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Measurement of thermal properties of a ceramic/metal joint by laser flash method

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ABSTRACT

This work describes the measurement of thermal diffusivity and the subsequent calculation of thermal conductivity and thermal contact resistance (from room temperature to 800 °C in N_2 atmosphere) at the material interface for carbon/carbon composites (C/C) joined to copper by using the laser flash method. According to these measurements, the thermal resistance at the interface, that is related to the heat transfer through the solid, is $<10^{-6}$ $m^2\,K\,W^{-1}$ up to 800 °C, indicating a high quality of the joint and no limitations for the thermal heat transfer during operation, e.g. in a nuclear fusion reactor. This measurement is proposed as an innovative non-contact and qualitative investigation technique to assess the ceramic/metal joint integrity.

1. Introduction

Carbon/carbon composites (C/C) will be part of the divertor, a complex and critical component of ITER (Latin for "the way") [1]. In particular, the divertor consists of a plasma facing material made of C/C which must be joined to a Cu-based alloy coolant structure. Between them there is a thin compliant layer of pure copper, able to accommodate thermally induced stresses and to provide high thermal conductivity, an essential parameter to remove the surface heat flux and to avoid overheating of the structures. Different design options for the joining of C/C to a Cu alloy have been developed [2,3].

Measurements of thermal conductibility and contact resistance of the C/C–Cu joint is helpful to understand and predict its in service thermal behavior.

If we consider two solids, in contact by a planar surface, as for C/C joined to copper, the thermal conductivity at the interface depends on the quality of the joint itself. As a result of a poor joining process, the surface can have a discontinuous interface and poor thermal contact. The heat flow across an interface takes place by conduction at the interface. Voids or porosity at the interface act as thermal resistance to the heat flow. Since this resistance is confined to a very thin layer between the surfaces, it is called "thermal contact resistance" [4].

The values of thermal contact resistance depend on several parameters: (1) the thermal conductivity at interface region; (2) the joining process which should lead to a flawless interface; (3) the materials to be joined (surface roughness, etc); (4) the induced thermal stress due to the coefficient of thermal expansion (CTE) mismatch.

High thermal contact resistance values are associated to a discontinuous interface and a poor quality of the joint; if the thermal contact resistance is low, the joined component can dissipate a significant quantity of heat for a given thermal gradient, thus confirming a good quality of the joint interface [5–7].

This paper reports on the measurement of thermal conductivity and thermal diffusivity of joined C/C–Cu samples by using the laser flash method, one of the most widely used techniques to measure thermo-physical properties of materials. Furthermore, this work analyzes the thermal contact resistance for this two-layered system (joined C/C–Cu) with and without calibrated defects and demonstrates that the measurement of the thermal contact resistance can be used as a qualitative technique to detect defects at interface that can impair the joined component performances appreciably.

For decades the laser flash method has been used to characterize the thermo-physical properties of thin solid materials. One of the most important advantages of this non-contact technique is the capability to analyze multi-layer samples by fitting the experimental data with an appropriate analytical model describing the transient heat transfer.

Theoretical models for the solutions of multi-layer systems were proposed by Lee [8]: they were based on adiabatic models that did not take into account any heat loss effects from the sample surfaces. Nevertheless, in order to achieve an accurate estimation of the contact resistance, a heat loss effect has to be included in the fitting model of experimental data. This procedure was developed by Cowan [9] for single material samples and later extended to multi-layer systems [10]. Several standards, referred to laser flash techniques, are applied to single layer samples or bulk samples, but not to multilayers [11].

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To the best of the authors' knowledge, no studies have been reported till now about measurements of thermal contact resistance between C/C composites and copper in joined samples.

2. Experimental

The geometry of the joined C/C–Cu component, which was cut from a parallelepiped in a disk-shaped plane-parallel sample, is shown in Fig. 1. The copper used for the joints is OFHC copper, i.e. Oxygen Free High Conductive copper (purity is 99.95%).

The used C/C (carbon fiber reinforced carbon matrix composite) is a high thermal conductivity 3D composite (CFC NB31) specifically developed for fusion application by Snecma Propulsion Solide, France; the preform is made from a needled texture composed of pitch- and PAN-based carbon fibres. This 3D texture is densified with pyrocarbon through a CVI process and then heat treated at high temperature to make it highly conductive and thermally stable. Fibers with maximum thermal conductivity are typically parallel to the direction of heat flux, i.e. perpendicular to the C/C surface to be joined to copper [12].

The CFC NB31 surface was modified by forming a chromium carbides layer and the joint between C/C and Cu is then obtained by Cu casting. The joining process is described elsewhere [13,14].

In order to assess the behavior of joined samples with detached interface, a C/C–Cu joined sample (disk-shaped, dimensions as above) was manufactured with a calibrated joint defect at the metal/composite interface; the defect has a circular cross section (1.5 mm diameter); it was manufactured in C/C at the interface between C/C and the copper layer by machining from the side surface. The reduction of the joint interface by making this defect was about 1.5%, which is a reasonable value that can be ascribed (in an effective joint) to a processing induced defect (i.e. porosity, not well wetted area at interface, etc).

A Laser Flash Apparatus LFA 457 MicroFlash, allowing measurements between room temperature and 1100 °C by a coupled furnace, was used to investigate the samples [15].

The thermal properties of non joined Cu and C/C samples and of C/C–Cu joints were measured from room temperature to 800 °C, in N_2 atmosphere with 80 ml/min flow; the used temperature ramp rate is 1 K/min up to 200 °C and 10 K/min up to 800 °C and the temperature stability during the measurement is 1 K/30 s. The number of shots at each temperature is 5. The laser parameters are the following: voltage = 1538 V, filter: 100% trasmissivity, energy = 11 J and laser pulse width = 0.5 ms.

The principle of operation is outlined in Figs. 2 and 3. In the laser flash test, a short laser pulse, generated by a Nd-glass laser with a duration of 0.5 ms, hits, guided by a mirror, the front side of a plan-parallel sample disk.

The samples were not coated with either graphite aerosol or other coating material before testing, even if this procedure represents a deviation from standard operating practice for flash diffusivity regardless of single layer or multilayer analysis; in any case, the lack of a graphite coating on the top surface is not expected to influence the results.

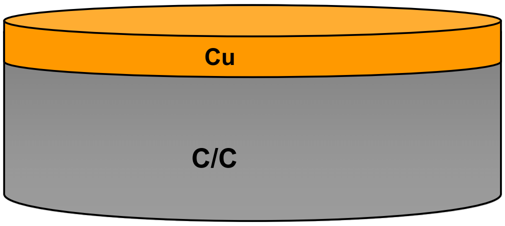

Fig. 1. Schematic of joined C/C–Cu sample with contacting surfaces (dimensions: height = 8.35 mm, diameter = 12.7 mm; C/C thickness = 6.014 mm Cu thickness = 2.335 mm; weight = 4.105 g).

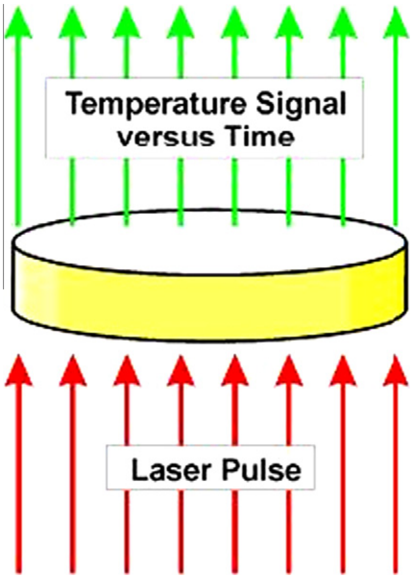

Fig. 2. Principle of operation of laser flash apparatus used in the experimental work.

The sample is placed on a carrier in the centre of a high temperature furnace capable to operate between room temperature and 1100 °C (Fig. 3). The pulse energy is absorbed in very thin layer on the surface and converted into a confined, bi-dimensional, heat source. The heat waves diffuse through the sample and lead to a temperature rise on the rear side of the sample. An indium antimonide infra-red detector, cooled by liquid nitrogen, is directed to the rear surface of the sample and generates a highly sensitive signal on response to the temperature transient of the sample. The detector electronic signals are sampled at different time windows, ranging from 700 ms (at sample temperature of 25 °C) to 2300 ms (at sample temperature of 800 °C). After acquisition, the data, i.e. the temperature rise versus time, are processed by a non-linear regression routine, allowing a curve fitting of the measurement results by the Cowan model [10] of the heat propagation through the multilayer sample.

The measurement of thermal diffusivity in combination with the determination of specific heat and bulk density allows, by calculation of their scalar product, a direct determination of the thermal conductivity according to:

$$k = \rho \cdot c_p \cdot a \qquad (1)$$

where k is the thermal conductivity, ρ is bulk density, c_p is specific heat, a is the thermal diffusivity.

The utilized model used by the instrument for the calculation of the thermal contact resistance requires the input of the value for the thermal expansion coefficient since as the temperature increases, the thickness of the layers forming the joint increases.

In addition, the C/C and Cu density values have been calculated using thermal expansion data; the only density values that were not obtained from CTE are the room temperature densities of both C/C and Cu: they have been measured for C/C and Cu (i.e. 1.958 g/cm^3 and 8.93 g/cm^3, respectively, determined by direct measurement of their weight and volume), from which the temperature-dependent densities were calculated using CTE data from Ref. [16]. The used CTE values for C/C materials take into account the 3-dimensional structure of the composites.

Preliminary experiments were carried out on non joined Cu and C/C samples, in order to measure thermal diffusivity, specific heat

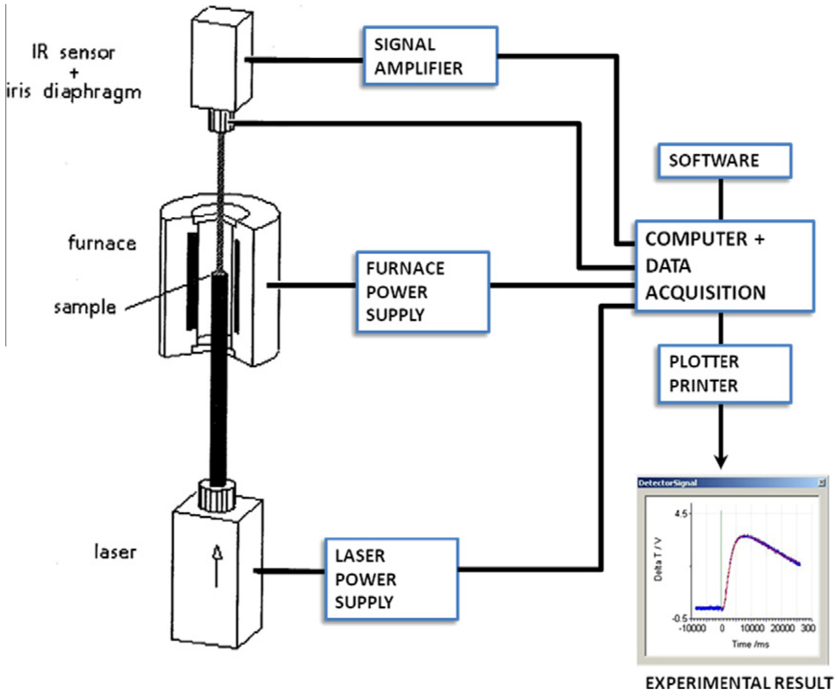

Fig. 3. Schematic design of the LFA 457 MicroFlash System.

and thermal conductivity of the two materials versus temperature. Two disk-shaped plane-parallel samples of C/C and Cu have been studied (thickness = 3.15 mm, diameter = 12.7 mm) as reference samples. The thickness deviation of the joined C/C–Cu sample is less than 0.01 mm, according to machining tolerance on flatness of reference samples.

Specific heat values of the two layers (Cu and C/C) forming the joined sample are obtained through comparative measurements with a reference standard (high purity iron, supplied and certified by Netzsch-Geratebau).

3. Results and discussion

Results on measured thermal diffusivity, thermal conductivity, specific heat and calculated density between 25 and 800 °C for non joined Cu and C/C are given in Table 1. The obtained values of c_p for Cu is higher than the literature value at room temperature, but the discrepancy is significantly reduced at higher temperature [17]. The calculated values for C/C fit well [18,19]; the reported uncertainty of the instrument on c_p calculation is about 5%. It confirms that laser flash method is less accurate then differential scanning calorimetry DSC measurement (which is the standard method for getting specific heat values), but it serves the purpose of this study.

An SEM backscattered image of the measured C/C–Cu joint is shown in Fig. 4. Two interfaces can be observed: C/C–chromium carbides and chromium carbides–Cu interfaces; the average thickness of the carbides layer is about 10–15 μm. The further discussion will be concentrated on the case of a 10 μm thick layer.

It can be supposed that, in the experimental conditions of the laser flash test, the Cr diffusion at interface is low and significant growth of the Cr carbides will not occur. In the following discussion it will be assumed that the physical state of the sample remains constant over the whole testing time.

The thermal diffusivity of the joined sample has been measured as described in the Section 2 for the non joined samples: the curve describing the thermal diffusivity is obtained by interpolation taking into account in the model the presence of two layers in contact. The joined sample has been modeled by two layers in contact through a contact resistance.

All thermal parameters reported in Table 1 (specific heat, thermal conductivity and thermal diffusivity) measured for the two materials have been used in the model. Thermal contact resistance is the only open parameter that can be changed in the instrument model.

Table 1
Measured thermal diffusivity, specific heat, thermal conductivity and density between 25 and 800 °C for non joined Cu and C/C.

	Copper				C/C			
T (°C)	Diffusivity (mm^2/s)	Specific heat (J/kg K)	Conductivity (W/m K)	Density (g/cm^3)	Diffusivity (mm^2/s)	Specific heat (J/kg K)	Conductivity (W/m K)	Density (g/cm^3)
25	110.8	447	442	8.920	216.5	695	294.4	1.959
150	104.5	455	424.3	8.860	133.8	965	253	1.959
300	98.7	468	412.2	8.790	90	1280	225.5	1.959
450	93.8	480	401.8	8.700	69.3	1508	204.2	1.958
600	89	493	391.5	8.616	56.7	1659	183.6	1.958
700	85.5	503	383.8	8.570	51	1710	170	1.957
800	82.2	511	374.8	8.483	47.1	1730	156.4	1.956

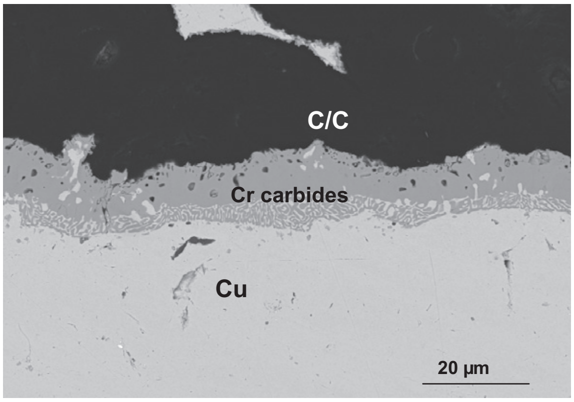

Fig. 4. SEM microstructure of C/C–Cu interface area; a chromium carbides layer is evident between the C/C and Cu.

The pulse propagation across the sample can be described by using an adiabatic model [10]. It states that the laser beam is evenly absorbed over the whole front of the test piece in an infinitely short time, the initial temperature of the test piece is constant and the test piece is homogeneous. No heat exchange with the surroundings occurs. An adiabatic heat transport (mathematically one-dimensional) takes place in the test piece if the above conditions have been fulfilled. At first, the temperature at the back of the test piece rises. It remains constant after reaching its maximum. It has been calculated [10] in this case that the thermal diffusivity α is:

$$\alpha = L^2/t_{1/2} \tag{2}$$

where L is the sample thickness and $t_{1/2}$ is the half rise time of the temperature versus time curve.

As explained before, the mathematical model can consistently evaluate the thermal contact resistance between two different materials, as long as the ratio

$$\frac{t_{\text{layer1}}}{t_{\text{layer2}}} \quad \text{with } t_i = \frac{L_i^2}{\alpha_i} \tag{3}$$

(where L is the thickness of the layer and α is the thermal diffusivity) is lower than 10.

In the present case, the layer 1 corresponds to C/C and the layer 2 to Cu, since the mismatch of the diffusivity and the thickness of the two layers is significant and the layer with the highest $t_{1/2}$, i.e. the C/C layer, dominates the temperature change as a function of time. For higher temperatures (>600 °C) the result accuracy is reduced because of lowered mathematical model fitting [10].

Fig. 5 represents the model of a two layer system, made of two solids in contact through a joint namely solid 1 (Cu) and solid 2 (C/C). The solids 1 and 2 have thicknesses x_1 and x_2 and thermal conductivities k_1 and k_2, respectively. The heat per unit of area that flows, assuming thermal equilibrium, from the front surface at temperature T_2 across the interface to the backside surface at temperature T_1, is given in (4)

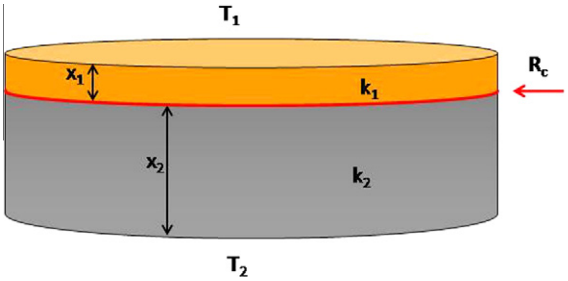

Fig. 5. Schematic view of an interfacial joint for two contacting interfaces, namely solid 1 (Cu) and solid 2 (C/C); solids 1 and 2 have thicknesses x_1 and x_2 and thermal conductivity k_1 and k_2, respectively. Upon heating, the heat flows from the surface at temperature T_2 to the surface at temperature T_1 across the interface.

$$Q(\mathrm{W/m^2}) = \frac{T_2 - T_1}{x_1/k_1 + R_c + x_2/k_2} \tag{4}$$

Therein R_c is the thermal contact resistance at the interface.

This equation is analogous to the Ohm's law for electric circuits where the charge flow (i.e. the current density) is replaced by the heat flow Q, $T_2 - T_1$ substitutes the electric potential difference and the denominator is the sum of three resistors; this justifies the choice to name x/k as thermal resistance and R_c as thermal contact resistance. It allows comparing the effect of the thermal resistance of the two layers to that of the contact.

The copper thermal resistance R_{Cu} and the C/C thermal resistance $R_{\mathrm{C/C}}$ are:

$$\begin{aligned} R_{\mathrm{Cu}} &= \frac{x_{\mathrm{Cu}}}{k_{\mathrm{Cu}}} \\ R_{\mathrm{C/C}} &= \frac{x_{\mathrm{C/C}}}{k_{\mathrm{C/C}}} \end{aligned} \tag{5}$$

To perform the analysis, the thermal expansion coefficient from Ref. [16], the measured specific heat and thermal conductivity versus temperature of the non joined Cu and C/C (Table 1) have been used. As the temperature increases, the sample changes its dimensions according to its CTE variations and the heat pulse propagation modeling is modified by the different sample geometry. Moreover, the density of the two layers is changed by the temperature increase and, according to Eq. (1), the thermal conductivity too. The CTE is required by the model to calculate the thermal parameters in the temperature range. It has been estimated that multilayer thickness correction due to thermal expansion affects the measurement uncertainty by less than 1% up to 1000 °C.

The C/C–Cu joints, with and without calibrated defects, have been tested at various temperatures between 25 and 800 °C in N_2 atmosphere. Measurement has been repeated five times for each value of temperature; Fig. 6 shows the obtained results for the C/C–Cu joints. The dots indicate the average of the contact resistance calculated at each temperature in each individual run; five shots at each temperature have been performed. The respective standard deviation are also introduced in the figure as bars.

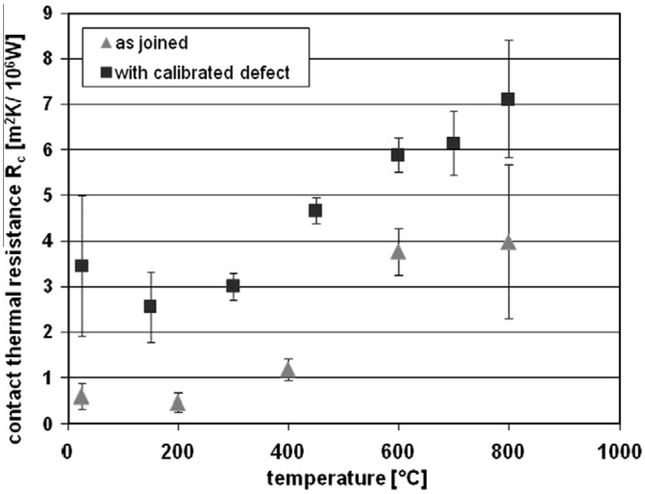

Fig. 6. Calculated contact thermal resistance an standard deviation for C/C–Cu joints as-joined and with calibrated defect at interface; the dots indicate the average of the contact resistance calculated on five shots per each temperature.

The trend of the two curves is similar: the thermal contact resistance of a defective sample is higher than that of the flawless one by about 3×10^{-6}. The thermal resistance of the joint is thus related to the integrity of the joint itself and is an indication of the actual heat transfer area. Thermal resistance is minimized by making the joint as continuous as possible by eliminating interstitial air and making sure that both surfaces are in intimate contact and flawless [20].

As it can be observed in Fig. 6, in the case of "as-joined" sample the difference between the R_c at room temperature and that at 200 °C falls into the uncertainty of the measurement. Regarding the sample with calibrated defect, a substantial variation of the sample geometry has been induced thus conditioning the sample modeling. The induced flaw in the defective joint can be described as a cylindrical hole; on the other hand, the fitting model is based on a thermal contact resistance area as a thin layer between the C/C and the Cu and does not refer to a local discontinuity.

At temperatures lower than 200 °C the accuracy of the model is affected by the difference between true (with local discontinuity) and ideal geometry (without any discontinuity) and the diffusivity values obtained at room temperature are no representative; at temperatures above 200 °C the model accuracy is not influenced by the local discontinuities.

If the contact between the two interfaces (copper–chromium carbides and chromium carbides–C/C, Fig. 5) is ideal, without defects and stresses, the thermal contact resistance can be ascribed to the chromium carbide layer ($R_c = R_{C,Cr-carbides}$). By assuming the average thickness of the chromium carbides layer to be about 10 µm, the thermal conductivity of the chromium carbides layer can be determined by readjusting Eq. (6) and compared with literature data.

$$R_{C,Cr-carbides} = \frac{x_{Cr-carbides}}{k_{Cr-carbides}} \tag{6}$$

The calculated value of thermal conductivity of chromium carbides of the as-joined sample is 16.6 W/m K at room temperature and about 21.4 W/m K at 200 °C. In Ref. [21] it is reported that the chromium carbides thermal conductivity is 19 W/m K at RT, comparable to the value achieved by our estimation. This means that the thermal quality of the as-joined sample is very good, without pores or discontinuities. In fact, the presence of only few pores in the joint area gives a significant increase of thermal resistance, due to the very low value of the thermal conductivity of air (0.026 W/m K at RT).

4. Conclusions

Experiments were performed using laser flash technique to measure thermal properties of a joint interface between C/C and copper.

The thermal contact resistance of the chromium carbides layer at interface provided useful information regarding the quality of the joint; this technique has the advantage of being fast and non-destructive, to require small samples and an easy sample preparation.

It can be used as an investigation method for characterizing thermal performance of ceramic/metal structures, especially for those components which cannot be correctly analyzed with traditional NDT [22]. Moreover, it can be one of the few suitable methods for the thermal analysis of multilayer components. While use of this technique is valuable to provide engineering data and information on detached interface at lab-scale, it does not replace the need for non-destructive examination of fabricated components, since it requires preparation of samples on the order of 10 mm in size and cannot probe for large areas of delamination or interface defects on the meter scale.

A preliminary study on influence of calibrated defect on the thermal contact resistance for a two-layered system has been carried out; further investigations will be addressed to correlate the result for the thermal contact resistance to the amount of defects. Further studies with different amounts of defects will be performed which then may lead to higher R_c values; the correlation between defects and reduction of thermal performance of the joints will be investigated.

References

[1] R. Tivey, E. D'Agata, V. Chuyanov, H. Heidl, Fusion Eng. Des. 75–79 (2005) 447–450.
[2] A.T. Peacock, M. Merola, M.A. Pick, R. Tivey, Phys. Scripta T128 (2007) 23–28.
[3] J. Linke, Fusion Sci. Technol. 49 (2006) 455–464.
[4] C.V. Madhusudana, Thermal Contact Conductance (Mechanical Engineering Series), Springer Verlag Inc., New York, 1996.
[5] A. Lahmar, T.P. Nguyen, D. Sakami, S. Orain, Y. Scudeller, F. Danes, Thin Solid Films 389 (2001) 167–172.
[6] C. Chiu, J.G. Maveety, Q.A. Tran, Microelectron. Reliab. 42 (2002) 93–100.
[7] J. Absi, D.S. Smith, B. Naït-Ali, S. Grandjean, J. Berjonnaux, J. Eur. Ceram. Soc. 25 (4) (2005) 367–373.
[8] H.J. Lee, Thermal Diffusivity in Layered and Dispersed Composites, PHD-Thesis, Purdue University, Indiana (USA), 1975.
[9] R.D. Cowan, J. Appl. Phys. 34 (1963) 926–929.
[10] Hubert M. James, J. Appl. Phys. 51 (9) (1980) 4666–4672.
[11] ASTM E1461 – 07 Standard Test Method for Thermal Diffusivity by the Flash Method.
[12] G. Pintsuk, J. Compan, J. Linke, P. Majerus, A. Peacock, D. Pitzer, M. Rödig, Phys. Scripta T128 (2007) 66–71.
[13] P. Appendino, V. Casalegno, M. Ferraris, M. Grattarola, M. Merola, M. Salvo, J. Nucl. Mater. 329–333 (2004) 1563–1566.
[14] P. Appendino, M. Ferraris, V. Casalegno, M. Salvo, M. Merola, M. Grattarola, J. Nucl. Mater. 348 (1–2) (2006) 102–107.
[15] S. Min, J. Blumm, A. Lindemann, Thermochim. Acta 455 (2007) 46–49.
[16] ITER Materials Properties Handbook, G AO FDR1 01-07-13 R1.0 Materials Assessment Report, Armour/Heat Sink Joining Technologies, Internal Project Document Distributed to the ITER Participants.
[17] ITER Materials Properties Handbook, ITER Doc. No. S 74 MA 2
[18] ITER Materials Properties Handbook, ITER Doc. No. G 74 MA 9 00-11-10 W 0.1.
[19] B. Hay, J.R. Filtz, J. Hameury, L. Rongione, Int. J. Thermphys. 26 (6) (2005) 1883–1897.
[20] S. Shaikh, K. Lafdi, E. Silverman, Carbon 45 (2007) 695–703.
[21] H. Pierson, Handbook of Refractory Carbides and Nitrides: Properties, Characteristics, Processing, and Applications, Noyes Publications, New Jersey, 1996.
[22] V. Casalegno, M. Salvo, M. Ferraris, F. Smeacetto, M. Merola, M. Bettuzzi, Fusion Eng. Des. 83 (2008) 702–712.