Diffuse Interface Field Approach (DIFA) to Modeling and Simulation of Particle-based Materials Processes

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Motivation

- Extend **phase field method** to model free-body solid particles

Outline

- Interesting and important issues in particle processes
  - Moving particles of **arbitrary shapes** and sizes in **close distance**: rigid-body translations and rotations
  - Short-range forces: mechanical contact, friction, cohesion, steric repulsion, Stokes drag (particle shape matters)
  - Long-range forces: electric charge, charge heterogeneity, electric double layer, electric/magnetic dipole, van der Waals (point-charge/point-dipole approximation inaccurate)
  - External forces: electric/magnetic field, gravity (field-directed self-assembly)
  - Multi-phase liquid: fluid interface evolution, capillary force on particles (surface tension, Laplace pressure via Gibbs-Duhem relation)
Model Formulation

- Diffuse interface field description: arbitrary particle shape, continuous motion on discrete computational grids, as desired for dynamic simulation

(a) diffuse interface field function on discrete computational grids

(b) centered at $X^*=86.95$, $Y^*=64$

(c) centered at $X^*=87.50$, $Y^*=64$
Model Formulation

- Short-range forces: mechanical contact, steric repulsion

\[ dF^{sr}(r; \alpha) = \kappa \sum_{\alpha' \neq \alpha} \eta(r; \alpha) \eta(r; \alpha') \left[ \nabla \eta(r; \alpha) - \nabla \eta(r; \alpha') \right] d^3r \]

action-reaction symmetry

\[ F^{sr}(\alpha) = \int_V dF^{sr}(r; \alpha) \]

soft-particle potential

\[ T^{sr}(\alpha) = \int_V \left[ r - r^c(\alpha) \right] \times dF^{sr}(r; \alpha) \]

torque

Arbitrary particle shapes without tracking interfaces

\[ F^{sr} = A \exp \left[ -B \left( \frac{r^*}{C} - 1 \right) \right] \]
Model Formulation

- Total force and torque acting on individual particle
  \[ \mathbf{F}(\alpha) = \mathbf{F}^{\text{sr}}(\alpha) + \xi^f(\alpha) \]
  \[ \mathbf{T}(\alpha) = \mathbf{T}^{\text{sr}}(\alpha) + \xi^t(\alpha) \]
  thermal noise for Brownian motion

- Particle dynamics in viscous liquid
  \[ \mathbf{V}_i(\alpha) = M_{ij}(\alpha) \mathbf{F}_j(\alpha) \]
  \[ \Omega_i(\alpha) = N_{ij}(\alpha) \mathbf{T}_j(\alpha) \]
  small Reynolds number \( \text{Re}<<1 \), Stokes drag (friction), mobility

- Equation of motion
  \[ \eta(\mathbf{r},t;\alpha) = \eta(\mathbf{r}^0,t_0;\alpha) \]
  \[ r_i = Q_{ij}(t;\alpha) \left[ r_j^0 - r_j^c(t_0;\alpha) \right] + r_i^c(t;\alpha) \]
  \[ r_i^c(t + dt;\alpha) = r_i^c(t;\alpha) + \mathbf{V}_i(t;\alpha)dt \]
  mapping without error accumulation
  \[ Q_{ij}(t + dt;\alpha) = R_{ik}(t;\alpha) Q_{kj}(t;\alpha) \]
  translation
  \[ R_{ij}(t;\alpha) = \delta_{ij} \cos \omega + m_i m_j (1 - \cos \omega) - \varepsilon_{ijk} m_k \sin \omega \]
  rotation
  incremental rotation
Simulation

- Particle sedimentation and stacking
Simulation

- Particle sedimentation and stacking
Simulation

- Phase field model of solid-state sintering: rigid-body motions
Model Formulation

- Long-range force: charged particles

\[
\rho(r, t; \alpha) = \rho(\alpha) \eta(r, t; \alpha)
\]

\[
\rho(r, t; \alpha) = \rho(\alpha) \eta(r, t; \alpha) \left[ 1 - \eta(r, t; \alpha) \right]
\]

\[
\rho(r, t) = \sum_{\alpha} \rho(r, t; \alpha)
\]

\[
E(r) = E^{\text{ex}} - \frac{i}{\varepsilon_0} \int \frac{d^3 k}{(2\pi)^3} \frac{\hat{\rho}(k)}{k} n e^{i k \cdot r}
\]

\[
F^{\text{el}}(\alpha) = \int_V E(r) \rho(r; \alpha) \, d^3 r
\]

\[
T^{\text{el}}(\alpha) = \int_V \left[ r - r^c(\alpha) \right] \times E(r) \rho(r; \alpha) \, d^3 r
\]
Model Formulation

- Long-range force: charged particles

\[
\rho(r,t;\alpha) = \rho(\alpha) \eta(r,t;\alpha)
\]

\[
\rho(r,t;\alpha) = \rho(\alpha) \eta(r,t;\alpha) \left[ 1 - \eta(r,t;\alpha) \right]
\]

body charge

\[
\rho(r,t) = \sum_{\alpha} \rho(r,t;\alpha)
\]

Surface charge

\[
E(r) = E^{\text{ex}} - \frac{i}{\varepsilon_0} \int \frac{d^3k}{(2\pi)^3} \frac{\tilde{\rho}(k)}{k} n e^{i k \cdot r}
\]

\[
F^{\text{el}}(\alpha) = \int_V E(r) \rho(r;\alpha) d^3r
\]

\[
T^{\text{el}}(\alpha) = \int_V \left[ r - r^c(\alpha) \right] \times E(r) \rho(r;\alpha) d^3r
\]

- Total force and torque acting on individual particle

\[
F(\alpha) = F^{\text{el}}(\alpha) + F^{\text{sr}}(\alpha) + \xi^f(\alpha)
\]

\[
T(\alpha) = T^{\text{el}}(\alpha) + T^{\text{sr}}(\alpha) + \xi^t(\alpha)
\]
Particles of same charge: repulsion
Particles of opposite charges: attractive self-assembly

Simulation

Mutually induced dipoles

dipolar

stable

$N^-:N^+ = 1:1$
$\rho^-:\rho^+ = 1:1$

$N^-:N^+ = 1:2$
$\rho^-:\rho^+ = 2:1$

$N^-:N^+ = 1:3$
$\rho^-:\rho^+ = 3:1$

$N^-:N^+ = 1:4$
$\rho^-:\rho^+ = 4:1$

$N^-:N^+ = 1:5$
$\rho^-:\rho^+ = 5:1$
Self-Assembly Mechanisms

- $\rho_{\text{surf}} = -2.0$
- $\rho_{\text{surf}} = +1.0$
- $N^-:N^+ = 1:2$

1. neutral & symmetric
2. induced dipole
3. attraction
4. repeated growth & dipolar

Neutral chain formation

Attractor region

Growth stopper
Self-Assembly Mechanisms

- \( \rho_{\text{surf}} = -2.0 \)
- \( \rho_{\text{surf}} = +1.0 \)
- \( N^-:N^+ = 1:1 \)

1. charged & dipole
2. alignment
3. attraction
4. repeated growth & charged chain formation,
   mutually repulsive,
   as straight as possible

repel at long distance
attract at short distance

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Simulation

- Particles of opposite charges: non-spherical shapes
Stacking of charged particles under external fields

Simulation
Model Formulation

- Long-range force: dipolar particles

\[ \mathbf{P}(\mathbf{r}, t; \alpha) = \mathbf{P}(t; \alpha) \eta(\mathbf{r}, t; \alpha) \]

\[ \mathbf{P}(\mathbf{r}, t) = \sum_{\alpha} \mathbf{P}(\mathbf{r}, t; \alpha) \]

\[ \mathbf{E}(\mathbf{r}) = \mathbf{E}^\text{ex} - \frac{1}{\varepsilon_0} \int \frac{d^3 k}{(2\pi)^3} \left[ \mathbf{n} \cdot \mathbf{\tilde{P}}(\mathbf{k}) \right] \mathbf{n} e^{ik \cdot \mathbf{r}} \]

\[ \mathbf{F}^\text{el}(\alpha) = \int_V \left[ \mathbf{P}(\alpha) \cdot \nabla \right] \mathbf{E}(\mathbf{r}) \eta(\mathbf{r}; \alpha) \, d^3 r \]

\[ \mathbf{T}^\text{el}(\alpha) = \int_V \mathbf{P}(\alpha) \times \mathbf{E}(\mathbf{r}) \eta(\mathbf{r}; \alpha) \, d^3 r \]

\[ + \int_V \left[ \mathbf{r} - \mathbf{r}^c(\alpha) \right] \times \left\{ \left[ \mathbf{P}(\alpha) \cdot \nabla \right] \mathbf{E}(\mathbf{r}) \right\} \eta(\mathbf{r}; \alpha) \, d^3 r \]
Model Formulation

- **Long-range force: dipolar particles**
  
  \[ P(r,t;\alpha) = P(t;\alpha) \eta(r,t;\alpha) \]
  
  \[ P(r,t) = \sum_\alpha P(r,t;\alpha) \]
  
  \[ E(r) = E^{ex} - \frac{1}{\varepsilon_0} \int \frac{d^3k}{(2\pi)^3} \left[ n \cdot \tilde{P}(k) \right] n e^{ik \cdot r} \]
  
  \[ F^{el}(\alpha) = \int_V \left[ P(\alpha) \cdot \nabla \right] E(r) \eta(r;\alpha) \, d^3r \]
  
  \[ T^{el}(\alpha) = \int_V P(\alpha) \times E(r) \eta(r;\alpha) \, d^3r \]
  
  \[ + \int_V \left[ r - r^c(\alpha) \right] \times \left\{ \left[ P(\alpha) \cdot \nabla \right] E(r) \right\} \eta(r;\alpha) \, d^3r \]

- **Long-range force: magnetic particles**

  \[ M(r,t) = \sum_\alpha M(t;\alpha) \eta(r,t;\alpha) \]
  
  \[ H(r) = H^{ex} - \int \frac{d^3k}{(2\pi)^3} \left[ n \cdot \tilde{M}(k) \right] n e^{ik \cdot r} \]
  
  \[ F^{mag}(\alpha) = \mu_0 \int_V \left[ M(\alpha) \cdot \nabla \right] H(r) \eta(r;\alpha) \, d^3r \]
  
  \[ T^{mag}(\alpha) = \mu_0 \int_V M(\alpha) \times H(r) \eta(r;\alpha) \, d^3r + \mu_0 \int_V \left[ r - r^c(\alpha) \right] \times \left\{ \left[ M(\alpha) \cdot \nabla \right] H(r) \right\} \eta(r;\alpha) \, d^3r \]
Simulation

- Dipolar particles: agglomeration
Self-Assembly of Arbitrary-Shaped Dipolar Particles
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Simulation

- Dipolar particles: *field-directed self-assembly*
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Processing-Microstructure Relationship

Mechanisms of Filler Particle Self-Assembly

- Strongly anisotropic force that can be tuned by external field
- Rigid-body motion (translation and rotation) of colloidal particles in liquids (water, organic solvent, polymer melt, etc.)
Simulation

- Phase field model of dielectric/magnetic composites

![Simulation Diagram]

- Directed particle self-assembly process during composite fabrication

![Matrix]

\[
\chi^{\text{eff}} = \chi_{\text{mat}} \begin{bmatrix}
\chi_{xx}^{\text{eff}} & \chi_{xy}^{\text{eff}} \\
\chi_{yx}^{\text{eff}} & \chi_{yy}^{\text{eff}}
\end{bmatrix}
\]

Effective susceptibility tensor

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Particle-Filled Polymer-Matrix Composites

- Alignment of irregular-shaped functional filler particles

- **Dielectric:** PZT fillers
- **Electro-Optic:** PbTiO$_3$ nanoparticles
- **Magnetostrictive:** Terfenol-D particles

*Or et al., J. Appl. Phys., 93, 8510, 2003.*
Model Formulation

Particles in multi-phase liquid: capillary forces

\[ F = \int \left[ f \left( \{c_\alpha\}, \{\eta_\beta\} \right) + \sum_\alpha \frac{1}{2} \kappa_\alpha |\nabla c_\alpha|^2 \right] dV \]

Landau polynomial

\[ f\left( \{c_\alpha\}, \{\eta_\beta\} \right) = A \left[ \sum_{\alpha=1}^{2} \left( 3c_\alpha^4 - 4c_\alpha^3 \right) + \sum_\beta \left( 3\eta_\beta^4 - 4\eta_\beta^3 \right) + 6 \left( \chi c_1^2 c_2^2 + \sum_{\beta} \sum_{\alpha=1}^{2} \lambda_\alpha c_\alpha^2 \eta_\beta^2 \right) \right] \]

Cahn-Hilliard

\[ \frac{\partial c_\alpha}{\partial t} = \nabla \cdot \left( M_\alpha \nabla \frac{\delta F}{\delta c_\alpha} \right) \]

Gibbs-Duhem

\[ dp = c_A d\mu_A + c_B d\mu_B \]

\[ p - p^0 = c_1 \mu_1 + c_2 \mu_2 \]

\[ \mu_\alpha = \frac{\partial f}{\partial c_\alpha} \]

Young-Laplace

\[ \Delta p = \frac{\gamma}{R} \]
Model Formulation

Particles in multi-phase liquid: capillary forces

Laplace pressure

\[ d \mathbf{F}^{LP}(r, \beta) = \kappa_p \nabla \eta(r, \beta) p(r) dV \]

Interfacial tension

\[ d \mathbf{F}^{IT}(\beta) = \kappa_T \left[ \nabla c \times (\nabla c \times \nabla \eta_\beta) \right] dV \]

\[ = \kappa_T \left[ (\nabla c \cdot \nabla \eta_\beta) \nabla c - |\nabla c|^2 \nabla \eta_\beta \right] dV \]

\[ \nabla c = c_1 c_2 \left( \nabla c_1 - \nabla c_2 \right) \]
Simulation

Irregular-shaped particle at curved fluid interface

(a)  
(b)  
(c)  

(a)  
(b)  
(c)  
(d)
Simulation

- Particle self-assembly directed by fluid interface: encapsulation

**Simulation**

- **Negative Pressure**
  - $t^* = 0$
  - $t^* = 3 \times 10^5$
  - $t^* = 6 \times 10^5$
  - $t^* = 1 \times 10^6$

- **Positive Pressure**
  - $t^* = 0$
  - $t^* = 1 \times 10^5$
  - $t^* = 5 \times 10^5$
  - $t^* = 1 \times 10^6$

- **Zero Pressure**
  - $t^* = 10^4$
  - $t^* = 10^5$
  - $t^* = 10^6$
Simulation

- Bijel: bicontinuous interfacially jammed emulsion gels

(a) $t^* = 4,000$
(b) $t^* = 20,000$
(c) $t^* = 40,000$
(d) $t^* = 60,000$
(e) $t^* = 200,000$
(f) $t^* = 400,000$
Simulation

- Bijel: bicontinuous interfacially jammed emulsion gels

(a) $t^* = 60,000$

(b) $t^* = 200,000$
Simulation

Capillary bridges for in-situ firming of colloidal crystals

100 nm, 10,000 Pa
Simulation

- Capillary bridges for in-situ firming of colloidal crystals

Time step=10,000 | Time step=50,000 | Time step=800,000

Time step=10,000 | Time step=50,000 | Time step=800,000
Acknowledgement

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