Ultrasound-Assisted Surface Modifications on Ceramic Reinforcement for Lead-Free Composite Solders: Short Review

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Abstract. The purpose of this paper is to review and examine the effect of ultrasound-assisted surface modifications of ceramic reinforcements on the properties of lead-free solders. The discussion will highlight the fundamental understanding, main parameters, configurations, and recent surface-modified ceramic reinforced composite lead-free solder developments. The review also identified and summarized the advantages, current trends, and significant findings in this field. The ultrasound-assisted surface modification was found to provide a crucial improvement on the wettability properties of molten solders as the matrix phase on the ceramic reinforcement. Further, the excellent distribution of ceramic reinforcement in solder matrix was also seen after the surface modification process. This has led to significant improvements in mechanical properties such as hardness and strength. The pinning of dislocation movement was seen as the reason for improving the mechanical properties. This positive impact in enhancing the ceramic reinforcement-solder interfacial reaction allows more explicit future research directions and opportunities for composite solder applications.

1 Introduction

Soldering is one of the most crucial and indispensable technology in electronic packaging. The main purposes of soldering are to provide physical continuity and electrical connectivity between different parts in electronic assembly [1]. Tin-lead (Sn-Pb) alloys have a long-standing history of application. The mature understanding of the characteristics of Sn-Pb alloys has allowed modern production lines to be optimized based on the properties of Sn-Pb. The Sn-Pb alloys are known to have a low melting temperature, physically and mechanically robust. At the same time, the availability of the raw materials is also high, at a much economical price [1, 2]. The presence of Pb was found to effectively negate the growth of dendritic Sn during solidification [1]. Sn-Pb alloys have a wide range of uses due to their many benefits over other metals. The harmful issue raised by the usage of Pb has sparked a
drive to limit its use over the previous two decades. This urges the use of Sn-Pb in electronic packaging to be replaced with new generation Pb-free solders.

In order to maintain high levels of reliability and processing consistency, many Sn-based alloys have been developed. These include tin-copper (Sn-Cu), tin-silver (Sn-Ag), tin-bismuth (Sn-Bi), tin-zinc (Sn-Zn), and ternary compounds such as Sn-Ag-Cu, among others. With the recent increase in interest for miniaturization in electronic devices, electronic assembly was forced to include the highest density of input/output (I/O) connections in a limited space [3]. This has become a severe issue for Pb-free solders to maintain the mechanical integrity at a much smaller joint size, as a single joint failure will cause malfunction of the whole electronic device.

One popular approaches to solving this problem is the production of composite solder by incorporating a nano-size ceramic reinforcement phase. Positive improvement in terms of microstructure refinement [4] and mechanical properties [5] have been reported from various types of ceramic reinforcement materials, such as graphene [6], silicon carbide (SiC) [7, 8], carbon nanotubes [9], titanium dioxide (TiO$_2$) [10] and zirconia (ZrO$_2$) [11]. However, the efficiency of adding this non-reacting reinforcement phase was hindered by segregation and non-wetting issues.

Aggregation is directly related to the dominant Van der Waals force between the particles [12]. As the particles aggregate, phase segregation took place in the composite, resulting in poor dispersion of the reinforcement. This causes the composite solder to have overall non-uniform properties. Meanwhile, the vast differences in coefficient thermal expansion between ceramic reinforcement and the solder triggered the non-wetting problem [13], aside from considerable differences in density [14]. Consequently, interfacial voids will be introduced (Fig. 1a). Non-wetting was also reported to produce a rough surface and introduce cracks on the composite solder due to expulsion of reinforcement [15]. To overcome this, void rigidification process was introduced to eliminate the void through interfacial bridging [16] (Fig. 1b-c). The use of ultrasound has been widely reported to produce interfacial bridging successfully [4, 17].

1.1 The concept of ultrasound in surface modification

Ultrasound is the sound waves with a frequency above the hearing threshold of humans, typically associated with the frequency range of 20 kHz to 500 MHz [18]. The frequency level is inversely proportional to the power output. Low-intensity, high-frequency ultrasound (in the megahertz range) does not alter the state of the medium through which it travels and is commonly used for nondestructive evaluation and medical diagnosis [19]. However, high-intensity, low-frequency ultrasound does change the form of the medium. Mechanical vibration will be introduced into the liquid when ultrasound passing through. In addition to this effect, ultrasound also generates acoustic streaming within the liquid [18].
the unique properties of ultrasound enables the amplification of reaction rates. This rate amplification technique is also known as sonochemistry and is widely used in various fields due to its versatility. Sonochemistry improves the yield, changes the reaction pathway, and initiates the reaction in biological, chemical, and electrochemical systems.

The fundamental of sonochemistry is known as the acoustic cavitation process. The term "cavitation" is from the Latin word "cavus," which means hollow. Cavitation is the formation of cavities (compressed gasses) inside a liquid, because of an abrupt decrease in the cohesive or intramolecular forces between the liquid, due to rapid pressure drop [20]. Ultrasonic irradiation of liquid generates oscillatory motion in the molecules transmitted through the liquid via pressure waves. Therefore, it induces rarefaction and compression waves to the molecular structure of the liquid. This causes the liquid molecules to vibrate around their position. An increased in the ultrasonic intensity within the liquid can increase the distances between the molecules and the loss of intramolecular forces within the molecular structures, as shown in Figure 2. The liquid molecules break down, and a cavity known as cavitation bubble is formed. These bubbles respond to the ultrasonic field within the liquid via expansion and contraction and finally collapse [21].

Fig. 2. The schematic diagram of the ultrasonic process

Ultrasound attracts attention because it offers low operating costs by removing various processes and reduce the synthesis steps. Besides that, ultrasound operates typically at low temperatures and pressures, eliminates the need to use special solvents, and reportedly increases end yields and catalyst activity [18]. It is also reported that ultrasound permits the use of lower reagents purity. However, undesirable physical effects such as shockwaves and turbulent flow need extra precaution from the user during the process [21]. The use of ultrasound in sonochemistry has been shown to deposit a wide range of nanomaterials on various substrates, including metals, ceramics, and semiconductors, among others. For example, successful deposition of amorphous nickel particles on micro-spheres of silica has been accomplished by a sonochemical [23], silver nanoparticles deposition on wool fabrics [24], and TiO\(_2\) on cotton fabric [25]. It is also worth noting that the ambient conditions of the system, such as the reaction temperature, pressure, and ultrasonic intensity or frequency, significantly affect the reaction rate [26].

The processes involved in depositing materials using the sonochemical route generally started with the mixing process of selected reinforcement materials with the source of the coating materials. In some instances, inert gas is needed during mixing to remove dissolved oxygen in the solution. For example, Liu et al. [27] flowed nitrogen gas for at least an hour prior to mixing cadmium sulfate with H\(_2\)PtCl\(_6\).6H\(_2\)O aqueous solution. The sonication process is typically implemented at frequency variations of 18-40 kHz, ranging from 30 minutes to a few hours, depending on the type of materials used [27, 28]. In some studies, the heating process may also be applied during sonication. Finally, the obtained precipitate will be washed with distilled or deionized water before drying in an air [28] or vacuum oven [27].
Cobley et al. [29] summarized the steps taken in depositing gold (Au) layer on silica (SiO$_2$) nanoparticles for Pb-free solder application. The crucial processes started with adding excess polydiallyldimethylammonium chloride (PDADMAC) solution into SiO$_2$ nanoparticles solution. Excess PDADMAC ensured a sufficient link or bond was made and introduced positive charges on the SiO$_2$ nanoparticles. The gold seeding process followed once the functionalization process finished. This includes ultrasonic and centrifugal processes to separate excess gold particles. Gold shell formation was the final step, where SiO$_2$ nanoparticles with gold seeds were added to the gold salt solution with formaldehyde as an additive to allow complete encapsulation of gold at the thickness estimated 2–3 nm.

1.2 Effect of surface modification on the reinforced particle to the solder properties

The agglomeration of reinforcement is a typical phenomenon in composite solder as it is directly associated with Van der Waals force [30]. Ahmad et al. [31] recently reported graphene agglomerates in different sizes and shapes at various locations within the SAC305 solder composite. The agglomerates of graphene were found to be bulkier in the vicinity of the solder/substrate interface than the agglomerates closer to the solder outer surface due to movement of the graphene reinforcement during the reflow process. Worse, the agglomerates on the outer surface have a higher tendency to be expelled from the solder. Consequently, this leads to pitting formation at the surface of composite solder [32]. A similar type of pitting appearance was also reported by other work [33]. Meanwhile, a slight increase in the average thickness of IMC at the interface of solder/reinforcement at the additions of 1 wt. % SiO$_2$ and ZrO$_2$ nanoparticles are undesirable as they reduce the shear strength of the joint produced [34].

Successful improvisation on the interface of tetra-needle-like zinc oxide (T-ZnO)-added Sn-1.0Ag-0.5Cu composite solder had been reported through the nickel oxide (NiO) surface treatment using ultrasonic agitation [4, 35]. The improved ductility and strength of NiO-decorated T-ZnO- Sn-1.0Ag-0.5Cu composite solder was directly contributed by the efficient load-transfer mechanism at the interface. It is known that the whiskers of T-ZnO possess excellent strength and modulus that permit the untreated composite solder to withstand high-stress loading. However, as the stress loading exceeded the critical strength, the whiskers fractured, and the force was transferred back to the solder matrix. Poor interaction between T-ZnO and the solder subsequently failed to stop the crack from propagating (Fig. 3a). For the NiO-decorated T-ZnO-Sn-1.0Ag-0.5Cu composite solder, the absence of air gaps and defects help to produce a bridging effect between NiO-decorated T-ZnO and Sn-1.0Ag-0.5Cu solder that inhibited the crack propagation and allows better load-transfer at the interfacial region (Fig. 3b).
Fig. 3. The schematic diagram of (a) T-ZnO-Sn-1.0Ag-0.5Cu and (b) NiO-decorated T-ZnO-Sn-1.0Ag-0.5Cu load transfer mechanism

The improved interfacial reaction between reinforcement and solder matrix also successfully impeded the growth of IMC. According to Huo et al. [36], good interfacial bonding can block the diffusion path of Sn and Cu during the reflowing process. It was demonstrated that the additions of sonicated reduced graphene oxide (rGO) restrained 39.3-64.4% growth of IMC in Sn-2.5Ag-0.5Cu solder as a result of depletion in the source of Sn and Cu. Furthermore, good interfacial bonding enables the reinforcement particles to successfully pin the grain boundaries' movement, producing a much smaller grain of IMC [17]. The pinning of grain boundaries movement, also known as the Zener pinning effect, also allows the composite solder to create higher strength due to the microstructure refinement. Elimination of aggregation also permits uniform distribution of reinforcement and allows the heterogeneous nucleation to commence on the surface of the reinforcement throughout the solder matrix. As witnessed by Ashayer et al. [37], sonication at 1 W produced higher nucleation sites for 1% gold-silica nanoparticles, albeit further increase in sonication power reduced the attachments of gold on the silica nanoparticles.

2 Conclusion

Ultrasound-assisted surface modification has shown promising impact in solving the agglomeration or aggregation issue of reinforcements that hindering the full potential of producing reliable composite solders. With a straightforward concept and ease of application, ultrasound-assisted surface modification has been shown to improve reinforcement/solder interfacial reaction that directly enhanced the distribution of the reinforcement. Additionally, this enables the reinforcement to act as a heterogeneous nucleation site for solidification, resulting in substantial microstructure refinement of the composite solder. Consequently, this increased the overall mechanical properties of the composite solders. This positive finding is hoped to open exciting opportunities in utilizing the vast prospect seen in composite solder in the future.
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