

## CHAOS-INDUCED MULTI-SCALE STRUCTURE DEVELOPMENT IN POLYMERIC SYSTEMS

This research utilizes self-similar microstructures of chaotic mixing to obtain unusual morphological forms in the blending of miscible and immiscible polymers and polymers with micro- and nanoscopic fillers.

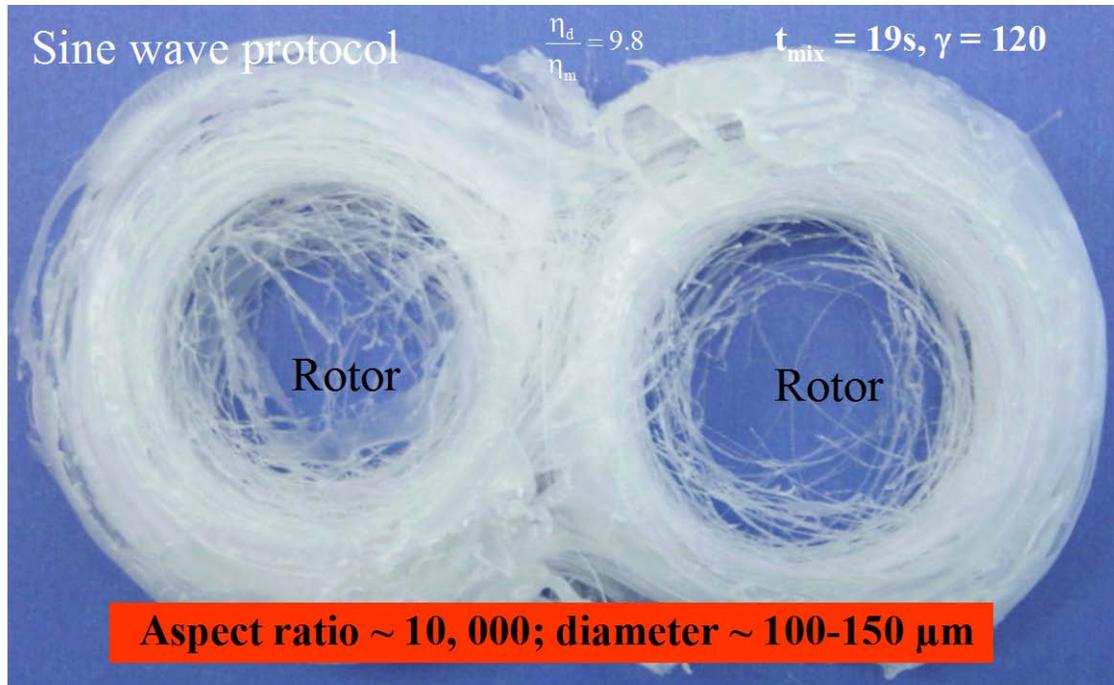
In our research chaotic mixing is produced by application of *piecewise shear flows*. Nevertheless, the overall mixing progresses at exponential rates, reminiscent of unbounded extensional flows. The repeated stretching and folding of material interfaces and alignment of material interfaces along the direction of local stretching are responsible for such exponentially rapid mixing. Other unique features of chaotic mixing are self-similarity of mixing microstructures which are produced and retained in the mixed materials. Many of these morphologies have not been seen previously in conventional blending in single and twin-screw extruders. Some wholesome attributes of chaotic mixing as highlighted by our work are listed below:

(1) A minor polymer component can be rendered into high aspect ratio ( $>1,000$ ) fibrils by chaotic mixing (**Figure 1**). Such structures augment dimensional stability of the blend materials and produce electrically conductive networks by double percolation. For example, a composite of 90 parts polyamide 6 (PA6) and 10 parts polypropylene (PP) is made electrically conductive with volume electrical conductivity of  $\sim 10^{-3}$  S/cm by using only 1 wt% Ketjenblack 300J carbon black (CB). Note that typically 20-30 wt% CB are needed to make PA6 electrically conductive. In this case, primary percolation of CB occurs in the PP-phase and secondary percolation is achieved due to continuous PP fibrils containing CB particles in major phase PA-6. Note, however, that such electrically conductive networks are produced only in flow direction and the blend does not conduct electricity in the thickness direction.

Publications:

1. Dharaiya, D., Jana, S.C., Lyuksyutov, S. 2006 Production of electrically conductive networks in immiscible polymer blends by chaotic mixing. *Polym. Eng. Sci.*, 46 (1), 19-28.
2. Dharaiya, D., Jana, S.C., Lyuksyutov, S. 2005 Production of conductive multiphase polymer systems via selective localization of carbon black under chaotic mixing conditions. SPE ANTEC 63, 2106-2110.

(2) The fibrillar structures in **Figure 1** break down by capillary instability upon further mixing and produce much smaller droplets compared to those found in conventional mixers and much smaller than those permitted by the equilibrium between interfacial and viscous forces. An analysis of flow kinematics in the chaotic mixer revealed negligible extensional flow component. Thus one would expect poor droplet deformation for polymer systems with viscosity ratio greater than  $\sim 4$ . However, our experiments on immiscible polymers with viscosity ratio varying between 1 and 30 show that droplet formation is possible at viscosity ratios higher than 10 due to a different pathway of breakup – *lamella to fibrils to droplets*. This is different from the well-studied droplet to fibrils to droplets pathway for breakup as in emulsions.



**Figure 1. Fibrillar morphology of PP-phase created by chaotic mixing of 10 wt% PP in PA6.**

Publications:

1. Dharaiya, D., Jana, S.C., 2005 Nanoclay-induced morphology development in chaotic mixing of immiscible polymers. *J. Polym. Sci., Part B: Physics*, 43(24), 3638-3651.
2. Perilla, J., Jana, S.C., 2005 Coalescence of immiscible polymer blends in chaotic mixers. *AIChE J.*, 51(10), 2675-2685.
3. Perilla, J., Jana, S.C., 2004 A time-scale approach for analysis of coalescence in polymer processing flows. *Polym. Eng. Sci.*, 44(12), 2254-2265.
4. Jana, S.C., Sau, M. 2004 Effects of viscosity ratio and composition on development of morphology in chaotic mixing of polymers. *Polymer*, 45(5), 1665-1678.
5. Sau, M., Jana, S.C. 2004 Effect of waveforms on morphology development in chaotic mixing of polymers. *AIChE J.*, 50(10), 2346-2358.
6. Sau, M., Jana, S.C. 2004 A study on the effects of chaotic mixer design and operating conditions on morphology development in immiscible polymer systems. *Polym. Eng. Sci.*, 44(3), 407-422.  
*Errata* "A study on the effects of chaotic mixer design and operating conditions on morphology development in immiscible polymer systems". *Polym. Eng. Sci.*, 44(7), 1403.
7. Sau, M., Jana, S.C. 2003 A study on the effects of chaotic mixer design and operating conditions on the development of morphology in immiscible polymer systems. *SPE ANTEC*, 61, 1556-1560.
8. Sau, M., Jana, S.C. 2003 Morphology development in PA6/PP system by chaotic mixing: effect of viscosity ratio and composition. *SPE ANTEC*, 61, 1508-1512.
9. Sau, M., Jana, S.C., 2002 Blending of immiscible polymer systems by chaotic mixing. *ANTEC 2002 Proceedings*, 60, 1431-1435.

10. Jana, S.C. 2002 Avenues of introducing chaotic mixing in single-screw extruders. *ANTEC 2002 Proceedings*, 60, 1436-1440.
11. Sau, M., Jana, S.C. 2002 Morphology development in immiscible polymer systems by baker's cut. *ANTEC 2002 Proceedings*, 60, 1446-1450.

(3) We found that droplets are stable against coalescence in the flow field of chaotic mixers. The chaotic trajectories of droplets deter coalescence due to frequent reorientation of the shear direction. Thus finer droplet morphologies generated by chaotic mixing are further stabilized against coarsening and coalescence due to the chaotic trajectories of the droplets. This attributes can be used in injection molding with the aid of chaos screws.

Publications:

1. Perilla, J., Jana, S.C., 2005 Coalescence of immiscible polymer blends in chaotic mixers. *AIChE J.*, 51(10), 2675-2685.
2. Perilla, J., Jana, S.C. 2004 Analysis of the effects of flow reorientation in chaotic mixing flows on coalescence in blends of polypropylene and polystyrene. *SPE ANTEC 62*, 2819-2823.