Effects of Droplet Size and Dispersion Homogeneity on the Dielectric Integrity of Liquid Metal Polymer Composites

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***Abstract*—With the goal of optimizing the dielectric integrity and resiliency, this work studies the effect of the average liquid metal droplet size and dispersion homogeneity on the breakdown properties of liquid metal polymer composites based on polydimethylsiloxane and galinstan. Two groups of composites with different average droplet size and varying galinstan concentration were fabricated, and the partial discharge inception electric field and dielectric strength were measured. Upon curing, the larger droplets of galinstan settled while the smaller droplets remained more homogenously dispersed throughout the material. This settling effect has a detrimental effect on the dielectric strength and partial discharge characteristics of these composites. Finite element analysis models show that this phenomenon is most likely caused by high electric field intensity between the liquid metal droplets leading to partial discharge and dielectric breakdown.**

Keywords—liquid metal, dielectric strength, partial discharge, finite element analysis

I. Introduction

In stretchable electronic applications, particularly wearable and implantable electronic devices, soft circuit components and dielectrics have demonstrated superior mechanical strength and reliability over non-deformable components due to their ability to be repeatedly stretched and compressed without mechanical failure [1], [2]. Liquid metal polymer composites (LMPCs) that are comprised of polydimethylsiloxane (PDMS) and eutectic gallium-indium-tin alloy (GaInSn or galinstan) have demonstrated high deformability, low toxicity and reactivity, and good effectiveness in stretchable electronic applications such as dielectric elastomers, sensors, and capacitors [1], [3]–[5]. The electrical and mechanical properties of LMPCs can be tuned by altering the concentration of liquid metal and incorporating additional fillers such as barium titanate or iron. LMPC dielectrics with relative permittivity greater than 150 can be achieved using a high concentration of liquid metal [3], [6], [7]. However, high concentrations of liquid metal or other conductive fillers, particularly iron, adversely affect dielectric integrity by causing partial discharge and dielectric breakdown at electric fields as low as 85 V/mm [6], [8].

While the geometry of conductive materials is known to have a profound effect on electric field, research on the effect of galinstan droplet size on the dielectric failure properties of LMPCs is lacking [9]. Information is also lacking on whether the homogeneity of liquid metal dispersions in polymeric matrices influences the electrical properties of the composites. In an effort to determine the effect of settling on the dielectric integrity of LMPCs, this study investigates the differences in dielectric strength and partial discharge inception electric field (PDIE) of LMPCs with different average galinstan droplet sizes. Two-dimensional approximations of each type of composite as well as other cases were modeled with respect to electric field distribution under AC voltage stress using finite element analysis (FEA).

II. Composite Fabrication

LMPCs were fabricated with two approximate galinstan droplet sizes. For the first set of samples, the droplet diameter was tuned to approximately 1 µm, and for the second set, the droplet diameter was tuned to approximately 10 µm. Digital microscopy was used to verify the droplet size in each specimen. To obtain the 1-µm droplet diameter, 50 vol% galinstan was dispersed in V41 PDMS with a kinematic viscosity of 1×10^4 cSt.

To obtain the 10-µm droplet diameter, 80 vol% galinstan was dispersed in a 1:3 ratio of V41 and T11 (kinematic viscosity: 10 cSt) PDMS. Both dispersions were diluted with additional PDMS to the final galinstan volume concentrations of 5 vol%, 10 vol%, 25 vol% and 50 vol%. Each composite was mixed using a high torque overhead mixer and poured into polytetrafluoroethylene (PTFE) molds with a diameter of 25 mm and a depth of 2.5 mm and cured for 72 hours at 82°C. After curing, the exact thickness of each sample was measured using digital calipers, taking care not to apply any force or compress the samples. Fig. 1 shows a specimen of each type of sample.

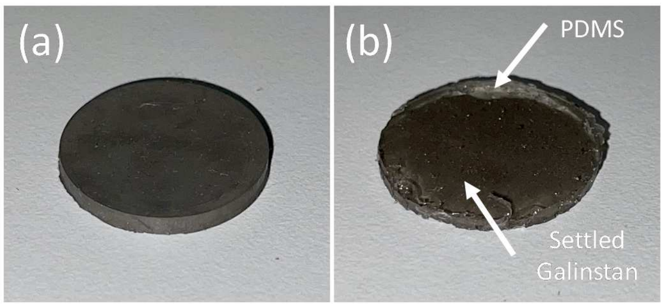


Fig. 1. (a) 10 vol% galinstan concentration at 1-µm droplet diameter and (b) 10 vol% galinstan concentration at 10-µm droplet diameter

After the samples were cured and removed from the molds, it was observed that the 10-µm diameter galinstan droplets had settled to form a layer at the bottom of the samples, while the 1-µm diameter galinstan droplets remained homogeneously dispersed throughout the material. Digital microscopy verified that the galinstan droplets in all samples remained distinct, and there was no coalescence of the galinstan droplets.

III. Experiment

The LMPC samples were analyzed experimentally using the testbed and circuit shown in Fig. 2 [6], [8].

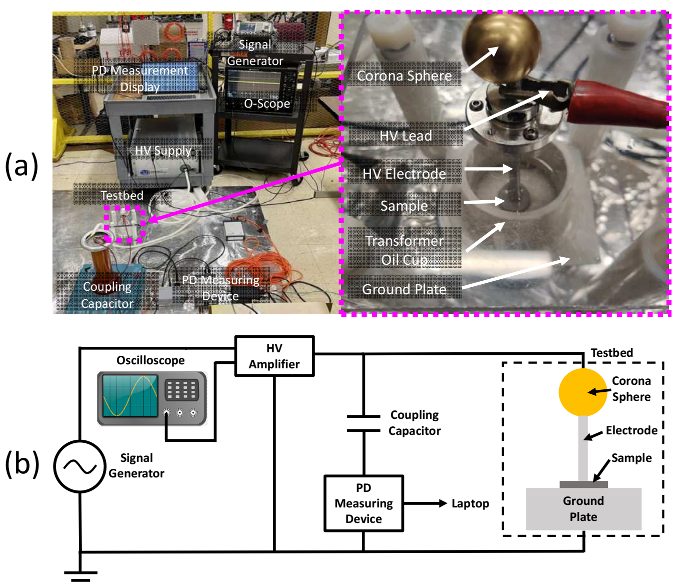


Fig. 2. (a) Experimental testbed and equipment and (b) test circuit (Note: Coupling capacitor and PD measuring device branch is only included for PD experiments.) [6], [8]

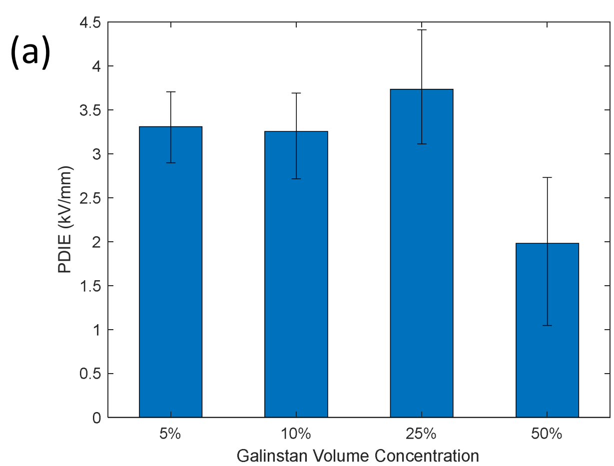
Each sample was placed in the testbed and submerged in transformer oil to suppress pollution of the measurements by surface discharge and surface flashover. Each sample was placed in firm electrical contact between the electrode and ground plate of the testbed, taking care not to compress or deform the sample. The 10-µm droplet diameter LMPCs were placed with the settled galinstan layer oriented upward.

For all experiments, the testbed electrode was connected to a power electronic driven high voltage amplifier, and the signal generator was set to output a 60 Hz sinusoidal signal. For partial discharge (PD) analysis, the system voltage was manually increased in small increments until PD was detected by the Omicron MPD800 PD measurement system. This voltage was recorded as the partial discharge inception voltage (PDIV) of the sample. The PDIV was then divided by the thickness of the sample to derive the PDIE assuming uniform electric field between the electrode and ground plate. To measure the dielectric strength of each sample, the coupling capacitor and Omicron MPD800 were disconnected, and the voltage was manually increased until the oscilloscope displayed a sinusoidal voltage collapse, indicating dielectric breakdown. This breakdown voltage (V_{bd}) was then divided by the thickness of the sample to derive the dielectric strength (E_{bd}). Six PD measurements and six breakdown measurements were performed for each type of sample, and the average values were recorded.

IV. Experimental Results

A. *Measurements for 1-µm Droplet Diameter LMPCs*

The experimental values obtained from the 1-µm droplet diameter LMPC samples are shown in Fig. 3. As shown in Fig. 3(a), the PDIE of the 1-µm droplet diameter LMPCs is nearly constant up to 25 vol% galinstan before decreasing dramatically between 25 vol% and 50 vol% galinstan. This behavior is likely due to the composites having a percolation threshold between 25% and 50% volume concentration of galinstan [10]. As shown in Fig. 3(b), the E_{bd} of the same group of composites exhibits a steady decrease with respect to volume concentration, which is characteristic of PDMS-based composites with highly conductive fillers [6].



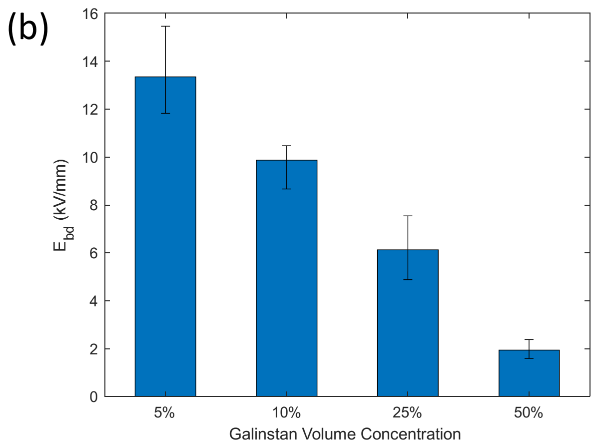


Fig. 3. (a) PDIE of LMPCs containing 1-μm diameter galinstan droplets and (b) E_{bd} of LMPCs containing 1-μm diameter galinstan droplets

B. *Measurements for 10-μm Droplet Diameter LMPCs*

The experimental values obtained from the 10-μm LMPC samples are shown in Fig. 4.

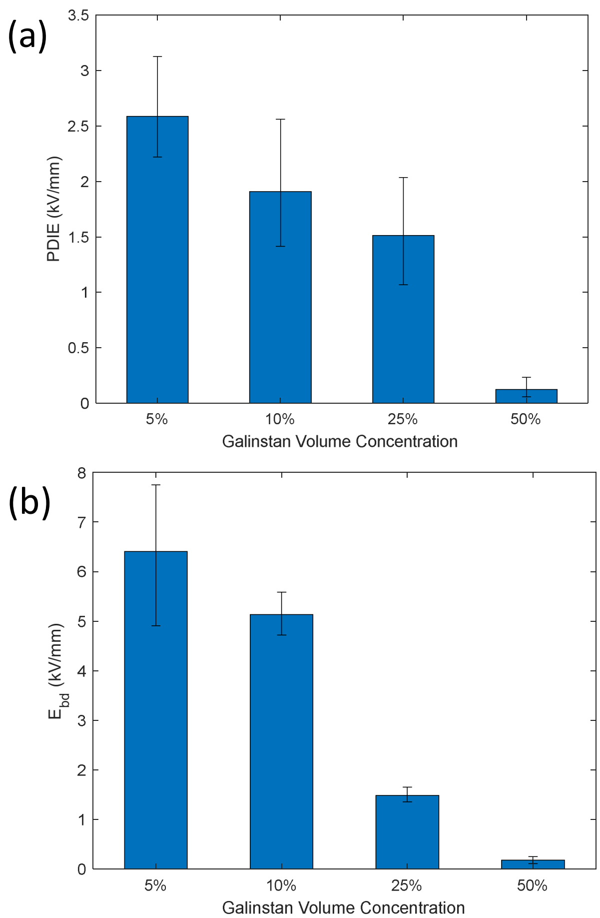


Fig. 4. (a) PDIE of LMPCs containing 1-μm diameter galinstan droplets and (b) E_{bd} of LMPCs containing 1-μm diameter galinstan droplets

As shown in Fig. 4(a), LMPCs containing 10-μm diameter galinstan droplets exhibit a steady decrease in PDIE and E_{bd}, unlike the composites with smaller galinstan droplets. At any given volume concentration of galinstan, LMPCs with 10-μm diameter galinstan droplets have lower PDIE and E_{bd} compared to those with 1-μm diameter galinstan droplets. This effect was magnified at higher galinstan concentrations.

V. Finite Element Analysis

In order to better understand the effect of droplet size and spatial distribution in LMPCs, two-dimensional qualitative FEA models were developed and analyzed using the electric currents module of COMSOL Multiphysics. Six models, all with an area galinstan concentration of 5%, were developed to study the effect of droplet size (small droplets and large droplets) and droplet arrangement (homogenously dispersed, settled on side nearest electrode, settled on side nearest ground plate). The galinstan droplets are represented by circles suspended in a rectangle of PDMS, and above each sample is a cross section of the aluminum electrode used in the experiments. For modeling purposes, aluminum is assumed to have a conductivity of 3.5×10^{7} S/m and a relative permittivity of 1. PDMS is assumed to have a conductivity of 1×10^{-13} S/m and a relative permittivity of 2.86, and galinstan is assumed to have a conductivity of 3.46×10^{6} and a relative permittivity of 1 at a frequency of 60 Hz[6]. The outside edge of the electrode is set to a 60 Hz sinusoidal voltage of 18 kV, and the bottom edge of the sample is set to 0 V.

Fig. 5 shows the 2D electric field distribution inside the LMPCs. Comparing Fig. 5(a) and Fig. 5(e), higher electric fields can be seen in the sample with a layer of large galinstan droplets adjacent to the electrode compared to the sample with homogenously dispersed small droplets. Since higher electric fields at a given voltage corresponds to a lower PDIE and E_{bd}, this model agrees with the experimental results. Based on the remaining models, droplet size alone has only a minor impact on electric field intensity, while droplet arrangement alone has a more significant impact on electric field intensity. In particular, a layer of galinstan droplets near the surface of the sample nearest the electrode leads to the highest electric fields of any scenario modeled.

VI. Discussion

This study finds that dispersion homogeneity and droplet size have a profound impact on the dielectric failure characteristics of LMPCs. In the 1-μm droplet diameter LMPC samples, PDIE is virtually constant at 25 vol% galinstan or less while being dramatically lower at 50 vol% galinstan. This is most likely because the percolation threshold is reached, forming discharge paths between the electrode and ground plate of the testbed [10]. Based on this behavior, the percolation threshold of these composites is most likely between 25% and 50% volume concentration of galinstan. The PDIE of the 10-μm droplet diameter composites and the E_{bd} of both composites exhibit a steady decrease with respect to galinstan volume concentration. This behavior is expected as higher concentrations of conductive filler increase the intensity of electric fields within the composite as voltage is applied, leading to PD and breakdown [6]. Furthermore, the droplet and dispersion characteristics can have a greater impact on PDIE and E_{bd} than overall galinstan concentration. Based on the FEA models shown in Fig. 5, there is only a slight difference in electric field intensity based on droplet size alone since internal

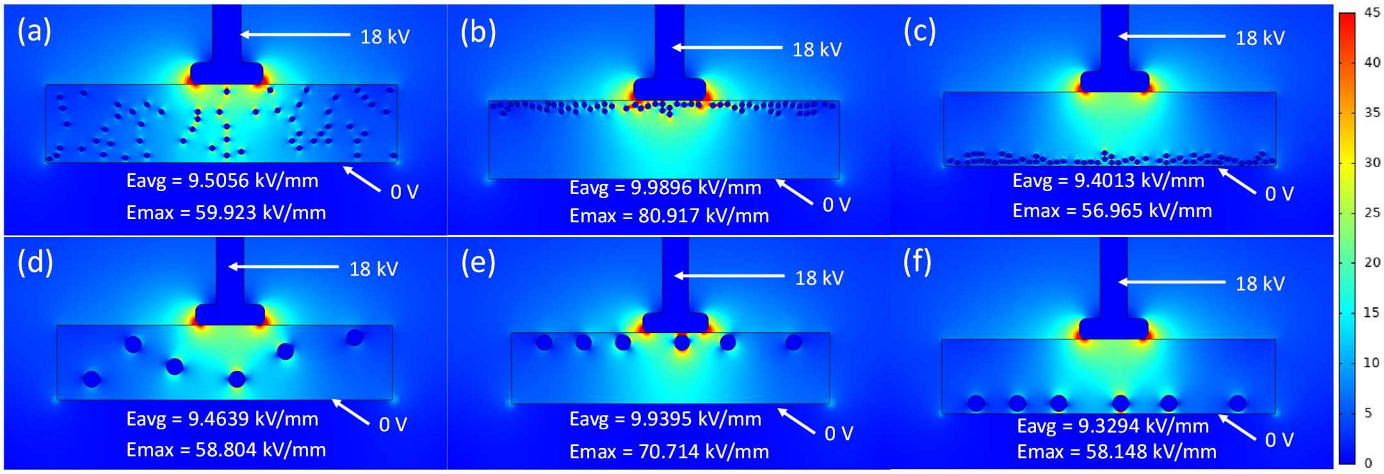


Fig. 5. FEA models of LMPCs showing the electric field distribution, average electric field, and maximum electric field within composites containing (a) small, homogenously dispersed galinstan droplets, (b) small galinstan droplets adjacent to electrode, (c) small galinstan droplets adjacent to ground plate, (d) large, homogenously µm dispersed galinstan droplets, (e) large galinstan droplets adjacent to electrode, and (f) large galinstan droplets adjacent to ground plate

electric field intensity at a given voltage is inversely proportional to dielectric strength. These models predict that if LMPCs are fabricated so that the 10-µm diameter galinstan droplets do not settle to the bottom of the PDMS before curing, 10-µm droplet diameter composites would have comparable PDIE and E_{bd} to 1-µm droplet diameter composites. The models shown in Fig. 5(b), Fig. 5(c), Fig. 5(e), and Fig. 5(f) predict that PDIE and E_{bd} exhibit a directional behavior in layered LMPCs. This is caused by high electric fields in the thin layer of PDMS between the galinstan droplets and the electrode and between individual galinstan droplets near the electrode.

VII. Conclusion

In this study, PD and breakdown measurements were conducted on PDMS-galinstan LMPCs of two distinct droplet sizes at varying galinstan concentrations. FEA was conducted to give further insight into the experimental results and to predict the behavior of additional droplet configurations. The results of this study suggest that galinstan droplet size and dispersion homogeneity have a great effect on the dielectric integrity of LMPCs, even more so than galinstan concentration. This study also predicts that a layer of galinstan droplets settled to one side of an LMPC would produce different PD and breakdown characteristics based on which side of the composite voltage is applied. This study suggests that a controlled galinstan droplet size and fabrication technique are necessary for tuning the dielectric properties, particularly the PD and breakdown characteristics, of LMPCs.

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