Influence of Entanglement State on Molecular Mixing of Intractable Polyolefins and Its Implications on Rheological Response and Mechanical Properties

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Ultra-high molecular weight (UHMW) polyolefins exhibit unique mechanical properties that allowed it to be used in a variety of applications. Although the high molar mass of UHMW polyolefins leads to their enhanced mechanical performance, it is in fact the main impediment in their implementation in industry. Above the critical molar mass, the melt-viscosity of the polymer increases dramatically due to the formation of entanglements, following the power law $\boldsymbol{\eta}_0 \; \alpha \; \mathbf{M}_w^{3.4}$, which in turn poses a major challenge in processing them using the conventional processing techniques. Thus, bimodal blends of polyolefins are attractive, especially when two extreme molar masses can be blended homogeneously at the molecular length scale. Such a blend allows ease in processability while securing the high mechanical properties.

This work focuses on the enhancement of mechanical properties of commercially available grades of polyethylene (HDPE) through the addition of low-entangled UHMW-PE components via industrially viable solvent-free processes, i.e., melt compounding. Blends of HDPE/low-entangled UHMWPE at different weight percentages were prepared. The homogeneity of the blends was investigated through studying the rheological response. The result showed that with increasing UHMWPE content, the crossover point shifts towards lower frequencies (**Fig. 1a**), indicating an increase in the reptation relaxation time ($\boldsymbol{\tau}_{rep}$) as a result of increased average molecular weight ($\mathbf{M}_w$), as predicted by reptation theory. The logarithmic relationship between the measured $\boldsymbol{\tau}_{rep}$ and the corresponding $\mathbf{M}_w$ showed a straight line with a slope of 3.24, which is in close agreement with the reptation theory. Tensile testing showed that by incorporating 10 w.t.% of UHMWPE in HDPE matrix, the average stress yield point and young's modulus increased by 37% compared to that of pure HDPE (**Fig. 1b)**. Additionally, shish-kebab crystals were detected in the blended samples by SEM (**Fig. 1c**).

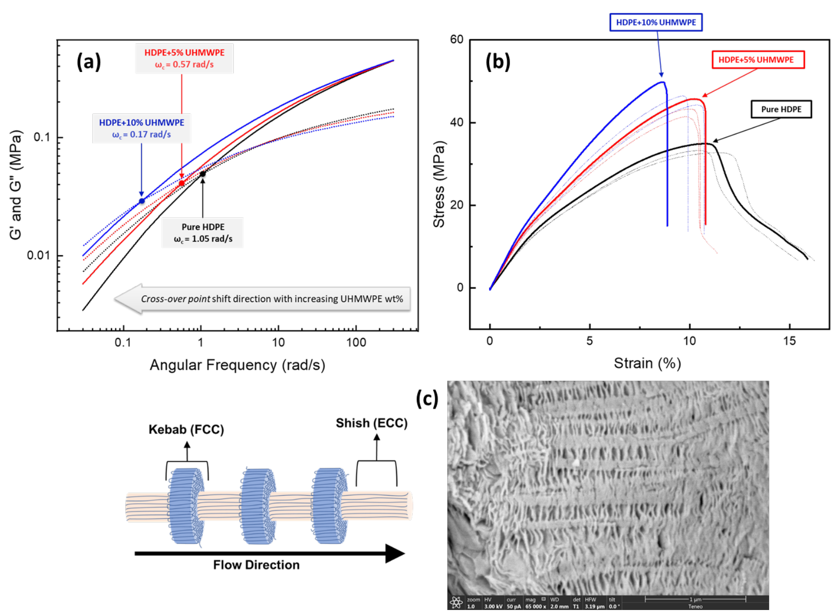

Fig 1: (a) Frequency sweep of HDPE/UHMWPE at different w.t. %. (b) Stress-strain curve of the blends. (c) Etched-SEM image of HDPE/UHMWPE that shows the formation of shish-kebab crystals.