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Article

Fundamental understanding on the dynamic reactions of liquid gallium with aluminum

Sarah Li[1], Kevin Li[1], Yajing Liu[1]
[1] Hamilton High School, Chandler, Arizona

SUMMARY

High-powered electronic devices require liquid gallium (Ga) and Ga-based alloys as highly conductive thermal interface materials (TIMs) to efficiently dissipate heat from devices into heat sinks, often made of aluminum (Al). However, Ga reacts rapidly with Al and severely damages the heatsink, which can significantly shorten the use life of the electronic devices. We hypothesized that adding a nanometer-thick coating, such as aluminum oxide (Al_2O_3), which is chemically stable due to its strong ionic bonds, or Iridium (Ir), which is chemically stable due to its full 6s and 4f outer electron shell, on the surface of Al can slow the reaction between liquid Ga and Al, thereby extending the heatsink's lifespan. To test this hypothesis, we studied the dynamic reaction of Ga with Al, Ga with nanometer Al_2O_3-coated Al, and Ga with nanometer Ir-coated Al at device operating temperature of 50°C using an in situ digital optical microscope. We observed that liquid Ga penetrates and diffuses faster into Al than into Al_2O_3-coated Al at 50°C, causing more severe surface damage on bare Al. Furthermore, Ir-coated Al prevents the chemical reaction between Ga and Al within the same reaction time frame, resulting in no obvious surface damage from liquid Ga. Our findings demonstrate that Ir-coated Al can potentially serve as an innovative heatsink material for system-level cooling architectures that use liquid Ga-based TIM in high-powered electronic devices.

INTRODUCTION

High-powered electronic devices, such as central processing units (CPUs) and graphics processing units (GPUs), generate significant amounts of heat as a byproduct of their operations. A heatsink made of aluminum (Al) is often applied onto the surface of these electronic devices to dissipate heat from the devices to the heatsink (1). A layer of thermal interface material (TIM) fills the gaps between the heatsink and the devices to improve overall heat dissipation efficiency. The devices, the TIM, and the heatsink are designed to work together to transfer heat from the devices to the heatsink, where the heat can be dissipated into the ambient environment.

The efficiency of thermal dissipation plays a crucial part in the performance and longevity of electronic devices (1, 2). For example, when heat is not efficiently dissipated to the ambient air, a personal computer (PC) often encounters thermal throttling, i.e., it powers off frequently due to overheating. In the current device market, thermal grease is typically the TIM applied between devices and the heatsink to dissipate heat due to its low cost (2). However, thermal grease has much lower thermal conductivity than gallium (Ga) and Ga-based alloys (3). Ga-based alloys can also reduce thermal interfacial resistance, but their use as TIM faces a major challenge: Ga reacts with common heatsink materials such as aluminum (Al) (Ga + xAl ---> GaAlx) and causes significant damage to the heatsink (4, 5).

Although the reaction of Ga with Al has been studied before, the fundamental mechanism of the dynamic reaction of Ga with Al and the kinetic reaction process is not well understood (6). We hypothesized that adding a nanometer coating, such as aluminum oxide (Al_2O_3), chemically stable due to its strong ionic bonds, or Iridium (Ir), chemically stable due to its full 6s and 4f outer electron shell, on the surface of Al can slow down the reaction between liquid Ga and Al.

To test this hypothesis, we investigated the mechanism of the dynamic reaction between Ga and Al with and without an Al_2O_3 coating at device operating temperatures over time. Furthermore, we also studied the reaction of Ga on nanometer Ir-coated Al. Our goal was to identify an approach that slows the reaction between Ga and Al to extend the use-life of the heatsink cooling solutions for high-power electronic devices.

RESULTS

We hypothesized that introducing a nanometer-thick, chemically stable coating on aluminum (Al), such as aluminum oxide (Al_2O_3), stabilized by strong ionic bonding, or iridium (Ir), stabilized by its filled 6s and 4f outer electron shells, would slow the reaction between liquid gallium (Ga) and Al and therefore extend the operational lifespan of Al-based heatsinks.

To test this hypothesis, we examined the interaction between liquid Ga and Al under controlled heating conditions, beginning with bare Al surfaces prepared by removing the native Al_2O_3 layer using sandpaper to expose fresh Al. We observed that Ga turns into liquid at temperatures above

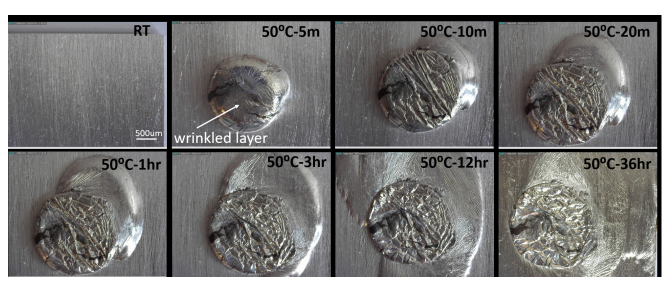

Figure 1. Fast Gallium (Ga) reaction on aluminum (Al) sheet at 50°C with thin aluminum oxide (Al_2O_3) polished off is observed. The sample is heated from room temperature (RT) to 50 °C. In situ optical imaging shows a wrinkled GaAlx layer forms on the surface of liquid Ga after 5 minutes of the reaction. Liquid Ga wets the surface of Al, and GaAlx segregates more to the surface of liquid Ga as time.

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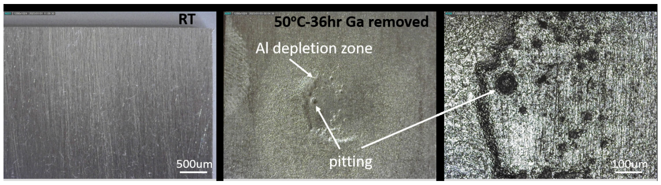

Figure 2. Optical imaging shows the surface pitting and corrosion on the aluminum (Al) with thin aluminum oxide (Al_2O_3) polished off at 50°C after 36 hours reaction. This indicates significant surface damage of the Al caused by fast liquid Ga reaction on Al.

29.5°C, i.e., the melting point of Ga. Liquid Ga reacted with Al immediately within 5 minutes at 50°C. Note that a thin wrinkled layer quickly forms at the surface of liquid Ga, and liquid Ga continuously wets, or spreads over, Al surface as time increases **(Figure 1)**. After 36 hours, the liquid Ga wet most of the Al surface with a thick, wrinkled layer segregated on the surface of liquid Ga. After we removed liquid Ga from Al surface, we observed significant damage, such as pitting corrosion and an Al depletion zone on Al surface **(Figure 2)**. We also measured the damage depth and area of liquid Ga on Al surface at 50°C through reaction time **(Figure 3)**.

The reaction of Ga on the surface of Al_2O_3-coated Al sheet at 50°C is first observed **(Figure 4)**. We assess that the reaction between Ga and Al_2O_3-coated Al sheet is slower than the reaction between Ga and Al. Liquid Ga does not wet the surface of the Al sheet at 50°C. The morphology of the Ga droplet on the Al_2O_3-coated Al surface remained largely unchanged for up to 36 hours at 50°C, indicating minimal wetting and limited interfacial interaction between Ga and the coated substrate. After the removal of the Ga droplet, the Al_2O_3-coated Al surface largely retained its original light-gray appearance; however, localized porous regions corresponding to minor pitting were observed, suggesting that while the Al_2O_3 coating substantially suppresses Ga-Al reactions, some localized degradation and Ga penetration may still occur. The depth of the damage (we suspect it is at the scale of µm) is not measurable with a micrometer. This further indicates that the reaction rate between Ga and Al_2O_3-coated Al is slower than Ga on Al, although pitting-like corrosion on both surfaces is observed.

To further validate the surface coating effect on Ga reaction with Al, we also studied the Ga reaction on Al with a nanometer-thick Ir coating. We observed no damage to Ga

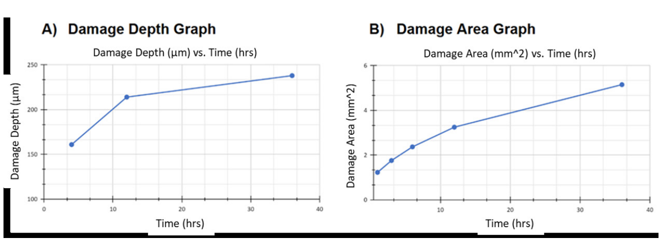

Figure 3. Damage to surface aluminum. The damage (A) depth and (B) area on the surface of aluminum were measured. Both increase as the reaction time between liquid gallium (Ga) and solid Al. This quantifies Al surface damage after the reaction between liquid Ga and solid Al at 50°C as time.

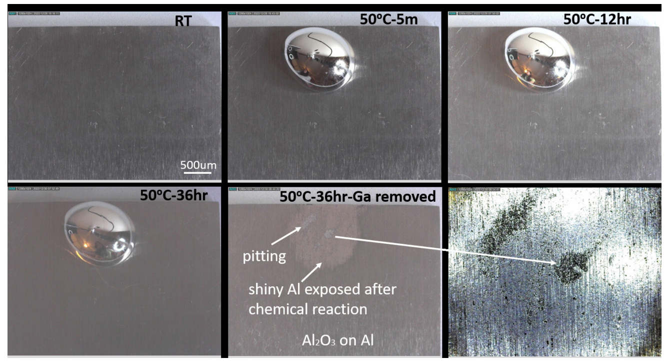

Figure 4. Gallium (Ga) reaction on aluminum (Al) sheet at 50°C with thin aluminum oxide (Al_2O_3) on the Al surface is less obvious than that on Al with Al_2O_3 polished off. There is only minor pitting and damage caused by Ga reaction with Al_2O_3-coated Al after 36 hours of reaction at 50°C.

on the Ir-coated Al sheet after 36 hours of liquid Ga on it at 50°C; therefore, there is no obvious reaction between Ga and Ir-coated Al that can cause damage to Ga on Ir-coated Al **(Figure 5)**. Overall, these results show that the reaction between liquid Ga and Al strongly depends on the surface condition of Al. Al reacts rapidly with liquid Ga, leading to extensive wetting and severe surface damage. The presence of an Al_2O_3 coating slows the reaction and limits Ga wetting, resulting in only minor surface damage after extended exposure. In contrast, a nanometer-thick Ir coating effectively prevents Ga-induced damage, with no observable reaction between Ga and Ir-coated Al under the same conditions.

DISCUSSION

In this work, we proposed that nanometer-scale, chemically stable surface coatings on aluminum, including Al_2O_3 and Ir, would inhibit gallium wetting and reaction kinetics, thus reducing Ga-induced damage and prolonging the functional lifetime of Al heat sinks. As our results show, the reaction rate and wetting behavior of Ga on Al sheet with and without the thin Al_2O_3 layer are much different. The reaction rate of Ga on Al (noticeable within 5 minutes) was much faster than that of Ga on Al with Al_2O_3 coating (noticeable after 36 hours). In practical thermal interface material applications, Ga-induced degradation occurring in the matter of minutes or even hours would be considered unacceptable, as it would lead to rapid failure in heatsink performance during normal device operation lasting years. This is due to Al_2O_3 having a more stable chemical form of Al ion than Al with valence electrons, so the chemical reaction of the liquid Ga on Al_2O_3 is much slower than that of liquid Ga on Al. Thus, extending the onset

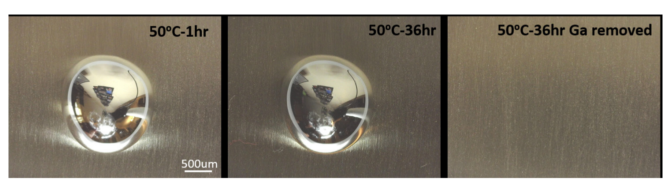

Figure 5. Gallium (Ga) reaction on aluminum (Al) sheet at 50°C with thin iridium (Ir) coating on the surface is not observed. There is no damage of liquid Ga on Ir-coated Al after 36 hours of reaction at 50°C

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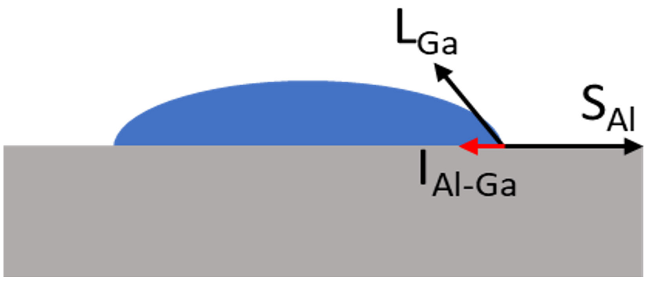

Figure 6. Young's equation illustration. The wetting behavior of liquid gallium (Ga) on the surface of solid aluminum (Al).

of visible reaction represents a meaningful improvement for device longevity toward real-world usability.

We also observed that liquid Ga wets more on Al than Al_2O_3. This phenomenon can be explained by the theory of Young's equation **(Figure 6)**. According to Young's equation, the wetting behavior of liquid droplet on the surface of a solid can be defined by the wetting angle or contact angle, which is set by the surface free energy of liquid droplet (Ga), solids (Al or Al_2O_3), and interfacial energy between liquid Ga and solids (Al or Al_2O_3). For example, in the case of liquid Ga on solid Al,

$$S_{Al}=L_{Ga}+ I_{Ga/Al} *\cos(\theta) \quad ....(\text{Equation 1})$$

Where S_{Al} is the surface free energy of solid Al, L_{Ga} is the surface free energy of liquid Ga, $I_{Ga/Al}$ is interfacial energy between solid Al and liquid Ga, and θ is the wetting or contact angle. From the literature, the surface free energies of liquid Ga, solid Al, and Al_2O_3 were studied, and their values are listed in **Table 1** (7). Since the surface free energy of solid Al is much higher than the surface free energy of solid Al_2O_3, the wetting angle of liquid Ga on Al is much lower than that on Al_2O_3, indicating a better wetting of liquid Ga on Al than on Al_2O_3, which also leads to faster reaction of Ga on Al due to much larger reaction areas. In thermal management systems, increased wetting directly correlates with increased interfacial contact area, accelerating atomic diffusion and chemical reaction area, which explains the severe damage observed on bare Al surfaces.

We observed another interesting phenomenon from the experiments: there was a wrinkled layer formed on the surface of liquid Ga when Ga reacts with Al rather than Al_2O_3. This can be explained by the dynamic reaction model of liquid Ga with Al_2O_3 and Al **(Figure 7)**. As for the case of Ga reaction with Al, the segregation of GaAlx phase at the surface of liquid Ga is defined by the following steps: Al atoms diffuse into liquid Ga and liquid Ga diffuses into solid Al at Ga and Al interface, then the lighter Al atoms diffuse toward to surface of liquid Ga due to its lower density (2.7 g/cm^3) than liquid Gallium (6.1 g/cm^3), and final there is a formation of stable GaAlx phase at the surface of liquid Ga. Such intermetallic phase formation is widely reported as a key failure in Ga-Al systems, as it both weakens the structural integrity of Al and sustains further Ga infiltration (5). In comparing the reaction of Ga on Al and Al_2O_3, our data clearly demonstrates that Al_2O_3 at the surface of Al can slow down the reaction between liquid Ga and Al due to the more stable Al^{3+} chemical form. Furthermore, we also demonstrated Ga has no damage on nanometer Ir-coated Al **(Figure 5)**. Therefore, we validated our hypothesis through this research work that chemically stable nanometer oxide or metal coating on Al can slow down (in the case of Al_2O_3 coating) and even prevent (in the case of Ir coating) the reaction between liquid Ga and Al up to 36hrs reaction time frame at 50°C. From just an application standpoint, a complete suppression of Ga-induced damage over this timescale suggests Ir coatings may be viable for long-term deployment in liquid-metal-based cooling systems.

The current study sheds light on the future research direction of applying nano-coating on metal heatsink to potentially prevent the reaction of liquid Ga with metals such as Al. Such an innovative Ir-coated Al heatsink applied for the high thermal conductive liquid Ga TIM is suitable for high-end and high-power electronic devices, compared to the grease TIM, which is more applicable for low-end devices due to its lower cost. In particular, high-performance computing and power electronics naturally demand thermal interfaces that remain chemically stable over prolonged operation, making coating-based mitigation strategies very important. To fully estimate the use life of Ir coated Al heatsink for system-level cooling architectures that use liquid Ga-based TIM, more experiments needs to be carried out: we need to collect the reaction rate data between Ga and Ir-coated Al up to 105°C, which is the device temperature running at stress testing, and we need to collect the reaction rate data between Ga and Ir-coated Al at longer time period up to months. Both sets of experimental data can be fit into the reliability model for better prediction of the device's use life.

We observed the Al surface damage area and depth increases as the time of Ga reaction with Al increases, and

Elements	Surface Energy (mJ/m^2)
Aluminum	840
Liquid Gallium	707
Aluminum oxide	169-638

Table 1. Surface free energy of liquid gallium (Ga), solid aluminum (Al), and aluminum oxide (Al_2O_3) (9). Liquid Ga has higher surface energy than Al_2O_3 but lower surface energy than Al, therefore, it wets the Al surface but does not wet the Al_2O_3 surface.

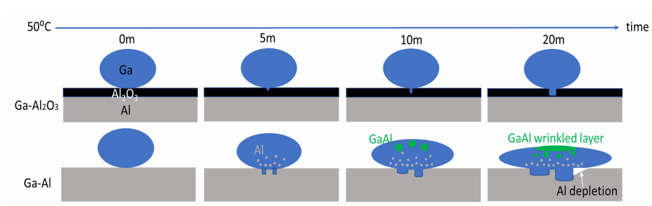

Figure 7. Dynamic reaction model of liquid gallium (Ga) with solid aluminum oxide (Al2O3) and aluminum (Al). Al2O3 slows down Ga reaction on Al. In the case of Ga reaction with Al without Al2O3 coating, lighter Al diffuses to the surface of liquid Ga forms GaAlx wrinkled layer, leaving significant Al damage (Al depletion zone) at Ga and Al interface.forms GaAlx wrinkled layer, leaving significant Al damage (Al depletion zone) at Ga and Al interface.

the Al depletion zone forms near the pitting holes, i.e., the diffusion pathway of Ga and Al into each other. This further demonstrates that the diffusion process between Ga and Al is the main cause of the damage to the Al sheet. In similar metal-liquid systems, even micro-scale pitting is known to significantly reduce mechanical strength and thermal conductivity, which highlights the severity of the damage that was observed. The hypothesis of whether such a diffusion process occurs through Al grain boundaries is still in investigation and validation. In addition, the correlation of the thickness of the Al_2O_3 to the reaction rate of the liquid Ga on Al will also be studied. The future work will involve a high-resolution scanning electron microscopy study of the Ga diffusion pathway into the Al sheet to further validate our diffusion models at the sub-micron scale.

In summary, we studied the dynamic reaction of liquid Ga with Al, Al_2O_3, and Ir-coated Al at 50°C under an in situ digital optical microscope. Lighter Al migrates to the surface of liquid Ga to quickly form GaAlx wrinkled layer, while liquid Ga diffuses into Al and causes Al surface damage, such as pitting. The surface damage area and depth increase as the time of Ga reaction with Al increases at 50°C. Liquid Ga wets Al but does not wet Al_2O_3 due to much higher surface free energy of Al than Al_2O_3. This also increases the reaction rate between Ga and Al due to the larger reaction area. Al_2O_3 slows down the Ga reaction with Al with minor damage on the surface observed, while Ir coating on Al prevents the Ga reaction with Al. Together, these findings position such nano-coatings as a promising solution towards enabling reliable liquid-Ga-based thermal management technologies.

MATERIALS AND METHODS

Sample preparation

In order to study liquid Ga reaction with pure Al, Al sheets and liquid Ga were ordered from Metals Depot (7) and Luciteria (8), respectively. Acetone was applied on the surface of all metal sheets to clean the incoming metal surface, and sandpaper with 350 grit size was used to polish off more than 100 µm metal surface to remove the 4 nm Al_2O_3 layer and expose the shiny Al surface. to beneath. Both Al sheets with/without Al_2O_3 layer with size of 30mm x 30mm x 2mm were placed on a hot stage with heating capability up to 300°C.

In situ optical imaging and data analysis

A digital Dino optical microscope was used to directly view the Ga surface. Optical images focusing on the surface of the Al sheets were taken at room temperature. Ga was then placed on the Al sheets with a pipette, and the optical images on the surface of Ga were taken to document initial morphology of Ga on the Al sheet. The samples were then heated to 50°C and held at 50°C for 36 hours. The temperature of 50°C is chosen since it is the typical personal computer heatsink temperature when it runs at normal condition. In situ optical images of Ga on Al with/without Al_2O_3 layer were taken at time intervals of 5 minutes, 10 minutes, 20 minutes, 1 hour, 3 hours, 6 hours, 12 hours, 24 hours and 36 hours.

After 36 hours of Ga reaction on Al sheet with/without Al_2O_3 layer at 50°C, the liquid Ga was removed. Optical images on the surface of Al sheets were taken to document the surface damage after reaction. The damage area was measured by the optical microscope, and damage depth (defined by the delta of the sheet thickness at the damage zone before and after the chemical reaction) was measured by a micrometer The experiments were then repeated on the nanometer Ir-coated Al sample. 1-2 nm Ir thin coating was sputtered on the surface of Al after polishing off the Al_2O_3 layer. The same data collection steps were taken to study the Ga reaction with the Ir-coated Al with an in situ digital optical microscope.

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