Inverse Problems, Design and Optimization Symposium
Rio de Janeiro, Brazil, 2004

ESTIMATION OF THE TRANSVERSE COEFFICIENT OF THERMAL EXPANSION ON CARBON FIBERS AT VERY HIGH TEMPERATURE

C. Pradère and J.C. Batsale
TREFLE
University Bordeaux1, UMR 8508
Esplanade des Arts et Métiers, 33405 Talence, France
christophe_pradere@yahoo.fr
batsale@lept-ensam.u-bordeaux.fr

S. Dilhaire
Centre de Physique Moléculaire Optique et Hertzienne (CPMOH)
University Bordeaux 1, UMR 5798
351 cours de la Libération, 33405 Talence, France
sdilhair@cpmoh.u-bordeaux1.fr

J.M. Goyhénèche and R. Pailler
Laboratoire des Composites Thermostructuraux (LCTS), UMR 5801
3 allée de la Boétie, 33600 Pessac, France
goyhenec@lcts.u-bordeaux.fr
pailler@lcts.u-bordeaux.fr

ABSTRACT

The main objective of this work is to present a new method for measuring the transverse Coefficient of Thermal Expansion (CTE) of fibers at very high temperature. The difficulty of the measurement is due to the thin fiber diameter (≈ 10 μm), the low variation with temperature of the fiber diameter (≈ 10-30 nm) and finally the important range of temperature (300-2800 K). An experimental device was designed for the measurement and an inverse method has been developed for identification. The characterisation, with a confidence interval lower than 8 %, of tungsten and carbon fibers allows the validation of this device and the related estimation method.

NOMENCLATURE

A	Identified parameter (magnitude)
d	Fiber diameter, m
D	Distance between fiber and sensor, m
I	Diffracted intensity, $W.m^{-2}.sr^{-1}$
L	Sensor length, m
R	Identified parameter (Δd/d)
x	Position along the sensor, m
x_o	Initial position of the sensor, m
α_d	Differential CTE, K^{-1}
α_m	Average CTE, K^{-1}
α_s	Specific CTE, K^{-1}
α_T	True CTE, K^{-1}
θ	Angular position of diffracted intensity, rad

INTRODUCTION

Carbon-carbon composite materials have very efficient properties at very high temperature : wide range of Young modulus, thermal conductivity, thermal expansion [1,2]. These properties are especially attractive for aeronautics (brakes), aerospace (heat shields) and military (nose-cones for reentry vehicles) applications. Further, design of carbon fibers composites with a numerical method needs a prior knowledge of the physical properties (thermal and mechanical) of each constituent, fiber and matrix. Properties of the constituents are needed at different observation scales : fiber and filament (≈ 10 μm), microcomposite (one fiber with matrix, ≈ 100 μm) or minicomposite (one thousand of fibers with matrix, 1 mm) and finally on composite (≈ cm).

Even if global macroscopic properties are generally sufficient in order to design and modelize the behaviour of those materials at large scale. Particular uses at very high temperature and very large thermal gradient need to investigate the properties of constitutive parts (fibers) of the materials. Very little data are available in the literature on properties of carbon fibers and matrices at high temperatures [3,4].

This work is included in a large program devoted to the global (mechanical, thermal and thermomechanical) characterisation of the carbon fibers [5-7] at very high temperature.

MEASUREMENT METHOD

Definition of the transverse CTE

First, it is important to define the different kinds of transverse Coefficient of Thermal Expansion (CTE) [8]. The specific CTE (1) represents the relative ratio between the fiber diameter at any temperature and the fiber diameter at room temperature.

$$\alpha_S(T) = \frac{d(T) - d(T_0)}{d(T_0)} \tag{1}$$

Then, the average CTE (2) represents the ratio between the specific one and the difference between any temperature and room temperature.

$$\alpha_m(T) = \frac{1}{T - T_0} \frac{d(T) - d(T_0)}{d(T_0)} \tag{2}$$

Finally, The true CTE (3) represents the derivative of diameter divided by fiber diameter at room temperature.

$$\alpha_T(T) = \frac{1}{d(T_0)} \frac{\partial d(T)}{\partial T} \tag{3}$$

All those CTE depend on the reference temperature T_0.

CTE measurement principles

There are two ways for measuring the transverse CTE : (i) with an indirect method [9,10] : global dilatation measurement of many thousands of fibers in a minicomposite ; (ii) with a direct method : measurement of the *fiber diameter* at different temperatures with a laser diffraction phenomenon [11,14] . Independently of the method used, micro scale and weak diameter variations ($\approx$ 10 nm) induce rather delicate measurement. Few articles related to this topic are available. Instead of the previous principles, the proposed method gives *a direct evaluation* of the CTE by using the diffraction phenomenon.

The fiber is warmed up (by Joule effect) at a temperature T, and irradiated by a laser *(figure 1)*. The diffraction phenomenon (electromagnetic interaction between fiber and laser) produces the beam dispersion into a plane where the diffracted figure, represented by the intensity I in the θ direction *(figure 1)*, takes form. Diffracted pattern depends on : (i) the incident beam intensity Io, (ii) the laser wavelength λ, (iii) the distance D between fiber and screen (where the diffracted pattern is projected), (iv) the position x, and (v) the fiber diameter d(T) at the temperature T.

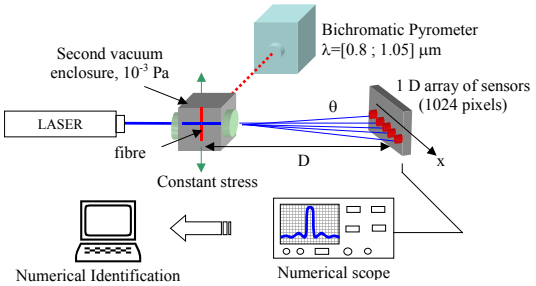

figure 1 : Experimental device

Fraunhofer model

Several electromagnetic theoretical studies [15] have been done in way to represent diffraction phenomenon (Fresnel, Mie, Fraunhofer). One of the most classical methods [11-14] is the Fraunhofer theory *(figure 2)*,which is represented by the following expression :

$$I(d_T, x) = I_o \left[\frac{\sin u}{u} \right]^2 \tag{4}$$

where :

$$u = \frac{\pi d_T}{\lambda} \frac{x}{\sqrt{x^2 + D^2}} \tag{5}$$

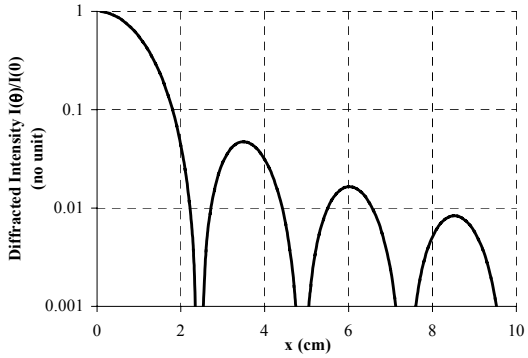

figure 2 : Fraunhofer model (4) logarithmic representation with λ = 488 nm, d =10 µm and D = 1 m

By an asymptotic expansion around zero of expression (4-5), authors [11,14] determined fiber diameter d_T by measuring the extreme positions

of the diffracted patterns. This method used at different temperatures allows determination of a monotonous variation of fiber diameter (d). By derivation of the measured diameter, they calculate the transverse CTE (1-3).

On the one hand, this technique is not very efficient because of the weak fiber diameter variations, only a few tenths of nanometers. This corresponds to a translation of 200 µm of the diffracted pattern at a distance D of one meter. On the other hand, the aspect of the diffracted pattern is strongly dependent of fiber roughness. Then, it is difficult to estimate the extreme positions. Further, diameter measurement errors are propagated on the calculation of different transverse CTE and increase the size of the confidence interval on the final results.

General principle of proposed method

In opposite with the previous ways, the data processing proposed in this work is based on a reduced knowledge of the model (4-5). It consists in a measurement of a first diffracted pattern at a temperature T, then another at a temperature $T+\Delta T$, in order to estimate with an inverse method the direct relative variation of the fiber diameter expressed as :

$$\left.\frac{\Delta d}{d}\right|_{T+\Delta T/2} = \frac{d(T+\Delta T)-d(T)}{d(T+\Delta T/2)} \qquad (6)$$

Then an additional transverse CTE (the differential one) can be defined and calculated at the temperature $T+\Delta T/2$:

$$\alpha_d(T+\Delta T/2) = \frac{1}{\Delta T}\left.\frac{\Delta d}{d}\right|_{T+\Delta T/2} \qquad (7)$$

The evaluation of α_d for a large range of temperature gives a monotonous variation with the temperature.

EXPERIMENTAL DEVICE

The experimental device (*figure 1)*, has been developed on a bench previously designed for mechanical characterisation [5]. The fiber is warmed up by Joule effect and assumed to be quasi-isotherm (T_{centre}-$T_{surface}$<2 °C, [5]). The fiber is maintained at very low pressure (P<10^{-3} Pa) in a silica glass enclosure. A bichromatic pyrometer measures the absolute temperature of the fiber, whereas an array of 1024 pixels (size of : 25×25 µm²) collects the diffracted intensity of an argon laser (488 nanometers). The diffracted patterns (*figure 3*) are visualised on a numerical oscilloscope and the signal is transferred to a computer with an IEEE 488–GPIB card where the numerical processing is done.

During the measurement, fiber stress is maintained at a small percent with a traction table controlled by a force sensor. The global device has been controlled in order to record a wide number of data to decrease the influence of signal noise. Typically the characterisation of a fiber from 300 to 2800 K takes 2 hours.

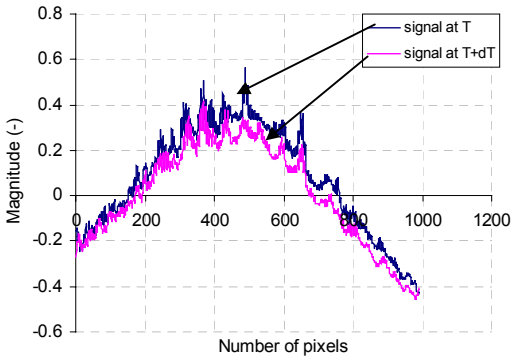

figure 3 : Measured signal at T and T+ΔT

DATA PROCESSING METHOD

Presentation

As detailed before, the Fraunhofer model (4-5) is often used to estimate fiber diameter in order to obtain the transverse CTE [11,14]. But, A.J. Perry et al. [12] showed that using a cardinal sine for determining the diameter $d(T)$ induced an error of about 4 % on the transverse CTE.

That is why, a method based on a reduced knowledge is proposed here. Due to weak variation of the fiber diameter with a raising temperature, it is possible to express an asymptotic expansion of the signal $I_{T+\Delta T}(d_{T+\Delta T}, x)$ measured at the temperature $T+\Delta T$ relative to the signal $I_T(d_T, x)$ measured at the temperature T, in order to obtain :

$$I_{T+\Delta T}(d_{T+\Delta T}, x) = \\ = A\left[I_T(d_T, x) + \frac{\Delta d}{d_T} d_T \frac{\partial I_T(d_T, x)}{\partial d} \right] \qquad (8)$$

where $\Delta d = d_{T+\Delta T} - d_T$. The expression (5), allows to express the differential term as :

$$d_T \frac{\partial I_T}{\partial d} = \frac{\partial I_T}{\partial x} \frac{x\left(x^2 + D^2\right)}{D^2} \tag{9}$$

which becomes independent of the fiber diameter. Soon, every term of the expression (8) can be determined experimentally : the method is autoregressive. The integration of (8) along the entire sensor gives :

$$\int_{x_o}^{x_o+L} \frac{D^2}{x\left(x^2 + D^2\right)} I_{T+\Delta T}\left(d_{T+\Delta T}, x\right) dx = A\left[\begin{array}{l} \int_{x_o}^{x_o+L} \frac{D^2}{x\left(x^2 + D^2\right)} I_T\left(d_T, x\right) dx + \\ \frac{\Delta d}{d_T}\left[I_T\left(d_T, x_o + L\right) - I_T\left(d_T, x_o\right)\right] \end{array}\right] \tag{10}$$

By using a least square method to solve this problem, a matrix system can be written [16] :

$$\beta = \left[S^t \cdot \boldsymbol{cov}^{-1}(\varepsilon) \cdot S\right]^{-1} \cdot S^t \cdot \boldsymbol{cov}^{-1}(\varepsilon) \cdot \left\{I_{T+\Delta T}\right\} \tag{11}$$

where, $\boldsymbol{\beta}$ represents the parameter vector : $\beta_1 = A$ and $\beta_2 = AR$ with $R = \Delta d / d_T$., $\boldsymbol{cov}(\varepsilon)$ the covariance matrix associated to the signal, $\boldsymbol{S}$ the sensitivity matrix (calculated with measured intensity $I_T(d_T,x)$) and $\{\boldsymbol{I_{T+\Delta T}}\}$ the measured vector signal. The differential CTE is calculated with :

$$\alpha_d\left(T + \Delta T/2\right) = \frac{2}{\Delta T} \frac{R}{2 + R} . \tag{12}$$

In order to express the transverse CTE (1-3) with the differential one (12), the specific CTE $\alpha_s(T)$ and true CTE $\alpha_v(T)$ can be calculated with an integration along the temperature range, such as :

$$\alpha_s(T) = exp\left[\int_{T_o}^{T} \alpha_d(T')\, dT'\right] \tag{13}$$

$$\alpha_v(T) = \alpha_d(T)\, exp\left[\int_{T_o}^{T} \alpha_d(T')\, dT'\right] \tag{14}$$

where T_o represents the room temperature (usually T_o=20 °C).

With this method ***no absolute measurement of the fiber diameter d is needed***. Measurement with low precision of fiber diameter is very difficult. Further, in classical methods [11,14] error sources are reported when transverse CTE $\alpha_d(T)$ are calculated. In this work, only expression (5) is used whereas the complete Fraunhofer model (4-5) is generally used in order to estimate $\alpha_d(T)$. Finally, the method only needs the diffracted pattern measured at T and $T+\Delta T$.

Validation

In order to test this method, different kinds of signals have been generated *(figure 4)* with the Fraunhofer model (4-5). Parameters are fixed at : 17.10^{-6} m for d, 0.053 m for x_0, 1 m for D, 0.0254 m for L, 10.10^{-6} K^{-1} for $\alpha_V(T)$ and 100°C for ΔT. A noise corresponding to a ratio of 1000 between signal and noise is added.

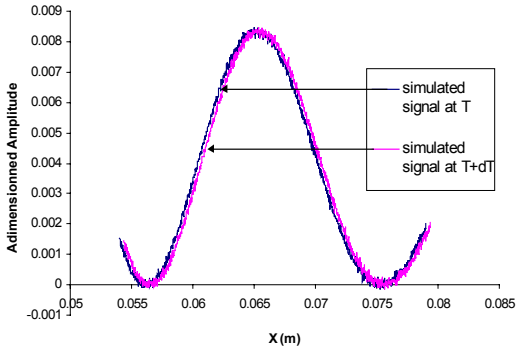

figure 4 : generated signal with noise at T and T+dT corresponding to a temperature gap (ΔT) of 100°C with a CTE of 10.10^{-6} K^{-1}

With this configuration the R true parameter to estimate is about 10.10^{-4}. With the method presented before, the identified parameter value is $10.011.10^{-4}$. This represents a relative difference of 0.11 % that is only due to signal noise. When a signal on noise ratio of 10 is taken, the relative error turns around 10 %. The confidence interval of this method is independent of parameter range. For example, even if the temperature gap is taken at 1°C which represents a diameter variation of 0.1 nm, identification gives the good value and the confidence interval stays correlated with signal noise. Experimentally, the limit of the method depend on the resolution of the array of detector.

ANALYSIS OF ERROR SOURCES

Main Experimental Error Sources

Fiber Roughness. The first error source comes from the roughness of the fiber which induces light dispersion. This phenomenon affects the diffracted pattern. This problem has been identified and modelised [7] for tungsten and carbon filament *(figure 5 and 6)*.

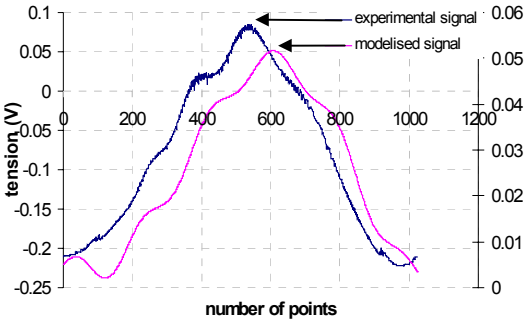

figure 5 : measured and modelised diffracted signal on carbon fiber

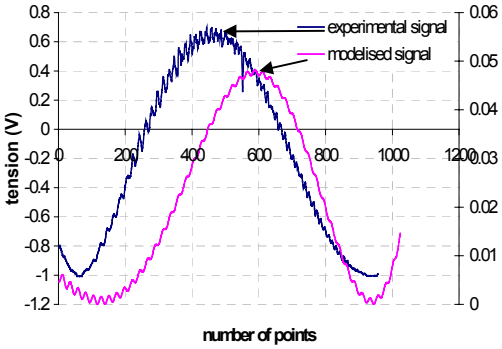

figure 6 : measured and modelised diffracted signal on a tungsten filament

In fact, this phenomenon induced a data processing error of 15 % on R. Even if this problem is attenuated by the method which compared directly the diffracted pattern at the temperature T and $T+\Delta T$, it is necessary to filter the measured signal in order to limit this kind of noise. The used filter is a moving average of N points. Confidence interval of the data processing method depends on the number of average points *(figure 7)*. Finally, working with a number of 80-100 points for the moving average reduced to 3 % the confidence interval on $\alpha_d(T)$ due to this phenomenon.

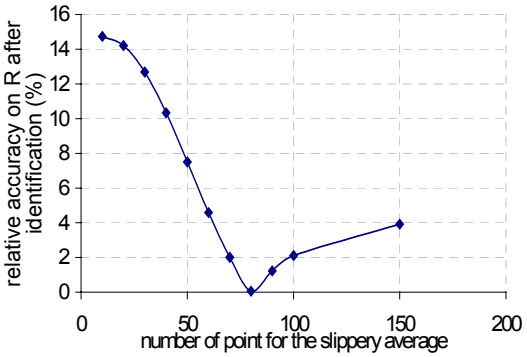

figure 7 : relative confidence interval on R with different number of points for the slippery average

Fiber Stress. The second error source is due to fiber stress. Without taking care, it gives an error of about 20 % on $\alpha_d(T)$. An active control of the fiber stress aiming to compensate longitudinal dilatation and environment vibration allows the reduction of this problem. Then, during a measure, an extension of 0.01 % to 0.1 % (depending on the fiber) is imposed with confidence interval of 3 %.

Resolution Of The Scanning Device. The third error source comes from sensor resolution. In fact, the distance D between sensor and fiber is fixed, and the sensor resolution (pixel dimension) too. So, for a good measurement of $\alpha_d(T)$ the temperature gap ΔT between two measurements must be sufficient to produce a measurable displacement of the diffracted pattern. The curve represented (*figure 8)* allows us to adjust those parameters before a measurement.

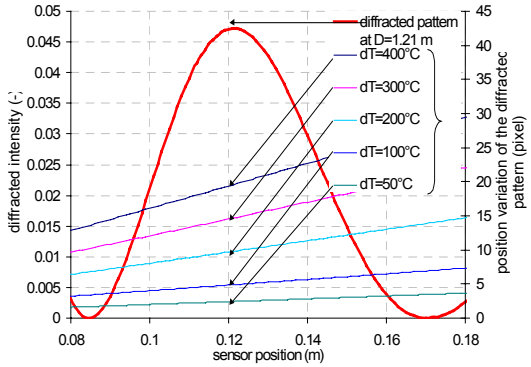

figure 8 : sensor confidence interval due to temperature variation for λ = 488 nm and $\alpha_m = 10.10^{-6}\ K^{-1}$

For example, if the sensor is located at about 1.21 m from the fiber which has a diameter of 10 µm, and if the approximate transverse CTE is about 10^{-5} K^{-1}, then, a minimum temperature gap (ΔT) of 100 K is necessary for a displacement of 5 pixels (≈ 1 nm on fiber diameter) of the diffracted pattern.

Estimation Of The Confidence Interval

Instead of previous works, a tentative calculation of the confidence interval of the estimation can be implemented. This can give some indication about the accuracy of the method.

Reproductibility Of The Measurements. First, the diffracted pattern is measured from an average of 256 periods of the signal. This operation decreases the noise influence. Then, 5 records of the diffracted pattern are made at each temperature (T and $T+\Delta T$) in order to identify 25 values of $\alpha_d(T+\Delta T/2)$. Finally, an average CTE and the standard deviation can be calculated. Generally, scattering of measurement never goes over 5 %.

Noise Propagation Estimation. Another method to determine the incertitude of a measure is to try to evaluate the data processing error on estimated parameters. This confidence interval is evaluated from the calculation of inverse covariance matrix (11) which varies like the inverse of the 1 D array measured signal *(figure 9)*.

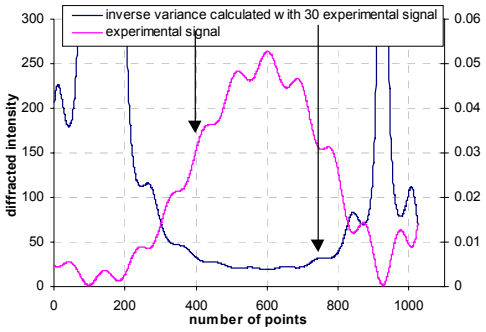

figure 9 : sensor confidence interval due to temperature

Then, the confidence interval on $\alpha_d(T)$ calculated with the expression :

$$cov(\beta) = \left[S^t \cdot \boldsymbol{cov}^{-1}(\varepsilon) \cdot S\right]^{-1} \qquad (15)$$

is always lower than the dispersion calculated before.

MEASUREMENT VALIDATION

Some measurements have been done on a tungsten filament ($d \approx 17$ µm). Since tungsten is isotropic, the results (*figure 10*), can be compared with those from the literature [11,14] obtained with massive sample usually measured with longitudinal devices. The difference is of about 2 %. So, the experimental and data processing method can be validated.

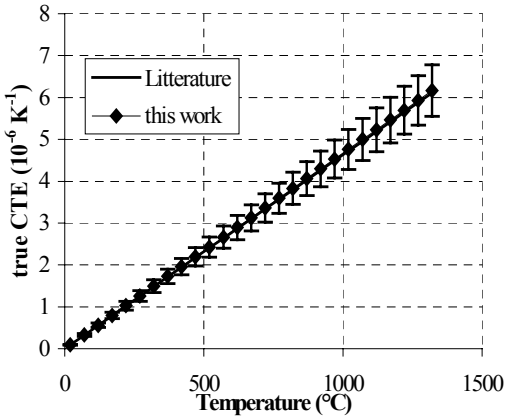

figure 10 : validation on a tungsten filament

A second test has been done on a carbon fiber XN05 (ex-pitch, Nippon Graphite Fiber, $\rho = 1610$ kg.m^{-3}, $d \approx 10$ µm.). The results prove a little dispersion, less than 8 %, (*figure 11)*.

Further, we can see that the longitudinal [5] and transverse CTE are very similar (*figure 12)*. This confirms that the fiber is isotropic.

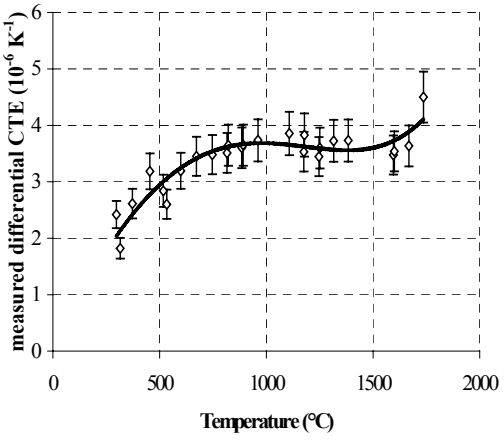

figure 11 : validation on a carbon filament

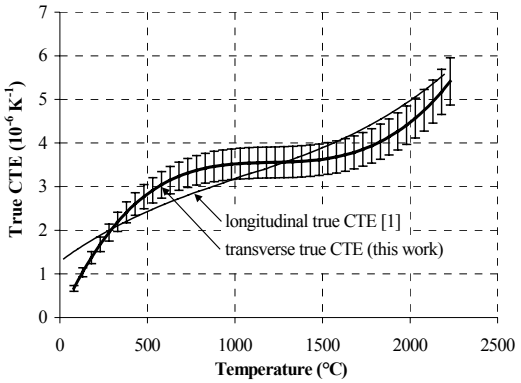

figure 12 : validation on a carbon filament, XN05 fiber heat treated at 2500 K.

CONCLUSION

A new method for measuring the transverse CTE at very high temperature has been developed in this work. Even if optical methods have been used previously, one of the main points of this work is to have developed special recommendations relative to the signal processing and the parameter estimation.

First, an optical method based on laser diffraction phenomenon at micro scale (fibers) is used.

Then, an original inverse method based on reduced knowledge of Fraunhofer model is proposed. The determination of the transverse CTE with a confidence interval lower than 8 % needs only the measurement of the diffracted pattern at the temperature T and $T+\Delta T$. This gives an autoregressive method.

Finally, an analysis of different kinds of error sources (fiber roughness, fiber stress, 1 D array sensor resolution) allows a validation of this work on isotropic fibers, like tungsten.

The perspectives of this work are to explore systematically the properties of the principal fibers used in the aerospace industry. Then, this estimation method will be implemented for the measurement of the Poisson ratio at very high temperature.

ACKNOWLEDGEMENTS

This work has been supported by the CNRS, the CEA and the Region Aquitaine.

REFERENCES

1. G.M. Savage, Carbon-Carbon composites, *Chemistry and Industry*, 1992, July, 525-529.

2. G.M. Savage, Carbon-Carbon composites, *Chapman and Hall*, London, UK,1992.

3. Tanabe Y., Yasuda E., Bunsell A. R. The strength of pitch-based carbon fibre at high temperature *J. Mat. Sci.*, **26**, 6, 1601-1604 (1991)

4. M. Trinquecoste, J.L. Carlier, A. Derré, P. Delahès, P. Chadeyron, High temperature thermal and mechanical properties of high tensile carbon single filaments, *Carbon,* **34,** 7, 923-929 (1996)

5. C. Sauder, Relation structure / properties at high temperature of carbon fibers and matrix, *PhD Thesis, n° 2477, University of Bordeaux 1, 2001.*

6. C. Sauder, J. Lamon, R. Pailler, *Composites Science and Technology*, 62, 3, 395-399 (2002).

7. C. Pradère, Thermal properties of carbon fibers at very high temperature, *Thesis, current work.*

8. M. Murat, « Dilatométrie », *Engineer Technical, vol P1, n°P1280.*

9. G. Wagoner, R. Bacon, Elastic constants and thermal expansion coefficients of various carbon fibers, *18th Biennial Carbon Conference, 1986.*

10. R.C. Fanning, J.N. Fleck, Transverse thermal expansion characteristics of graphite fibers, *Journal of Carbon 10 (3) (1972) 331.*

11. J.F. Villeneuve, Study of thermal expansion and thermocinetic behaviour in traction and twisting of ceramic fibers, *PhD Thesis, n° 661, University Bordeaux 1, 1997.*

12. A.J. Perry, B. Ineichen, B. Eliasson, Fibre diameter measurement by laser diffraction, *Journal of Materials Science 9 (1974) 1376-1378.*

13. P.M. Sheaffer, Transverse expansion of individual carbon and graphite filaments, *18th Biennale Conference on Carbon, 1986.*

14. W.C. Morgan, "Thermal expansion coefficients of graphite crystals", *Carbon*, **10** (1972), 73-79.

15. E. Hecht, Optics, *Addison-Wesley Publishing Company, 1987.*

16. J.V. Beck, K.J. Arnold, Parameter estimation in engineering and science*, John Wiley & Sons, New York, 1997.*