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Thermal response of two-layer systems: Numerical simulation and experimental validation

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Abstract

Heat transfer through a two-layer system constituted of different materials has been studied. We focus on the overall apparent thermal conductivity in the direction perpendicular to the planes of the layers and consider the case of two materials, alumina and glass, which exhibit a strong difference in individual thermal conductivity. Numerical simulations using finite element analysis have been performed for two situations: transient response corresponding to characterisation of the thermal conductivity by the laser-flash experiment and steady-state response corresponding to the behaviour of two resistors in series. The calculations show that the two methods give similar results up to a volume fraction equal to 0.5 of the alumina phase. However, for alumina volume fractions above 0.5 the laser-flash experiment overestimates the overall thermal conductivity predicted by the steady-state series model. This is explained by a limit to the validity of a homogeneous approximation to the two-layer heterogeneous system, typically used in the analysis of laser-flash data. Experimental results, obtained on alumina/glass two-layer samples of varying relative thicknesses, support these deductions, though the thermal resistance of the contact between the layers should also be taken into account.

Keywords: Thermal conductivity; Al_2O_3/glass; Simulations; Laminates

1. Introduction

Two-layer systems are encountered in a wide variety of industrial applications.[1,2] For example, in mechanical engineering the high temperature performance (>400 °C) of moving solid surfaces in contact can be improved by the addition of a layer of material with high resistance to wear.[3] In electronics, two-layer or multilayer systems are assembled in order to remove excess local heat resulting from the Joule effect. Knowledge, of the thermal behaviour and characteristics of such systems, is therefore important for achieving optimum operating efficiency.

In this paper, we focus attention on the overall thermal conductivity of a two-layer system consisting of a glass layer joined to an alumina layer. These materials exhibit significantly different values of thermal conductivity and we consider the heat flow direction perpendicular to the planes of the layers. In theory a steady-state method, exploiting Fourier's law, can be used to obtain an experimental value for the overall thermal conductivity. In practise, transient methods, and in particular the laser-flash method, are preferred to reduce errors due to thermal contact resistance and heat losses.[4,5] The laser-flash method measures directly the overall apparent thermal diffusivity of the two-layer system. Bulmer and Taylor[6] have developed an analytical model for this situation and demonstrated its validity for two-layer systems where the volumes fractions are not too different. The steady-state thermal conductivity can then be calculated assuming that the system is a homogeneous medium. Strictly, in the case under study, this is not true. Consequently, the paper examines the validity of such a homogeneous approximation to a heterogeneous situation. First, we compare numerical simulations of the transient behaviour of a two-layer system in the flash experiment to a simple steady-state model based on two resistors in series. We explore the role of the volume fractions of each layer to determine when the approximation is valid and when it is not. Then, the computer-simulated results are compared to real experimental data for the alumina/glass two-layer system.

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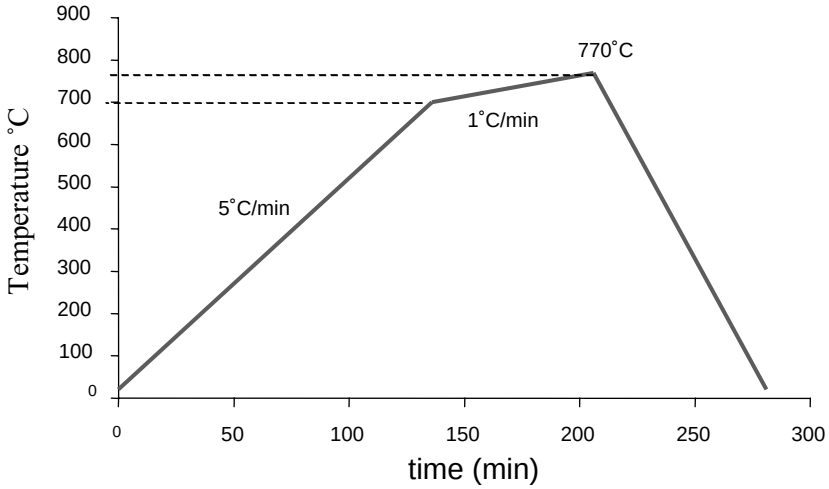

Fig. 1. Heating cycle used in the fabrication of the two-layer system.

2. Experimental procedure

2.1. Sample preparation

Two-layer samples of alumina joined to glass were made with a total thickness of approximately 3 mm. Initially, the layers of glass and alumina were prepared separately. The composition of the glass was chosen so that its coefficient of thermal expansion was close to that of alumina, which was measured with a value of $6.8 \times 10^{-6}\,K^{-1}$ at room temperature. Hence, 3 wt.% of Na_2O was mixed with a glass frit (Ferro-BC832). A glass slab was then obtained by firing at 940 °C. The corresponding alumina layer was obtained by uniaxially pressing fine powder (P172 SB, Pechiney) into a 25 mm disc and firing at 1600 °C for 4 h. After polishing the contact surfaces, joining was then achieved by pressing the two layers together during a heating cycle at 700 °C (Fig. 1). The two-layer samples were then cut and polished into discs of 8 mm in diameter suitable for the laser-flash experiment. A batch of two-layer samples was made with variation of the volume fraction (relative thickness) between the two solid phases (Table 1). Individual samples of each solid phase were prepared for the characterisation of the basic parameters which were needed for the analytical calculations and simulations. The quality of the interface was also examined by scanning electron microscopy. Fig. 2 shows that it is well formed and devoid of significant air gaps resulting from separation of the two layers.

2.2. Characterisation

The density of each type of layer was evaluated using the method based on Archimedes' principle. For the two-layer samples an overall value of density, ρ_a, was calculated using an expression based on the rule of mixtures:

$$\rho_a = \frac{\rho_1 v_1 + \rho_2 v_2}{v_1 + v_2} \quad (1)$$

where ρ_1, ρ_2 refer to the measured values of density for each layer. The volume fractions v_1, v_2 were estimated from the relative thicknesses of each layer, measured using the translation table of an optical microscope. In a similar way, a value for the overall specific heat, C_a, was evaluated with the expression:

$$C_a = \frac{C_1 \rho_1 v_1 + C_2 \rho_2 v_2}{\rho_1 v_1 + \rho_2 v_2} \quad (2)$$

where C_1 and C_2 refer to the specific heats of the alumina and glass solid phases at room temperature.

The laser-flash method was used to determine, via the thermal diffusivity, the overall thermal conductivity of the samples. A short pulse (0.5 ms) of a laser beam (assimilated in theory to a Dirac function) is used to heat up the front face of the cylindrical sample. The absorbed heat diffuses throughout the sample and an infra red detector is used to monitor the evolution of the back face temperature. Samples

Table 1
Characteristics of the batch of two-layer samples

Two-layer samples	Total thickness (m)	% volume of alumina
1	3.6E − 03	9.2
2	2.9E − 03	15.5
3	3.2E − 03	16.2
4	2.4E − 03	23.5
5	3.0E − 03	32.2
6	3.7E − 03	33.3
7	2.8E − 03	35
8	3.3E − 03	44
9	3.9E − 03	48.7
10	3.5E − 03	49
11	2.8E − 03	49.1
12	2.6E − 03	49.2
13	2.9E − 03	66.3
14	1.8E − 03	67.4
15	2.6E − 03	80.8
16	2.3E − 03	90.2
17	2.3E − 03	90.5

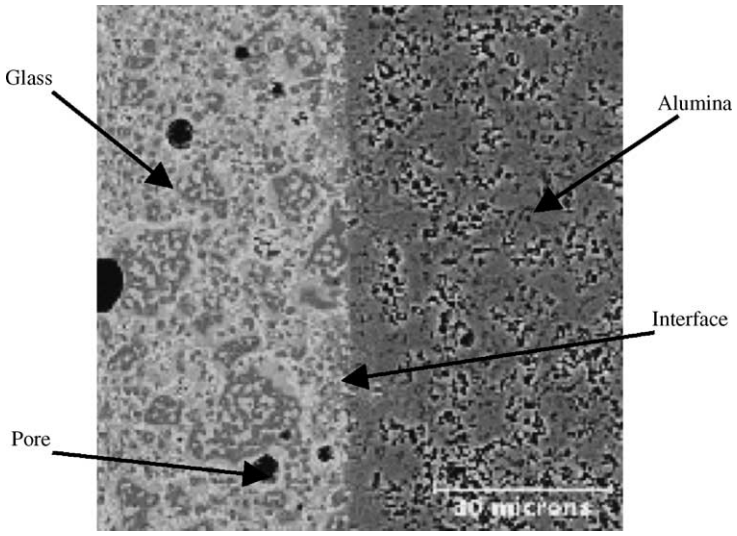

Fig. 2. Micrograph of the alumina/glass interface.

were coated with a thin graphite layer to improve laser beam absorption and also the emitted signal from the back face. The back face temperature can be analysed with Parker's original expression:[4]

$$\alpha = \frac{0.139e^2}{t_{1/2}} \tag{3}$$

where e is the sample thickness and $t_{1/2}$ the time taken by the back face to increase its temperature to one-half of the maximum value. However, the thermal diffusivity value will be overestimated if heat losses occur. This discrepancy is due to the deviation from the ideal temperature–time curve for a homogenous sample described in the form of a universal curve in a dimensionless plot. A number of procedures have been developed to provide corrections for non-ideal behaviour.[7–11] Similar to Clark and Taylor's method for handling heat losses,[10] we have used Degiovanni's expression to calculate experimental values of thermal diffusivity:[11]

$$\alpha = \frac{e^2}{t_{5/6}}\left[0.8498 - 1.8451\frac{t_{1/3}}{t_{5/6}} + 1.0315\left(\frac{t_{1/3}}{t_{5/6}}\right)^2\right] \tag{4}$$

where $t_{1/3}$ and $t_{5/6}$ refer to times for temperature increases to 1/3 and 5/6 of the maximum value, respectively.

A further advantage to this approach is that non-linearity in the conversion of the detector signal into recorded voltage is also taken into account. The apparent thermal conductivity λ_a is then given by the relation:

$$\lambda_a = \alpha\rho_a C_a \tag{5}$$

where ρ_a is the density and C_a is the specific heat of the sample which is assumed to be a homogenous medium. The overall specific heat and density values for each two-layer sample were evaluated using the data in Table 2 and Eqs. (1) and (2). Experimental error in α is estimated at 5%.

3. Numerical simulation

With respect to the laser-flash method, the calculation of the overall conductivity of a two-layer system should be based on the transient heat transfer behaviour. Analytical calculations for heat propagation in a two-layer system have been made involving a Laplace transformation on the general heat equation in the form

$$\frac{\partial T}{\partial t} = \alpha\frac{\partial^2 T}{\partial x^2} \tag{6}$$

Assuming a perfect contact, Bulmer and Taylor[6] used a computer routine to solve a complicated series of equations and predict the overall temperature–time behaviour. Bouayad et al.[12] extended the approach to take into account the thermal resistance of the contact between the layers. Given the difficulties of the analytical approach, a convenient and efficient alternative is provided by numerical simulation. We have therefore modelled the transient response of the two-layer system in conditions corresponding to the laser-flash experiment using a finite element analysis software package called ABAQUS. The operations, which are necessary for the simulation, can be divided into three major parts: model construction, calculation and interpretation of the results.

The sample symmetry in the form of a disc and the reception of a uniform heat flow on the front face allow a two-dimensional model to be used. The apparent thermal conductivity of this system was studied for the transient con-

Table 2
Characteristics of individual materials

	Glass	Alumina
Density [kg/m^3]	2760	3230
Specific heat [J/kg K]	744	766
Conductivity [W/m K]	1.5	14

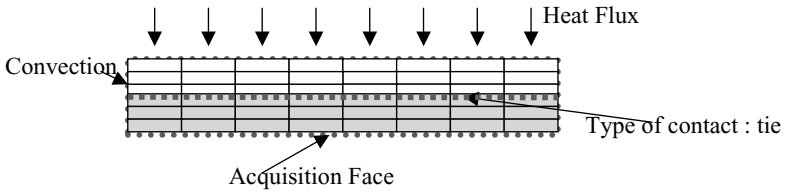

Fig. 3. Model of a two-layer sample in the laser-flash experiment.

ditions corresponding to the laser-flash experiment and also in steady-state conditions after application of a constant temperature difference across the sample (Fig. 3). The thermal contact between the two layers is assumed to be perfect (no thermal resistance).

3.1. Steady-state conditions

A temperature difference is applied across the sample and the heat flow leaving the back face as a function of time is simulated. The apparent thermal conductivity λ_a is then calculated from Fourier's law and the steady-state value of the heat flow with:

$$\lambda_a = \frac{\phi e}{T_1 - T_2} \tag{7}$$

where ϕ is the back face heat flow density ($W\,m^{-2}$) and e is the total thickness of the sample. The situation is simulated with a single calculation step after time duration of 5–10 s depending on the sample thickness. Heat loss to a convection environment at 20 °C is fixed at the curved sample surface of the cylinder.

3.2. Transient conditions

The laser-flash experiment is simulated first, by a step in which the laser beam flash is modelled by a heat flow density at the front face of $1.2 \times 10^8\,W\,m^{-2}$ lasting 0.45 ms, and second, by a step of heat propagation through the sample. The total calculation time, allowing for achievement of thermal equilibrium, was 10–20 s depending on the sample thickness. In both these steps the time and space increments are chosen so as to give maximum information while respecting reasonable calculation times.

4. Results and discussion

4.1. Steady-state calculation

The values of thermal conductivity predicted by numerical simulation for a two-layer system in the steady state are shown in Fig. 4. They are plotted as a function of the volume fraction of the alumina phase and agree very closely with the solid line calculated by the combination of two thermal resistances in series:

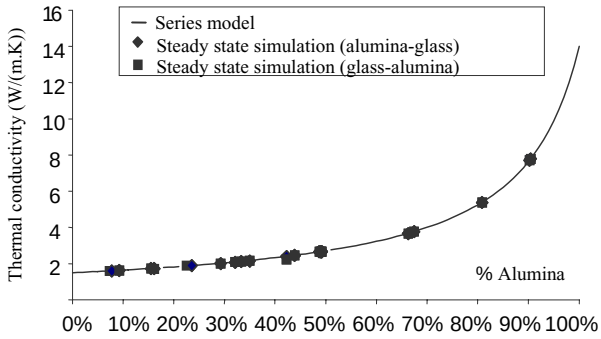

Fig. 4. Thermal conductivity calculated by numerical simulation (steady state) and by a series model using Eq. (8) vs. volume fraction of alumina.

$$\frac{e_1 + e_2}{\lambda_a} = \frac{e_1}{\lambda_1} + \frac{e_2}{\lambda_2} \tag{8}$$

where e_1, e_2 refer to the thicknesses of the individual layers and λ_1, λ_2 refer to their thermal conductivities. The numerical simulation is therefore validated. A further check was performed by reversing the temperature difference across the simulated sample and this again revealed no difference.

4.2. Transient conditions (laser-flash experiment)

The laser-flash experiment was simulated numerically on each two-layer sample by predicting the corresponding temperature–time behaviour at the centre of the back face. Parker's expression and the Degiovanni method were both used to calculate the thermal diffusivity and then the corresponding thermal conductivity values were obtained with Eq. (5). In Fig. 5, the results are compared with the steady-state series model given by Eq. (8). Both sets of

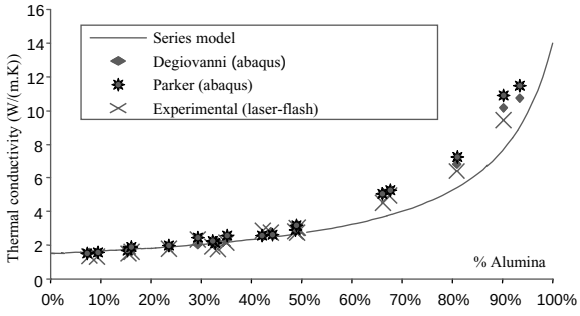

Fig. 5. Thermal conductivity determined by numerical simulation (transient regime), by the series model and by the laser-flash technique vs. volume fraction of alumina.

data show a similar trend as a function of alumina volume fraction. However, above 50% of alumina, a significant difference is observed between the steady-state values and those obtained by the transient calculations. For large volume fractions of alumina (the conducting phase) the transient values are up to 15% higher than the steady-state counterparts. Furthermore, there is also a slight separation of the Parker and Degiovanni values.

The differences could be explained by:

- a problem in the accuracy of the numerical simulation of the transient regime,
- the influence of heat losses which deform the temperature–time curve.

The close agreement of the numerical calculations for samples up to 50% volume fraction of alumina with the steady-state curve suggests that the accuracy of the procedure is not in question. Further support to this idea is given by the transient calculations for 100% alumina where the values coincide with the steady-state value. Heat losses can also lead to an over estimation of the thermal diffusivity by the Parker expression and hence an increase in the thermal conductivity value. Though this would explain the separation between the Parker and Degiovanni values the argument is not consistent. First, such an effect should be more pronounced for the more insulating lower alumina volume fraction samples, which is not the case. Second, the Degiovanni values, essentially independent of heat losses, should lie on the steady-state curve which is not observed between 65 and 95% volume fractions of alumina. We note also that the Parker and Degiovanni values coincide for 100% alumina.

We therefore ask whether there is a limit to the validity of the homogeneous approximation used implicitly in Eq. (5). For strongly heterogeneous samples constituted of a thin insulating layer joined to major conducting phase, Eq. (5) overestimates the steady-state thermal conductivity. In essence, the laser-flash experiment measures the overall thermal diffusivity ($\alpha = \lambda/\rho c$), a sort of inverse thermal time constant, which is only slightly modified by the addition of the glass layer. In contrast, the glass layer strongly reduces the overall steady-state thermal conductivity.

More evidence is given by the experimental results measured with laser-flash method which are compared to the calculations in Fig. 5. For samples with less than 50% volume fraction of alumina, good agreement is found between experiment and theory. Above 50% volume fraction of alumina this agreement is no longer true. The experimental values are found between a lower limit given by the steady-state series combination of resistors and an upper limit given by the numerical simulation of the laser-flash experiment. The difference from the steady-state behaviour increases with the volume fraction of alumina. This supports the interpretation that there is a limit to the validity of a homogeneous approximation to a two-layer heterogeneous system.

Another manifestation of the heterogeneous response of the two-layer system can be found by examination of the temperature–time behaviour in Fig. 6. The plots are given in dimensionless units of temperature and time

$$T^* = \frac{T - T_0}{T_{\max} - T_0} \tag{9}$$

where T_0 is ambient temperature, $T_{\max}$ the maximum temperature achieved by the sample after reception of the laser pulse in adiabatic conditions, and

$$\omega = \frac{\pi^2 \alpha t}{e^2} \tag{10}$$

where t is the time, e is sample thickness and α is the apparent thermally diffusivity of the sample assuming homogeneous response. The curve for 100% alumina gives the reference homogeneous response for a solid and is essentially independent of the value of thermal diffusivity. The simulated curve for the two-layer system with 10% in volume of glass exhibits a significant deviation from this homogeneous response especially for times greater than $t_{1/2}$ (Fig. 6a). It can be seen that the two curves cross over at about $t_{1/2}$ meaning that the Parker analysis is almost unaffected. In contrast, the Degiovanni analysis, based in part on $t_{5/6}$ will apply a correction to push $t_{5/6}$ back onto the homogeneous curve. This increases its value and hence decreases the calculated thermal diffusivity. The separation of the Parker and Degiovanni values for the high alumina volume fraction samples is thus explained. We note that the Parker and Degiovanni values coincide within calculation errors for 100% alumina (Fig. 5) and also for the lower alumina volume fraction samples. Fig. 6b shows that for a 10% volume fraction of alumina the temperature time curve exhibits almost no deviation from homogeneous response. We deduce that for the alumina/glass samples under study the homogeneous approximation via Eq. (5) gives access to a satisfying value for the overall thermal conductivity in the steady state for alumina volume fractions up to 0.5. This corresponds simply to two resistors in series. However, it is no longer the case for the higher alumina volume fraction samples where the laser-flash data should be interpreted with a more complex transient model of the two-layer system.

Finally, the difference between the experimental points and the simulated values can be explained by the additional thermal resistance of the contact which is certainly less than perfect.[13] Even a very good contact at an interface can be considered to have a thermal resistance. For example, the thermal resistance of a grain boundary in a dense sintered ceramic has been evaluated at $10^{-8}\,\mathrm{m^2\,K\,W^{-1}}$ in alumina[14] or $5 \times 10^{-9}\,\mathrm{m^2\,K\,W^{-1}}$ in zirconia.[15] The attribution of this value to the interface in the two-layer system yields no significant effect. At the other extreme the thermal resistance of a poor contact, such as two aluminium blocks simply held together by pressure, is of the order of $10^{-4}\,\mathrm{m^2\,K\,W^{-1}}$. It is reasonable to assume that alumina/glass interfaces of our samples lie between these two limits. In another piece of work concerning these samples, we have shown that the effect of a thermal contact resis-

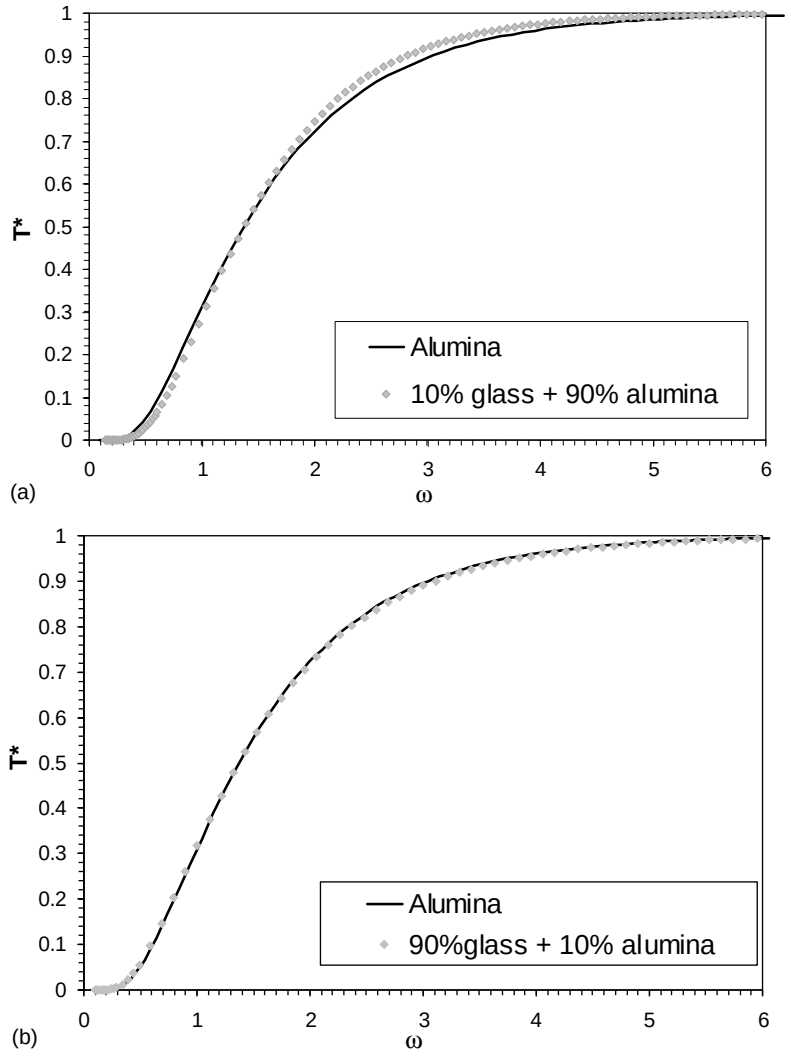

Fig. 6. (a) Dimensionless plot of back face temperature history for single-layer and two-layer (10% glass and 90% alumina) systems. T^* and ω are defined in Eqs. (9) and (10). (b) Dimensionless plot of back face temperature history for single-layer and two-layer (90% glass and 10% alumina) systems.

tance of 10^{-6} to $10^{-5}\,m^2\,K\,W^{-1}$ on the simulated values is sufficient to obtain agreement with experiment.[13] This value applies to all volume fractions in the two-layer system but its effect becomes less significant for samples exhibiting greater thermal resistance (low volume fraction of alumina).

5. Conclusion

The thermal response of a two-layer system has been studied for the case of a strong difference in thermal conductivity of the respective layers. In particular, the overall thermal conductivity value of the system, in the direction of heat flow perpendicular to the planes of the layers, given by a transient measurement of the thermal diffusivity is compared to that obtained in the steady-state situation. The assumption of the laser-flash method is that the sample behaves as a homogeneous medium and hence the thermal conductivity λ_a can be calculated with the expression:

$$\lambda_a = \alpha \rho_a C_a$$

where α is the measured thermal diffusivity, and ρ_a and C_a are overall density and specific heat values for the two-layer system. We have studied the validity of this approximation as a function of relative volume fraction between the two solid phases. Numerical simulation shows that both approaches yield very similar results up to an alumina volume fraction of 0.5. With alumina volume fractions greater than 0.5, the transient method overestimates the value of overall thermal conductivity compared to that obtained in a steady-state experiment. The difference from steady-state behaviour increases with the volume fraction of the more conducting phase and exhibits a maximum value of 15% at an alumina volume fraction equal to 0.9. This is explained by deformation of the temperature–time behaviour of the two-layer system compared to the response of a homogeneous medium. Experimental results for alumina/glass two-layer samples support

these observations, though it is also necessary to take into account the thermal resistance of the contact between the two layers.

Finally, we conclude that for the case of a thin thermally resistive layer on a conducting substrate, a model which correctly describes the transient response of the two-layer system, should be used for a precise analysis of the data obtained in a laser-flash experiment.

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