Organic eutectics: characterization, micro-structural evolution and properties.

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Eutectics are well-known multi-component systems used in various day-to-day applications. However, they are enigmatic in terms of structural organization (interactions and packing, the two prime features of a crystalline entity), despite having a long history. At the microstructural level, they are phase-separated (multi-phasic) solid solutions i.e. they are heterogeneous crystalline materials composed of homogeneous (single phase) but multiple solid solutions [1]. This phase heterogeneity in the structural integrity is what makes them complex-to-understand materials. Although research has been done in understanding the eutectic structural organization particularly in inorganic systems using advanced techniques such as atomic pair distribution function (PDF) analysis, X-ray microtomography and atomic force microscopy (AFM), no comprehension of eutectic microstructural integrity was achieved [2]. Furthermore, the structural and microstructural arrangement of organic eutectic systems has not been addressed so far in the literature [3,4]. This complexity in organic eutectic systems is augmented by several aspects such as 1) the constituents are primarily C, H, N and O which makes them soft materials, 2) atomic number contrast essential to image the microstructure is lacking, 3) frequent existence of polymorphism, 4) occurrence in lower structural symmetry. In this regard, one can transfer the knowledge of inorganic eutectics to organic eutectics or can verify the organic eutectics with competent experimental techniques in search of a improvised understanding from *molecular perspective*. Here, we manage to solve the microstructural features of organic eutectics through *in-situ* variable temperature (VT) PXRD experiments, DSC experiments with multiple heating and cooling cycles, *in-situ* VT Raman spectroscopic studies, gas phase energy calculations using Gaussian09 and electron microscopy imaging technique on a series of systems. We observe for the first time, the evolution of eutectic systems through formation of multi-domain eutectic particles at higher temperature. The eutectic particles melt altogether near the melting point of the eutectic system as showed in DSC experiments, via thermal energy induced heteromolecular interaction through the domain boundaries as confirmed from VT-Raman studies.

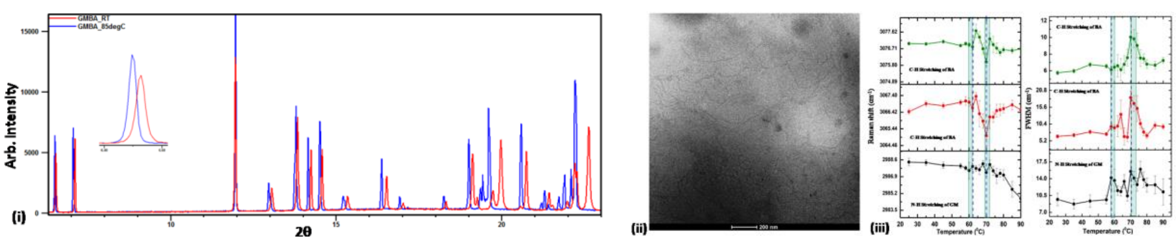

Figure 1. Characteristics of Gluterimide-Benzoic Acid eutectic system. (i) Overlay of room temp. (RT) (red) and high temp. (HT) (blue) PXRD patterns. Inset shows the increment of peak heights of Bragg's peaks at HT which indicates the increase in crystallinity. (ii) Electron microscopic (120 keV) image of the eutectic particle formed at HT shows domain boundaries. (iii) Raman shift and FWHM variation of molecular vibrational modes with temperature show transitions at HT indicating micro-structural evolution in the organic eutectic system .

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