated polished AISI 304 piece.

3.3. Stress and Young's modulus measurement

The experimental set-up is depicted in Fig. 4. The strip was heated in a oil-filled chamber. The oil (Dow Corning 550) has a flame temperature of 300°C which sufficiently covered the desired temperature range (20°C – 150°C). The oil temperature was measured using three thermocouples (K-type). The thermocouples were placed sufficiently far from the oil to ensure uniform temperature in the whole oil bad. The strip's temperature was assumed to be equal to the oil temperature. The temperature was increased in a stepwise manner and the curvature was measured at each temperature step. For the curvature measurement, a calibrated CCD camera, a PC equipped with a frame-grabber card and an image-processing software package (TIM Windows) was used. At each temperature step an image was grabbed by the framegrabber card. This results in a digitized 768×512 pixels image that can be stored on the hard disk. An original image, as shown in Fig. 5a contains 256 different gray levels. A sequence of image processing operations was applied to the original image until the neutral line of the

Fig. 7. The Young's modulus and residual stress of TiN versus the I/I_0 set point.

strips was left (Fig. 5b). This neutral line is one pixel width and contains only one gray level. The radius of curvature of the strips are found by fitting a circle through the remaining pixels.

The residual stresses is calculated from the strip's curvature at room temperature. The coating's Young's modulus is evaluated by fitting the Eq. (1) to the measured point.

4. Results and discussion

A typical experiment is depicted in Fig. 6 showing the relation between the strip temperature and the change in curvature for three strips sectioned out of the same coated stainless steel foil ($I/I_0 = 80\%$). The measured curves show both the heating and the cooling curvature indicating that the response of the strips essentially is linear and that a non-linear effect (plastic deformation of the substrate) is absent in this temperature regime or not detectable with this experimental technique. Using Eq. (1) to calculate the modulus yielded an average modulus of 220 ± 20 GPa.

The Young's modulus and the initial stresses as a function of the Ti line set point are shown in Fig. 7. As can be seen, the measurements show the general trend that was expected with two scattered point at a Ti set point of 60% and 50%. The Young's modulus increases with a decrease in Ti set point with a maximum at 55% Ti set point. Beyond this point, the Young's modulus decreased slightly.

In a previous study [10], it was shown that stoichiometric TiN is produced with a Ti line set point of 55%. Thus the highest Young's modulus is expected here; a value of 527 GPa was found. The cause of scattered points in Fig. 7 may be attributed to the deposition process itself.

The internal stress of the coating is calculated with the calculated Young's modulus using the Brenner and Senderoff formula. The stress varied from 4.3 GPa for a high Ti intensity set point to 6.7 GPa at the stoichiometric set point. Beyond this set point, the stress is dropped slightly.

It should be noted that all calculations assume a constant linear expansion coefficient of 9.4×10^{-6} ($^{\circ}\text{C}^{-1}$) for the TiN coating. Since this value linearly enters Eq. (1), serious errors might be introduced if a different value is found for the linear expansion coefficient for the different TiN coatings produced above. The proposed method should therefore be extended to measure this value as well.

5. Conclusion

In this article, the thin strip stress measurement method has been extended. It has been shown that the Brenner and Senderoff relation yields excellent agreement with the non-linear theory up to a certain width-to-length ratio. Although the non-linear theory is probably too complicated to be practical, it can be used to design experiments in the range over which the linear relation is valid. The major advantage of the proposed methods is the occurrence of large curvature, which considerably reduces the efforts involved with the curvature measurements as well as the requirements put on the substrate and the measurement equipment. Furthermore a new method was introduced to measure the elasticity modulus of thin hard coatings as well. This method can be carried out very easily. Also, the possibility to measure several samples at the same time provides a fast and practical tool for the job coating industry for purposes of quality control.

References

- [1] R.O.E. Vijgen and J.H. Dautzenberg, *Thin Solid Films*, 270 (1995) 264–269.
- [2] A.J. Perry, J.A. Sue and P.J. Martin, *Surf. Coat. Technol.*, 81 (1996) 17–28.
- [3] G.G. Stoney, *Proc. R. Soc. Lond. A*, 82 (1909) 172–175.
- [4] A. Brenner and S. Senderoff, *J. Res. Natl. Bur. Stand. (U.S.)*, 42(2) (1949) 105–123.
- [5] C.B. Masters and N.J. Salamon, *Int. J. Eng. Sci.*, 6 (1993) 915–925.
- [6] B.D. Harper and C.P. Wu, *Int. J. Solids Struct.*, 5/6 (1990) 511–525.
- [7] C.C. Chiu, *J. Am. Ceram. Soc.*, 73 (1990) 1999–2005.
- [8] C.H. Hsueh and A.G. Evans, *J. Am. Ceram. Soc.*, 68 (1985) 241–248.
- [9] D.G. Teer, *Surf. Coat. Technol.*, 39/40 (1989) 565.
- [10] R.O.E. Vijgen, *Ph.D. Thesis*, Eindhoven University of Technology, 1994.