

# Modulated IR radiometry as a tool for the thickness control of coatings

S. Chotikaprakhan<sup>a</sup>, F. Vaz<sup>b</sup>, R.T. Faria Jr.<sup>c,d,e</sup>, A.C. Fernandes<sup>b</sup>,  
P. Kijamnajsuk<sup>a,d</sup>, J. Gibkes<sup>d</sup>, B.K. Bein<sup>d</sup>, F. Macedo<sup>e</sup>

<sup>a</sup> Department of Physics, Kasetsart University, 10900 Bangkok, Thailand

<sup>b</sup> Physics Department, University of Minho, Campus de Azurém, 4800-058  
Guimarães, Portugal

<sup>c</sup> Physics Sciences Laboratory, Norte Fluminense State University, 28013-602  
Campos, RJ, Brazil

<sup>d</sup> Solid State Spectroscopy, Physics & Astronomy, Ruhr-University, D-44780  
Bochum, Germany

<sup>e</sup> Physics Department, University of Minho, Campus de Gualtar, 4710-057 Braga,  
Portugal

E-mail: fmacedo@fisica.uminho.pt      sutharat.c@ku.ac.th

**Abstract.** The thickness of coatings can be determined using the data measured by Modulated IR Radiometry for sets of coatings, produced under specific controlled conditions: - Keeping constant all deposition parameters except the deposition time, coatings of approximately constant thermal transport properties, but different thickness are produced. The modulated IR phase lag signals measured for the coatings are calibrated with the help of signals obtained for homogeneous opaque reference samples of smooth surface. Quantitative results for the thermal transport properties are obtained using the inverse solution of the 2-layer thermal wave problem by which direct relations are established between the *relative extrema* of the inverse calibrated thermal wave phase signals measured as a function of the heating modulation frequency and the thermal coating parameters, the ratio of the effusivities coating-to-substrate, the coating's thermal diffusion time, and the coating thickness. The coating thickness values obtained by Modulated IR Radiometry are compared with the values measured by standard microscopic methods, and relative errors of 3 - 4% have been found for the coating thickness of a set of TiCO coatings on steel, presented here as an example.

## 1. Introduction

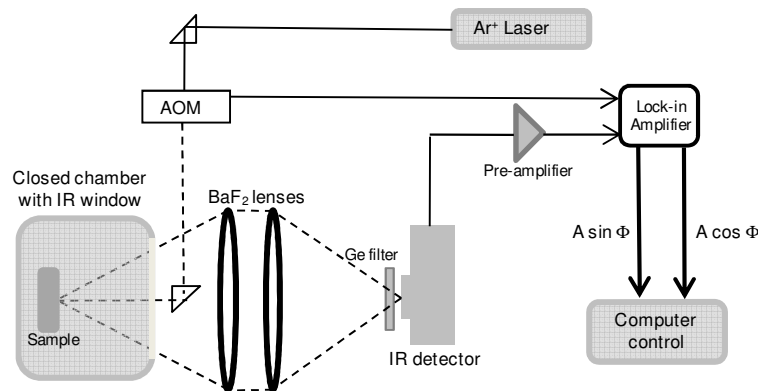
Thermal characterization of thin films and coatings remains an open issue, mainly due to the continuous development of new coating materials and increasing demands of performance in multiple aspects. The ability of photothermal techniques to see “into depth” and the fact that these techniques are non-destructive, represent a high potential of application in different scientific-technological areas, e.g. vacuum technology, welding of steel sheets, and wear of cutting tools [1,2,3]. In a highly competitive market, the potential industrial application of new coating materials [4] strongly depends on a rapid and reliable characterization that can clearly prove the added value of each new coating

material, and thermal analysis is playing an increasing role during the last few years, as the demand for more complex film systems has been growing.

In Sect. 2 a brief description of the setup of Modulated IR Radiometry is given, which can be applied both for high-temperature measurements in vacuum chambers and in coating deposition devices and basics of the quantitative determination of the thermal parameters are reviewed. In Sect. 3 some results are presented, including a few details on the coating deposition process, thermal transport properties as well as thickness determination. In Sect. 4 the main conclusions are presented.

## 2. Principles of modulated IR radiometry – interpretation with respect to thermal parameters and coating thickness

The measurement system (Fig. 1) used for the excitation and non-contact detection of thermal waves mainly consists of four components: - (1) The beam of an Ar<sup>+</sup> laser (Spectra Physics 2000), intensity-modulated periodically by means of an acousto-optical modulator (ISOMET Corp 1205C -2), is used to excite small periodical temperature oscillations at the sample surface. The heating modulation frequency, varied in the range of 0.03 Hz - 100 kHz, allows depth resolved measurements in the range from about 2 mm to thin films of less than 1 μm. - (2) IR optics of short focusing length (≈ 15 cm) consisting of two large-diameter BaF<sub>2</sub> lenses (10 cm) and an IR filter limiting the detectable IR wavelength interval (2 - 12 μm) serve to focus the IR emission of the sample surface on a MCT IR detector (Judson-Infrared JD15-D12). - (3) A two-phase Lock-in amplifier (Stanford 830 DSP) is used to filter the small periodical variations of the IR emission related to the temperature oscillations of the sample - at the modulation frequency of excitation - from the comparatively high radiation and temperature background. From the in-phase and out-of-phase components of the modulated IR signals supplied by the two-phase Lock-in amplifier the thermal wave amplitude  $A$  and phase lag  $\Phi$  relative to the modulated excitation are determined. - (4) The measurement process is computer-controlled, with the amplitude and phase lag data registered as a function of the heating modulation frequency  $f$ .



**Figure 1.** Schematic of the modulated IR radiometry set-up.

Based on the described detection system, thermal wave amplitudes of about 44 μK, 15 μK, and 9 μK have been detected in a high-temperature cell at average background temperatures of 300 K, 400 K, and 500 K, respectively [5]. In the reflection configuration, with the thermal waves excited and detected at the coated surface, depth-resolved measurements of the thermal transport properties of thin films and coatings on thermally thick substrates can be done. To interpret the measured signals quantitatively, these are calibrated with the help of thermal wave signals measured for homogeneous opaque reference solids of smooth surface under the same conditions of heating and detection. The frequency characteristics of the various components of the measurement system, e.g. of IR detector and pre-amplifier, are also eliminated by this calibration process.

The *effusivity ratio*  $g_{cb}=(e_c/e_b)$  and the *thermal reflection coefficient*  $R_{cb}= -(1-g_{cb})/(1+g_{cb})$  coating-to-substrate as well as the coating's *thermal diffusion time*  $\tau_c=d_c^2/\alpha_c$  are determined according to the *Extremum method* [6] as mathematically unique, analytical solutions

$$R_{cb} = \mp \sqrt{\frac{1 - \tan\{0.5 \cdot \arccos[(\tan \Phi_{n \min})^2]\}}{1 + \tan\{0.5 \cdot \arccos[(\tan \Phi_{n \min})^2]\}}} \cdot \exp\{0.5 \cdot \arccos[(\tan \Phi_{n \min})^2]\} \quad (1)$$

$$\tau_c = \{0.5 \arccos[(\tan \Phi_{n \min})^2]\}^2 / (4\pi f_{\min}) \quad (2)$$

of the relative minimum of the inverse calibrated phases  $\Phi_n(f) = \Phi_{ref} - \Phi_c$ . The two equations (1) and (2) allow to determine the thermal transport properties of the coating-substrate system directly from the two quantities  $\Phi_{n \min}$  and  $f_{\min}$ , namely the phase value and the modulation frequency at the position of the relative minimum. As  $g_{cb} = (e_c/e_b)$  only depends on the *thermal reflection coefficient*  $R_{cb}$  coating-to-substrate, the thermal effusivity of the thin film or coating can be calculated, once the substrate's effusivity  $e_b$  is known.

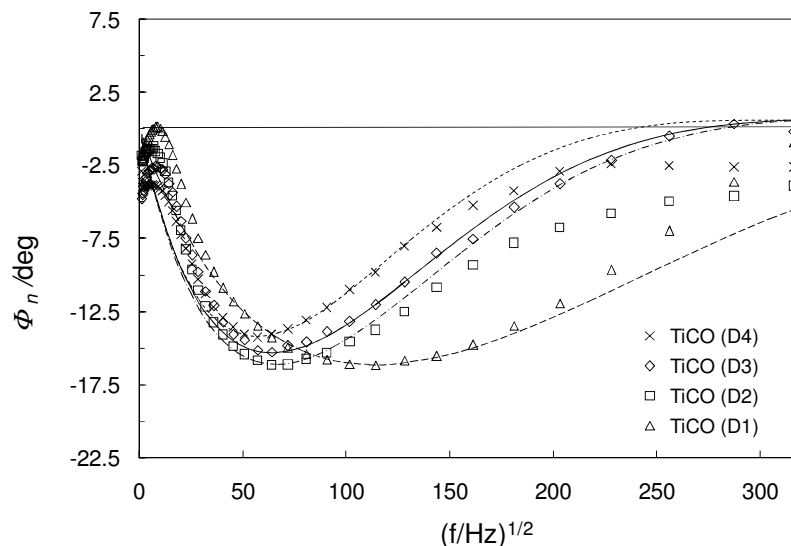
If the film thickness  $d_c$  is known, the thermal diffusivity  $\alpha_c = d_c^2 / \tau_c$  can immediately be calculated, and inversely the film or coating thickness can be determined

$$d_c = \sqrt{\alpha_c \cdot \tau_c} \quad (3)$$

if the thermal diffusivity of a set of thin films of approximately constant thermal properties, prepared under constant deposition conditions, is known. This gives the possibility to control the thickness of thin films and coatings even during deposition, once constant deposition conditions are maintained.

### 3. Results of coating thickness – modulated IR radiometry compared to a destructive method

The results presented here are from a set of titanium oxycarbide (TiCO) thin films, prepared by reactive DC magnetron sputtering on high speed steel (AISI M2) substrates, using a laboratory-size deposition system. Deposition conditions were kept constant [7], with the deposition time the only independent variable parameter. The films were prepared with the substrate holder (in grounded condition) positioned at 70 mm from the target. The depositions were carried out at 200 °C, using a DC current density of 100 A·m<sup>2</sup>. The Ti target had 12 cylindrical carbon pellets (10 mm diameter) placed in the preferential eroded zone, serving as C source. The argon flow was kept constant at 60 sccm in all depositions. The thickness of the coatings varied from about 2 to 6 μm, whereas their composition, structure (all films were crystalline, developing a face centred cubic-type lattice), and thermal transport properties were approximately constant [7].



**Figure 2.** Inverse calibrated IR phase lag signals measured for TiCO films of approximately equal effusivity on high speed steel, compared to opaque two-layer approximations.

The quantitative interpretation of the measured relative minima (Fig. 2) is summarized in Table 1. One of the thicker coatings in Table 1, TiCO(D3), for which the phase signals can well be

approximated by the opaque two-layer model (Fig. 2), was chosen for thickness calibration, taking into account the thickness value obtained by the destructive microscopic measurement [7]. From the known thickness value of that film, the thermal diffusivity was determined and that value  $\alpha_{c0}$  was then used as reference for the whole set of films of approximately constant thermal transport properties, prepared under constant deposition conditions.

**Table 1.** Relative phase minima and thermal parameters for a set of titanium oxycarbide coatings of approximately constant thermal transport properties and differing coating thickness, determined by Modulated IR Radiometry and microscopic destructive measurements (CALOTEST) [7].

Sample	$(f_{\min}/\text{Hz})^{1/2}$	$\Phi_{n\min}$ [deg]	$\tan \Phi_{n\min}$	$R_{cb}$	$(e_c/e_b)$	$\tau_c$ [ $10^{-6}$ s]	$\alpha_{c0}$ [ $10^6 \text{m}^2/\text{s}$ ]	$d_{c\text{MIRR}}$ [ $\mu\text{m}$ ]	Rel error of $d_{c\text{MIRR}}$	$d_{c\text{CALOTEST}}$ [ $\mu\text{m}$ ]
TiCO(D1 $\Delta$ )	113.87	-16.14	0.2894	0.431	0.40	3.39		2.9	3.6%	2.8
TiCO(D2 $\square$ )	66.30	-16.10	0.2886	0.430	0.40	10.01		5.0	3.3%	4.84
TiCO(D3 $\diamond$ )	64.13	-15.33	0.2740	0.410	0.42	10.8	2.50	5.2		5.2
TiCO(D4x)	57.01	-14.19	0.2528	0.380	0.45	13.9		5.89	1.2%	5.82

#### 4. Conclusions and outlook

TiCO films produced by reactive magnetron sputtering on steel substrates, keeping constant all deposition parameters except the deposition time, were studied here using Modulated IR Radiometry. The thermal transport parameters, controlled by means of the relative phase minima giving information on the coatings' thermal effusivity, were found to be approximately constant. From the obtained results we can conclude that this non-contact and non-destructive method gives reliable results for the coating thickness, when applied to thin films of approximately constant effective thermal transport properties. Thus, modulated IR radiometry seems to be a promising tool for the on-line control of coatings, even during the deposition process.

However, some limitations arise when dealing with thin films, for which the relative minima are found at high modulation frequencies, or when the effusivity ratio coating-to-substrate is rather small [7], as is often the case with Silicon as substrate material: - (1) The larger errors observed for measurements on rather thin coatings can be due to the fact that at high modulation frequencies, corresponding to small thermal wave amplitudes, the detectable signals are already affected by IR background fluctuations [5] and that only small deviations found for the inverse calibrated phase lag signals  $\Phi_n(f)$  can lead to larger shifts of the modulation frequency  $f_{\min}$  found for the relative minimum  $\Phi_{n\min}(f=f_{\min})$  [7]. - (2) Another source of larger errors for thin films and coatings can be due to the semi-transparency, affecting thinner coatings more than thicker coatings. Such effects of semi-transparency can be identified by larger positive values of the inverse calibrated phase lag values, considerably exceeding the Zero line  $\Phi_n=0$  at higher modulation frequencies. In such cases, the errors of the thickness measurement based on Modulated IR Radiometry are due to the opaque two-layer model used for interpretation [7], whereas a two-layer model with IR semi-transparency would be more appropriate [8]. - (3) In addition to the limitations for thin coatings at high modulation frequencies, related to IR background fluctuations and to semi-transparency, there are also limitations for coatings of very low thermal effusivity, for which the relative phase minima are measured in the range of  $-45 \text{ deg} < \Phi_{n\min} < -38 \text{ deg}$ . These limitations can mathematically be described by the error propagation of the two-layer thermal wave problem [7] and can physically be interpreted: the modulated heat applied at the coating's surface will easily pass into the substrate of higher effusivity and will be distributed there, whereas only a small thermal wave response can be sensed at the coating surface.

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