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Data Article

Dataset for additional information on the evaluation of thermal properties of thin films by IR radiometry using a comprehensive set of Zr—O—N thin films

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ABSTRACT

This paper provides additional negative data regarding the paper *Evaluation of thermal properties of thin films by IR radiometry using a comprehensive set of Zr—O—N thin films* [1]. In that paper, a matrix of samples was prepared to evaluate the so-called Extremum method for the analysis of Infrared (IR) radiometry data. Such matrix was composed by 3 types of films with 4 different thickness in 3 types of substrates, totalizing 36 samples in total. The data of this paper can be divided into three separate categories: i) lack of adhesion of the films deposited on Teflon, simultaneously to the films deposited on other substrates. ii) Improvement of the signal and signal-to-noise ratio on samples that did not present an extremum (minimum or maximum) using the initial (more conventional) way of measurement. iii) It is also presented a failed fitting of the IR radiometry data created with entangled material parameters. All this data is relevant for researchers devoted to measurement of thermal properties of thin films by IR radiometry that employ the two layer model and Extremum Method.

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Specifications Table

Subject	Materials Science > Surfaces, Coatings and Films
Specific subject area	Evaluation of thermal properties of films by IR radiometry
Type of data	Table Image Graph
How data were acquired	Data was acquired with IR Radiometry, with a laser beam of 532 nm. The set-up consists of a laser Dream Lasers SDL-532-1000T with a maximum output of 1100 mW and a beam radius of ap. 1.5 mm. The acousto-optical modulator AOM-40 from IntraAction can go to up 40 MHz. The reference signal for the AOM is provided by the lock-in amplifier (SR-830). A small prism redirects the modulated beam towards the sample placed in a mechanical sample holder with a hole in the center, in order to avoid any contributions from the sample holder material. Two BaF2 lenses with a focal distance of 15 cm were used in order to collect and focus the IR emission from the sample into the HgCdTe detector (J15D12-M204-S04M-60, from Teledyne Judson) spot. The information from the detector was first pre-amplified and then sent to the lock-in for synchronous detection. All the steps of each experimental run are controlled by dedicated software. Each run took about 15 minutes, ranging from 9.6 up to 100 kHz. Fittings were performed in an ad-hoc Microsoft Excel® sheet.
Data format	Raw Analyzed
Parameters for data collection	The samples were constituted by three types of films deposited on four types of substrate with four different thickness, in order to evaluate the influence of each parameter. All samples were measured by IR Radiometry using with the same operational conditions.
Description of data collection	Laser (532 nm) output power of 600 mW. Less than 100 mW in the sample surface. A lock-in time constant of 300 ms was used. A delay of 3 s was introduced between measurements for lock-in stabilization. Each run takes about 15 minutes, ranging from 9.6 kHz up to 100 kHz.
Data source location	City/Town/Region: Guimaraes, Braga Country: Portugal
Data accessibility	Repository name: Mendeley data Data identification number: https://doi.org/10.17632/z2r9gkndsy.1 Direct URL to data:
Related research article	C.I. da Silva-Oliveira, D. Martinez-Martinez, F.M. Couto, L. Cunha, F. Macedo, Evaluation of thermal properties of thin films by IR radiometry using a comprehensive set of Zr−O−N thin films, Appl. Surf. Sci. 498 (2019) 143,666, https://doi.org/10.1016/j.apsusc.2019.143666

Value of the Data

- This paper shows negative data including: i) lack of film adhesion on Teflon substrates, ii) Re-measurement of inverse calibrated IR phase lag signals for samples that showed no extremum (minimum or maximum) and iii) unsuccessful fittings of the inverse calibrated IR phase lag signals with the so-called extremum method using entangled parameters
- This data is of relevance for researchers interested on measurement of thermal properties on thin films by IR radiometry, and maybe also for those interested in the deposition on thin films on Teflon.
- This data can be used to advance on the measurement of thermal properties of thin films with IR radiometry on 3 directions: i) improve the design of samples, ii) adjust the best conditions for measurement, and iii) to realize the limitations of the Extremum Method and provide hints for its improvement.
- This data illustrate that measurements of IR radiometry vary depending on the acquisition conditions, and how the signal and signal-to-noise ratio can be enhanced.
- This data shows that a well-designed matrix of samples sharing different parameters helps validating the robustness of a model.

1. Data description

The purpose of the original paper [1] was to evaluate the applicability and robustness of the so-called extremum method [2] to measure thermal properties of thin films from data obtained by Infrared (IR) radiometry. To do so, a matrix of different samples was designed, where all the parameters influencing the model (thermal properties of substrate and film and its thickness) were systematically

controlled. Thus, 3 films prepared in 3 different sputtering regimes (Metallic, Reactive and Poisoned) were coated with 4 different thicknesses (1, 2, 3 and 4 hours of deposition time) by reactive magnetron sputtering on substrates of different nature; steel, glass, silicon and Teflon were selected as 4 materials with very different chemical nature. Unfortunately, films deposited on Teflon suffered from systematic lack of adhesion, as it can be noted for the films deposited for 4 hours in Fig. 1 (similar images are provided for films deposited during 1, 2 and 3 hours in the repositorium). Therefore, films deposited on Teflon were not subjected to IR radiometry.

During the measurement of the inverse calibrated IR phase lag signal, we observed that some of the samples presented a lack of extreme (maximum or minimum). Therefore, we re-measured those samples (summarized in Table 1) with different conditions (cf. next section for details), and the data are presented in Fig. 2. Some samples presenting minima (Fig. 2i, j and o) were also re-measured for control (i.e. verify that the data was maintained in the new measurement conditions). It can be seen that in most of cases a minimum appears in the new measurement conditions, although in few cases this is not the case (Fig. 2e, h and p).

The extraction of the thermal parameters of films from the measured IR data was carried out using the so-called extremum method, which is described in detail elsewhere [1,2]. In short, each plot can be simulated using 4 material parameters; 1 of them corresponds to the thermal diffusivity of the substrate (e_s), while the other three corresponds to the thickness (d_c), thermal conductivity (k_c) and volumetric heat capacity ($(\rho C)_c$) of the coating. In the original paper [1], the thermal diffusivity of the substrates and thickness of the films were introduced in the calculations in order to obtain the thermal parameters of the films, which was done fitting each inverse calibrated IR phase lag plot individually. However, considering the nature of the matrix of samples designed for that paper, there are several parameters that would be shared among films, provided that the thermal properties of the films would be the same regardless of thickness and substrate. Such parameter sharing could be used as constraints during the fitting of data, which is summarized in Tables 2–4. Thus, as summarized in Table 2, the thickness of each film (M, R and P) would be controlled just by one parameter (the thickness of the thinnest film), while the others would be that parameter times a factor (2, 3 and 4). Table 3 illustrates that the thermal properties of the films should be equal regardless of the substrate and thickness, and

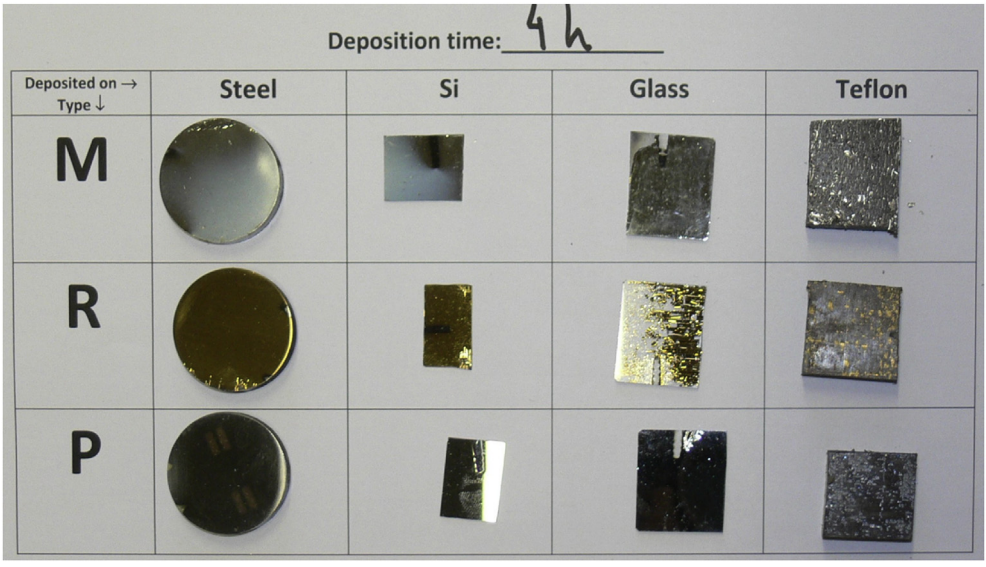

Fig. 1. Films deposited on Metallic (M), Reactive (R) and Poisoned (P) sputtering modes deposited for 4 hours on different substrates. Note the lack of adhesion for films deposited on Teflon.

Table 1
Samples whose inverse calibrated IR phase lag signals were re-measured.

Substrate	Film M				Film R				Film P			
	M1	M2	M3	M4	R1	R2	R3	R4	P1	P2	P3	P4
Steel	✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	✓
Glass												
Silicon	✓			✓	✓			✓	✓			

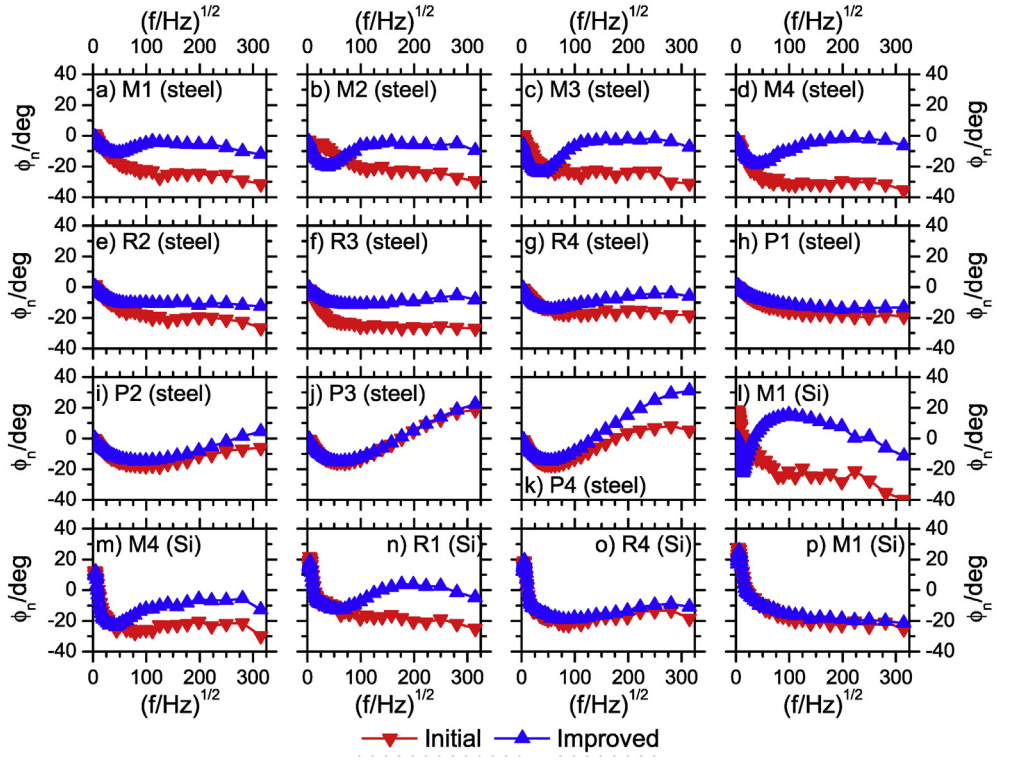

Fig. 2. Re-measurement of inverse calibrated IR phase lag signal for samples summarized in Table 2 in improved measurement conditions.

Table 2
Constraints employed on the thickness of the films (d_c).

Substrate	Film M				Film R				Film P			
	M1	M2	M3	M4	R1	R2	R3	R4	P1	P2	P3	P4
Steel	d_c(M)	$2d_c$(M)	$3d_c$(M)	$4d_c$(M)	d_c(R)	$2d_c$(R)	$3d_c$(R)	$4d_c$(R)	d_c(P)	$2d_c$(P)	$3d_c$(P)	$4d_c$(P)
Glass												
Silicon												

Table 4 indicates that the thermal effusivity of the substrate (steel, glass and Si) is the same regardless the characteristics of film deposited.

As a consequence, we could analyse the 36 IR plots (one per sample) with just 12 parameters: 3 values of e_s (one per substrate) plus 9 values of d_c, k_c, $(\rho C)_c$ (one triad per type of film). This number is

Table 3
Constraints employed on the thermal conductivity (k_c) and volumetric heat capacity of the coating ($(\rho C)_c$).

Substrate	Film M				Film R				Film P			
	M1	M2	M3	M4	R1	R2	R3	R4	P1	P2	P3	P4
Steel Glass Silicon	k_c(M) $(\rho C)_c$(M)				k_c(R) $(\rho C)_c$(R)				k_c(P) $(\rho C)_c$(P)			

Table 4
Constraints employed on the thermal effusivity of the substrate (e_s).

Substrate	Film M				Film R				Film P			
	M1	M2	M3	M4	R1	R2	R3	R4	P1	P2	P3	P4
Steel Glass Silicon	e_s (steel) e_s (glass) e_s (silicon)											

significantly lower than the maximum number of parameters, which would be 144 (36 samples x 4 parameters per sample), or even 72 if we fix *'ex-situ'* the values of thickness of each film and the thermal properties of each sample. In fact, since now we have less parameters than plots to fit, we would be in a situation where we have more data than unknowns, leading to averaged parameters among all the data. Furthermore, if the assumptions made so far would be true, we would obtain the values of thickness of all the films and also the thermal effusivity of all the substrates as outputs of the calculations.

These fittings are depicted in Fig. 3. R4 and P films on glass and M1 on Si have been excluded from the analysis due to the strange shape of the plots, following the same approach than in the reference paper [1]. It can be seen that the fittings are not successful, since the plots from the modelling differ quite a lot from the experimental data. In addition, the thicknesses of the films and thermal effusivity of the substrates differ strongly from the expected values. This lack of agreement is in line with the observed variation of film parameters depending on thickness and substrate when performing individual fittings [1], which invalidates the assumption of constant properties of the film regardless substrate and thickness. One of the reasons is that the microstructure of the films evolves with deposition time. Other 'softer' fittings, e.g. excluding some samples also failed.

2. Experimental design, materials, and methods

Zr–O–N thin films were deposited onto (111) silicon pieces (1.5 cm × 1.5 cm), glass (2 cm × 2 cm), Teflon (2 cm × 2 cm) and mirror-polished high-speed steel (HSS) cylindrical substrates (⌀ = 3 cm × 0.5 cm) by reactive direct current magnetron sputtering in a laboratorial size deposition equipment. The substrates were first cleaned with ethanol and etched in a Zepto Plasma System (Diner) equipped with a 40 kHz/100 W generator. During the etching process, the power used was 100 W and the Ar pressure was approximately 80 Pa. For the depositions, the substrates were clamped in a rotating holder (5 rpm) placed at 75 mm from the magnetron head. The base pressure was always below 2.6×10^{-3} Pa. The depositions were performed by sputtering of a Zr target (99.6% at., 10×20 cm^2) using Ar as working gas and N_2 and O_2 as reactive gases. Three different sputtering regimes (Metallic, Reactive and Poisoned) were chose by proper selection of the flow of reactive gases (low, medium and high flows, respectively). The specific synthesis conditions and characteristics of each sample are summarized in Ref. [1].

Thermal properties determination was carried out using data obtained by Modulated IR Radiometry (MIRR) using laser beam (532 nm) irradiation. The created "thermal waves" were detected with an IR HgCdTe detector, connected to a two-phase Lock-in amplifier (SR830), used to filter and amplify the small periodical variations of the detected IR emission caused by the time and space small temperature oscillations occurring in the samples. Detailed information on the experimental setup can be found

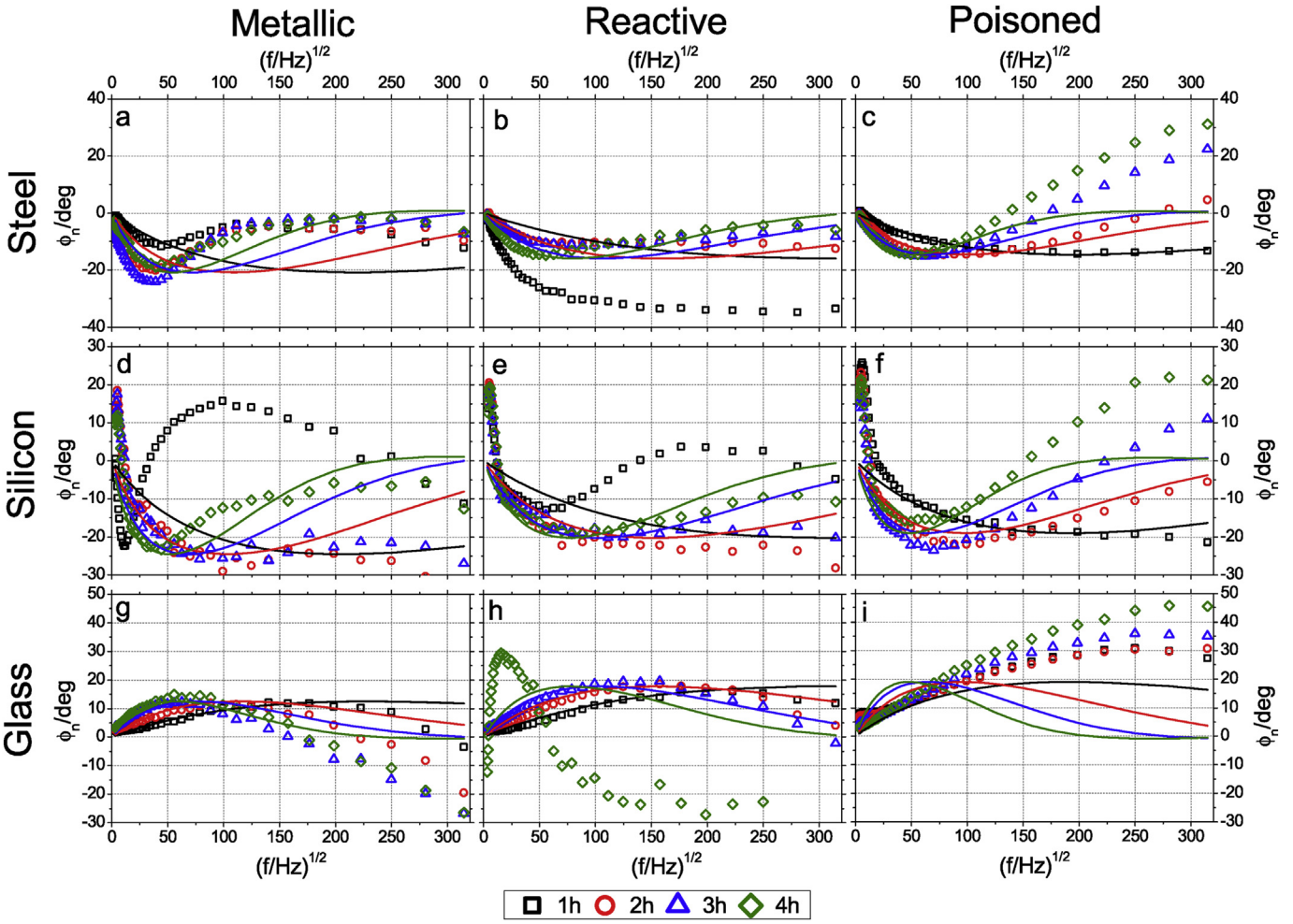

Fig. 3. Inverse calibrated IR phase lag signals measured for three types of films (M, R and P), on three different substrates (steel, silicon and glass) with four different deposition times, using the constraints of parameters described in Tables 2–4. The points and solid lines correspond to experimental measurements and the opaque two-layer approximations, respectively.

elsewhere [3]. For several samples, particularly those deposited on steel and silicon substrates under metallic and reactive regimes, it was observed that the signals detected were too low at high frequencies, compromising the accuracy of the measurements. In many of those samples it was not possible to detect the extremum (minimum) expected, and needed for the calculations.

The main reason for this initial behaviour is related to the specific characteristics of the samples. As a general rule the initial experimental conditions are always very broad in order to protect the samples concerning possible damages from high laser beam power excitation. Once we were dealing with samples with high reflectivity, some optical transparency and using a "low" laser power, all this led us to very weak signals or no useful signals at all. To avoid those problems the experimental conditions were improved after the first measurements. The laser power was slightly increased, the very small prism used to redirect the laser beam towards the sample was deviated from the path of the IR detector and instead of the two Ge filters just before the IR detector we used just one. With this we got a significantly increase in the signal/noise ratio and we were able to get reliable measurements, once our control sample maintained the same behaviour with increasing definition.

The fitting of the inverse calibrated IR phase lag signals to the opaque two-layer approximation was performed using a dedicated excel sheet (enclosed in the repositorium). The general expression of $\Phi_n(f)$ was calculated, and also its difference with the experimental values in the vicinity of the extremum. For each sample, the sum of absolute differences was calculated and added. Finally, that sum was minimized while varying the 12 possible parameters, using the 'solver' tool in Excel.

Acknowledgments

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Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.dib.2020.105291.

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