

# Cohesive and Interfacial Fractures in Particulate Polymer Composites: Reflow-induced Filler Densification and Epoxy-Filler Adhesion Loss

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**Abstract**— *Heterogeneous viscoelastic materials such as silica or silver particle-filled epoxy glues are important adhesives in the semiconductor and electronics industries. Systems requiring both materials adjacent to each other to satisfy conductive and insulative package requirements are rare, and studies on their compatibility, thermomechanical behavior and reliability are scarce. Herein, we report the analysis of the cohesive and interfacial fracture observed in such systems subjected to mechanical stress, after varying assembly conditions. Filler densification in the conductive glue resulting in an early fail as compared with the non-conductive glue, manifested as an open failure in the simultaneous electrical and mechanical test.*

**Keywords**— *Filler, epoxy, adhesion, electrical failure, mechanical stress.*

## I. INTRODUCTION

The adhesion between materials is an important concept in many industrial applications such as in the microelectronics, biomedical, automotive and aerospace industries. The adhesive joint formation and its integrity determine the reliability of the resulting product [1].

Particulate polymer composites (PPCs) generally consist of metallic or non-metallic particles dispersed in either organic or inorganic matrices [2]. Composite matrices are often thermosets because of their high stiffness, strength and glass transition temperature relative to the thermoplastics [3]. Epoxy-based polymers are the most important class of thermosets for structural composites due to their relatively lower cost and ease in processing. One glaring disadvantage of epoxy-based composites is their relative brittleness, often resulting in structural damage due to poor crack growth resistance [4]. A common approach to improve damage tolerance is to incorporate secondary phases, as filling polymers with particles of different size-scale, aspect ratio, stiffness, and filler-matrix interfacial strength offers a cost effective way of functionalizing a polymer [5].

Electrically conductive adhesives (ECA) are composites containing electrically conductive fillers dispersed in a polymer. ECAs bind two surfaces and provide continuous conductive electrical paths, allowing the simultaneous electron or charge transfer. The fillers are highly conducting metal such as gold, silver, copper, and nickel. The selection of the type of metal filler is equally as crucial as the matrix selection because the filler may provide a reinforcing effect on the ECA

mechanical properties because of a higher hardness and modulus of elasticity compared to the polymer matrix [6-7].

Herein, we report the observed early failure in the conductive glue after unit reflow, exhibiting both cohesive and interfacial fracture. Imaging and elemental analyses show filler densification and epoxy-filler adhesion loss resulting in the electrical failure during mechanical stress.

## II. EXPERIMENTAL DETAILS

Polyamide (S1) and silica (S2) surfaces were attached using a conductive glue (CG, Ag-filled epoxy) and non-conductive glue (NCG, SiO<sub>2</sub>-filled epoxy) as shown in Fig. 1. The unit containing these surfaces undergoes standard reflow process for SAC305 solder because of a separate component attach. Mechanical stress was performed to assess the integrity of the adhesive joint. The mechanical stress is done by applying an incremental amount of force to push S2 from S1, and simultaneously monitoring the electrical signal to check for the integrity of the S1-CG-S2 interfaces. Lateral and cross-sectional imaging were performed using a Hitachi S4800 Scanning Electron Microscope (SEM). Elemental analysis was performed using the Energy Dispersive X-ray Spectroscopy feature of the SEM equipment. Thermogravimetric Analysis (TGA) was done on a TA TGA Q500.

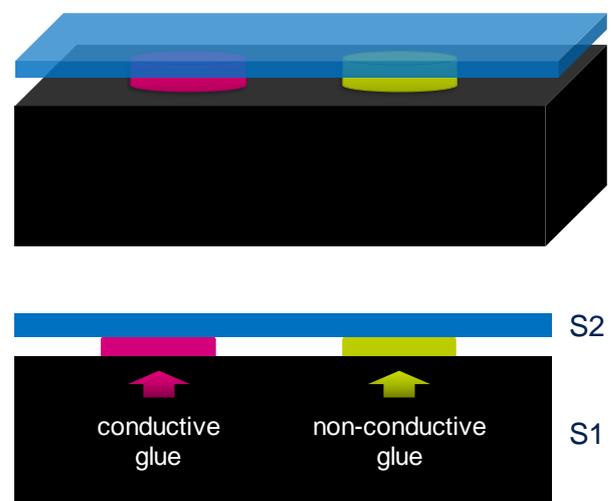


Fig. 1. Schematic representation of the attachment of S2 to S1 using CG and NCG.

### III. RESULTS AND DISCUSSION

The simultaneous mechanical and electrical testing of the attached surfaces revealed that the integrity of the adhesive joint is limited by the weakness of the S1-CG and S2-CG interfaces. Test results shown in Fig. 2 show that the early joint failure occurs in the CG area (orange data), and the failure occurs only in samples that underwent reflow (red box). This prompted a systematic failure analysis to ascertain the cause of this failure mode.

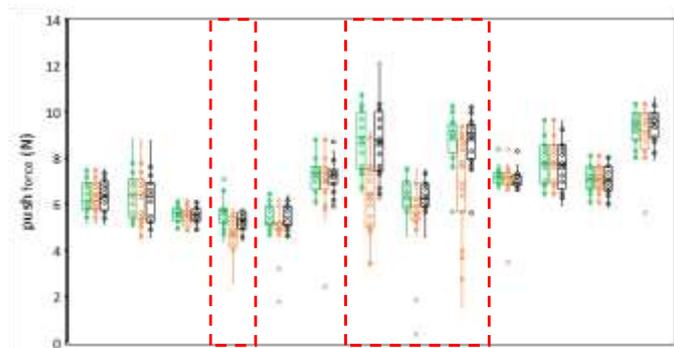


Fig. 2. Push test results showing early electrical failure in the interfaces created by the CG. Data enclosed in red box are samples that underwent reflow process. The orange bar shows push force where electrical failure is encountered indication fracture within or along the CG region.

Preliminary analysis considers the glue material as a potential source of the failure, possibly due to excessive outgassing resulting in crack initiation and propagation, or an out of control filler concentration resulting in less epoxy content, and hence weaker adhesion. TGA profiles (Fig. 3) reveals that there is no significant difference in the outgassing behavior and filler content between samples with and without reflow treatment. These results eliminate the possibility of the CG material as the culprit of the observed failure.

Micrographs of the fractured surface (Fig. 4) after the push test reveal important clues on the mechanism of the failure. The CG has a stronger adhesion with the S1 surface as indicated by the larger volume of the CG remnant on this surface. The fractured surface revealed that the spherical Ag fillers populate the surface. Chunks of smooth surfaces on the fractured surface indicate that interfacial fracture is also present, in addition to the more dominant cohesive fracture [6-7]. The interfacial fracture suggests the failure of the epoxy to develop a robust intercalated polymer network, which acts as the anchor of the glue on the surface of S2. The fractured surface of the reflowed sample is more planar, as compared with the sample without reflow, indicating that crack initiation and propagation is lateral and very close to the interface [8]. The higher density of Ag fillers on the fractured surface for the samples that underwent reflow suggests a possible densification of the fillers near the interface. In contrast, the non-uniform fracture plane in samples without reflow suggests that the fillers are more dispersed in this system, preventing a clean and lateral crack propagation [9]. A closer look on the CG remnants on the S2 clearly show more pronounced imprints of the Ag fillers in samples that underwent reflow.

These imprints are epoxy materials that have weak interaction with the fillers. These results indicate that the reflow process induced severe degradation in the strength of adhesion of the epoxy on the spherical fillers, resulting in the total separation of the epoxy and the filler during crack propagation.

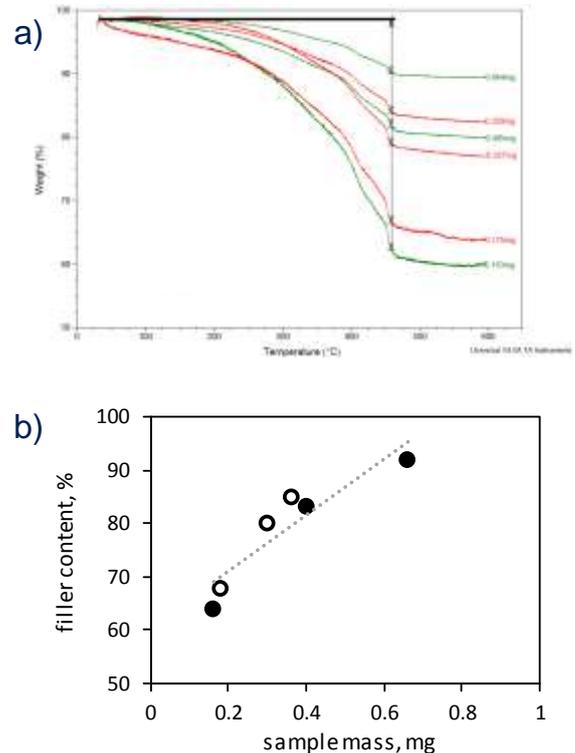


Fig. 3. a) TGA profiles of different sample mass for systems with (red) and without (green) reflow. b) Comparative plot of filler content of samples with (●) and without (○) reflow.

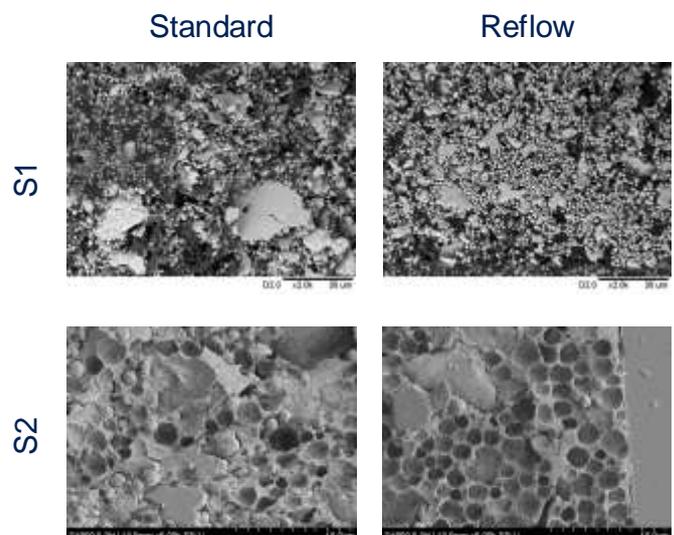


Fig. 4. SEM micrographs of the fractured surface after the push test.

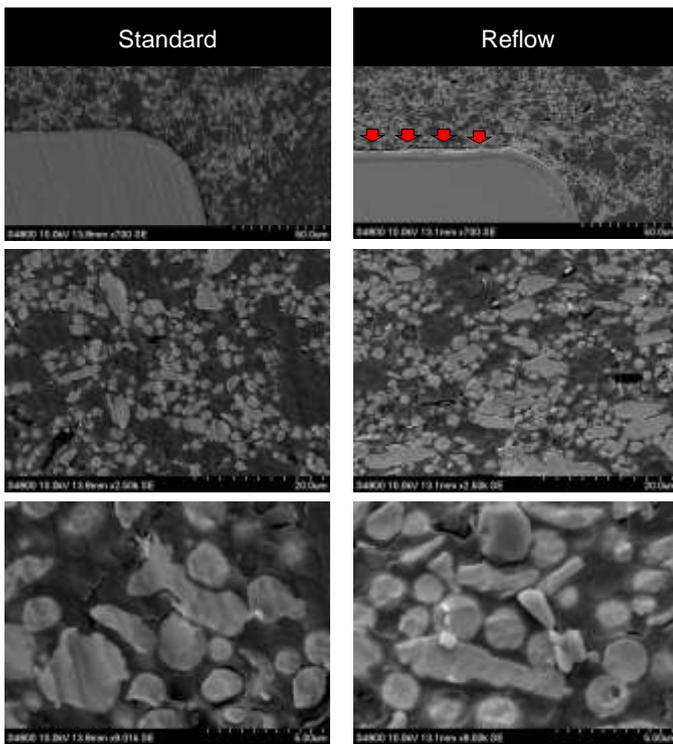


Fig. 5. Cross-sectional SEM micrographs showing higher filler density in the sample that underwent reflow.

Cross section SEM micrographs support the densification observed, revealing that samples that underwent reflow has  $34 \pm 7\%$  higher filler content near the interface as compared with samples without reflow. The higher filler content means that the epoxy content is lower, and hence less material that acts as dispersing medium. In addition, the filler-to-filler contact is higher resulting in weaker resistance to mechanical stress [10-11].

EDS analysis of the fractured surface (Fig. 6) confirmed the propensity of the Ag fillers along the fracture plane. Elemental map and line scan show the low organic content (epoxy) on the fractured surface and the periphery of the Ag fillers. This indicates the adhesion loss between the epoxy and filler as induced by the reflow process.

#### IV. CONCLUSION

Cohesive and interfacial fractures were observed in a system that involves attaching two surfaces via two epoxy adhesives, wherein one is conducting and the other is non-conducting. The adhesive joint strength is limited by the weakness of the S1-CG-S2 area where filler densification near the interface and filler-epoxy adhesion loss were observed. The reflow condition resulted in the degradation in the strength of adhesion of the epoxy on the spherical fillers, resulting in the total separation of the epoxy and the filler during crack propagation.

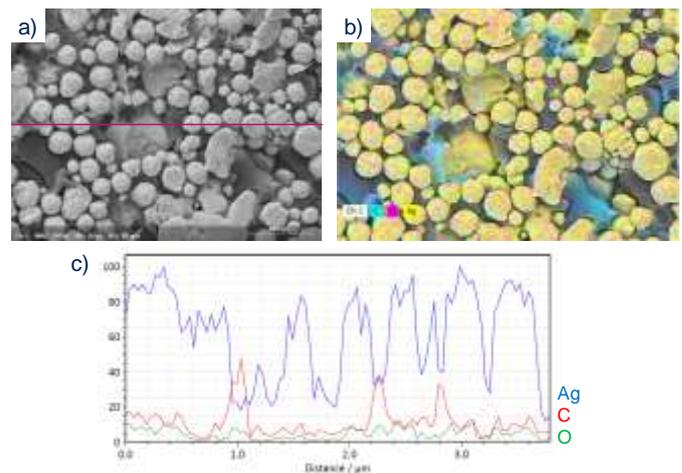


Fig. 6. EDS analysis of the fractured surface of the sample with reflow. a) Secondary ion image, b) elemental composite map, and c) line scan spectrum (elemental composition taken along the pink line in a).

#### ACKNOWLEDGMENT

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