Resolution of an inverse heat conduction problem with a non-linear least square method in the Hankel space.
Application to photothermal infrared thermography.

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Abstract. Integral transforms (Laplace, Fourier, Hankel) are widely used to solve the heat diffusion equation. Moreover, it often appears relevant to realize the estimation of thermophysical properties in the transformed space. Here, an analytical model has been developed, leading to a well-posed inverse problem of parameter identification. Two black coatings, a thin black paint layer and an amorphous carbon film, were studied by photothermal infrared thermography. A Hankel transform has been applied on both thermal model and data and the estimation of thermal diffusivity has been achieved in the Hankel space. The inverse problem is formulated as a non-linear least square problem and a Gauss-Newton algorithm is used for the parameter identification.

1. Introduction
A large amount of works, combining experimental and mathematical developments, has been devoted to the determination of thermophysical properties, such as thermal diffusivity or conductivity. Two-dimensional models of heat transfer in homogeneous samples have been proposed and several relevant analyses have been realized, for example Balageas [1] or Degiovanni [3], while experimental methods have been developed like “flash”, “pulsed” or ”modulated” methods. Maillet et al [7] investigated heat conduction problems with analytical models using integral transforms and proposed an estimation procedure in the transformed space with contrast determination, yielding opportunities for measurement noise filtering and data reduction. Following this approach, the minimum of deviation between theoretical and experimental curves achieves quickly. The inverse problem becomes well-posed, resulting in better reliability and efficiency for thermal diffusivity estimation.

The present work aims to apply such a procedure to characterize black coatings, which are necessary for thermographic studies on metals, in order to enhance and homogenize the sample absorptivity and emissivity. Two investigations are presented, the first one dealing with a common black coating, the Krylon™ Ultra Flat black paint, and the second with a thin film of amorphous carbon deposited by magnetron sputtering.

For identification procedure, on the one hand, a mathematical model has been developed, based on a Hankel transform of zero order applied to the heat diffusion equation. On the other hand, a discrete Hankel transform has been applied to the experimental temperatures. Finally, a Gauss-Newton parameter estimation procedure in the Hankel space has been proposed. The estimation possibilities

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being essentially influenced by the sensitivity coefficient to each single parameter, a sensitivity study has been achieved for the main ones. The coating thermal diffusivity and absorption coefficient are identified, together with their confidence intervals.

2. Inverse procedure for parameter estimation

For the inverse problem considered here, a Gauss parameter estimation procedure is used, about which complete relevant information can be found in Beck and Arnold [2]. The technique is based on the minimization of the difference between the measured and the corresponding theoretical values, as calculated by the mathematical model.

As far as some parameters of the applied model can be estimated easier than others, the sensitivities to the unknown parameters have to be calculated. On the one hand, they give information on the identification feasibility then, on the other hand, they play an important role in the identification procedure. Sensitivities to the most important parameters, unknown or a priori known, are investigated. The main ones are the thickness, the thermal conductivity, the optical absorption coefficient of the coating and the thermal conductivity of the substrate.

The matrix $X$ of sensitivity coefficients can be written:

$$X = \left[ \begin{array}{ccc} \frac{\partial \bar{T}_1}{\partial \vartheta_1} & \cdots & \frac{\partial \bar{T}_1}{\partial \vartheta_p} \\ \vdots & \ddots & \vdots \\ \frac{\partial \bar{T}_n}{\partial \vartheta_1} & \cdots & \frac{\partial \bar{T}_n}{\partial \vartheta_p} \end{array} \right]$$

(1)

where $\bar{T}$ is the value of the Hankel transform of the temperature, as calculated by the model, $n$ is the number of measured values, $\vartheta_i$ is the $i$th estimated parameter ($i=1,\ldots,p$) and $p$ is the number of parameters under estimation.

A qualitative study has been realized, using the reduced form of the sensibility coefficient, which is usually defined as:

$$X^*_j = \vartheta_i \frac{\partial \bar{T}_j}{\partial \vartheta_i}$$

(2)

These reduced sensitivities are more convenient for comparison, and indicate which parameters could be simultaneously or separately estimated. Their values are calculated by use of an approximate formula:

$$X^*_j = \vartheta_i \frac{\partial \bar{T}_j}{\partial \vartheta_i} \approx \vartheta_i \left( \bar{T}_j (\vartheta_1, \vartheta_2, \ldots, \vartheta_i + \Delta \vartheta_i, \vartheta_{i+1}, \ldots, \vartheta_p) - \bar{T}_j (\vartheta_1, \vartheta_2, \ldots, \vartheta_p) \right) / \Delta \vartheta_i$$

(3)

where $\Delta \vartheta_i$ is fixed here at $10^{-3} \vartheta_i$.

The estimation of $\vartheta = [\vartheta_i]_{i=1}^p$ consists in minimizing the Ordinary Least Square criterion:

$$S(\vartheta) = \sum_{j=1}^{n} \left( T^*_{\text{discrete}} - \bar{T}(\vartheta) \right)^2$$

(4)

where $T^*_{\text{discrete}}$ is the matrix of the discrete values of the Hankel temperature [$n \times 1$], calculated from the measured temperatures. The model solution $\bar{T}(\vartheta)$ being not linear with respect to $\vartheta$, the minimum $\tilde{\vartheta} = \arg \min S(\vartheta)$ is computed according to the Gauss-Newton algorithm. The iterative process is described in [2].
3. Characterization method

3.1. Experimental set-up
The technique applied here to characterize the coating is photothermal infrared thermography under modulated laser irradiation. The coating is deposited on a URB25 steel substrate (thickness: 2.5 mm, density: 8000 kg.m\(^{-3}\), thermal conductivity: \(k = 13\) W.m\(^{-1}\).K\(^{-1}\), specific heat: \(C_p = 480\) J.kg\(^{-1}\).K\(^{-1}\)). The sample surface is heated by a diode pumped YAG laser (DPSS) at the wavelength 532 nm. The modulation of the laser beam is realized by an acousto-optic modulator driven by a sinusoidal function under Labview\textsuperscript{TM} program. For the lock-in procedure, a photodiode gives the reference signal, allowing amplitude and absolute phase measurements. An infrared camera, CEDIP IRC 320-4LW, records series of images of the thermal responses. A home-made software, developed under Labview\textsuperscript{TM}, allows the numerical lock-in detection of real and imaginary parts of the complex temperature.

3.2. Theoretical model
The thermal problem can be written by using the heat diffusion equation for a bi-layer medium and assuming the geometry as 2D axis-symmetrical:

\[
\frac{\partial^2 T(r, z)}{\partial r^2} + \frac{1}{r} \frac{\partial T(r, z)}{\partial r} + \frac{\partial^2 T(r, z)}{\partial z^2} - jw \frac{a}{k} T(r, z) = - \frac{q(r)}{k} \quad (5)
\]

The thermal response can be searched as Green harmonic functions [4,5]:

\[
\frac{d^2 H(\lambda, z; z')}{-\delta(\lambda - \lambda')} = - \frac{\delta(z - z')}{k} \quad (6)
\]

Then, the Hankel transform of the temperature in the layer 1 (black coating) is expressed as:

\[
\overline{T_1}(\lambda, z) = \int_0^l H_{11}(\lambda, z')q_1(\lambda, z')dz' + \int_0^d H_{12}(\lambda, z')q_2(\lambda, z')dz' \quad (7)
\]

The following boundary and interface conditions are considered:

At the surface of black coating (\(z=0\)):

\[
- k_1 \frac{\partial T_1(r, z)}{\partial z} \bigg|_{z=0} = -hT_1(r, z) \bigg|_{z=0} \quad (8)
\]

- At the rear face of the sample (\(z=d\)):

\[
- k_2 \frac{\partial T_2(r, z)}{\partial z} \bigg|_{z=d} = hT_2(r, z) \bigg|_{z=d} \quad (9)
\]

- Continuity of the heat flux at the interface between substrate and black coating (\(z=l\)):

\[
- k_2 \frac{\partial T_2(r, z)}{\partial z} \bigg|_{z=l} = - k_1 \frac{\partial T_1(r, z)}{\partial z} \bigg|_{z=l} \quad (10)
\]

- Perfect contact between the coating and the substrate. So, the contact thermal resistance is considered as negligible and the last boundary condition can be written as:

\[
T_1(r, z) \bigg|_{z=d} - T_2(r, z) \bigg|_{z=l} \quad (11)
\]

The analytical expressions for the Hankel transforms of the source in each layer are:

\[
\overline{q_1}(\lambda, z) = \frac{(1 - \rho)P\beta_1}{4\pi} \exp\left[-\left(\frac{\lambda r_0}{2}\right)^2\right] \exp[-\beta_1 z] \quad (12)
\]

\[
\overline{q_2}(\lambda, z) = \frac{(1 - \rho)P\beta_2}{4\pi} \exp[-\beta_1 z] \exp[-\beta_2 (z - l)] \exp\left[-\left(\frac{\lambda r_0}{2}\right)^2\right] \quad (13)
\]

4. Results and discussion

4.1. Influence of the deposit thickness on the estimation results
Two sensitivity studies have been achieved. The first one has been realized for paint parameters and the second one for amorphous carbon data.

The sensitivity curves (figures 1a and 1b) for the black paint show that the reduced sensitivities on the imaginary part are much weaker than those on the real part. Among the four parameters selected ($k_1$: thermal conductivity of black paint, $\beta_1$: optical absorption coefficient of black paint, $l$: thickness of black paint and $k_2$: thermal conductivity of substrate), it appears that the deposit thickness is the most influent on the estimation results. Thus, to improve the accuracy of estimation, this parameter needs to be measured. Practically, the coating thickness, here about 20$\mu$m, is taken as an average of 10 measurements, given by a POSITECTOR 6000 (allowing a resolution of 1$\mu$m). Moreover, the curves illustrate that the sensitivities to parameters $k_1$ and $\beta_1$ are not correlated on the spatial frequency domain. Then, a simultaneous identification becomes possible, whereas in the real space, it did not succeed.

Regarding the amorphous carbon thin film, the thermophysical properties have been found in the literature ($k=1.6 \text{ W.m}^{-1}\text{.K}^{-1}$, $\rho=1950 \text{ kg.m}^{-3}$, $C_p=710 \text{ J.kg.K}^{-1}$) [6]. The thickness of the deposit is quasi-constant, around 1$\mu$m, all across the sample surface and the accuracy on this value is 0.05$\mu$m.

The sensitivity study shows that, around this coating thickness, the model is not sensitive to the coating properties but to the substrate properties (figures 2a and 2b). Then, this technique should allow the characterization of the substrate, without knowing precisely the thermal properties of the coating.
4.2. Optical and radiative characterization of the coatings

Samples have been tested with different thicknesses of Krylon paint but no variation of emissivity was found for thicknesses between 20 and 40 μm. Consequently, we considered the Krylon™ films as opaque.

For the amorphous carbon film, the optical index necessary for the radiative characterization depends on the depositing conditions. Two important parameters are the magnetron power ($P_{\text{magnetron}} = 5\text{ kW}$) and the sample temperature during the process ($T_{\text{sample}} = 400\text{°C}$). Some optical index values were found in the literature [8, 9 and 10].

In the visible spectrum, the optical indexes are $n = 1.8$ (refraction index) and $k = 0.72$ (extinction index). With these values, and for a thickness of 1 μm, the optical thickness calculation shows that the coating can be considered as opaque. In the far infrared spectrum, the optical indexes become $n = 2.38$ and $k = 0.65$. The layer appears here to be semi-transparent. Then, a correction could be applied to the experimental values, the multiplying factor being the global emissivity of the sample, provided that the layer is isotherm. A calculation with the thermal model (Table 1) shows the evolution of the temperature as a function of the depth.

| Thickness                | Modulus | Phase shift (degrees) |
|--------------------------|---------|-----------------------|
| 1 μm (Amorphous carbon layer) | 0.97 → 0.95 | -11.35 → -11.60 |
| 20 μm (Black paint layer) | 0.97 → 0.5 | -11.35 → -20 |

These results confirm the isothermal hypothesis for the carbon layer, ensuring that a global emissivity can be validly calculated (table 2) from the integration of the radiative transfer equation. Then:

$$
\varepsilon_{\text{global}} = \frac{(1 - \rho_{0})(1 + \rho_{1}e^{-\beta_{\text{IR}}l})(1 - e^{-\beta_{\text{IR}}l})}{1 - \rho_{1}\rho_{0}e^{-2\beta_{\text{IR}}l}}
$$

where $\rho_{0}$ is the reflectivity at the air-carbon interface; $\rho_{1}$ is the reflectivity at the carbon-steel interface; $\beta_{\text{IR}} = (4\pi k)\lambda_{0}$ is the infrared absorption coefficient; $l$ is the amorphous carbon thickness (here 1.7 μm).

| $\beta_{\text{IR}} (m^{-1})$ | $\rho_{0}$ | $\rho_{1}$ | $\varepsilon_{\text{global}}$ |
|-----------------------------|-------|-------|------------------|
| 816 814                     | 0.12  | 0.73  | 0.66             |

In order to confirm this value, an experimental method has been applied. It consists in raising the temperature of a steel sample recovered partially with two coatings, the emissivity of one of them being known. The infrared camera records the radiative intensities.

On the amorphous carbon part of the sample, at 50°C:

$$
L_{\text{measured}}-\text{Carbon} = \varepsilon_{\text{sample}}L_{0}(T_{\text{sample}}) + (1 - \varepsilon_{\text{sample}})L_{\text{environment}}
$$

On the black paint part of the sample, at 50°C:

$$
L_{\text{measured}}-\text{Black paint} = \varepsilon_{\text{Black paint}}L_{0}(T_{\text{sample}}) + (1 - \varepsilon_{\text{Black paint}})L_{\text{environment}}
$$

Then, the emissivity of the sample with its amorphous carbon coating is derived from:

$$
\varepsilon_{\text{sample}} = \frac{L_{\text{measured}}-\text{Carbon} - L_{\text{environment}}}{L_{0}(T_{\text{sample}}) - L_{\text{environment}}}
$$

with:

$$
L_{0}(T_{\text{sample}}) = \frac{L_{\text{measured}}-\text{Black paint} - (1 - \varepsilon_{\text{Black paint}})L_{\text{environment}}}{\varepsilon_{\text{Black paint}}}
$$

Using this experimental method, the emissivity was evaluated here at 0.68.
4.3. Estimation procedure for thermal diffusivity

The first step consists in replacing the detector pixels arranged in a Cartesian system as a function of their radial position (figure 3). A process of localisation of the map centre determination must be also realized by fitting the amplitude image with a bidimensional Gaussian function [5, 11]. For this particular inverse method, a Levenberg-Marquardt algorithm is used. With this approach, the estimation results are extremely accurate, even if the image is disturbed with a great amount of measurement noise.

![Figure 3. Rebuilt temperature profile (URB25 + 20µm black paint)](image)

The second step refers to the evaluation of the discrete Hankel transform of the data. A normalized discrete Hankel transform of the temperature data series is calculated as:

$$T_i^*(\lambda, z) = \frac{2}{r_0} \sum T(r_i, z) J_0(\lambda r_i) r_i \Delta r_i$$

(19)

Practically, the radius step $\Delta r_i$ being non-constant, a numerical integration, using a polynomial approximation, is carried out with the help of a dedicated toolbox available in Mathematica™.

The third step is the determination of the laser beam radius. An experimental method, “the sliding edge method”, associated with an inverse method using again the Levenberg-Marquardt algorithm, allows to identify $r_0$ [5].

The fourth step achieves the estimation of the thermal conductivity $k_i$ simultaneously with $\beta_i$ for the black paint. Here, the layer volumic heat was taken at a nominal value of $\rho C_p = 2 \times 10^6$ J.m$^{-3}$.K$^{-1}$. However, the identified parameters remain in fact the thermal diffusivity $a_i = k_i / \rho C_p$ and $\beta_i$. The estimation should be performed in a spatial frequency band $[\lambda_{\min}, \lambda_{\max}]$, together with a spatial frequency step. Let $u$ and $v$ be the classical Fourier variables, that are the spatial frequencies along $x$ and $y$. Since $\lambda^2 = 4\pi^2(u^2+v^2)$, we chose $u_{\min}$ corresponding to the inverse of the window width of the infrared camera and $u_{\max}$ to the inverse of the inter-pixel length. Then, $\lambda_{\min} = 2\pi\sqrt{2} u_{\min}$ and $\lambda_{\max} = 2\pi\sqrt{2} u_{\max}$. In this way, we attempted to overcome the classical sampling problems (aliasing and windowing). Here, $\lambda_{\min} = 3$ 000m$^{-1}$ and a sampling step $\Delta \lambda$ of 100 m$^{-1}$ have been chosen. The sampling in the real space is close enough to ensure $\lambda_{\max} = 300$ 000m$^{-1}$, far away from the useful spatial frequency range. The results of the fits on the experimental measurements in the Hankel space are shown on figure 4.
The Gauss-Newton algorithm converges easily towards a unique value. The results of the estimation are listed in table 3. It is to be pointed out that the confidence intervals given here concern only the fitting procedure.

**Table 3**: Results of the estimation for Krylon™ black paint

| Steel substrate | Thermal conductivity $k_i$ [$W.m^{-1}.K^{-1}$] | Optical absorption coefficient $\beta_i$ [$m^{-1}$] |
|-----------------|--------------------------------------------|-----------------------------------------------|
| URB 25          | 0.15 ± 0.02                                 | $(11 \pm 1).10^4$                             |

However, further improvements of the method should imply the study of the convolution effects induced by the sampling window. Oscillations on the imaginary part reveal these effects in the Hankel space (figure 4). A deconvolution procedure would surely be more efficient than the crude truncation used here. Obviously, such a procedure should be applied in the Fourier space, since the windowing and sampling effects actually take place in the Cartesian geometry. The whole procedure would then benefit from the numerous existing developments of algorithms devoted to two-dimensional Fourier analysis, such as fast Fourier transforms.

5. Conclusion

The identification from theoretical and experimental spatial frequency profiles in the Hankel domain has allowed a convenient way for the estimation of the thermophysical properties of layers on steel substrate. Identifications of the thermophysical properties of the sprayed black paint well succeeded, but the low diffusivity ($7.5 \times 10^{-8} \text{ m}^2\text{s}^{-1}$) and the large thickness (at least 20 µm) bring low sensitivities to the substrate properties.

Preliminary studies have been realized with amorphous carbon coatings, before undertaking the identification of thermophysical properties of metals under such thin films. These coatings reveal to be sufficiently thin (around 1 µm) and conducting, in order to bring far better sensitivities to the metal substrate properties, when compared with those to the paint layer properties. Indeed, the carbon layer is quite isotherm along its depth, which furthermore is very uniform across the sample surface. However, depending on the deposition process, the layer could become semi-transparent in the infrared range, requiring the determination of the sample global emissivity. The main advantage of the amorphous carbon coating is the low sensitivity of the photothermal method to its own thermal properties.
6. References

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