

Liquid Metal Integrated Multifunctional Elastomeric Materials For Soft Technological Applications

Pratip Sankar Banerjee¹, Soumili Chakraborti, Dhiraj Kumar Rana, and Shib Shankar Banerjee*

Department of Materials Science and Engineering, Indian Institute of Technology Delhi, Hauz Khas, New Delhi-110016, India ¹Presenting author e-mail: <u>msz218022@iitd.ac.in</u>; *Corresponding Author



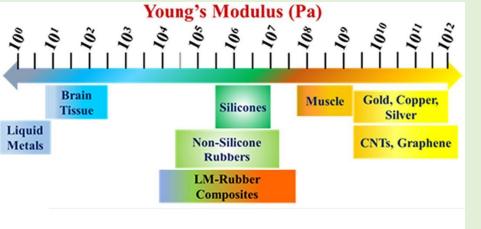
Abstract

Liquid metal (LM) integrated elastomeric composites are a breakthrough in soft multifunctional materials (SMMs) having immense transformative potential leveraging innumerable possibilities towards high-end engineering applications. LMs like eutectic gallium-indium alloy mitigate the long-withstanding filler-matrix modulus mismatch issues of a rigid filler-based composite while imparting immense functionality. In this work, viable strategies of introducing LM into different elastomeric matrices such as block copolymeric thermoplastic elastomers and silicone rubbers, bearing efficient interactions have been attempted via liquid state and vacuum processing which facilitates a homogeneous droplet-matrix morphology addressing significant phase-incompatibility and sedimentation of the LM phase often encountered in such materials. The elastic behaviour of the LM inclusions synergistically interacts with the matrix rubbers imparting outstanding mechanical features like, ultra-stretchability, and reduced hysteresis loss over an unfilled elastomer while inducing multifunctional attributes like remarkable enhancement of dielectric permittivity, thermal conductivity, piezoconductivity, and IR response. The experimentally determined results have been also rigorously analyzed and validated using theoretical models like Eshelby's and modified Eshelby's approach, modified Cole-Cole model, and Bruggeman formulation. The theoretical analysis also delves deep into the exciting outcome of LM inclusion size-driven softening or stiffening of the elastomeric matrix. Henceforth, this work provides a facile and scalable pathway to establish LM-elastomer interactions towards soft, multifunctional composite materials ratifying their instrumentality towards the possibility of smoothening the human-machine interface suited for next-generation soft robotics and wearable devices.

Introduction: Multifunctionality and the Possibilities □ Soft and compliant elephant's trunk exhibiting immense load bearing capacity

☐ Incredible conformability of cephalopods like octopuses The viable material possessing necessary compliance of the biological world? Elastomers??

And what about the functional fillers?





□Addressal of modulus mismatch issues experienced in conventional rigid functional fillers □Elastomer-like reversible deformability of LM fillers

Objectives

- □ Development of soft multifunctional binary and ternary composite materials by integrating LM with elastomeric matrices
- □ Designing of the composite material bearing high dielectric functionality, high thermal conductivity, magnetic response while maintaining a very high compliance
- □ Fabrication of compliant composites bearing skin-like softness for smoothening the human-machine interface

Experimental Section

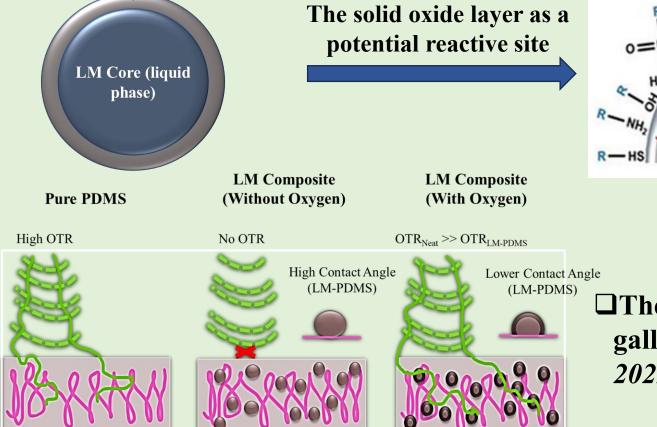
Part A: Multifunctional Block Copolymeric Composites

- □Styrene-Ethylene-Butylene-Styrene (SEBS); Density: 0.90 gm/cm³
- □Maleic Anhydride grafted Styrene-Ethylene-Butylene-Styrene (SEBS-g-MA); Density: 0.91 gm/cm³
- □Liquid Metal: Eutectic Gallium-Indium Alloy; Density: 6.44 gm/cm³; M.P.: ~ 18 °C
- ☐ Carbonyl Iron Powder (CIP); Density: 7.87 gm/cm³

Ga₂O₃ shell (rigid solid phase)

- Part B: Liquid Silicone Rubber Matrix ☐ Pt-catalysed two-part Liquid Silicone Rubber (LSR); Density: 1.01 gm/cm³
- □Liquid Metal: Eutectic Gallium-Indium Alloy; Density: 6.44 gm/cm³; M.P.: ~ 18 °C

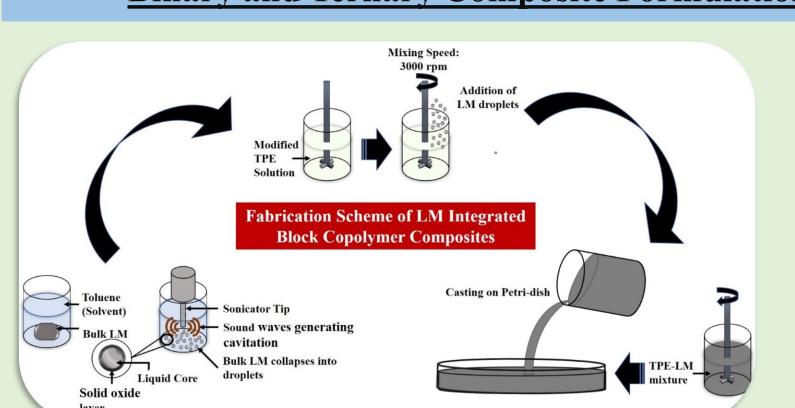
Possibilities of Interaction : The Solid-Oxide Layer



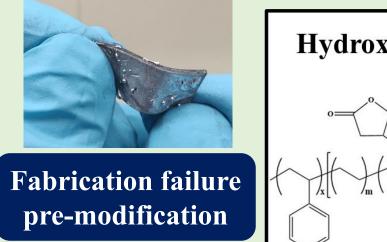
☐ The adhesion energy between surface oxidised eutectic gallium-indium and PDMS is $\sim 400 \text{ mJ/m}^2$ (Xie et al.,

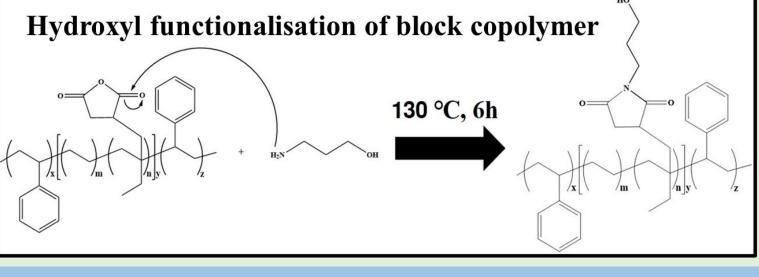
Part A: Multifunctional Block Copolymeric Composites

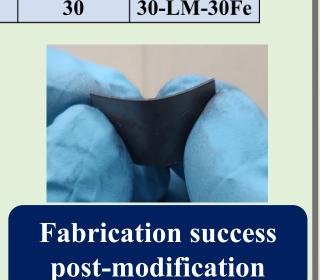
Binary and Ternary Composite Formulation and Fabrication Scheme



Modified SEBS-g-	EGaIn (vol%)	CIP (vol%)	Sample Name
MA (vol%)			
100	0	0	Neat
100	5	0	5-LM
100	10	0	10-LM
100	20	0	20-LM
100	30	0	30-LM
100	40	0	40-LM
100	30	10	30-LM-10Fe
100	30	20	30-LM-20Fe
100	30	30	30-LM-30Fe



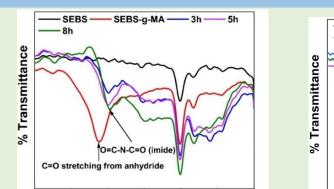


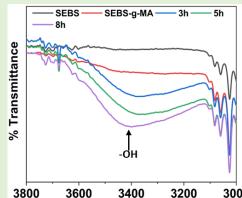


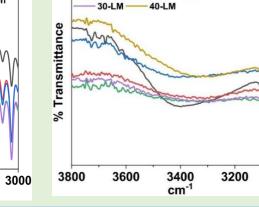
Functional Groups

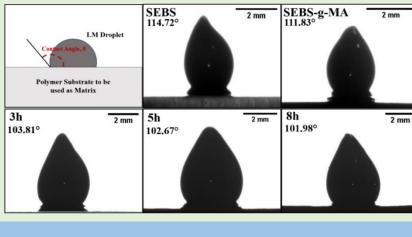
LM-5

H-Bonding Interaction at the LM-Block Copolymer Interface

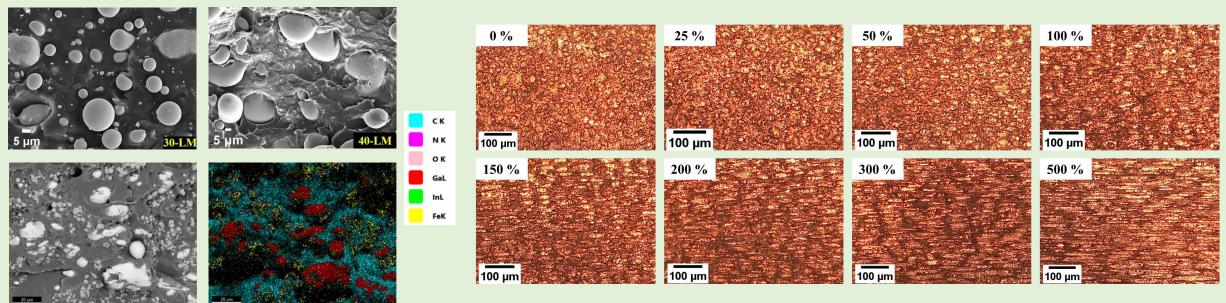








Morphological Investigation: Visualization of Adaptative Deformation



- ☐ Lumpy droplets and agglomeration at 40vol% LM loading
- ☐ Increasing tensile strain indicates the adaptative deformation of LM inclusions in a 30-LM specimen which also indicates the formation of a percolation pathway

Mechanical and Functional Properties 30-LM-10Fe \square Resistivity of the neat block copolymer $\sim 1.7 \text{ pS/m}$ Under compression mode the σ reaches up to ~ 0.005 S/m, in tension mode the maximum σ is ~ 0.15 S/m, and for dual strain mode it reaches to ~ 42 S/m

Part B: Ultra-Soft Composites with Skin-like Softness

Binary Composite Fabrication: Challenges and Solutions



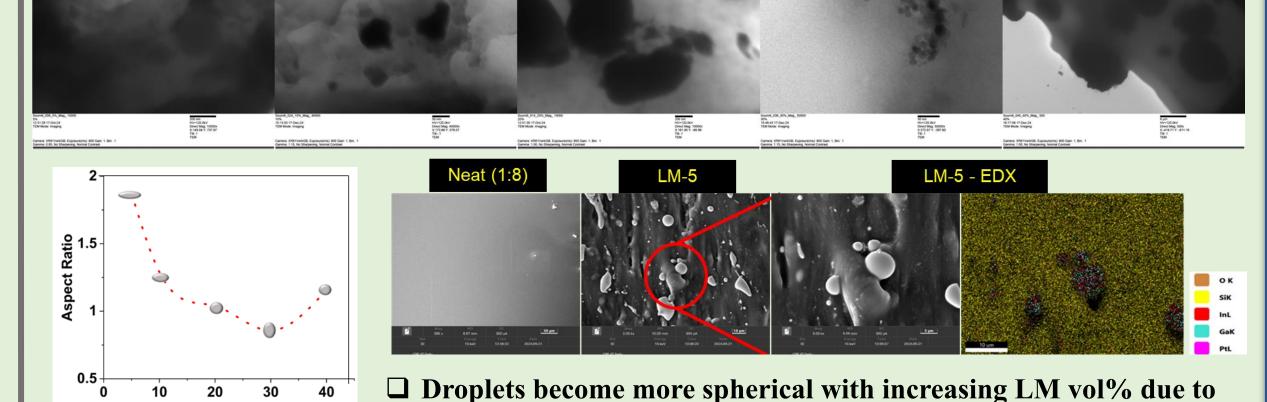
- ☐ Failure in mixing of Neat with 5 vol% LM in High Shear Mixer due to vortex formation and bubble entrapment;
- ☐ Shifting to vacuum mixing for obtaining results with minimal sedimentation even at high loadings of LM

LM-20

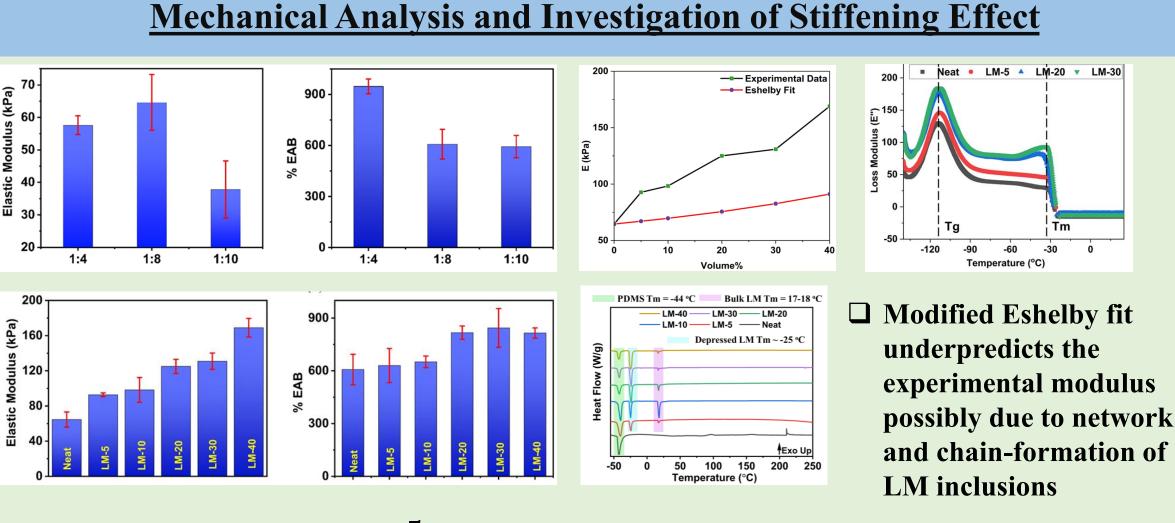
☐ Curing conditions: 16h@140 °C

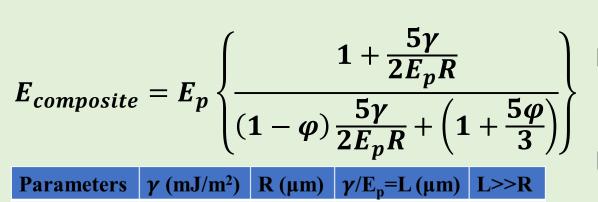
LM-10

Morphological Investigation: Effect of LM Concentration on Inclusion Shape



lesser available space within the matrix





2.45

Modified

Eshelby Fit

158.338

☐ Size-scale reduction of LM inclusions below the elastocapillary scale (L) renders matrix stiffening as surface tension of LM dominate ☐ The reduction of size scale of LM is also

LM-40

evident from the depressed Tm of LM from **DMA** and **DSC** analysis

Conclusion

3.5

- ☐ LM sedimentation and phase separation in a biphasic LM-block copolymer has been mitigated with reasonable success
- □Enhanced dielectric, thermal, electrical and magnetic properties could be achieved suitable for energy harvesting and thermoregulation devices
- ☐ Ultra-soft and skin-like binary composites have been fabricated by infusing LM into LSR with reasonable softness ☐ Unlike the block copolymer matrix, the LSR-based composites exhibits a stiffening effect of
- LM inclusions owing to size-scale driven phenomena

Acknowledgements







Magnetics and Advanced Ceramics Lab, IITD

- □ Banerjee PS, Banerjee SS. Liquid-Metal Infused Soft and Multifunctional Elastomeric Smart Thermal Management Device. Adv. Mater. Technol. 2025 (recently accepted)
- □ Banerjee PS, Banerjee SS. Interfacial H-bonding enabled liquid metal integrated multifunctional elastomeric architectures. J. Mater. Sci. 2023: 58, 14009 □ Banerjee PS, Rana DK, Banerjee SS. Influence of microstructural alterations of liquid metal and its interfacial interactions with rubber on multifunctional properties of soft composite materials. Adv. Colloid Interface Sci. 2022: 308, 102752