Molecular understanding of mechanical properties of Archimedean tiling through star terpolymer thin film

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The eleven Archimedean tilings, suggested by Johannes Kepler [1], are described by the number of edges of polygons meeting at each vertex. These tiling patterns rarely occur naturally but can be replicated in specific synthetic polymers. The 3-miktoarm star terpolymers of different arms can form these tiling patterns based on the arm length ratio, displaying rich self-assembled morphologies [2].

This study utilized many-body dissipative particle dynamics simulations [3] with coarse-grained molecular models to investigate the relationship between morphologies and mechanical properties. Simulations were performed on single and binary systems of miktoarm star terpolymers with varying arm length ratios (Fig. 1a).

Morphology: Archimedean tiling patterns appear when the arm length ratio (x) is between 0.5 and 3.0, with broader interfaces than lamellae structures. These interfaces are widest at x = 1.0 and 1.5, corresponding to specific tiling patterns.

Mechanical Properties: Under tensile deformation, systems with broader interfaces showed more robust mechanical responses. This is due to the junctions connecting distinct domains, similar to cross-links, which resist void growth. Systems with smaller homogeneous domains exhibit less void formation, leading to improved mechanical properties.

Fracture Behaviors: Two fracture modes were identified: simple void enlargement and aggregation into larger cavities. Systems with Archimedean tiling experienced constrained void expansion, resulting in numerous small voids that eventually merged (Fig. 1b). A void-fibril network formed during deformation in binary mixtures with semi-tiling structures, preventing large cavities.

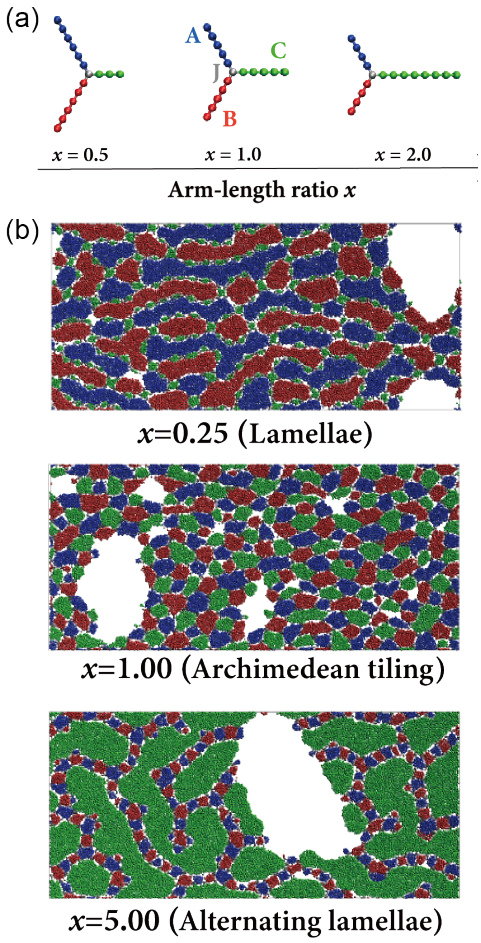

Figure 1 (a) Schematic representation of coarse-grained molecular models with different arm length ratios (b) Representative snapshots of fracture behaviors at strain 120%, which is before the fracture finishes

In summary, the mechanical properties of miktoarm star terpolymer thin films are closely related to the morphology, specifically the interface area. Archimedean tiling patterns offer superior mechanical properties compared to lamellae structures. The addition of different star terpolymers can either weaken or strengthen the material, depending on the morphology changes they induce. These findings provide insights into designing polymeric materials with desired mechanical properties by controlling the molecular arrangement and morphology.

[1] B. Grünbaum, G. C. Shephard, *Tilings and Patterns*, Courier Dover Publications (1986).
[2] T. Gemma, A. Hatano, T. Dotera, *Macromolecules*, **35**, 3225 (2002).
[3] P. B. Warren, *Phys. Rev. E*, **68**, 066702 (2003).