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Accelerated aging and thermal cycling of low melting temperature alloys as wet thermal interface materials

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ABSTRACT

This paper focuses on developing an effective thermal interface material (TIM) using low melt alloys (LMAs) containing gallium (Ga), indium (In), bismuth (Bi) and tin (Sn). The investigation described herein involved the thermal performance evaluation of LMAs after accelerated life cycle testing, which included isothermal aging at 130 °C and thermal cycling from -40 °C to 80 °C. Three alloys (75.5Ga/24.5In, 100Ga, and 51In/32.5Bi/16.5Sn) were chosen as candidate LMA TIMs. The testing methodologies followed ASTM D5470 protocols and the performance of the alloys was compared to commercially available thermal grease and liquid metal TIMs. To understand the LMA-substrate interactions, the alloys were applied to different surfaces (bare copper, nickel coated copper and tungsten coated copper). It was found that the proposed alloys between bare copper substrates were able to survive as long as 2700 h of aging at 130 °C and 1400 cycles from −40 °C to 80 °C without significant performance degradation.

1. Introduction

As the power density of microelectronics continues to escalate and is expected to exceed 100 W/cm^2 (1 W/mm^2) [1–3], the thermal management is becoming more challenging. Therefore, international electronics manufacturing initiative (iNEMI) declared thermal management a research priority in 2013 [4]. One of the crucial thermal management concerns in electronics cooling involves reducing the thermal resistance between the microprocessor chip and its accompanying heat sink by use of a thermal interface material (TIM). The function of the TIM is to transfer the heat effectively from the silicon die to the sink while incurring as small a temperature drop as is possible. For lower power applications (<30 watts of power, typically used in laptops), the silicon die is directly attached with the heat sink *via* TIM. However, for medium to high power (>30 watts of power, used in desktop and server applications), the die is connected with the heat sink *via* an integrated heat spreader (IHS) [5–7]. In the second case, two TIMs are used. One is placed in between the die and IHS referred to as TIM1 (or a level-1 TIM) and the other is in between IHS and the heat sink, known as TIM2 (or level-2 TIM) [3,8]. An ideal TIM would offer low thermal resistance with a thin bond line thickness (BLT), a high thermal conductivity, conformability at low to moderate pressures, good wetting properties, ease of manufacturing, and low cost while also being environmentally and health friendly [3,9,10]. In addition to those, compliant TIMs also must be able to withstand the mechanical stresses result from the coefficient of thermal expansion (CTE) mismatches between the adjoining materials (*e.g.* silicon – copper). If the CTE strain overwhelms the mechanical properties of the TIM, the joint will ultimately fail. Therefore, high performing compliant TIMS are a desirable design option for better thermal performance and improved reliability. Several factors such as surface roughness, flatness (waviness) and contact pressure affect the performance of TIMs [9,11]. Other factors such as non-uniformity of the heat flux [7] and pressure and die warpage [6] are major concerns in actual TIM applications.

Traditional TIMs include greases, phase change materials (PCM), gels, and pads, which are polymer based materials loaded with conductive particles (metal or ceramic), to enhance the thermal conductivity [5–12]. Greases are the most widely used TIMs and offer thermal resistances in the range of 0.1 to 1 cm^2 °C/W [6,12,13]. Chung [14] reported that a polyethylene glycol (PEG) based thermal paste can offer thermal resistance as low as 0.053 cm^2 °C/W when mixed with boron nitride. However, greases are messy, difficult to apply and remove during rework and have reliability issues such as pump out, phase separations, and dryout, which limit the use of grease as an efficient TIM over a nominal lifespan of use [6,9,11,12]. Another class of TIM, solders, can offer thermal resistance as low as 0.05 cm^2 °C/W [12,14], however, reworkability, high temperature processing, thermal stress development, and void formation the major concerns with solders [6,11,12]. Carbon-

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based materials such as carbon nanotubes (CNTs) (used directly or as a filler in composites) [1,2,8,15] and graphene [16,17] have been investigated by many researchers as TIMs. Cola [2] presented the thermal resistances of CNT based TIMs from different investigators, which all fall in the range 0.01 to 0.19 cm^2 °C/W, even though the thermal conductivity of CNTs is extremely high (6600 W/m °C and 3000 W/m °C and for individual single-walled and multi-walled CNTs) [2]. Also, design complexity and higher costs are associated with CNT based TIMs [17]. CNT-based TIM products have yet to appear as commercially available products.

In this work, the performance of low melt alloys (LMAs) as a TIM was investigated. Several researchers [3,10,18,19] have encouraged using LMAs as efficient TIM. LMAs possess high thermal conductivity (an order of magnitude higher compared to the traditional TIMs [3]) and they offer extremely low thermal resistance at small contact pressures and at a thin bond line. Eutectic alloys of indium, bismuth, gallium, and tin are preferable to use as TIMs [3,18,19]. Mercury, lead and cadmium based alloys are hazardous and usually should be avoided due to their toxicity and environmental issues [11,19]. Martin and Kessel [3] reported the thermal resistance of an undisclosed liquid metal TIM to be as low as 0.02 cm^2 °C/W with a thermal conductivity of 31 W/m °C. LMAs can be used either as a thin foil or in combination with a substrate [18,19] or as a filler material in a composite matrix [20]. Investigators are also looking into using LMAs in combination with carbon nanotubes [21] for TIM applications. Webb and Gwinn [18] measured the thermal resistances of alloy 117 (44.7Bi/22.6Pb/19.1In/8.3Sn/5.3Cd) with a copper (Cu) substrate and alloy 19 (51In/32.5Bi/16.5Sn) with or without a Cu substrate as 0.058 cm^2 °C/W and 0.071 cm^2 °C/W, respectively, with an uncertainty of about 30%. They [18] also observed performance degradation of alloy 117 upon thermal cycling from room temperature (RT) to 80 °C. Hill and Strader [10] reported the thermal resistance of a Cu-LMA of 130 μm thick (developed by coating alloy 19 on both sides of a 70 μm nickel plated Cu disk) as 0.017 cm^2 °C/W at 10 psi. Webb and Gwinn [18] observed that cycling 20 °C above the melting point resulted in significant increase in thermal resistance. However, Hill and Strader [10] did not find any notable performance degradation after cycling 90 °C above the melting point of the Cu-LMA TIM they tested. Therefore, a profound understanding of the reliability of LMAs as TIMs is missing from the literature. Finally, Hamdan *et al.* [13] reported the thermal resistance of liquid mercury micro droplets deposited onto gold plated silicon die is to be as low as 0.00253 cm^2 °C/W. However, mercury should be avoided unless its use is absolutely required.

Although the LMAs offer very low thermal resistances, there are several concerns such as oxidation/corrosion, intermetallic growth, dryout, dewetting and migration. Several investigators provided different ways to mitigate these problems [3,10,18,19]. For example, oxidation/corrosion of LMAs can be mitigated by providing a hermetic seal, and the formation of intermetallics can be prevented by applying a diffusion barrier coating [19]. Hill and Strader [10] found that the use of a gasket reduced oxidation significantly and facilitated alloy containment.

2. Experimental

2.1. Materials and test methodology

In this research, three commercially available alloys (alloy 60, alloy 14, and alloy 19) were chosen to test the thermal performance. The properties of these alloys are presented in Table 1. The reasons for choosing these three alloys are that they have a wide range of melting temperatures (from 16 °C to 60 °C) spanning the range of interests for most application in today's markets and various compositions.

In order to characterize the performance of the TIM, a standardized testing methodology was needed for the current investigation. The performance of any TIM is quantified by measuring the temperature jump across the joint (interface) per the level of heat flux traveling through the interface. Alternatively, this can also be described as the thermal resistance of the interface. The lower the temperature drop incurred across the thermal interface, the higher the performance of the TIM is regarded. Obviously, for high density computing applications, TIMs with the highest performance (lowest thermal resistance) are sought.

Table 1
Properties of the LMAs with melting temperature ranges from 16 °C to 60 °C.

Alloy no. (indium corporation)	Composition (% by mass)	Melting point (°C)	Density* (gm/cm^3)	Specific heat* (J/g °C)
60	75.5 Ga, 24.5 In	16	6.35	0.34
14	100.0 Ga	30	5.90	0.37
19	51 In, 32.5 Bi, 16.5 Sn	60	7.88	0.20

* The densities were obtained from the manufacturer data and the specific heats were calculated using Kopp–Neumann's law.

Several techniques such as steady state (ASTM D5470), transient laser flash, synthesized dynamic models, thermal test dies, and modified hot wire are available to quantify the thermal performance of TIMs [23]. The method chosen for measuring the performance of the TIMs investigated under this effort was ASTM D-5470, which is a standard, widely accepted method for testing the thermal performance of TIMs. According to the standard [24], the testing apparatus consists of two meter bars (hot & cold). Electrical heat is supplied through one bar as the other bar is cooled. The sample (TIM) is placed between the meter bars. Each meter bar is equipped with several temperature sensors to measure the drop across the sample. Several assumptions are made such as the sample thickness being uniform at the interface and the resulting heat flow is uniform, perpendicular to the test surfaces and purely one dimensional with no lateral heat spreading [24].

2.2. Description of the apparatus used

As previously stated, the thermal performances of all TIMs reported herein were generated using an ASTM D5470 standard TIM tester. The tester is commercially available through Analysis Tech. The detailed specifications of the apparatus used can be found in [25]. In this set-up (Fig. 1a and b), heat flows through the upper meter bar and the lower meter bar is cooled with a chiller. This tester uses a linear variable differential transformer (LVDT) sensor to measure the *in situ* thickness of the TIM joint. An applied pressure can be controlled from 5 to 380 psi using several different pressure kits. For the testing of LMAs, a pressure sensor was used which is accurate to ±2.5 psi in the range 5–95 psi. The electronic thickness measurement accuracy is ±1 mil (25 μm). The test surfaces of this testing device are a highly smooth, nickel polished finish with a flatness within 7–8 μm. The meter bars are thermally insulated to minimize the heat loss to the surroundings.

2.3. Test-rig modification

To avoid any contamination of the TIM tester surfaces, the LMAs were tested by placing them between Cu disks (alloy 110, highly smooth and flat within 7–8 μm) of 3.18 mm thick and 33 mm in diameter. The resulting disks' assembly was then placed under the tester, shown in Fig. 1b. Silicone oil (Xiameter PMX-200, viscosity 1000CS) was applied on the top and bottom surfaces to make a better and more reproducible contact between the test surfaces of the TIM tester and the Cu disks. The temperature differential (ΔT) across the LMA TIM was measured by inserting two high precision thermistor probes (1 mm diameter, accuracy within 0.05 °C) in a 1.2 mm diameter hole (16.5 mm depth, radius of the disk) drilled in the middle of the Cu disk. The hole was filled with thermal grease (Laird Tech. Tgrease 880) to reduce the contact resistance of the probe in the hole.