Journal of Alloys and Compounds 736 (2018) 246–254

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Interface engineering in ferromagnetic high-thermal conductivity iron-diamond/metal composites for electric conversion applications

J.M. Molina [a], [b], [c], *, E. Louis [a], [c], [d]

[a] Instituto Universitario de Materiales, Universidad de Alicante, Apdo 99, E-03080, Alicante, Spain
[b] Departamento de Química Inorgánica, Universidad de Alicante, Apdo 99, E-03080, Alicante, Spain
[c] Departamento de Física Aplicada, Universidad de Alicante, Apdo 99, E-03080, Alicante, Spain
[d] Unidad Asociada del Consejo Superior de Investigaciones Científicas, Universidad de Alicante, Apdo 99, E-03080, Alicante, Spain

ARTICLE INFO

Article history:
Received 24 July 2017
Received in revised form
31 October 2017
Accepted 2 November 2017
Available online 8 November 2017

Keywords:
Metal matrix composites
Diamond
Iron
Ferromagnetic
Electric conversion

ABSTRACT

The objective of this work is to investigate whether any combination of metal and magnetic particles may fit the specifications of electric conversion applications, which require, among other properties, sufficiently high magnetic permeability and thermal conductivity and a low (adjustable) thermal expansion coefficient. After having explored a wide variety of combinations, guided by both chemical and physical considerations, it was decided to investigate composites fabricated by gas pressure infiltration of Ag or Ag3wt%Si alloys into compacts of bimodal mixtures of diamond (high thermal conductivity) and iron particles (high magnetic permeability). Three average particle sizes of each component were used to fabricate the composites, namely, diamond particles of 230, 285 and 295 μm and iron particles of 30, 42 and 398 μm. In addition the volume fraction varied in the ranges 0.1–0.59 (diamond) and 0.12–0.43 (iron). In order to avoid alloying with the infiltrating metal and iron-diamond reaction, iron particles were coated with amorphous carbon. The results indicate that only composites containing a volume fraction of carbon-coated iron particles higher than 0.4 showed properties (a thermal conductivity higher than 200 W/mK and a relative magnetic permeability above 0.3) within the range valid for electric conversion applications. Composites containing non-coated iron particles reached in almost all cases very low values of both properties.

1. Introduction

Some of the high-end applications in the field of electric conversion have reached their technological limits because of the impossibility of finding materials with acceptable ferromagnetic properties and, at the same time, capable to dissipate the excessive heat generated in their normal running [1–5]. Monolithic materials like carbon or steel have been traditionally used in the cores of the converter units. However, their thermal conductivity is extremely low and cannot accomplish the future demands of power control, since overheating of the systems would pose problems for keeping the structural integrity of the whole equipment. There is, in consequence, a need to identify new developments of ferromagnetic materials with improved thermal conductivity.

Composite materials with ferromagnetic properties have been since long fabricated by incorporating ferromagnetic materials as finely divided inclusions (like iron powders or fibers [2,4], barium ferrites [4], strontium particles [6], flakes of Fe-Cr [7], SmCo5 [8], iron nitride [9], iron oxide [10], CoFe [11] and ferrites [12,13]) into different non-magnetic metal matrices (Zn-22Al, Cu-ZnAl, $Nb_{0.33}Cr_{0.67}$, silver and copper). These soft magnetic powder-based composites (FPC), often fabricated by the established procedures of powder packing and ulterior densification by liquid infiltration with an appropriate matrix, were firstly developed aiming to replace some steel parts, subjected to time-varying magnetic fields, in electromagnetic devices of high power converter units and electric motors [1–5]. However, this class of FPC composites likely have excessively low thermal conductivity, still far from being auspicious for proper heat dissipation.

Other family of FPC materials fabricated with iron or ferrite powders and polymeric matrices, do not accomplish the current needs of thermal management for the mentioned applications. Aiming to increase the thermal response of these composites, it was

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* Corresponding author. Instituto Universitario de Materiales, Universidad de Alicante, Apdo 99, E-03080, Alicante, Spain.
E-mail address: jmmj@ua.es (J.M. Molina).

https://doi.org/10.1016/j.jallcom.2017.11.010
0925-8388/

explored the route of embedding a mixture of a ferromagnetic powder and dispersed heat conductive particles (SiC, AlN, BN, BeO, etc.) in a polymeric matrix; these composites, however, did not either show promising properties [14,15].

The present work is addressed to develop new ferromagnetic-high thermal performance three-phase composite materials through another approach that consists of combining high thermally conductive diamond particles with iron powder in a bimodal filler architecture, subsequently consolidated with silver matrix by gas pressure infiltration. In order to make effective the high thermal conductivity of metal and diamond particles, interfacial engineering must be applied, with the aim of achieving the proper heat transfer between the different phases.

Silver has been selected as metal matrix for their high thermal conductivity. In contrast to aluminium, another non-ferromagnetic metal often selected as matrix for composite materials, pure silver does not react with diamond and, in consequence, the interfaces are weakly conductive. Additions of silicon into silver have proven to increase the thermal conductivity of silver/diamond composites [16]. Regarding the ferromagnetic filler, iron particles reacts with silver and silver-silicon alloys, forming inter-metallics with poor thermal conductivity and low magnetic permeability. For this reason, along with the fact that iron, at temperatures of composites processing, may catalyse the transformation of diamond surface into graphite at the points where these two phases enter into contact, a protective carbon coating was applied to iron particles.

In essence, new FPC materials have been herein fabricated by gas pressure assisted liquid metal infiltration of silver and silver-3% silicon alloy into densely packed diamond-iron powder preforms. The thermal conductivity and magnetic permeability are discussed with the help of modelling schemes and scanning electron microscopy that allows a direct access to a close view of the different interfaces. Unfortunately, most of the available literature does not provide enough data and/or information to allow a detailed comparison with the results discussed in this work. In particular, we have not found any reliable work presenting both thermal and magnetic properties of a given material.

2. Materials and procedures

2.1. Materials

Infiltrations were done with pure Ag 99.9% and Ag-3wt%Si alloy. Pure silver was supplied by Heraeus S.A. (Madrid, Spain). The silver-silicon alloy, in its eutectic composition (Ag-3wt%Si), was prepared in the laboratories of the University of Alicante, in an induction furnace designed for this purpose that works under inert argon atmosphere. The silicon used for the preparation of this alloy was purchased in lumps shape from Goodfellow (Cambridge Limited, England). According to specifications purity is higher than 99,9%.

Diamond particles of different average diameters (225–540 μm) and ISD 1700 quality were purchased from Iljin Diamond Co., Ltd. (Korea). Iron powder (purity >99%) with several average particle diameters (15–400 μm), purchased from Goodfellow (Cambridge Limited, England), was used throughout the experiments. Diamond particles were highly regular both in size and shape while Fe particles had a very irregular shape and a variable size (see Fig. 1). Table 1 shows the thermal conductivity of the four materials used in this work, the average diameters of the particulates and the codes used to denominate all of them.

2.2. Coating of iron particles

In order to avoid alloying with the infiltrating metal, iron particles were coated with amorphous carbon by pyrolysis of polyvinylpyrrolidone (PVP) polymer. For that sake, 2 g of PVP were dissolved in 60 ml of ethanol under vigorous stirring conditions. Afterwards, iron particles were added to the dissolution and the stirring was kept for hours. Powder was filtered and dried in a heating muffle at 60 °C. Thereinafter iron particles were subjected to heat treatment in order to achieve pyrolysis of the polymer. The pyrolysis treatment was done in a closed chamber in which two subsequent vacuum-argon purging steps were done. In each step vacuum reached 0.01 mbar and argon was inserted in the chamber up to a pressure of 1 bar. The final vacuum, prior to the heat treatment, was 0.001 mbar. The remaining oxygen in the chamber was not measured as carbon-based samples at high temperatures (at 1000 °C as in the present case) can act as an oxygen getter. So, the trace amounts of oxygen would most probably be consumed in sample combustion. Anyhow, the absence of grooves or rough surfaces in samples containing coated iron particles is an indication of low oxygen contents. Fig. 1 shows an iron particle already coated plus an amplification of its surface. Coating significantly decreases the surface roughness (see Fig. 1c) of the particles practically eliminating the sharp corners that can be clearly noted in Fig. 1b. A detail of the surface shown in Fig. 1d reveals the smoothing produced by carbon coatings.

2.3. Liquid metal infiltration

The composites were prepared by gas pressure assisted liquid metal infiltration into densely packed preforms. Mixing of large (small) diamond particles with small (large) iron particles and packing of both mono- and bimodal powders were done mechanically. Graphite crucibles were used as infiltration moulds. An ingot of solid metal was placed on top of the packed preform. Prior to melting, vacuum was applied until a pressure of 0.1 mbar was reached.

Heating was done in an electrical furnace that was set at a very low rate (2 °C/min) until 250 °C to allow for slow desorption of humidity and gas adsorbed on particles surface. Subsequently, heating was continued up to 1075 °C at a rate of approximately 5 °C/min. The maximum temperature was maintained for 20 min. The liquid metal was forced into the preform using inert argon gas at a pressure of approximately 4 MPa. The chamber was kept under pressure until metal was directionally solidified. Finally, the pressure was released and the sample demoulded.

2.4. Characterization of composite materials

Thermal conductivity of composites was measured by means of a relative steady-state (equal-flow) technique, in an experimental set up assembled in our laboratories following the ASTM E1225-04 International Standard (see Ref. [14] for a detailed explanation). Sample and reference, in contact across their cross sections, were clamped between a room temperature water-cooled block (reference end) and a block connected to a thermally stabilized hot water bath (sample end). The temperature gradient in the sample was measured, and compared to that in the reference, by means of two thermocouples in each of them. The linearity of the temperature gradient in the reference was measured, by means of a third thermocouple, and was typically within ±1%. Accounting also for uncertainties related to a variety of factors (geometry among others) in the sample and in the reference (diameter after grinding, precise position of the thermocouples, thermal conductivity of the reference, etc.) we estimated the overall uncertainty of the measured thermal conductivities to be within ±5%.

The metal-diamond interfaces were characterized by following a coupled procedure that implies: i) direct access to the interface by a preparative method of electro-etching that preserves any