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A new solder matrix nano polymer composite for thermal management applications

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ABSTRACT

The increasing integration of microelectronics, raising the need for effective heat dissipation, requires new and improved composite materials technologies. For both thermal interface and die attach materials, a major challenge is to combine low thermal resistance joints with sufficient thermomechanical decoupling and reliability. In this paper, we present the fabrication and characterisation of a new type of solder matrix nano polymer composite (SMNPC) aiming to address these challenges. The SMNPC is fabricated into preforms by liquid-phase infiltration of a Sn–Ag–Cu matrix into a silver nanoparticle coated electrospun polyimide fibre mesh. The composite is demonstrated to possess high heat transfer capability, close to that of a direct soldered interface, lower elastic modulus compared to pure Sn–Ag–Cu alloy, and reliable thermomechanical performance during thermal cycling. Taken together, the results indicate that the developed SMNPC can be a useful composite alternative compared to conventional solders and polymer matrix materials for thermal management applications.

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1. Introduction

With the increasing integration density in the electronics industry, the development of improved materials for high density electronic packaging are required [1–3]. In particular, the thermal management of power dense microsystems, such as microprocessors, power electronics, LEDs, MEMs and thermoelectrics, is challenging and demands new die attach and thermal interface material (TIM) technologies that can combine effective heat transfer with high thermomechanical reliability [4–7].

Die attach materials have the function of mechanically and thermally connecting the IC to the substrate. Conventional die attach materials include solders and curable polymer matrix composites loaded with filler particles [8]. In assembly, some of the most important mechanical attributes for die attach materials are: strong adhesion towards joining surfaces; sufficient mechanical strength to keep the die in place; high impact resistance; and to impart minimum stress onto the joined surfaces during thermal expansion [8,9]. For power dense applications, die attach materials are also considered to play an important role in the heat dissipation [10]. The thermal conductivity of composite die attach materials based on polymer matrices with conductive fillers are typically limited to <5 W/mK, due to low effective heat transfer through multiple particle–particle contacts. By engineering filler type and size, many studies have attempted to improve the thermal properties, aiming for higher thermal conductivity [11–13]. However, with increasing filler content the die attach materials modulus increase, which can have negative effects for a die attach material. Firstly, it has been shown that higher modulus can drastically increase die/substrate warpage as well as stresses in both the die attach material and die [14]. Secondly, as reduced modulus and increased loss factor leads to increased energy dissipation, low modulus has been identified as one of the key material properties governing high impact resistance [15].

Solder based Thermal Interface Materials (STIMs) are attractive as they offer thermal conductivities which are approximately one order of magnitude higher than conventional particle laden polymer matrix materials [5]. Even though there is a wide range of solders used in conventional applications, such as interconnect and die attach applications, the solder alternatives viable for thermal interface applications are limited. The limitations are related to the assembly process temperature, environmental and health restraints and the mechanical compliance requirements causing

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reliability issues [16]. More specifically, STIMs are restricted to use alloys with high resistance to crack formation and propagation, which otherwise quickly lead to severe degradation of the thermal performance. Since resistance to cracks, under dynamic and thermally induced loads, is associated with high ductility, alloys with high modulus, such as tin based alloys, can be problematic to use as interface materials [17].

Furthermore, insufficient mechanical decoupling, through high modulus, puts geometrical constraints on the bond line thickness (BLT) of STIMs. In TIM1 application, i.e. between the die and the integrated heat spreader, solders require sufficiently high BLT in order to keep acceptable levels of stress transfer onto the silicon die during warpage [16]. It has for instance been found [17], that out of several viable solder candidates for TIM1 in microprocessor package, indium solder, with a BLT of 200–300 μm, is the only configuration that allows reliable assembly process and operation without crack formation. The finding was attributed to the softness of indium and its ability to deform under the stress caused by CTE mismatch between a silicon die and copper heat spreader.

In addition, high joint stiffness also limits the area that can be soldered. This is due to the combination of thermal expansion mismatch and solidification shrinkage, which cause increasing strain and inelastic dissipated energy at larger distance to the neutral point [18]. With the trend towards larger die sizes, thinner packages [8], and higher power dissipation, there is a need for thermal interface and die attach materials that can combine low elastic modulus with high thermal conductivity, to minimize thermal and bending stresses while offering reliable high heat dissipation capability.

Metal matrix composites (MMCs) have an important role in thermal management of microelectronics, such as processors, power semiconductors and packaging for microwave devices [19]. In this area, the MMCs developed are typically Al or Cu based with additions of SiC, W, Be, Mo, Al_2O_3 carbon or diamond [20–25]. The general aim with these composites is to achieve high thermal conductivity at low weight and tailor CTE to minimize the thermal mismatch [26]. Aiming for solder applications, there are a few MMCs, on the market, with indium matrices reinforced with copper, stainless steel meshes or random-fibre glass cloth [27]. For these composites the reinforcement meshes have higher modulus than the indium matrix, with the purpose to increase strength.

Despite the promising features of the electrospinning process for composite applications, very limited research on electrospun fibres as matrix reinforcement have been reported in the literature [28]. A few studies have investigated the incorporation of electrospun fibres as reinforcement in epoxy [29,30] and aluminum matrices [31]. However, recent work in our group has investigated and shown promising thermal characteristics for indium alloy based matrices in electrospun polymer fibre carriers, aimed for thermal interface applications [32–37].

In this paper, we present the concept, fabrication and thermomechanical characterisation of a new type of solder matrix nano polymer composite (SMNPC) preform, aimed for die attach and TIM applications. The introduced SMNPC is composed of a nano silver (Ag) surface functionalized highly porous electrospun non-woven polyimide fibre network, embedded in a Sn95.5–Ag3.8–Cu0.7 alloy matrix. When assembled as an interface or die attach material and reflowed, the SMNPC forms a structure that has continuous heat transfer paths through the Sn–Ag–Cu matrix and a fibre phase that can act to lower the stiffness of the joint.

2. Materials and methods

2.1. Fabrication

The fabrication of the SMNPC starts with electrospinning of a nanofibre polyimide network, followed by coating with Ag nanoparticles through a process based on polyimide cleavage in combination with an ion exchange. The Ag coated polymer network is then infused with a Sn95.5–Ag3.8–Cu0.7 alloy matrix in molten stage using a pressure assisted infiltration process to fabricate the composite preforms.

2.1.1. Electrospinning

The PI fibrous non-woven mesh was formed by electrospinning a solution of 16 wt% benzophenone-3,3′,4,4′-tetracarboxylic dianhydride 5(6)-amino-1- (4′-aminophenyl)-1,3- rimethylindane (BTDA–AAPTMI) polyimide (Matrimid 5218, Huntsman Advanced Materials) in N,N-dimethylacetamide (DMAC, Sigma–Aldrich). Electrospinning was carried out using 18 kV, 2 mL/h feeding rate and a distance of 20 cm between cannula and collector in a commercial electrospinning unit (Nanofibre Electrospinning Unit, Kato Tech). The mesh was spun to a thickness of 50 μm and heated to 320 °C for 1 h to extort any residual solvent.

2.1.2. Surface modification

For the surface modification the polyimide fibre network was rinsed in absolute ethanol followed by DI water to wash of organic residues. The mesh was then immersed in 4 M KOH aqueous solution of at 80 °C during 30 min to induce imide-ring cleavage and form potassium polyamate layer in the polymer chains on surface of each fibre. After rinsing with DI water, the fibres were immersed in 100 mM $AgNO_3$ aqueous solution at room temperature for 30 min to induce an ion exchange process in which the bound potassium ions are replaced by silver ions. Following another rinsing with DI water, the Ag^+ ions were reduced to form a silver coating on the fibre surface by immersion in 1 mM DMAB aqueous solution at room temperature for 30 min. After a final rinsing in DI water, the residual layer of polyamic acid was re-imidized through a thermal treatment in nitrogen atmosphere at 350 °C for 40 min.

2.1.3. Infiltration

The Sn–Ag–Cu matrix was introduced into the Ag coated fibrous network through liquid phase infiltration. The surface modified fibre film was first placed in a sealed mould cavity which was then heated to 250 °C and evacuated using a vacuum pump to a pressure of around 20 kPa. Molten alloy was filled into the cavity and the pressure was increased to 30 MPa to commence infiltration of the melt into the fibrous network. Finally a mould insert was used to define the final composite thickness before cooling and solidification.

2.2. Characterisation

The surface morphology of the fibre, composite preforms and fracture surfaces was characterised with scanning electron microscopy (SEM) (JSM6301F, JEOL). Samples that were prone to charging were first sputter coated with approximately 5 nm gold.

To investigate the Ag surface modification on the fibres, ultrathin sections were prepared using a diamond knife in an ultramicrotome (PowerTome XL, Boeckeler) from samples cast in epoxy. The sections were then placed on a 400 mesh copper grid for inspection using transmission electron microscopy (TEM) (Tecnai T20, FEI).

Attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) (Spectrum Two, PerkinElmer) was used to follow the transition to and from polyamic acid (PAA) during the surface modification. The spectra were normalized against the aromatic species at 1510 and 1488 cm^{-1} which are not affected by the treatment.

The elastic modulus was evaluated on samples with dimensions 15 mm × 5 mm × 0.05 mm that were cut out from fabricated

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