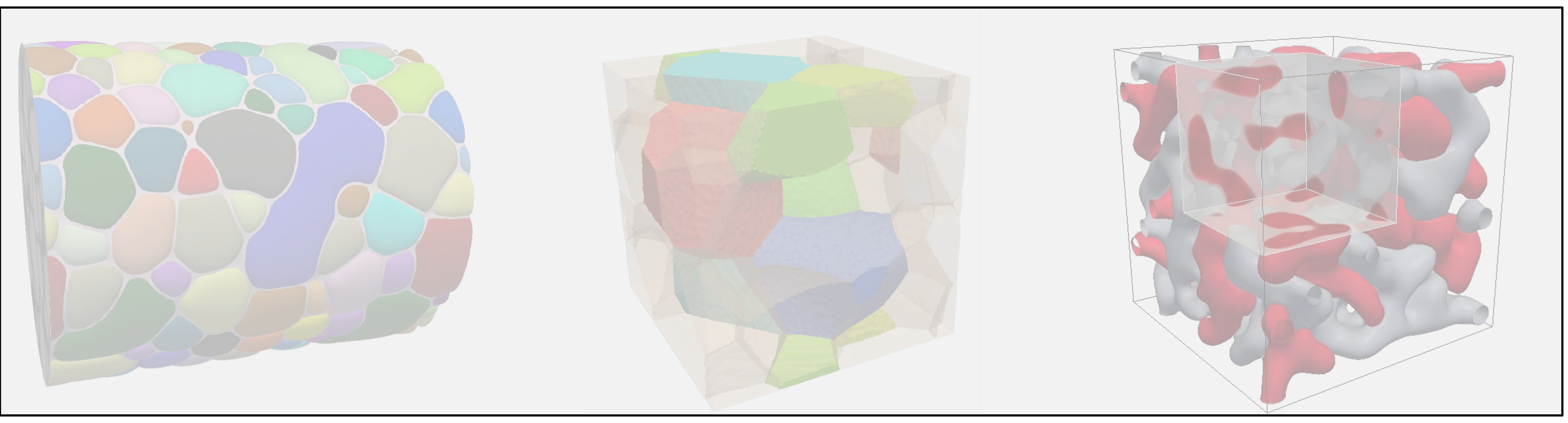
Exceptional service in the national interest



Mesoscale Modeling of Materials Microstructures

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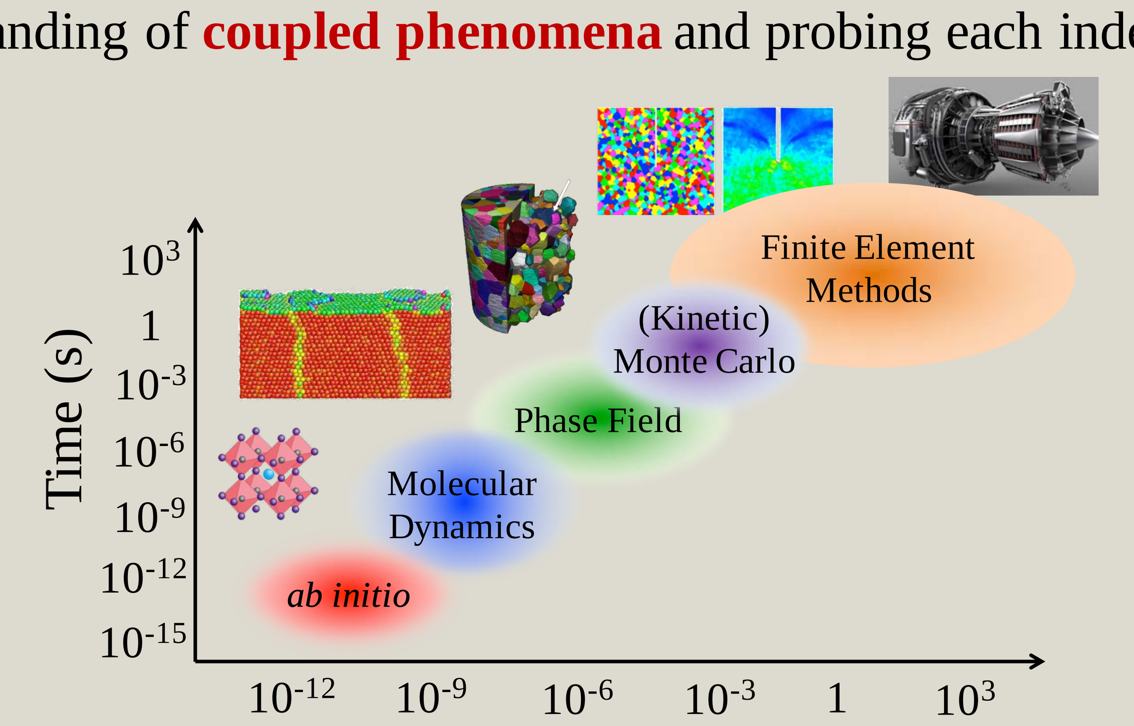
The Role of Microstructure

(Microstructure, composition are at the origins of degradation)

- Heterogeneous materials are the forefront of next generation structural, electrical and energy applications
- Observed materials properties are the manifestation of evolving morphologies and associated **features at multiple scales**
- Theoretical and computational models help in gaining an understanding of **coupled phenomena** and probing each independently

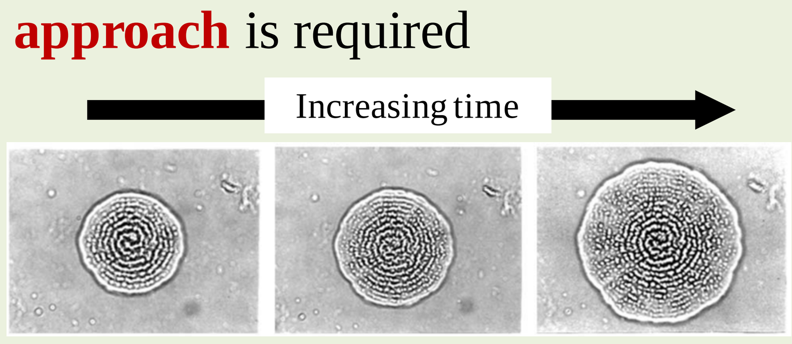


Evolving Morphologies in Polymers

(The root of aging and degradation)

- In PV polymeric systems, thermo-physico-chemical-environmental factors influence phase stability and several non-equilibrium processes, such as phase separation, self assembly and crystallization
- The structural and morphological landscape is greatly influenced by such processes, which in turn affects **processability characteristics** and **functional properties**
- In order to understand and predict the stability of morphologies and their time evolution, a **mesoscopic approach** is required

Ex. 1: Optical micrograph showing the time sequence of the evolution of spiral spherulite undergoing breakup of the spiral arms in a blend of PVDF/EVA during isothermal crystallization at 170 ºC [From Xu *et al.*, Phys. Rev. E (2006)]



Spherulite crystallization of PVDF/EVA blend

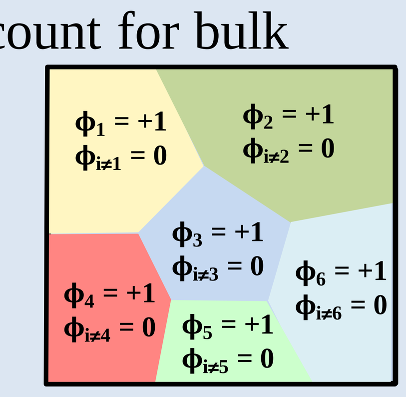
Ex. 2: Microscopy observation of phase separation into SBS- and bitumen-rich domains during isothermal annealing at 180 °C [From Zhu *et al.*, Materials & Design (2016)]

Phase separation in polymer modified bitumen (PMB)

Phase Field (PF) Frameworks

- PF (or Ginzburg-Landau) is a **mesoscale** framework that incorporates both atomic scale information and evolving microstructures
- Order parameters (OPs) are defined (concentrations, phases, mass density, …)
- A coarse-grained energy functional is proposed in terms of the OPs to account for bulk thermodynamics and interfacial attributes

$$\mathcal{F}_{tot}[c,T] = \int d\mathbf{r}\,[f_{bulk}(c,\phi_i,T) + f_{int}(|\nabla c|,|\nabla \phi_i|)]$$



- Dynamics are driven by minimization of the energy

$$\frac{\partial c}{\partial t} = \nabla \cdot \left[M\nabla\left(\frac{\delta \mathcal{F}_{tot}}{\delta c}\right)\right]$$ Conserved Cahn-Hilliard Eq.

$$\frac{\partial \phi_i}{\partial t} = -L_i\left(\frac{\delta \mathcal{F}_{tot}}{\delta \phi_i}\right)$$ Non-conserved Allen-Cahn Eq.

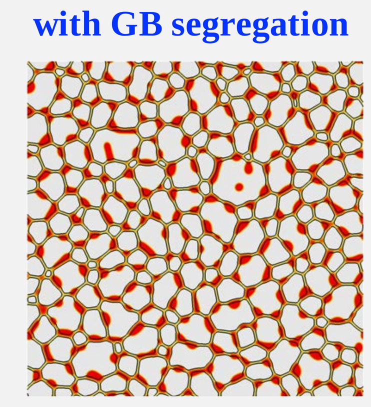
- As inputs, PF leverages **atomistic information** for the construction of the free energy functional along with the kinetic coefficients (diffusivity of species, interface mobility, anisotropies, attachment kinetics, …) ➔ Collaboration with **NREL**

Nanocrystalline Materials

- Nanocrystalline materials (NCMs) exhibit a unique combination of properties
- NCMs are unstable against homogenization (grain growth) processes at low temperatures
- Grain boundary (GB) solute segregation has been proposed to stabilize NCMs

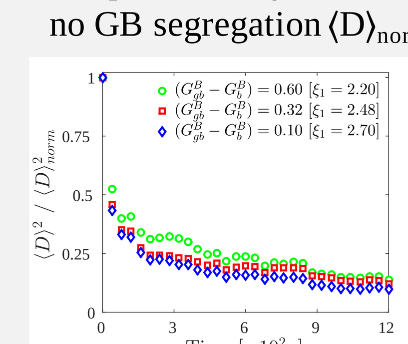
No GB segregation

with GB segregation

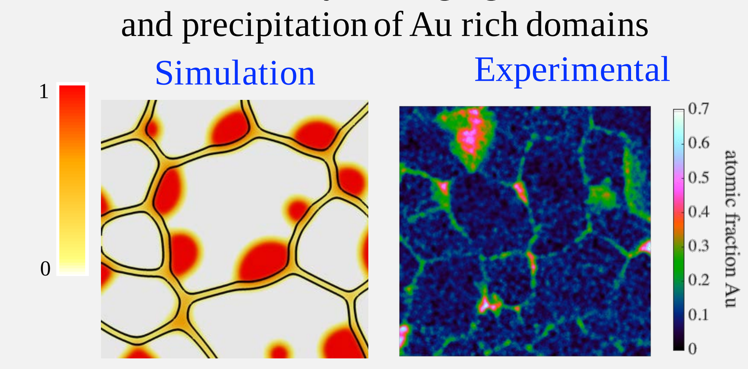


Abdeljawad and Foiles, Acta Mater. (2015)
Abdeljawad *et al.*, Acta Mater (under review) (2016)

Average grain diameter ⟨D⟩ compared to a system with no GB segregation ⟨D⟩$_{norm}$



For a Pt-Au alloy, Au segregation to GBs and precipitation of Au rich domains



Argibay *et al.*, Submitted to Nature Mater. (2016)

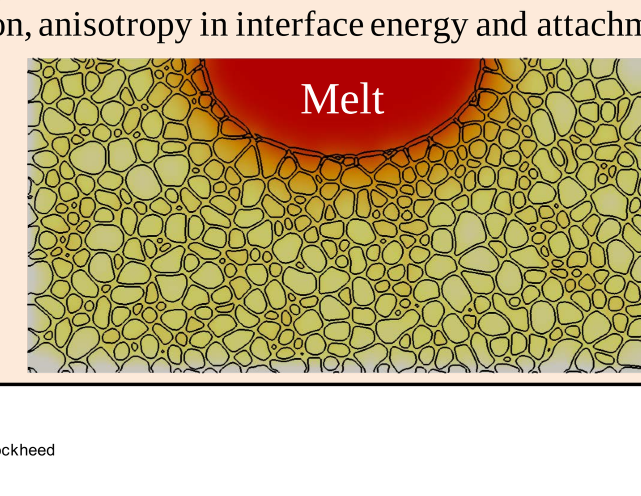
Additively Manufactured (AM) Materials

(Degradation mechanisms can often be traced to manufacturing)

- AM techniques rely on making objects by building the material layer upon layer
- AM involves coupled processes, such as solidification, sintering, thermal and fluid flow
- The ability to predict evolving microstructures in AM is key to determining **properties**

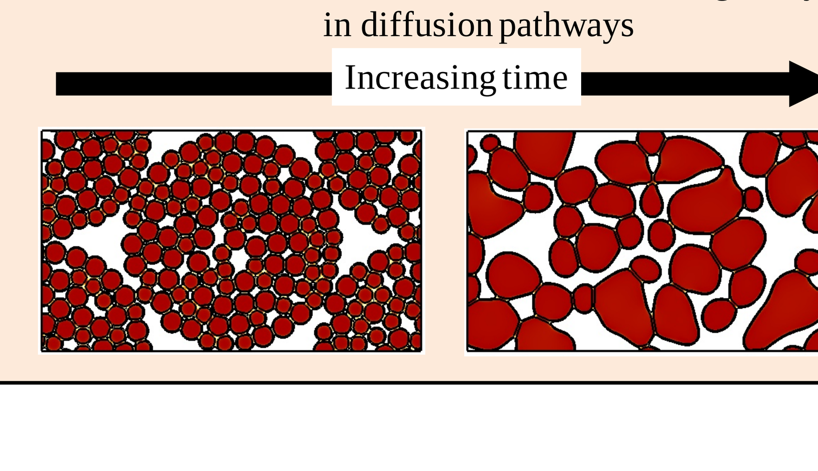
Solidification from melt

Coupled microstructure, thermal diffusion and latent heat generation, anisotropy in interface energy and attachment kinetics



Ceramic sintering

Grain boundaries, free surfaces, inhomogeneity in diffusion pathways



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