



Investigation into the application of low melting temperature alloys as wet thermal interface materials



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ABSTRACT

The application of low melt alloys (LMAs) containing In, Ga, Sn, and Bi as compliant high performance thermal interface material (TIM) is investigated. The investigation described herein involves *in situ* thermal performances of the LMAs as a function of applied pressure and interface temperature, performance evaluation after accelerated life cycle testing, and guidelines to improve the cycle life. Testing methodologies follow ASTM D5470 protocols, and measurements are validated by testing well established, commercially available TIMs and comparisons are made to values reported in open literature as well as manufacturers' data. Measurement shows that LMAs can offer thermal interfacial resistances as low as $0.005 \text{ cm}^2 \text{ }^\circ\text{C/W}$, and their performance is highly contingent upon the quality of the mating surfaces. The issue of LMA containment and dewetting are discussed along with the solutions to mitigate them.

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1. Introduction

Thermal management plays a key role in electronics cooling as the power density continues to escalate and is expected to exceed 100 W/cm^2 (1 W/mm^2) [1–3]. Therefore, international electronics manufacturing initiative (iNEMI) declared thermal management a research priority in 2013 [4]. One of the thermal management areas in electronics involves reducing the thermal resistance between the microprocessor chip and heat sink using a thermal interface material (TIM). The function of the TIM is to transfer the heat effectively from the silicon die to the sink. For low 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] where two TIMs are used. One is placed in between the die and IHS referred

to as TIM1 and the other is in between IHS and the heat sink, known as TIM2 [3,8]. An ideal TIM should possess high thermal conductivity, low thermal resistance at a thin bond line thickness (BLT), conformability at low to moderate pressure, good wetting properties, ease of manufacturing, and low cost while also being environment and health friendly [3,9,10]. 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 application.

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 resistance in the range of 0.1 to $1 \text{ cm}^2 \text{ }^\circ\text{C/W}$ [6,12,13]. Chung [14] reported polyethylene glycol (PEG) based thermal paste can offer thermal resistance as low as $0.053 \text{ cm}^2 \text{ }^\circ\text{C/W}$ when mixed with boron nitride (BN). However, greases are 'messy', difficult to apply and remove due to their high viscosity, and have reliability issues such as pump out, phase separations, and dryout, which limits the use of grease as an efficient TIM [6,9,11,12]. Solders can offer thermal resistance as low as $0.05 \text{ cm}^2 \text{ }^\circ\text{C/W}$ [12,14], however, re-workability, high

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temperature processing, thermal stress development, and void formation are the major concerns with solders [6,11,12]. Carbon based material 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 carbon nanotube based TIMs from different investigators, which fall in the range 0.01 to 0.19 cm² °C/W even though the thermal conductivity of CNTs are extremely high (6600 and 3000 W/m °C and for individual single walled and multi-walled CNTs) [2], however, high cost is associated with carbon nanotube based TIMs [17].

In this paper, the performance of LMAs as TIMs has been investigated. Several researchers [3,10,18,19] have encouraged using LMAs as an efficient TIM. LMAs possess high thermal conductivity (an order of magnitude higher compared to the traditional TIMs [3]) and they offer extremely low thermal resistances at small contact pressure 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 usually avoided due to their toxicity and environmental issues [11,19]. Hamdan et al. [13] reported the thermal resistance of liquid mercury micro droplet (deposited on gold plated silicon die) as low as 0.0025 cm² °C/W. However, mercury should be avoided unless its use is absolutely necessary. Martin and Kessel [3] reported the thermal resistance of a liquid metal as 0.02 cm² °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 (copper) [18,19] or as a filler material in composite [20]. Investigators are also looking into using LMAs in combination with carbon nanotubes [21]. Webb and Gwinn [18] reported the thermal resistance of alloy 117 (which contains lead and cadmium) with a copper substrate and alloy 19 (with or without a copper substrate) as 0.058 and 0.071 cm² °C/W, respectively, with an uncertainty of about 30%. Hill et al. [10] provided the thermal resistance of a copper-LMA (developed by coating alloy 19 on both sides of a 70 micron nickel plated copper disk) of thickness 130 micron as 0.017 cm² °C/W at 10 psi.

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 those 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. In this work, we have presented ways to reduce the escaping of molten metal and to enhance the wetting with the mating surfaces.

2. Experimental

2.1. Materials and test methodology

In this research, three alloys (alloy 60, alloy 14, and alloy 19) of Indium Corporation were chosen to test the thermal performance. The properties of these and other potential alloys that can be used as TIMs are presented in Table 1. The reasons for choosing these three alloys are that they have wide a range of melting temperatures (from 16 °C to 60 °C) and are of 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

Table 1

Candidate LMAs with melting temperature ranges from 11 °C to 81 °C.

Alloy no. (Indium corporation)	Composition (% by mass)	Melting point (°C)	Density (gm/cm ³)	Specific heat (J/g°C)
51	62.5 Ga, 21.5 In, 16 Sn	11	6.50	0.32
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
162	66.3 In, 33.7 Bi	72	7.99	0.20
174	57 Bi, 26 In, 17 Sn	79	8.54	0.17
27	54 Bi, 29.7 In, 16.3 Sn	81	8.47	0.17

* Bold entries designate alloys tested and reported for TIM applications herein. The densities were obtained from the manufacturer data and the specific heats are calculated using Kopp–Neumann's law [22].

density computing applications, TIMs with the highest performance (lowest thermal resistance) are sought.

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. A typical ASTM D5470 standard setup is presented in Fig. 1(a). 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 reference [25]. In this set-up (Fig. 1(b)), heat flows through the upper meter bar and the lower meter bar is cooled with a chiller. The 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 micron). The test surfaces of this testing device are a highly smooth, nickel polished finish with a flatness within 7–8 micron. 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 copper disks (alloy 110, highly smooth and flat within 7–8 micron) of 3.18 mm thickness 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 copper disks. The temperature differential (ΔT) across the LMA TIM was measured by inserting two high precision thermistor probes (1 mm diameter,

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