METHODOLOGY FOR EVALUATING THE UNCERTEAINTY OF THERMOPHYSICAL PROPERTY MEASUREMENTS BASED ON THE LASER FLASH METHOD

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Abstract. *The Laser Flash method is the most popular method to measure the thermophysical properties of solid materials. Despite its several advantages, some experimental obstacles have been found due to the difficulty of supplying the ideal and boundary conditions required by this method. The present work presents a methodology for estimating uncertainty of thermal diffusivity measurement, thermal conductivity and specific heat of Pure Iron and Inconel 600, at 25 ºC. A probabilistic mathematical modeling has been developed, where Finite Volumes techniques, Inverse Heat Conduction Problems (IHCPs) and the Monte Carlo Method (MCM) for uncertainty evaluation were applied in a hybrid manner. The main objective is estimating the influence of the uncertainty components due: pulse time; laser power; thermal exchanges; absorptivity, emissivity, sample thickness, sample specific mass, and additionally due to the temperature measurement system dynamic response. The results have also supplied a sensitivity analysis of the modeled parameters, from which a parameter hierarchyzation was possible, establishing which input quantities need to be known with greater accuracy and less measurement uncertainty. Therefore, it is possible to reduce the measurement uncertainties associated to the determination of thermophysical properties based on the Laser Flash technique.*

Keywords*: Laser Flash Method, measurement uncertainty, finite volumes, thermophysical properties, Monte Carlo Method.*

1. INTRODUCTION

A methodology for probability distribution propagation by means of probabilistic mathematical models developed for measurement processes in transient regime has been developed in the present work. Such methodology is based on the application of the Monte Carlo Method (MCM), a central model of thermal diffusion in the transient regime, in solid medium, considering initial and real boundary conditions of a physical model. As results, the material thermophysical properties (models with multivariate output) and their respective associated uncertainties are assessed.

A hybrid probabilistic modeling structure, using the Monte Carlo Method, Optimization Algorithms and methods for Inverse Heat Conduction Problems solution, performs the propagation of the probability distribution functions (PDF) estimated for the input parameters or quantities, generating resultant PDFs for multiple modeling output quantities.

The validation of the proposed methodology has been performed using standard samples and the estimated values for the physical model parameters, the test bench based on the Laser Flash Method (Parker et al., 1961) of the *Laboratório de Medições de Propriedades Termofísicas - LMTP of the Centro de Desenvolvimento da Tecnologia Nuclear - CDTN.*

The result of the present work is the definition of a coherent and sound methodology to assess the statistic properties involving models with multiple input parameters and multiple outputs. Generically, uncertainty propagation of a central mathematical model of any phenomena or real processes can be done by the methodological structure presented.

2. TEST BENCH DESCRIPTION

Figure (1) schematically presents the LMPT-CDTN test bench. The bench consists of a CO_2 laser, a sample holder placed inside a small tubular oven and a temperature measurement system.

The CO_2 laser (wavelength of 10,6 μm, power of up to 25 W, continuous emission and beam diameter of 8mm) is responsible for the application of the energy pulse onto the front face of the sample (minimum duration of 0,1 s). To ensure the unidimensional heat flow, the sample must have the same diameter of the laser beam (8 mm).

The oven has a sample supporting system that can resist temperatures of up 1700°C. The oven temperature control is set by a PID controller that feeds a platinum heating resistance with 30 % Rhodium. Thus, the measurement of the material thermal properties as a function of temperature is possible.

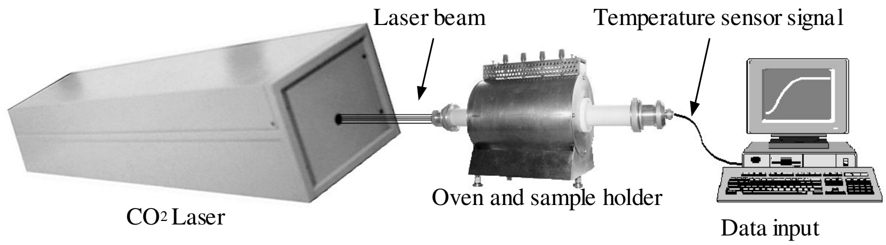

Figure 1 - Thermophysical Property Measurement System for solid material through the Laser Flash Method of the LMPT-CDTN.

Grossi, Ferreira and Andrade (2001) and Grossi et al. (2001) present more detail of both the bench and the process of thermophysical property measurement used by the LMPT-CDTN.

3. MAIN SOURCES OF UNCERTAINTY

The main sources of uncertainty associated to measurement are due to the measurement instruments used, the sample material itself, the measurement method and the medium (neighborhood), where the sample is inserted. Those have been determined according to the methodology of the guide to the Expression of Uncertainty in Measurement (ISO/BIPM – GUM, 1995).

Measurement Instruments

The uncertainties of the measurement instruments are related to the evaluations of the physical quantities involved in the measurement process (sample thickness, L, sample initial temperature, T_0, environment temperature, T_{amb}, temperature measurement system dynamics, SMT, deadtime, t_d, energy delivered to the sample, Q, measurement noise level, ε^*, time resolution, t etc).

Materials

The main sources of uncertainty relative to the materials are associated to their geometrical characteristics (flatness and parallelism) and to their optical, chemical and thermophysical properties (thermal dilatation coefficient, isotropy, homogeneity, emissivity, ε, absorptivity, α_{abs}, and transmissivity, τ, thermal conductivity, k, specific heat, c_p, specific mass, ρ and the variation of thermophysical properties with temperature).

Measurement Method

The uncertainties relative to the measurement methods are due to the differences between the real and the experimental conditions, assumed as input parameters by the model (finite-pulse time, τ, pulse energy shape applied by the laser, pulse energy reduction factor, pulse uniformity factor, assessment of the real power emitted by the heating system), to the SMT identification process (structure of the estimator used for identification, data quality), and to the structures adopted by the models to obtain the numerical solutions for the thermal diffusion process (temporal and spatial meshes adopted, convergence criteria, cut-off, numerical method used etc).

Medium (Neighborhood)

The influences of the medium on the measurement process, or the uncertainties introduced by the neighborhood/environment that involve the sample, correspond to the experimental conditions, and they are basically induced by the characteristics of sample heating (temperature homogeneity and stability), internal and external atmosphere characteristics (medium transmissivity and baffles), and all the parameters relatives to thermal exchanges (contact resistances, thermal exchange coefficient through conduction, convection and radiation).

Table 1 shows the values of the respective uncertainty components considering the LMPT-CDTN experimental bench.

Table 1 - Summary of the influence quantities at the Measurement System of the LMPT – CDTN, their components of combined standard uncertainty, uncertainties expanded to 95%, effective degrees of freedom and PDF type.

Uncertainty Table of the Influence Quantities at the LMPT-CDTN Measurement System							
Symbol	Uncertainty source	Unity	Type	PDF	$u_c(x_i)$ (%)	$U_{95\%}(x_i)$ (%)	$\nu_{eff.}$
ρ	(Sample specific mass)	$g\ mm^{-3}$	A	Normal	1,81	3,54	4,05E+12
L	(Sample thickness)	mm	A	Normal	0,47	0,93	1551
τ	(Finite-pulset time)	s	A	Normal	4,50	10,17	9
ϕ	(Pulse shape)	-	-	Normal	-	-	-
P	(Laser power)	W	A	Normal	5,16	11,15	13
α_{abs}	(Sample absorptivity)	-	B	Rectangular	1,37	2,69	∞
τ_{meios}	(Medium transmissivity)	-	B	Rectangular	0,20	0,39	∞
ε	(Sample emissivity)	-	B	Rectangular	1,37	2,69	∞
U	(Overall heat transfer coefficient)	$W\ m^{-2}\ k^{-1}$	B	Normal	2,00	3,92	∞
FRE	(Energy reduction factor)	-	A	Normal	2,72	5,69	21
L*	(Thermal analogue: thickness)	m	A	Rectangular	1,44	2,83	∞
ρ^*	(Thermal analogue: specific mass)	$kg\ m^{-3}$	A	Rectangular	1,44	2,83	∞
c_p^*	(Thermal analogue: specific heat)	$J\ kg^{-1}\ k^{-1}$	A	Rectangular	1,44	2,83	∞
k*	(Thermal analogue: conductivity)	$W\ m^{-1}\ k^{-1}$	A	Rectangular	1,44	2,83	∞
ε^*	(Measurement noise)	K ou ^oC	A	Normal	0,53	1,03	19999

4. PROBABILISTIC MODEL OF THE THERMOPHYSICAL PROPERTIES DETERMINATION SYSTEM BASED ON THE LASER FLASH METHOD

Probabilistic models are adopted as effective methodologies for uncertainty propagation (JCGM, 2007). However, a methodology for uncertainty propagation in models with multiple input parameters and multiple output parameters are not yet established. The probabilistic modeling proposed in the present work was based on Grossi (2003). However, a higher number of influence quantities in the thermal diffusion process have been considered. This modeling is divided into two steps:

1. Acquisition of the direct numerical solutions to the Thermal Diffusion process in transient regime, through the Finite Volume Method. Such solutions are obtained considering the range of values of the model parameters that show conditions close to those experimentally found. This reduces the degree of model simplification, providing an improved physical meaning to the results and uncertainty reduction;
2. Solutions for Inverse Heat Conduction Problem through the application of a nonlinear programming algorithm, based on the coordinate descent method and on the Golden Section method (Luenberger, 1984). Thus, a objective function characterized by the deviation between the experimental temperature transients and the numerical solutions is minimized, considering value ranges for the initial, boundary and experimental conditions that better adapts themselves to the physical model of application of the Laser Flash Method. An optimal inverse solution for the problem is obtained for determined input parameters, generating as main output Thermal Diffusivity values (α), Volumetric Calorific Capacity (ρc_p) and Thermal Conductivity (k), characterizing the thermophysical properties of the analyzed material.

The Monte Carlo Method for uncertainty propagation has been adopted (Cox et al., 2002). The developed probabilistic structuration was based on Inverse Heat Conduction Problems solved by using the deterministic structure through the Finite Volume Method with the application of the Laser Flash Method. The final model obtained is characterized as a probabilistic, implicit, dynamic, nonlinear one, with multiple input and output parameters.

4.1. Probabilistic Model Structure

The probabilistic method was implemented in Fortran language using a main software that implements the MCM and solutions for Inverse Heat Conduction Problems, IHCP, (FLASH.for), and auxiliary subroutines for thermal diffusion process numerical solution (CONDUCT.for), input quantity evaluation and insertion, initial and boundary conditions (ADAPT.for), reevaluation of the quantity search values (ARRANJO.for) and generation of random sampling with Uniform, Gaussian or Triangular distributions (NORMAL.for), as shown in Figure 2. The expected mean values and uncertainties associated with the multiple output parameters (k, c_p, ρ and α,) were obtained as final results and according to the LMPT – CDTN experimental bench.

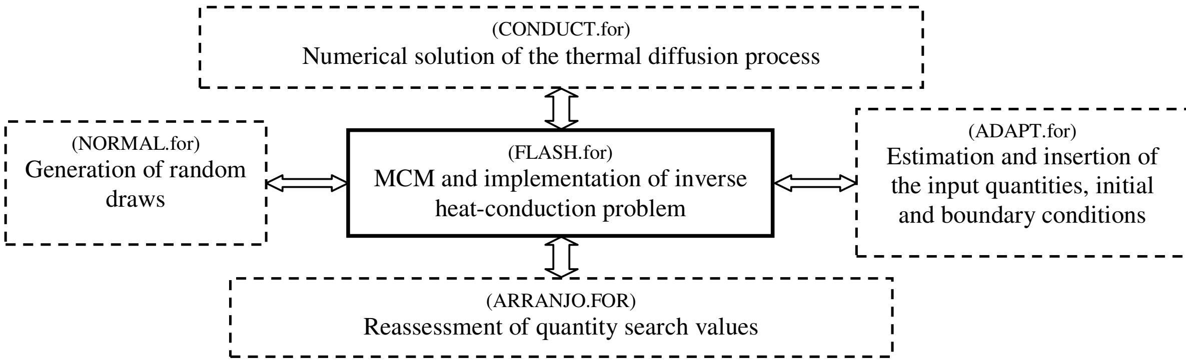

Figure 2 – Schematic diagram of the probabilistic model structure implemented in the Compaq Visual Fortran environment: main software and subroutines.

The detailed structure of the probabilistic model of Monte Carlo Method application for the solution of Inverse Heat Conduction Problem is divided into the following steps.

1 – Initialization of the simulation process

- A set of real thermal transients are obtained (transients obtained from measurements by the Laser Flash method under repetitivity conditions);
- The N input quantities of the model are defined and assessed, estimating their mean values, associated standard uncertainty and PDF type (Table 1);
- The M number of Monte Carlo Simulations to be performed is established
- The value search range of the R output quantities is established, ensuring the physical meaning of the solution of the Inverse Heat Conduction Problems;

2 - Beginning of Loop 1: Monte Carlo Method

- A vector containing a set of N random values for each of the model input N parameters is generated in accordance with their mean values, standard deviation and associated PDF. This step is repeated M times, so that for each new interaction, new random values are supplied;

3 – Beginning of Loop 2: Inverse Heat Conduction Problem

- Four values within the search range for the output parameters are selected (maximum, minimum values, and two intermediate points, 38,2 and 61,8 %, according to the Golden section method);
- With the values generated in steps 5 and 6, direct numerical solutions for the sample thermal diffusion process are obtained, additionally modeling the temperature measurement system (deterministic modeling);
- For each time (discrete values), the mean value of the experimental transients (sampled in step 1) and that of the set of numerical transients are calculated (generated in step 7). The mean transients are compared (quadratic mean deviation among the curves) and the convergence criterion is verified;

4 – End of Loop 2

- The optimal values (numerical solutions and model output quantities) are stored. If the number of optimal solutions is equal to the number M of Monte Carlo simulation M is estimated, go to the next step, else, a new iteration of the Monte Carlo Loop is performed;

5 – End of Loop 1

- M values for the R output quantities are estimated as a model response when excited by each of the M random vectors of the N input quantities;
- The Optimal Parameters Matrix, MPO, is created, grouping the M input vectors and their respective output quantities (model responses), for each of the simulations (iterations) of the Monte Carlo Method, according to the Equation (1);
- The mean values of the model input and output quantities are estimated;
- The Optimal Parameter Matrix Corrected, MPO*, is created, where the individual M values of each quantity in the MPO are subtracted from its respective mean (calculated in the previous step). Thus, the MPO* represents the value deviations used or obtained in each optimal simulation relative to the mean values of each quantity;
- The Covariance Matrix, MCOV, or Uncertainty Matrix associated to the output (and also input) quantity estimates is obtained through the Equation (2);

$$MPO = \begin{bmatrix} y_{1,1} & \cdots & y_{1,M} \\ \vdots & \ddots & \vdots \\ y_{R,1} & \cdots & y_{R,M} \end{bmatrix} = f \begin{bmatrix} x_{1,1} & \cdots & x_{1,M} \\ \vdots & \ddots & \vdots \\ x_{N,1} & \cdots & x_{N,M} \end{bmatrix} \tag{1}$$

$$MCOV = \frac{1}{M-1} MPO^{*}(MPO^{*})^{T} \,. \tag{2}$$

- The value of the output quantities and standard uncertainties associated is estimated, according to the Equations;

$$\bar{y}_R = \frac{1}{M}\sum_{m=1}^{M} y_R(m) \qquad \text{and} \qquad u(y_R) = s(\bar{y}_R) = \sqrt{\frac{1}{M\,(M-1)}\sum_{m=1}^{M}\left[y_R(m) - \bar{y}_R\right]^2} \,. \tag{3}$$

- The output quantity values are ordered in an increasing sequence and classified, thus obtaining a discrete representation of the PDFs (Cox et al., 2002);
- The mean value and uncertainty associated to the output quantities are estimated for a 95% coverage interval from the discrete obtained PDFs (Cox et al., 2002).

Figure 3 presents the computer algorithm flowchart developed and implemented using the Compaq Visual Fortran platform.

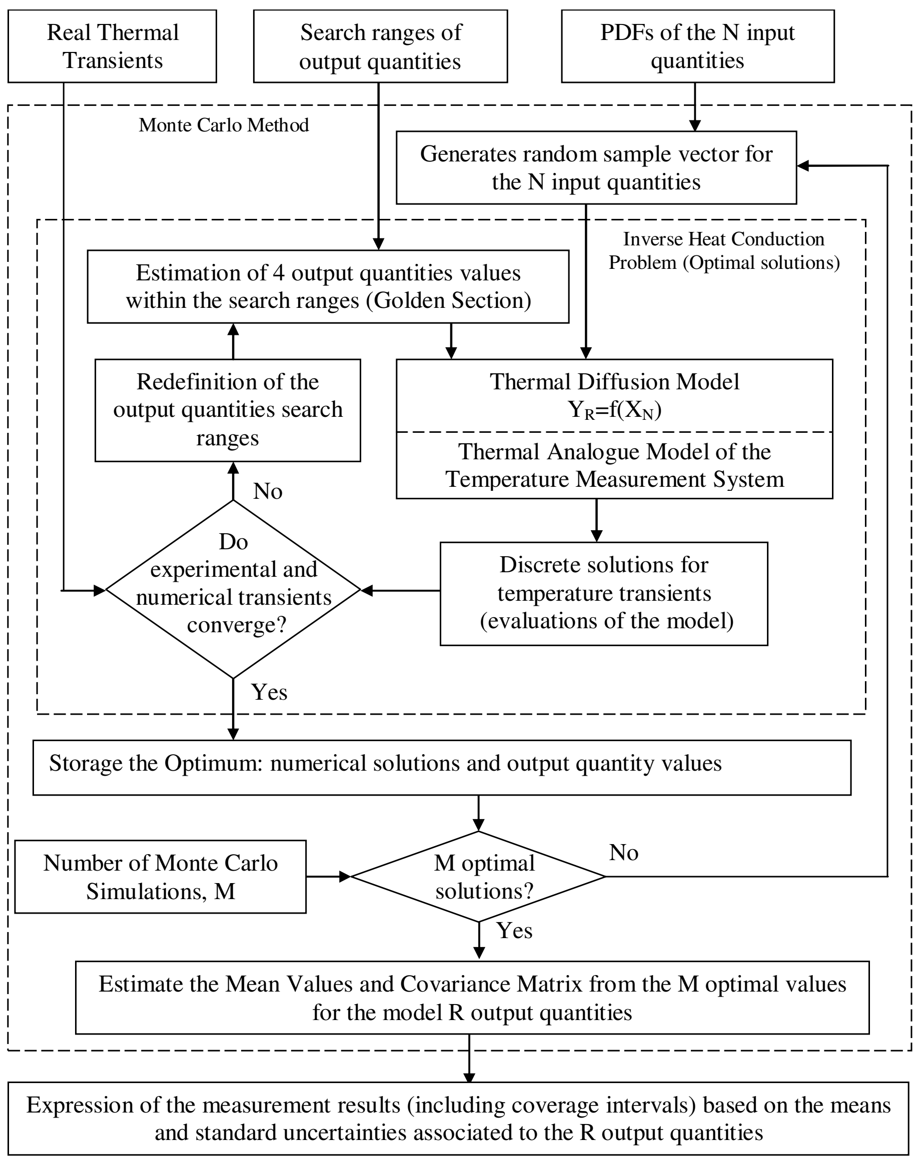

Figure 3 – Flowchart of the application of the MCM to solve the Inverse Heat Conduction Problem.

5. RESULTS

At first, the mean values and the standards uncertainties for all the R output parameters of the probabilistic model of application of the Monte Carlo Method for Thermal Diffusion Inverse Problem solution (flowchart depicted in Figure 3), were obtained considering the uncertainties associated to the LMPT experimental bench and the values of the reference sample properties declared in the certificates of the respective samples. In this phase, the fine adjustment of the model parameters to the experimental apparatus is performed. Fifteen numerical simulations for each output parameter of the Probabilistic Model were carried out, considering the two standard samples of BSC Pure Iron and Inconel 600.

From the parameters modeled as input parameters (including the thermophysical properties, until then considered as output parameters), random values were generated according to the PDFs identified for the parameters, and direct numerical solutions were obtained as output for the thermal transients using only the modeling deterministic structure. Therefore, a second application of the MCM is carried out with the purpose of numerically and graphically validating the obtained values. These results were compared with those experimentally obtained from the 15 measurements made with each sample (Figures 4 and 5). The convergences between the experimental and numerical results indicating the consistency of the mathematical modeling are verified.

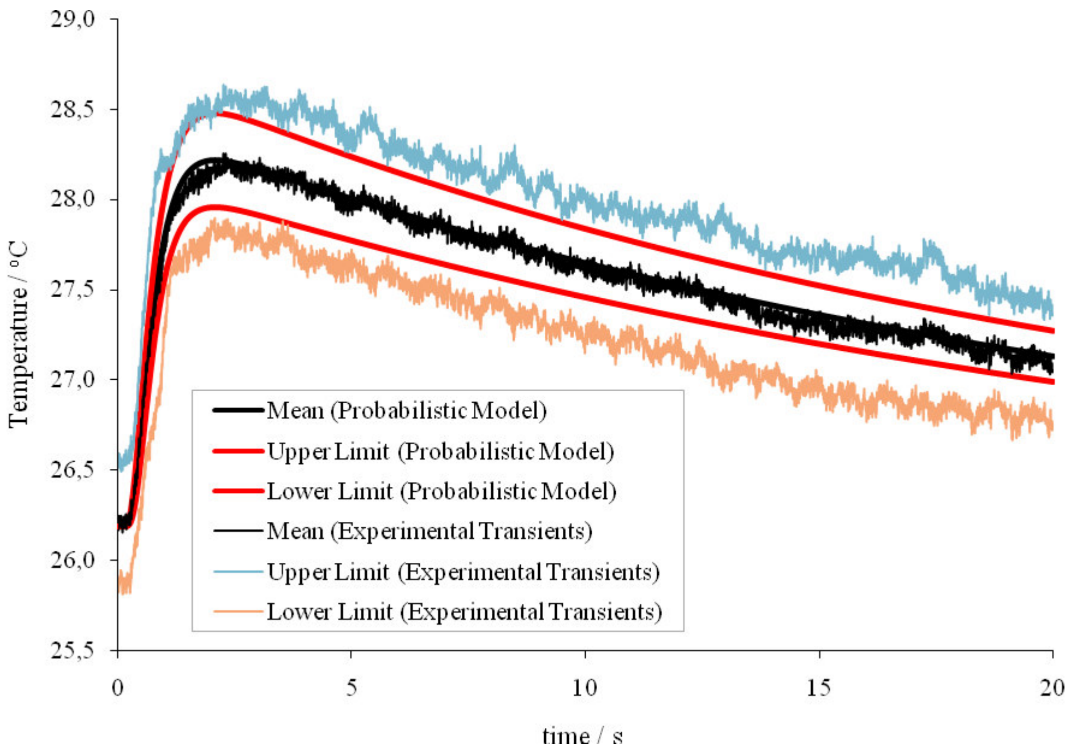

Figure 4 – Upper and lower limits for a 95 % coverage interval considering the experimental transients and the Probabilistic Modeling transients for the standard sample of Inconel 600.

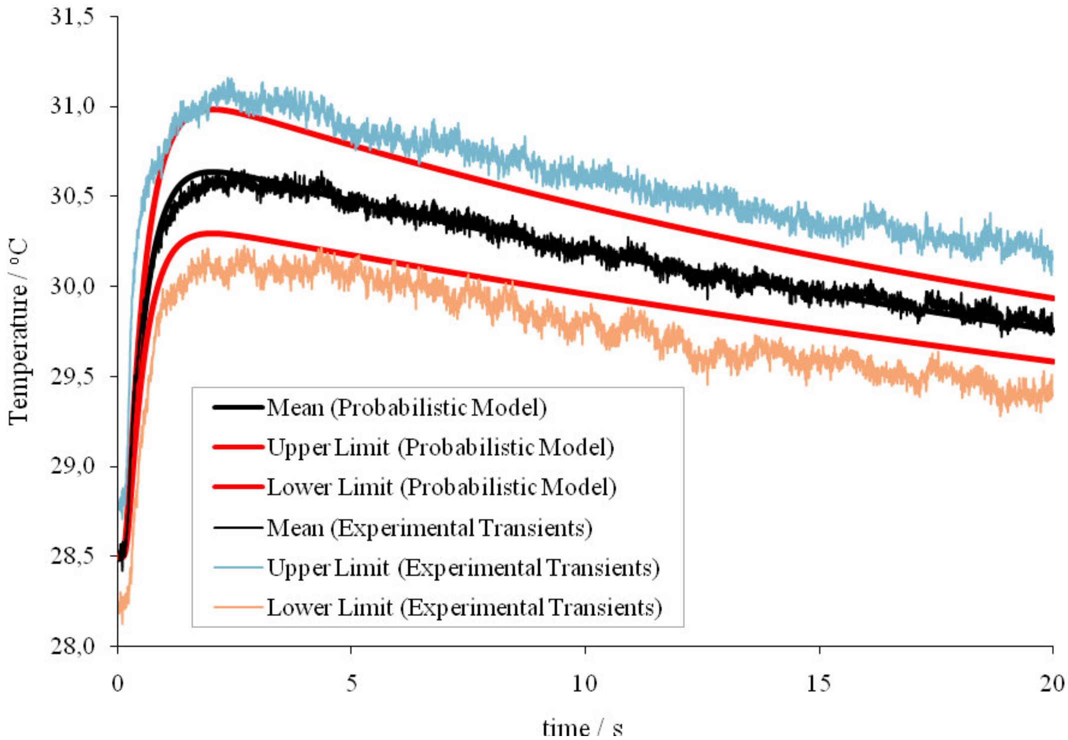

Figure 5 - Upper and lower limits for a 95 % coverage interval considering the experimental transients and Probabilistic Modeling transients for the standard sample of Pure Iron BSC.

Assessment of Figures 4 and 5 also makes clear the convergence of the Probabilistic Model proposed to the experimental transients (measurements) considering the standard sample of Inconel 600 and BSC Pure Iron.

Table 2 presents the mean values and uncertainty of standard samples thermophysical properties at 25 ºC, evaluated by the probabilistic model.

Table 2 – Mean values and uncertainty of standard samples thermophysical properties at 25 ºC evaluated by the probabilistic model.

Standard samples		**α(m^2 s^{-1})**	***k* (W m^{-1} K^{-1})**	**c_p (J kg^{-1} K^{-1})**
Inconel 600	Mean Value	3,458	12,805	444
	u(x)	6,323 %	5,320 %	6,168 %
	Coverage Interval (95%)	[3,11 - 4,01]	[11,7 – 14,5]	[386 - 494]
Pure Iron BSC	Mean Value	21,6	76,60	450
	u(x)	8,912 %	6,774 %	5,362 %
	Coverage Interval (95%)	[17,47 – 25,05]	[66,13 – 86,86]	[409 - 506]

6. CONCLUSIONS

The Probabilistic Modeling developed has presented itself as an effective approach methodology of the two most relevant issues addressed in the present work:

- Propagation of probability distributions associated to input parameters through dynamic probabilistic models with multivariate output;

- Assessments of the uncertainties associated to the thermophysical properties determined at the **Laboratório de Medição de Propriedades Termofísicas - LMPT** of the **Centro de Desenvolvimento da Tecnologia Nuclear - CDTN**.

The Joint Committee for Guides in Metrology (JCGM) has been developing the Supplement 2 to the Guide to the expression of uncertainty in measurement: Models with any number of output quantities, which aims the uncertainty propagation through methods with several output quantities. Such an approach coincides with the purpose of the present work, showing its relevance and ineditism for the solution to problems at the knowledge frontier.

The high uncertainty values associated to the output parameters are due to the propagation of uncertainties associated to the inputs, which are relatively even more elevated. Therefore, the suggestions for the minimization of such values are:

- Substitution of the thermal excitation source (CO_2 Laser) responsible for high values of combined standard uncertainty of the emitted power and pulse duration values. Modern systems generate more stable beams, with shorter duration and higher power levels;
- Utilization of a greater number of standard samples, in order to validate the Probabilistic Modeling by a higher number of value ranges, which should be close to the thermophysical properties to be measured. This causes the reduction of the uncertainties associated with several input parameters, more effectively dealing with the nonlinearity characteristics of the system;
- Substitution of the temperature measurement system, responsible for high values of dynamic uncertainties and thermal inertia;
- Reassessment of the interface structure of the three modules in Figure 1, allowing for a better assessment of the energy reduction factor and thermal exchange global coefficient.

The following are recommendations of this work and suggestions for future works:

- Application of the methodology shown in the present work for uncertainty assessment in models with any number of output quantities (a methodology still in its development stage by the JCGM that will be presented no Supplement 2 to the GUM);
- Utilization of Deterministic and Probabilistic Models presented for the revision of the ASTM-E-1461-92 standard measurement method of thermal diffusivity of solids by the Laser Flash method. This will allow that the standard encompasses the assessment of thermal conductivity and specific heat, efficient and simultaneous implementation of corrections for all the experimental problems associated to the Laser Flash Method implementation and, finally, assessment of uncertainties associated to the values estimated for such properties;

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