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Guiding heat in active thermal management: one-pot incorporation of interfacial nano-engineered aluminium/diamond composites into aluminium foams

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ABSTRACT

Traditional metal foams, which are incapable to meet the heat dissipation demands of today's electronics industry, were recently reformulated by the incorporation of thermal inclusions of graphite flakes that enhance their heat dissipation efficiency. In this work we manufacture, by means of a one-pot liquid metal infiltration synthesis, new layer-structured multicomponent materials that consist of aluminium foam components alternated with aluminium/diamond composite components. The thermal conductivity of aluminium/diamond components can substantially vary with processing conditions, which allow the nano-dimensional control of reaction products at the aluminium-diamond interface. The experimental and analytical results, plus finite element modelling, revealed that the low thermal resistance interface between components derived from the one-pot synthesis route, allowed multicomponent materials to significantly improve the thermal dissipation of traditional foams since aluminium/diamond components acted as effective thermal guides capable of transferring heat to the aluminium foam components where dissipation was achieved by forced convection.

KEYWORDS: A. Foams; A. Metal-matrix composites (MMCs); B. Interface; B. Thermal properties.

1. Introduction

The current increasing heat dissipation needs of the electronics industry oblige material science researchers to find solutions to this problem, which is actually responsible for more than 50% of catastrophic system

failures [1]. The exponential increase in the electrical powers used in electronic systems entails conventional heat-dissipating materials not constituting a serious proposal to this ongoing problem. In an attempt to develop new heat-dissipating materials, two main heat release mechanisms have been considered to date: i) passive dissipation and ii) active cooling. With time, the first has led to the development and processing refinement of metal/ceramic composite materials, among which aluminium/diamond and silver/diamond systems represent the current state-of-the-art technology. Although silver/diamond is a very promising system given its high thermal conductivity [2,3], aluminium/diamond is even more interesting thanks to its lower cost and density, and the possibility of adjusting its thermal conductivity by properly controlling the reaction at the aluminium-diamond interface during processing [4–9]. These materials transport heat by a conductive mechanism to other materials that can release it to the environment. The ultimate materials, which demonstrated being able to accomplish active cooling, essentially consist of open-pore foams through which a fluid is passed so that the heat conducted by the foam's solid structure is transferred by solid-fluid convection to the fluid to then be dissipated to the surrounding environment. Since Zaragoza et al. [10,11] announced that the future of these materials lies in foams with high thermal conductivity, i.e. foams with not a very high fraction of pores (roughly $< 70\%$), several attempts have been made to demonstrate the applicability of foams of various material natures, such as carbon, graphite and pure metals and alloys in thermal active cooling. In an attempt to increase the thermal conductivity of foams and, thus, their thermal performance, the most successful strategy has been the incorporation of highly conductive materials into foam structures. In a first approach, multicomponent materials have been developed by the stack packing of alternating layers of metal foams and sheets of that same metal (the so-called finned foams) [12–14]. These materials offer a limited increase in their thermal performance as the heat transmission between components is restricted by their high thermal resistance interfaces, generated by mere physical contact or by using adherent resins with low thermal conductivity [15]. Another more recent strategy has consisted in incorporating high thermal conductive inclusions, such as diamond or graphite flakes into the foam material. It is worth mentioning the development of magnesium/diamond composite foams [16], whose thermal conductivities go up to $82 \text{ Wm}^{-1}\text{K}^{-1}$, and the recent aluminium foams that incorporate graphite flakes oriented in random or alternating layer arrangements [17], whose recorded thermal conductivities go up to $290 \text{ Wm}^{-1}\text{K}^{-1}$ and with much higher power dissipation densities than conventional aluminium foams.

In this article, the authors present a new proposal to develop highly conductive thermal foams that consists in combining the passive dissipation properties of aluminium/diamond composites with the high active cooling capacity of aluminium foams. For this purpose, multicomponent layer-structured materials were manufactured in a one-pot liquid state processing by liquid metal infiltration as it allows a continuous metallic phase in the overall material to be developed. The thermal conductivity in aluminium/diamond components can be managed over a relatively wide range by nano-dimensionally controlling the reaction products at the interface. Unlike the aforementioned finned foams, the current materials exhibit a much superior thermal performance given the high thermal transmission between the foam and composite components caused by metal phase continuity. The recorded thermal conductivities reached outstanding values of up to $435 \text{ Wm}^{-1}\text{K}^{-1}$ and the materials showed power dissipation density values that were 130% higher than those shown by the conventional aluminium foams used for the aforementioned active thermal management purpose. All the experimental results on thermal characterisation are discussed within the framework of both available analytical models and computational fluid dynamics-CFD simulations.

2. Experimental procedures

2.1 Materials

High purity aluminium (99.999 wt.%) was purchased from Alfa Aesar (GmbH & Co KG-Karlsruhe, Germany). Two sets of particles were used: cubo-octahedral diamond particles of ISD1700 quality (60/80, 40-50 and 20/30 mesh with average Feret diameters of 215, 395 and $640 \mu\text{m}$, respectively), purchased from Iijin Diamond (S. Korea), and quasi-spherical NaCl particles of analytical grade (99.5% purity) acquired from Panreac Química S.L.U (Barcelona, Spain). NaCl particles, originally within the $300 \mu\text{m}$ -1 mm size range, were sieved to obtain fractions of narrower particle size distributions, from which those within the 500 - $700 \mu\text{m}$ range were selected. The morphologies of the diamond and NaCl particles are shown in the SEM micrographs in Figures 1a-b and 1c, respectively.

2.2 Fabrication of aluminium foams and aluminium/diamond composites

Aluminium foams and aluminium/diamond composites were manufactured by liquid metal gas pressure infiltration.

To manufacture the aluminium foams, NaCl particles were packed into graphite crucibles (38 mm high and 10 mm internal diameter) at volume fractions within the 0.58-0.63 range by following a compaction procedure of alternative strokes and vibrations (see [18] for more details). To manufacture the aluminium/diamond composites, porous preforms of packed diamond particles were achieved by the same compaction procedure. Diamond particles acquired a packing volume fraction within the 0.60-0.61 range. A solid aluminium piece was placed on top of the packed preforms and the whole configuration was placed inside an equipment designed for gas pressure infiltration [18–20]. Vacuum was applied to the infiltration chamber up to a value of 0.2 mbar and temperature was raised at a speed of 4 °Cmin⁻¹ up to 760 °C by an electrical furnace installed inside the chamber. After 5 minutes, time necessary to achieve temperature homogeneity, the infiltration chamber was pressurised with argon gas up to a pressure of 9.5 bar. This allowed liquid metal to be infiltrated into the preform. For the aluminium/diamond composites, it is very important to control the time from the point at which the maximum pressure in the chamber is reached. This time, during which the liquid metal remains in contact with the diamond particles before the solidification of the metal and the herein so-called contact time (t_c), determines the system's reactivity and, therefore, the quality of the generated interface [5]. Contact times (t_c) of 0, 15 and 45 minutes were selected. Immediately after the contact time chosen for each case was reached (t_c equalled 0 for the infiltrations of the NaCl particulate preforms), the infiltration chamber was rapidly cooled from its base at a cooling rate of 190 °Cmin⁻¹ by displacing it into a cool water bath to achieve fast directional solidification of the metal. Since aluminium was 100 °C above its melting point, this cooling rate means that the metal was solidified only 30 seconds after starting the cooling process. Such short cooling times do not significantly affect the nominal contact times explored in the manuscript ($t_c=0, 15$ or 45 minutes) during aluminium/diamond composites manufacture. Subsequently, the sample was extracted and any excess metal surrounding the samples was machined away. There was a final step in the fabrication of aluminium foams that consisted of eliminating the NaCl particles by dissolution with water at room temperature following the two-step process detailed in [17].

2.3 Fabrication of multicomponent materials in one-pot liquid-state processing

The multicomponent materials were manufactured by one-pot liquid metal pressure infiltration, following the replication technique [21–24], by a similar procedure to that explained in [17]. One of the most delicate

steps of the entire manufacturing process was preform preparation which, according to the diagram in Figure 1d, was achieved by the alternative compaction of suitable quantities of 40/50 mesh diamond particles and NaCl particles in a mould with the help of a manual piston. In any case, vibrations were avoided to prevent possible segregation effects due to the presence of particles of different density and size. Preforms were produced with two different layer orientations in relation to the sample's largest dimension: i) longitudinal, with layers in parallel to sample length; ii) transversal, in which layers were arranged perpendicularly to sample length. The total number of diamond compaction layers interspersed in the salt compacts was 5 in the longitudinal arrangement and 15 in the transverse arrangement. The thicknesses of each component layer varied in order to define distinct component composition percentages. Rectangular graphite moulds (10×10×50 mm) were used for the longitudinal arrangement while circular graphite crucibles (38 mm high and 10 mm internal diameter) were used for the transversal orientation. Prior to packing, both moulds and crucibles were coated with BN spray coating (ZYP Coatings Inc., Oak Ridge, USA) to facilitate demoulding. Alumina paper with several holes, each of about 0.5 mm, was added to top of the compacted preform to avoid the liquid metal and preform coming into contact before infiltration commenced.

The infiltration procedure and NaCl dissolution were followed as described in the previous section. The resulting multicomponent materials comprised alternating layers of both open pore aluminium foams and aluminium/diamond composite components.

2.4 Microstructural and interfacial characterisation

As the materials containing diamond particles cannot be polished by conventional metallographic techniques, two different strategies were followed to obtain information on structural featuring. The overall microstructure of the multicomponent materials (succession of component layers) was observed under an optical Olympus PME-3 microscope after samples were fractured by controlled impacts. For details of the interfaces in the aluminium/diamond composite components the method proposed in [4,25] was followed. It basically consists of etching the metal by an electro-chemical procedure, which has the advantage of preserving the interfacial products generated by chemical reactions during manufacture processing. The interfaces were characterised with the help of a SEM-Hitachi S3000N scanning electron microscope operating at 20.0 kV.

2.5 Thermal conductivity – measurement and modelling for aluminium foams, aluminium/diamond composites and multicomponent materials

The thermal conductivity of the materials was measured in an experimental device based on a relative steady-state (equal-flow) technique. The equipment, assembled at the University of Alicante laboratories, follows international standard ASTM E-1225-04 [26]. Samples were placed between two blocks, with the layers oriented in parallel (longitudinal measurements) or perpendicularly (transversal measurements) to the direction of the heat flux. The bottom of the sample remained in contact with a cylindrical cooled brass base (achieved by a room temperature water flux), and the top of it came into contact with a brass reference block at 70 °C. This temperature was stabilised with the help of a water bath connected to the brass block. Three thermocouples were connected to the reference material and two more to the sample. This configuration allowed the temperature gradients be measured to derive thermal conductivity with an uncertainty of $< \pm 5\%$.

An estimation of the thermal conductivities in the multicomponent materials was made by applying two theoretical models that took into account the specific microstructures of the materials and the orientation of the layers in relation to the heat flux. The next sections describe how the two modelling schemes were developed and considered for the longitudinal and transversal thermal conductivities. For the time being, comments are made only on the modelling framework for the composite materials consisting of randomly distributed monodispersed particles in a metal matrix, which is properly accounted for by the well-known expression of the Generalized Differential Effective Medium Scheme (GDEMS) [2,25,27,28]:

$$\int_{K_m}^{K_{com}} \frac{dK}{K \sum_i X_i \frac{-(K - K_r^{eff})}{(K - K_r^{eff})p - K}} = -\ln(1 - V) \quad (1)$$

where K is thermal conductivity and subscripts com and m refer to the composite and matrix, respectively. X_i is the fraction of the i -inclusion type in the total amount of inclusions of the composite (in the composites containing only one type of inclusions $i=1$, and hence $X_1=1$). V is the total volume fraction of the inclusions and p is the polarisation factor of an inclusion (equal to $1/3$ for spheres, as modelled in this study). K_r^{eff} is the effective thermal conductivity of an inclusion which, for spherical geometries, is related to its intrinsic

thermal conductivity, K_r^{in} , the matrix/inclusion interface thermal conductance h , and the radius of the inclusion r , by

$$K_r^{\text{eff}} = \frac{K_r^{\text{in}}}{1 + \frac{K_r^{\text{in}}}{hr}} \quad (2)$$

In general, the integral on the left-hand side of Equation (1) has no analytical solution and needs to be numerically solved with appropriate mathematical software.

Equation (1) is also valid to estimate thermal conductivities in foam materials (K_f) by considering that pores are inclusions of zero nominal thermal conductivity ($K_r^{\text{in}}=0 \text{ Wm}^{-1}\text{K}^{-1}$). Then Equation (1) becomes:

$$K_f = K_m(1 - V_p)^{\frac{1}{1-p}} \quad (3)$$

where K_m is taken as $237 \text{ Wm}^{-1}\text{K}^{-1}$ for pure 99.999 wt.% aluminium [29], V_p refers to the volume fraction of pores in the two-phase foam material and p is now the polarisation factor of the pores (equal to the polarisation factor of the NaCl particles from which the pore structure of the foam derived as the replication method maintains the morphological characteristics of the leachable particles in the pores of the final material): $p=1/3$ for spherical particles again [16].

2.6 Power dissipation density and pressure drop – measurement and modelling/simulation for aluminium foams and multicomponent materials

The testing of materials for the active thermal management applications should emulate the realistic working conditions used in electronic systems. As no standardised methodology exists, the experimental equipment presented in [16] and also used in [17] was herein applied. In this device, air flow was forced to pass through a porous sample, which remained in contact with a hot surface so that air could take part of the heat transferred to foam by conduction and remove it by forced convection. The base of the sample touched a heated brass block that led heat from a heating source. The contact between both surfaces was improved by using a small amount of silicon-free heat transfer compound (nominal thermal conductivity of $2.5 \text{ Wm}^{-1}\text{K}^{-1}$, Electrotube, UK). The brass block contained two thermocouples, t_1 and t_2 , distanced at 36 mm and with t_1 located 1 mm far from the surface of the brass block that came into contact with the sample.

The heating system operated to control the temperature of the t_1 thermocouple at 80 °C. At the opposite end of the sample, room temperature air was forced to flow inside the sample pore structure. Air was regulated by a ± 0.1 lmin⁻¹ flowmeter within the 1-15 lmin⁻¹ range. Having set a specific air flow, the system was allowed to reach the steady-state conditions for 15 min, after which the thermal gradient in the brass block was obtained by the readings of thermocouples t_1 and t_2 . This thermal gradient could be related to the sample's power dissipation density P (Wm⁻²) by the following equation (an equal heat flux through the reference and sample, and negligible heat losses were assumed, provided that the reference was thermally isolated, and that both the reference and sample had the same contact areas):

$$P = \frac{K_{\text{ref}}(T_2 - T_1)}{\Delta x} \quad (4)$$

where K_{ref} is the thermal conductivity of the reference material (in the present case K_{ref} for 99.99 wt.% brass equalled 109 Wm⁻¹K⁻¹), T_1 and T_2 are the temperatures measured in the brass block with thermocouples t_1 and t_2 , respectively, and Δx is the distance between both thermocouples.

The pressure drop was measured with the help of a homemade air permeameter, already presented in [17] and equipped with ± 0.001 bar precision manometers at both ends. The air flow was followed with an air flowmeter that operated within the 0-30 lmin⁻¹ flow range (Ki Key Instruments, Trevoze, UK).

Finite elements computational fluid dynamics (CFD) simulations were carried out with the ANSYS Fluent software package (free academic resource version). This software incorporates a porous media model that can effectively represent the open pore foam systems herein proposed [30,31]. The pressure drop for the fluid passing across a sample can be accounted for with the well-known Darcy-Forchheimer equation, which combines two terms associated with viscous and inertial losses (first and second terms, respectively, on the right-hand side of the following equation):

$$\frac{\Delta P}{\Delta L} = Av + Bv^2 \quad (5)$$

with

$$A = \frac{\mu}{K_1} \text{ and } B = \rho \cdot \frac{1}{2} C_2 \quad (6)$$

where $\Delta P/\Delta L$ is the pressure drop along the porous material, K_1 and C_2 are constants for a given pore microstructure, v expresses superficial velocity, μ is fluid dynamic viscosity and ρ is fluid density. $1/K_1$ and C_2 are the viscous and inertial resistance coefficients, respectively (K_1 is better known as material permeability). The results obtained for the aforementioned coefficients were then employed in ANSYS Fluent to model the power dissipation densities of both the aluminium foam and the multicomponent materials.

2.7 Flexural modulus measurement

The flexural strength of some of the resulting materials was measured using the Instron 4411 universal testing machine in three-point-bending test mode, configured with 30 mm span and 0.5 mm/min crosshead speed. Prior to the bending tests, pure aluminium (99.999 wt.%) flexural modulus was measured under the above testing conditions for the calibration of the equipment. The flexural modulus obtained for pure aluminium was 69 GPa, fully consistent with literature values.

3. Results and discussion

3.1 Preparation of preforms – packing restrictions

The volume fractions of NaCl, diamond and pores for each preform (the equivalent to the volume fractions of pores, diamond and metal, respectively, in the multicomponent materials) are shown in Table 1 and are plotted on a ternary composition diagram in Figure 2.

The ternary diagram shows that preforms can only be manufactured in a narrow band delimited by the upper and lower limits of the packing volume fractions for NaCl particles (within the 0.58-0.63 range) and for diamond particles (0.60-0.61). Within these intervals, the lower values correspond to the minimum compaction achieved by the natural arrangement of particles when they are subjected to vibrations. The upper value corresponds to the compaction value when particles are carefully packed with the help of a manual piston, whose striking action helps to better accommodate particles in a packed bed. The use of other particles with different characteristics (morphology, size and size distribution) can lead to changes in the allowed region as packing volume fractions can largely differ.

3.2 Microstructure of multicomponent materials and interface engineering of aluminium/diamond composite components

Figure 3 provides micrographs that illustrate the structure of the KK-4B^L sample in its longitudinal arrangement and the effect of the contact time between the liquid aluminium and diamond particles on the interface evolution in its aluminium/diamond components. The first two micrographs (a-b) reveal a laminated structure of alternating aluminium foams and aluminium/diamond composite layers that exhibits some irregularities as regards perfect stack-pack geometry. These irregular features, which came about because no perfect component layering existed, are probably caused because diamond particles are smaller (295-400 μm) than NaCl particles (500-700 μm). Under this scenario, at the limit of separation between components, the diamond particles were rearranged by partially filling the gaps left between the larger NaCl particles. This effect generated a slight decrease of about 30% in the thickness of each aluminium/diamond component compared to that expected for its normal compaction. Despite this, and given the relatively large thicknesses of the components, no significant variations in the volume fractions of the respective constituents were observed in each component.

Figures 3c-h illustrate the reactivity in the aluminium/diamond component system, which was evidenced by the growth of Al_4C_3 crystals at the aluminium-diamond interface as the contact time between the liquid aluminium and the diamond particles increased during the manufacturing process. It was proved that the nature of the crystals was Al_4C_3 by means of XRD analyses, not included in this work because it is a product of the aluminium-diamond interfaces of composite materials processed by infiltration of liquid aluminium already well characterized in bibliography. Similar results have been previously published in works co-authored by one of the authors of this paper where the relationship between the presence of Al_4C_3 crystals and the interfacial thermal conductivity in the aluminium/diamond composite materials has been demonstrated, which determines their final thermal conductivity [4–6,32]. The three contact times explored in this work were chosen based on the findings published in [5], which display that for the pure aluminium and diamond of ISD1700 quality, the optimum contact time that generated the highest interfacial conductance was around 15 minutes ($t_c=15$ min) at the infiltration temperature of 760 °C. At this contact time, both the main (100) and (111) diamond faces were partially covered by Al_4C_3 crystals.

At $t_c=0$, Al_4C_3 crystals appeared almost exclusively on (100) faces due to their greater reactivity because the terminal carbon atoms on these faces were only 3-fold bonded. For long contact times ($t_c=45$ min), both

the (100) and (111) diamond faces were fully covered with Al_4C_3 crystals of different growth orientations depending on each face (see [33] for details).

3.3 Thermal conductivity of aluminium foams, aluminium/diamond composites and multicomponent materials – experimental results

The experimental thermal conductivity values of aluminium foams, aluminium/diamond composites and multicomponent materials are found in Table 1. Some aspects are mentioned to evaluate the resulting data. Aluminium foams (samples A-1, A-2 and A-3) present thermal conductivities that can be interpreted with the help of Equation (3). A representation of $\log(K_f)$ vs. $\log(1-V_p)$ displays a linear relationship (Figure 4a) from the slope of which the polarisation factor p was derived. For these aluminium foams, parameter p equalled 0.33, which was consistent with the quasi-spherical geometry of their pores given the close-to-spherical morphological characteristics of the raw NaCl particles.

The aluminium/diamond composite materials herein manufactured displayed thermal conductivity values that were closely dependent on the interfacial products generated under the processing conditions. Figure 4b represents the experimental results of the 9 aluminium/diamond composites samples shown in Table 1, in which the size of the diamond particles and the processing contact time t_c varied. For comparison purposes, Figure 4b depicts a line that represents the results of similar experiments reported in [5]. The herein obtained results corroborated the thermal conductivity vs. contact time profile in [5]. The optimal contact time was around $t_c=15$ min with an infiltration temperature of 760 °C for which a thermal conductivity value of $604 \text{ Wm}^{-1}\text{K}^{-1}$ was recorded when 40/50 diamond particles were used. In [5], the thermal conductivities at 0 and 15 min were slightly higher than those presented herein, which was probably due to slight variations in the quality of the diamond particles as they were purchased from the same supplier, but belonged to different production batches. Another salient feature of Figure 4b is that the aluminium/diamond composites had greater thermal conductivity when they incorporated larger diamond particles, which falls in line with Equation (2) and the general findings of [6].

Regarding the thermal conductivities of the fabricated multicomponent materials, the experimental results for the longitudinal arrangements gave values up to $435 \text{ Wm}^{-1}\text{K}^{-1}$ when the proportion of aluminium/diamond component in relation to that of the aluminium foam was 70:30. As expected, the thermal conductivities of samples KK increased with the volume fraction of diamond ($V'D$) (see Table 1)

or, in other words, with the volume fraction of the composite component, and consequently lowered with the global porosity content. These effects are observed in Figures 4c and 4d, where the longitudinal thermal conductivities in Table 1 are plotted according to $V'D$ and $V'p$, respectively. Once again, the effect of the infiltration processing conditions on the final thermal conductivity had to be considered. The three samples fabricated with the same components composition (KK-1) at the 0, 15 and 45 min contact times (KK-1A^L, KK-1B^L and KK-1C^L, respectively) illustrate the same tendency as the aluminium/diamond composites did, with maximum thermal conductivity at $t_c=15$ min. Figure 4c also contains a theoretical curve calculated with Equation (3) for the aluminium foams. It can be easily understood that the newly developed porous materials fabricated in this work can provide much higher thermal conductivities than conventional aluminium foams for a given global pore volume fraction. The possibility of managing interfaces by nano-engineering has the benefit that the high thermal conductivity values reported can be achieved with low diamond particle content. The small diamond content explored, together with the relatively low cost of ISD1700 quality diamond particles [6], renders these materials a high performance/cost ratio.

3.4 Thermal conductivity – modelling longitudinal and transversal orientations

The modelling of thermal conductivity for anisotropic systems formed by the stack-packing of alternating semi-infinite component layers has already been covered in the bibliography [17,34]. Models are based on an electrical analogy of a combination of series and parallel resistances, as shown in Figure 5.

3.4.1 - Longitudinal arrangement

In a longitudinal arrangement (the equivalent to an electrical distribution of resistors in parallel), thermal conductivity can be calculated by bearing in mind the rule of mixtures approximation:

$$K_c^L = K_{com} \cdot V'_{com} + (1 - V'_{com}) \cdot K_f \quad (7)$$

where K_c^L is the longitudinal thermal conductivity of the multicomponent material and V'_{com} is the volume fraction of the aluminium/diamond component in the overall material.

Equation (7) is derived by considering that heat is transported in each component independently and with no heat transfer among them. This is roughly true when materials are subjected to important temperature gradients and also when components display dissimilar heat transport mechanisms.

K_{com} in Equation (7) can, in turn, be calculated with the GDEMS model (Equation 1) for which the appropriate values of the intrinsic conductivity of the diamond particles and interfacial conductance h must be known. Both values K and h are specific for each diamond particle type and each particular interface, respectively. Therefore, h is dependent on processing conditions while K_r^{in} is determined mainly by the quality of the diamond particles. By taking into account Equation (2), $1/K_r^{eff}$ vs. $1/r$ can be represented for samples D-i in Table 1 for each processing condition (contact times $t_c=0$ min, $t_c=15$ min and $t_c=45$ min). Figure 6a shows the fitting lines obtained for each processing condition while Table 2 shows the derived values of K_r^{in} and h .

With an average K_r^{in} value derived to equal $1536 \text{ Wm}^{-1}\text{K}^{-1}$, the longitudinal thermal conductivity for the multicomponent materials can be represented according to the percentage fraction of the aluminium/diamond component for the wide range of h values found in Table 2, characteristic of each processing condition (Figure 6b). It was verified that the experimental and calculated values closely agree.

3.4.2 - Transversal arrangement

A transverse component arrangement is the equivalent to an electrical distribution of series resistors. Heat is forced to cross the interfaces between components and the transversal thermal conductivity (K_c^T) can be estimated by [34]:

$$\frac{1}{K_c^T} = \frac{V'_{com}}{K_{com}} + \frac{(1 - V'_{com})}{K_f} + \frac{2}{dh_{com}} \quad (8)$$

where V'_{com} and K_{com} are defined identically as in Equation (7), d is the sum of the widths of each component layer and h_{com} is the interfacial thermal conductance of the interface between the aluminium foam and aluminium/diamond components. The samples for which transverse conductivity was measured are limited to those indicated in Table 1. These samples were manufactured at the optimum contact time ($t_c=15$ min).

The transverse thermal conductivity values, both those estimated with Equation (8) and the experimental ones, can be correlated by a graph in which both results are plotted according to the thermal conductance of the interface between the aluminium/diamond and aluminium foam components (Figure 7). As shown in Figure 7, thermal conductivity and the thermal conductance scale with the same trend and the highest

values were consequently found for the sample with the highest aluminium/diamond component content (KK-4B^T). The horizontal dotted lines allowed the value of thermal conductance that could be assigned to the interface between components to be known. To gain an idea of the magnitude of values found, two extra samples with the same components composition as sample KK-4B^T were prepared for this study. One of them was comprised of packaging of alternated layers of composite D-2B and aluminium foam A-1, joined only by physical contact. The other was packed by joining layers by means of small quantities of silicone-free heat transfer compound (with a nominal thermal conductivity of 2.5 Wm⁻¹K⁻¹, as specified by the supplier Electrotube, UK). Figure 7 reveals a significant difference between the thermal conductance values of sample KK-4B^T, manufactured by the one-pot liquid-state procedure, and the two new extra samples. The fact that the KK-4B^T sample had a continuous metallic matrix enhanced the heat transfer by conduction from component to component. On the contrary, the samples whose layers were packed by physical contact and/or by using thermal paste presented a worst thermal interface which, in practical terms, can be considered an effective resistance to heat transfer between components and, therefore, reduced the possibilities of a thermal synergy between the components in the heat dissipation mechanism. Conversely, in the samples manufactured by the one-pot liquid-state procedure, the thermal interface between components was favourable for heat transfer to not only occur longitudinally along each component, but an important contribution was made by the transverse heat transfer from the aluminium/diamond component of high thermal conductivity to the aluminium foam component where active dissipation occurred.

3.5 Power dissipation density and pressure drop

The characteristic viscous (1/K₁) and inertial (C₂) resistance coefficients of the foam and multicomponent materials were obtained by the quadratic regression of the experimental data on pressure drop across samples versus air flow according to Equations (5) and (6), which can be expressed in fluid flow terms as:

$$\frac{\Delta P}{\Delta L} = \frac{1}{60 \times 10^3} \frac{\mu}{Z K_1} Q + \left(\frac{1}{60 \times 10^3} \frac{1}{Z} \right)^2 \frac{C_2}{\rho} Q^2 \quad (9)$$

where Z and Q are the cross-sectional area (in m²) and fluid flow (in lmin⁻¹) in the foam component. Air viscosity and density were taken at 20 °C to be equal to 1.82×10⁻⁵ kgm⁻¹s⁻¹ and 1.20 kgm⁻³, respectively. Figure 8 depicts the experimental results and their corresponding second-degree polynomial fittings

obtained for samples A-1, KK-1B^L and KK-4B^L (samples KK-1B^L and KK-4B^L were processed at $t_c=15$ min and contained the highest and lowest foam component percentage, respectively). The results of $1/K_1$ and C_2 for the foam components (the only air-permeable component) are shown in Table 3.

Overall, we noted that the results for the different samples showed a relatively good agreement for the $1/K_1$ and C_2 values for the foam components, which is a uniformity test of the structures of the foams in the A-1 aluminium foam sample and those present in the multicomponent KK-1B^L and KK-4B^L materials. The slight differences between the values of samples, which were especially noticeable in the permeability data (K_1), could be a phenomenological manifestation of the lower pore connectivity near the ends of each foam component layer, which translates into a gradual loss of permeability as the thickness of the foam layers are reduced.

These values of the viscous and inertial resistance coefficients were implemented into the CFD modelling tool to estimate the power dissipation density under conditions that emulated those described in the setup explained in Section 2.6, used to take measurements. The ANSYS Fluent platform allowed two thermal working conditions in porous media to be considered, namely thermal equilibrium (LTE) and non-thermal equilibrium (LTNE), which represent that the fluid and solid in the porous media were locally at the same or at a different temperature, respectively. For low permeability porous media (low pore volume fraction foams), with relatively large dimensions and considered for relatively high air flows, which was our case, both modelling conditions LTE and LTNE offered very similar results (see Figure 9a), where the calculated and experimental power dissipation densities are represented in relation to the air flow for the aluminium foam (A-1 sample). For the sake of simplicity, thermal equilibrium conditions (LTE model) are used from now on.

The CFD calculations predicted much higher air flow dissipated powers in the multicomponent materials than in the aluminium foam (Figure 9b). In addition, power dissipation density increased with the multicomponent materials manufactured at the optimal contact time of 15 min. Figure 9c represents the power dissipation densities of the materials studied in relation to pressure drop. The heat dissipation behaviour of the multicomponent materials was superior to that of the aluminium foam in all cases as they were capable of dissipating higher powers for the given pressure drop values. The validity of these conclusions is endorsed by Figure 9d, which shows the values calculated using CFD vs. those measured

according to the experimental setup described in Section 2.6 for samples that were 30 mm long. The results showed a very good correlation, with 6% errors at the most.

The temperature contours obtained with the LTE model and simulated with ANSYS Fluent are shown in Figure 10, where cross-sections at $z=5$ mm are drawn. The multicomponent materials diagrams indicate significant differences in temperature profiles compared to the conventional aluminium foam. As previously mentioned, the high thermal conductivity of the aluminium/diamond component layers allowed the heat to be transported over large distances, which improved the transversal heat transfer to the foam components and, hence, the air-metal convection. Negligible temperature gradients were observed for these components as a result of the high conduction that the material promoted. Velocity streamlines are also represented in Figure 10 as grey lines. It was verified that the mean air velocity came close to zero when it had crossed approximately three quarters of the sample length. This fact supports the notion that transport by conduction plays an important role in heat dissipation, enhanced by the presence of highly conductive phases like the aluminium/diamond components under study, which acted as effective heat guides in the herein developed multicomponent materials.

3.6 Flexural modulus

The flexural moduli of the components (aluminium foam and aluminium/diamond composites) and the multicomponent material with the highest thermal conductivity (sample KK-4B^L) were measured in order to evaluate their mechanical properties for the intended applications. The measured flexural modulus for aluminium foam (sample A-1) was 8 GPa, consistent with the Young's modulus obtained by compression or tensile tests for aluminium foams produced by the replication method [35–37]. In addition, the flexural moduli of aluminium/diamond composites manufactured at contact times $t_c=0$, 15 and 45 minutes were measured to be 173, 228 and 210 GPa, respectively. Interestingly enough, the flexural moduli scale in the same order as the thermal conductivity in these materials. Therefore, producing an adequate thermal interface suggests greater mechanical bonding strength between metal and diamond particles. This was previously reported in the literature, specifically in aluminium/diamond composites manufactured by squeeze casting [38] in which the contact time between liquid metal and diamond varied. Adequate extended contact times (optimized squeeze casting) with regard to conventional squeeze casting conditions (very short contact times) resulted in significant improvements in both thermal conductivity and flexural properties. Thus, samples produced by squeeze casting offered values of $321 \text{ W m}^{-1}\text{K}^{-1}$ and 184 GPa for

thermal conductivity and flexural modulus while samples derived from optimized squeeze casting conditions offered higher values for both properties: $606 \text{ Wm}^{-1}\text{K}^{-1}$ and 238 GPa , respectively. Considering the multicomponent materials anisotropy, flexural moduli corresponding to two different orientations were measured. For the KK-4B¹ sample, 154 GPa and 31 GPa were obtained for configurations where the components remain parallel and perpendicular to the direction of applied force, respectively. These values were found to be halfway those of their components, suggesting a component-to-component interface that can efficiently transfer stress, probably due to its continuous nature. Since low-pore density aluminium foams (like those obtained by replication) do not pose flexural failure problems when integrated in heat dissipation architectures, it can therefore be concluded that the multicomponent materials developed in this study far exceed the minimum mechanical requirements of these applications.

4. Conclusions

In this work, new multicomponent materials that combine aluminium/diamond and aluminium foam components structured in layers were manufactured by the replication method in a one-pot liquid-state procedure. The processing conditions were carefully controlled to nano-engineer the aluminium-diamond interface to obtain optimal thermal conductivity values ($604 \text{ Wm}^{-1}\text{K}^{-1}$). This anisotropic system allowed the study of the heat transport through and between components with two different layer orientations: longitudinal and transversal arrangements. The high longitudinal thermal conductivity of the multicomponent materials fell within the $137\text{-}435 \text{ Wm}^{-1}\text{K}^{-1}$ range while the transversal thermal conductivities reached $156 \text{ Wm}^{-1}\text{K}^{-1}$. The results suggest that the presence of high thermal conductive aluminium/diamond components can significantly improve the heat dissipation performance of conventional aluminium foams because aluminium/diamond components act mainly as effective thermal guides that conduct heat and transfer it to the aluminium foam components where dissipation is achieved by forced convection. The comparison of the experimental power dissipation density results with those obtained by CFD simulations shows a good correlation and values up to 130% higher in relation to conventional aluminium foams. In view of the aforementioned results, the new aluminium foam-aluminium/diamond multicomponent materials proposed herein can be considered serious candidates for effective active thermal management of current and upcoming electronic systems.

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Credit author statement

Both authors (L.P. Maiorano and J.M. Molina) contributed equally to the published work in the following categories: conceptualization, methodology, software, validation, formal analysis, investigation, writing-original draft, writing-review&editing and visualization. J.M. Molina also contributed to: resources, supervision, project administration and funding acquisition.

Figure 1. Scanning electron microscopy (SEM) micrographs of the ISD1700 diamond particles (40-50 mesh) (a-b) and the NaCl particles (500-700 μm) (c). (d) is a schematic drawing of the layered preform microstructure alternating packed beds of both the NaCl and diamond particles.

Figure 2. Composition ternary phase diagram for the preforms containing NaCl particles and/or diamond particles from which the aluminium foams, the aluminium/diamond composites and the multicomponent materials herein fabricated were derived. The shaded area represents the region allowed for the preforms fabricated by the presented manufacturing method.

Figure 3. Optical (a-b) and SEM (c-h) micrographs showing the microstructure of the KK-4B^L sample ($t_c=15$ min) (a-b) and the interfacial products in the aluminium/diamond component in the KK-1 samples

corresponding to: (c-d) the KK-1A^L samples fabricated at $t_c=0$ min; (e-f) the KK-1B^L samples fabricated at $t_c=15$ min; (g-h) the KK-1C^L samples fabricated at $t_c=45$ min.

Figure 4. (a) A graph of $\log(K_f)$ vs. $\log(1-V_p)$ for samples A. The straight line fitting the data is: $\log(K_f) = 1.478 \times \log(1-V_p) + 2.354$ and the regression coefficient is $R^2=0.996$; (b) bar diagram representing the experimental thermal conductivities of the aluminium/diamond composites according to the contact time and its correlation with the bibliographic data reported in [5]; (c-d) the experimental longitudinal thermal conductivities of samples in Table 1 versus the volume fraction of the diamond (c) and volume fraction of pores (d).

Figure 5. Schematic view of the layers' arrangements of the multicomponent materials, where R1 and R2 represent the two different resistances as the two components herein proposed.

Figure 6. (a) Linear correlation of $1/K_r^{\text{eff}}$ vs. $1/r$ for the aluminium/diamond composite materials fabricated under different processing conditions ($t_c=0, 15$ and 45 min), from which K_r^{in} and h were derived following Equation (2). For $t_c=0$, the fitting linear equation is $y=2.52 \times 10^{-7}x+6.44 \times 10^{-4}$ with $R^2=9.95 \times 10^{-1}$, $y=5.68 \times 10^{-8}x+6.48 \times 10^{-4}$ with $R^2=9.57 \times 10^{-1}$ when $t_c=15$ and $y=9.11 \times 10^{-8}x+6.61 \times 10^{-4}$ with $R^2=9.54 \times 10^{-1}$ when $t_c=45$ min.; (b) is a contour diagram of the experimental and theoretical longitudinal thermal conductivities of the multicomponent materials plotted according to h and the percentage of the aluminium/diamond composite

component in the multicomponent material. The volume fractions of the pores in the aluminium foam component and of diamond in the aluminium/diamond composite components were set at 0.61 and 0.60, respectively.

Figure 7. Thermal conductivity calculated for the multicomponent materials in the transversal arrangement found in Table 1 according to interfacial thermal conductance (h) for the interface between the aluminium/diamond and aluminium foam components. Intersections between the calculated curves and horizontal lines (experimental values) allowed h to be estimated.

Figure 8. Experimental results and second-degree polynomial fittings of the pressure drop versus fluid flow for samples A-1 ($y=10090x^2-4544x$; $R^2=0.9999$), KK-1B^L ($y=14711x^2+11072x$; $R^2=0.9975$) and KK-4B^L ($y=35686x^2+70195x$; $R^2=0.9990$).

Figure 9. Comparison of the experimental and calculated values of the power dissipation densities (P) performed with both the LTE and LTNE models plotted according to air flow for sample A-1 (a); the power dissipation densities calculated (P_c) with the LTE model vs. air flow (b) and pressure drop (c) for samples A-1, KK-1A^L, KK-1B^L, KK-4A^L and KK-4B^L; (d) is a graph of the power dissipation densities calculated (P_c) with the LTE model vs. the experimental results (P_{exp}) for multicomponent samples KK-1A^L, KK-1B^L, KK-4A^L and KK-4B^L.

Figure 10. Fluid and solid temperature distributions and velocity streamlines (grey lines) at 15 lmin^{-1} for sample A-1(a), KK-1B^L (b) and KK-4B^L (c) obtained with the LTE model.

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Table 1. Thermal conductivity K ($\text{Wm}^{-1}\text{K}^{-1}$) of the aluminium/diamond composites, aluminium foams and multicomponent materials herein fabricated. $V'D$, $V'p$ and $V'm$ denote diamond particles, pores and metal volume fractions, respectively. $V_{\text{com}}:V_{\text{foam}}$ is the compositional percentage ratio of the aluminium/diamond composite and aluminium foam components. Subscripts L and T indicate the longitudinal and the transversal configuration, respectively. Samples D-1 correspond to those fabricated with a diamond particle size of 60-80 mesh, D-2 with 40-50 mesh and D-3 with a 20-30 mesh particle size.

Material type	Sample code	t_c	$V'D$	$V'p$	$V'm$	$V_{\text{com}}:V_{\text{foam}}$	K
Aluminium/ diamond composites	D-1A	0					280
	D-1B	15	0.61	-	0.39	100:0	534
	D-1C	45					451
	D-2A	0					383
	D-2B	15	0.61	-	0.39	100:0	604
	D-2C	45					569
	D-3A	0					440
	D-3B	15	0.60	-	0.40	100:0	668
	D-3C	45					589
Aluminium foams	A-1	-	0	0.63	0.37	0:100	52
	A-2	-	0	0.59	0.41	0:100	60
	A-3	-	0	0.58	0.42	0:100	63
Multicomponent materials	KK-1A ^L	0	0.16	0.45	0.39	27:73	137
	KK-1B ^L	15	0.16	0.45	0.39	27:73	196
	KK-1C ^L	45	0.16	0.45	0.39	27:73	183
	KK-1B ^T	15	0.16	0.45	0.39	27:73	68
	KK-2B ^L	15	0.21	0.40	0.39	35:65	244
	KK-2B ^T	15	0.21	0.40	0.39	35:65	79
	KK-3B ^L	15	0.33	0.27	0.40	55:45	351
	KK-3B ^T	15	0.33	0.27	0.40	55:45	111
	KK-4A ^L	0	0.42	0.18	0.40	70:30	279
	KK-4B ^L	15	0.42	0.18	0.40	70:30	435
	KK-4B ^T	15	0.42	0.18	0.40	70:30	156

Table 2. Material parameters used in the interface thermal conductance calculations by Equations (1) and (2) and a comparison with the data available in the literature.

Material	K_r^{in} ($\text{Wm}^{-1}\text{K}^{-1}$)	h ($\text{Wm}^{-2}\text{K}^{-1}$)
Aluminium/diamond composites ($t_c=0$)	1553	4.0×10^6 [5] 4.0×10^6 (this work)
Aluminium/diamond composites ($t_c=15$)	1543	1.9×10^7 [5] 1.8×10^7 (this work)
Aluminium/diamond composites ($t_c=45$)	1513	1.2×10^7 [5] 1.1×10^7 (this work)

Table 3. Parameters $1/K_1$ and C_2 obtained by analogy between Equation (9) and those obtained from the second-degree polynomial fittings of the pressure drop vs. fluid flow experimental results.

Sample code	$1/K_1$ (m ⁻²)	C_2 (m ⁻¹)
A-1	1.91×10^9	633648
KK-1B ^L	1.94×10^9	642201
KK-4B ^L	2.08×10^9	640214