Chaotic Advection in Multi-component Melts for the Manufacture of Composite Materials

Presented to a Session on Complex Fluids
Meeting of the Southeast Section of the American Physical Society
October 20, 2011
by

D. A. (Dave) Zumbrunnen
Warren H. Owen-Duke Energy Professor

Department of Mechanical Engineering
Clemson University
zdavid@clemson.edu
864-656-5625
Dr. Hassan Aref: The father of chaotic advection and the person who most profoundly influenced my research. Smart blending was truly enabled by chaotic advection.
Aref’s blinking vortex model

Particles were tracked to give 1-d striations. I saw layers. I had an interest in layer stability as well as chaos. I wondered about multilayer stability in a melt and structural outcomes.
Contents of this seminar

- Chaotic advection and its application to controllable in situ structure formation (smart blending)
- Computational modeling and physics of in situ structure formation
- Physical properties of some novel plastic materials produced
  - Toughness
  - Electrical conductivity
  - Permeation reduction (barrier)
- Other applications (manufacturing and rheology)
- Suggested areas for further study
- Discussion

- Controlled release
- Solvent resistance
- Patterning (wood grains)
- Not covered
Acknowledgements

**Financial support:** NSF, DARPA, US Dept. of the Army, US Dept. of Commerce, 3M Company, Appleton, Kuraray America, ILC Dover, Dow Chemical Company, Pliant Corporation


*Lattice Boltzmann modeling that is presented in this talk was performed by Abhijit Joshi in conjunction with his doctoral studies.*

**Post-doctoral research associates:** R. Danescu, O. Kwon, B. Kulshreshtha, X. Jin
**Co-extrusion**: To put together only in one manner.

**Mixing**: To put together indiscriminately or confusedly
(Ref: American College Dictionary).

**Observation**: Plastics that combine different polymer types or polymers and solids are either co-extruded or mixed. Structural outcomes are limited and are not necessarily associated with optimal results.
Progressive structure (morphology) development

Processing conditions where fine-scale shapes among melt components or arrangements among solid additives are formed progressively in situ. A variety of polymer blend morphologies are obtained via sequential morphology transitions.

*Chaotic advection is enabling to progressive structure development and thereby smart blending.*
Chaotic advection: What is it?

**Chaotic motion of passive particles in a fluid.**

What is its relevance to smart blending?

The collective chaotic motions of melt domains cause successive shear deformations and reorientations. A layered morphology first arises that may lead to other morphologies. Morphology development can be controlled.

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Characteristics of chaotic advection

1: Stretching and folding

Multilayers in plastics

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Characteristics of chaotic advection

2: Sensitivity to initial conditions

Initial Particle Cluster

Electrically conducting plastics

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Early example using batch chaotic advection blender of incipient development of a PE layer in a PS matrix due to stretching and folding of both melt components about one another.

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Laboratory smart blender test bed

Various dies can be installed to produce structured materials of essentially any shape.

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Smart blender operation  

*On-line* control of in situ structure formation

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Progressive structure development in PP/LDPE 70/30% blends

Droplets finally result such as obtaining by conventional processing.

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Structured fibers (Structure in extruded fibers is tunable on-line.)

Internal fiber in fiber

Internal fiber and fiber imprints in matrix revealed upon fracture

Cryogenic fracture of monofilament exposing internal fibers having internal fibers (novel hierarchical structure)
Experimental examinations have pointed to the central importance of rupture formation and growth in multi-layers to structural changes.

PP/ 20% EPDM, smart blender
Shape of the hole interface resembles the stable catenoid shape, but does not have fixed edges.

Soap film between two rings forms a stable catenoid surface under certain conditions.

- A single hole formed in a fluid layer can **NEVER BE IN STABLE EQUILIBRIUM**.
- The hole either grows or shrinks.
- How and why do holes form in thin layers?
Conjoining pressure causes instability  (Kheshgi & Scriven, 1991)

\[ P = \frac{A}{h^3} \]

- \( P \) = conjoining pressure
- \( h \) = layer thickness
- \( A \) = Hamaker constant
Instabilities in freely standing fluid layers

- Can be unstable
- Usually stable

Interfacial tension is a restoring force and promotes flatness.
Van der Waals force is the destabilizing agent and can lead to rupture.
A single hole can grow if the hole diameter $d$ is greater than twice the layer thickness $h_1$. Interfacial force $F_2$ dominates $F_1$ under this condition.
Local modeling by Lattice Boltzmann Method (LBM)

3D analog to more familiar dewetting of a surface by a liquid layer

- Growth in ruptures of multi-layers occurs in tandem with melt redistribution and morphology changes.
Fiber formation at low minor component compositions (single component continuity)

At low compositions, little layer interaction occurs and fibers emerge from rupture growth and coalescence.
Formation of interpenetrating blend morphology (dual component continuity)

At intermediate compositions, layer interaction occurs as ruptures grow to give sponge-like blend morphologies.

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Formation of interpenetrating blend morphology with initial random rupture locations
Formation of sieve-like structures and small droplets

Droplet diameter is related to the parent layer thicknesses so very small diameters can result.

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Impact toughness improvements in PP/EPDM 80/20 blends

Interconnected multilayer morphology is shown subsequent to cryogenic fracture and solvent removal of EPDM. Coalesced layers via ruptures increase toughness.

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Impact toughness improvements in polypropylene

In addition to toughness increases, the impact failure of PP-EPDM films was qualitatively changed by blend morphology. Crack propagation was suppressed.

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Electrically conducting films with addition of carbon black powder

Optical micrographs of translucent sub-micron thick sections taken from extruded films with addition of 3% by weight carbon black. Interconnectivity of carbon black is controllable to impart distinct electrical properties.
Electrically conducting films with addition of carbon black powder

Resistivities pertain to the extrusion (or machine) direction for various overall carbon black compositions. Electrical properties can be selected or tuned via on-line process control.

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Multilayer nanocomposites with silica platelets

Permeant pathways are reduced or blocked.

Inefficient structure: unoriented and randomly located platelets obtained by conventional blending.

Effective placement and alignment of platelets in multi-layers.
Multilayer nanocomposites with silica platelets

Recursive horseshoe mappings (or baker’s transformations), platelets become oriented within the layers that contain them which also decrease in thickness to nano-scales.

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Novel structured nanocomposites

- Oriented platelets
- Platelets localized in numerous layers
- Platelet-rich and virgin polymer layers have nano-scale thicknesses $\ell$ and $L$.
- Hierarchical structure
- Structure can be tailored for outcome.
“Ultra-dispersion” of nano-platelets (TEM image)

Chaotic advection can be continued until layer thicknesses are thinner than the nano-platelet dimensions. The result is an ultra-dispersion where the nano-platelets are shown obliquely.

Can you improve on this dispersion by moving a nano-platelet?
In situ structuring rheometer
(Chaotic advection applied to rheology)

Applications: Rheological properties of polymer blends, nanocomposites, mixtures, biological fluids, percolating networks in melts........

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Industrial smart blenders
(Cast film or sheet, general purpose)

Research has resulted in industrial smart blenders that expand greatly the variety and properties of plastic products producible. Various dies can be attached.

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Suggested areas of further study:

1. Chaotic advection in multicomponent flows with interfacial effects
2. Multiscale (i.e., time and size) models of in situ structure formation
3. Mechanisms for hole formation and interactive growth in multilayer melts
4. Further clarifications of progressive structure development and morphology transitions
5. Viscoelastic and shear thinning effects on progressive structure development and chaotic advection
6. Morphology changes in extrusion steps
7. Properties of structured nanocomposites
8. Utilization as a rheological tool