Bridging atomistic to continuum scales for designing energy materials: challenges and opportunities

Sreekanth Pannala
Senior Research Staff Member
Computer Science and Mathematics Division
Oak Ridge National Laboratory

pannalas@ornl.gov

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Energy Trends

If we want to change these trends, it is critical to make key investments today and simulations can/should play a big role.

Source: Annual Energy Outlook 2008, Early Release
Outline

• Introduce the multiscale/multiphysics problems in energy materials (fuel, extraction and efficiency) where bridging atomistic to continuum scales is needed for predictive simulations
  – Nuclear fuel coating process
  – Coal/Biomass gasification
  – Catalysts for efficient combustion systems – chemical looping
  – Carbon nanotubes and nanofibers
  – Thin film Si deposition on powders and Si production

• Current set of models used at various scales

• Need for multiphysics coupling

• Compound wavelet matrix method (CWM), dynamic CWM, time parallel CWM

• Future work, opportunities and challenges?
Nuclear fuel coating process

Coated fuel particle (small scale)
- Si-C
- Amorphous C
- Kernel
- Inner Pyrolitic C

Spouted bed coater (device scale)
- Ballistic zone
- Transport reaction zone (~10^6-10^-2s)
- Pickup zone (~10^-3 m)

- Design challenge: Maintain optimal temperatures, species, residence times in each zone to attain right microstructure of coating layers at nm scale
- Truly multiscale problem: ~O(13) time scales, ~O(8) length scales

- Coating at high temperature (1300–1500°C) in batch spouted bed reactor for ~10^4 s
- Particles cycle thru deposition and annealing zones where complex chemistry occurs

- 0.5- to 1-mm particles
- Coating encapsulates fission products
- Failure rate < 1 in 10^5
- Quality depends on surface processes at nm length scale and ns time scales

Predictive simulations need innovative multiscale algorithms for peta-/exascale computing

SP et al. (CVD, 2007)
Coal/Biomass Gasification

Coal Particle (small scale)
- ~ mm particles
- Complex flow: gas phase, gas phase in char, pyrolysis front, unreacted coal
- Wide range of species
- Surface processes at nm length scale and ns time scales

Coal Gasifier (device scale)
- ~ m in size
- Gasification at high temperatures (1000–2000°C) in reactor with large residence times ~100 s
- Coal particles cycle thru wide range of conditions where complex chemistry occurs

Design challenge:
Maintain optimal temperatures, species, residence times in each zone to attain right gasification

Truly multiscale problem:
~O(13) time scales, ~O(10) length scales

Materials challenge:
Design/understand material properties for the biomass/coal pellets/particles at μm/nm scale
  - Size
  - Porosity
  - Integrity
  - Composition
  - Binders?
  - Mix of biomass and coal

SP et al. (CFD in CRE, 2008)
**Other Applications**

Chemical Looping Combustion

- Similar to hemoglobin in our blood
- Higher efficiency with lower entropy losses
- No thermal NOx
- Separated CO2 stream for sequestration
- Potential carbon-negative technology if used with biomass
- Challenges
  - Catalysts with fast oxidation and reduction
  - Material durability
  - Cost

**Carbon Nanotubes, Nanofibers and Nanostructures**
- Light weight and high strength
- Supercapacitors
- Challenges about bulk production with desired chirality, diameter, number of walls etc.

**Thin film Si deposition on powders and Si production**
- Modify material properties (strength, corrosion resistance, tribology etc.)
- Reduce cost for PVs

**Reactive flows through fibrous media**
- Light weight, low-cost and high-strength composites
- Fuel cell components

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Wood, SP et al. (PRB, 2007)
Multiphysics heterogeneous chemically reacting flows for energy systems

Goal: Building a suite of models for unprecedented capability to simulate multiphase flow reactors

• Through support from various DOE offices (FE, EERE, and NE) we have developed suite of models for unprecedented capability to simulate heterogeneous chemically reacting flows

• Hybrid methods to couple two physical models (e.g. MFIX DEM)
Flow over a catalyst surface

- Chemically reactive flow over a surface is a basic building block that is central to many energy-related applications.
- Illustrative benchmark to demonstrate the capability to integrate scales of several orders of magnitude.

Figure adapted from Succi et al., Computing in Science and Engineering, 3(6), 26, 2001.
So what needs to be done for multiphysics coupling?

- Can we rewrite the equations or the solution methods so that only relevant information is propagated upward from fine- to coarse-scales (upscaling) and coarse- to fine-scales (downscaling) in a tightly coupled fashion?
  - New mathematics, theory and analysis
  - Unification of governing equations across several scales
    - Lattice based methods across all scales?
  - Long-term

- If that is not possible, can we take the information from different methods and perform this in a post priori fashion with various degrees of coupling?
  - Interpolation and extrapolation between the regions
    - Typical multiphysics coupling approach: FSI, BEM-Level Set, Inviscid/Viscous-BL for external aerodynamics, fluid-particle/droplet etc.
    - Usually invoked as a boundary condition or a source term
    - This is done almost implicitly in various methods we already use: grid stretching, multiblock, AMR, Adaptive basis
  - One can do better by transferring higher order statistics rather than just averages
    - Use stochastic processes to transfer the information
      - Use UO process to drive a stationary isotropic turbulence problem
General Problem Definition

\[ \dot{y}_c = g(x, t, y_c), \quad y_c(x, 0) = y_0(x), \quad x \in \mathcal{B}, \quad t \in [0, T] \]
\[ \dot{y}_f = f(x, t, y_f), \quad y_f(x, 0) = y_0(x), \quad x \in \mathcal{B}, \quad t \in [0, T]. \]

where \( g \) describes the coarse field, \( f \) describes the fine field

- We seek solution of the form

\[ y(x, t) = \Phi [y_c(x, t), y_f(x, t)] \]

- Map takes the solution of the coarse-field over the entire domain and the fine-field over a subset of the domain to obtain a good approximation to \( y_f \).

- The algorithms should be amenable to parallel implementation in both space and time
CWM (Compound Wavelet Matrix) and dCWM (dynamic CWM) Algorithms

![CWM](image)

1: Given: \( y_c(t) \) and \( y_f(\tau) \) with \( t \in [0, T] \) and \( \tau \in [0, T_f] \)
2: Compute wavelet transforms: \( y^W_c = \mathcal{W}[y_c(t)] \) and \( y^W_f = \mathcal{W}[y_f(\tau)] \)
3: Apply window filter: \( y^{H \circ W}_c = \mathcal{H}_{(a, b)}[y^W_c] \) and \( y^{H \circ W}_f = \mathcal{H}_{(b, c)}[y^W_f] \)
4: Compute CWM: \( y_{CWM} = y^{H \circ W}_c \oplus y^{H \circ W}_f \)
5: Compute inverse wavelet transform: \( y(t) = \mathcal{W}^{-1}[y_{CWM}] \)

![dCWM](image)

1: \textbf{for} \( n = 0 \) to \( N - 1 \) \textbf{do}
2: Given: \( y_c(t) \) and \( y_f(\tau) \) with \( t \in [T_n, T_{n+1}] \) and \( \tau \in [T_n, T_n + \delta T_f] \)
3: Compute wavelet transforms: \( y^W_c = \mathcal{W}[y_c(t)] \) and \( y^W_f = \mathcal{W}[y_f(\tau)] \)
4: Apply window filter: \( y^{H \circ W}_c = \mathcal{H}_{(a, b)}[y^W_c] \) and \( y^{H \circ W}_f = \mathcal{H}_{(b, c)}[y^W_f] \)
5: Compute dCWM: \( y_{dCWM} = y^{H \circ W}_c \oplus y^{H \circ W}_f \)
6: Compute inverse wavelet transform: \( y(t) = \mathcal{W}^{-1}[y_{dCWM}] \)
7: \textbf{end for}

SP with Frantziskonis et al. (IJMCE, 2006)  SP with Mishra et al. (LNCS, 2008)
SP with Mishra et al. (IJCRE, 2008)  SP with Muralidharan et al. (PRE, 2008)
tpCWM (Time Parallel CWM)

Schematic of the TP and CWM methods. (a) The TP method. The fine method instantiates at several temporal “nodes” typically for a period $\delta t$ that covers time until the next node. (b) The temporal CWM. The fine method is employed for a fraction of the coarse method. (c) The CWM reconstruction updates the mean field. (d) The CWM reconstruction updates the temporal statistics.
Example: tpCWM applied to Lotka-Volterra predator-prey equations

\[
\begin{align*}
X + Y_1 & \xrightarrow{\alpha} 2Y_1 \\
Y_1 + Y_2 & \xrightarrow{\beta} 2Y_2 \\
Y_2 & \xrightarrow{\gamma} Z
\end{align*}
\]

Lotka-Volterra System

\[
\begin{align*}
\frac{dY_1}{dt} &= \alpha XY_1 - \beta Y_1 Y_2 \\
\frac{dY_2}{dt} &= \beta Y_1 Y_2 - \gamma Y_2
\end{align*}
\]

Factor of computational savings, X as a function of the ratio \( r \) (number of processors/number of iterations) and the fraction \( f \) (fraction of KMC time used in each assigned time interval).

Three orders of magnitude savings (time-to-solution) can be achieved by \( r \) in the range of 20 and \( f \) in the order of 1/64.
General Multiphysics Framework Requirements

- Flexible and adaptive
  - To handle various multiphysics codes operating at various scales in an hierarchical fashion

- Scalable

- Automatic optimization based on the architecture
  - Load balance, memory access, IO etc.

- Automated profiling options

- Automatically chose optimal physical models and numerical methods to give the required accuracy with given resources
  - Choosing scalable algorithms – these might not be the best algorithms on small set of processors

- Optimization of the code at run-time

- Ensure all conservation laws are valid at the coupling scales
  - e.g. conformance to second law of thermodynamics (entropy)

- Uncertainty propagation through various models and quantification
Opportunities

• Revolutionize the way simulation tools are used in the design process
  – Move away from the current edisonian approach
  – Design new industrial scale devices at a very short turn-around
    – Today depending on the system it can take 10-20 years from concept to lab-scale to pilot-scale to industrial-scale

• Develop new designs exploiting the efficient paths at the molecular scales
  – All reactions and processes happen at the atomic and molecular scales
  – Today the design process is totally decoupled – data is handed over from a group working at one scale to the other group at another scale in a sequential iterative process
    – Some designs are 5-6 decades old

• Develop feedback control systems to run devices in most optimal fashion
Computational Science Challenges

- Bringing a broad set of researchers working on materials related processes together to get their buy-in
  - Academia, Research labs, Industry
  - Agree on common codes, interfaces, data standards etc.

- The future architectures (with millions of cores and 100s of cores to a processor) are more conducive to locally coupled simulations
  - Many physical processes are globally coupled
  - Running multiple codes would need large and fast data movements across the processors/cores
    - Need to have smart algorithms to overlap communications and computations

- Validation and verification
  - Most validation is at steady state or subset of time-space-trajectories
    - Very difficult to get all the data required to verify all the components of the simulations
    - Considerable investments need to be made in non-intrusive experimental techniques to obtain enough data
    - March towards the integration of “Theory, Experiment and Simulation”

Coal Gasification
Courtesy: Chris Guenther, NETL
Summary

• Energy crisis is real and we need tomorrow’s technology today
• Integrated experiments and simulations at scale can revolutionize the design of energy materials and devices
  – Include all relevant scales so that molecular scale interactions are included when designing device scale
  – Cut down the current 20-30 year design cycle
  – Break cultural barriers
• Develop computations based feedback control systems to run devices in most optimal fashion
  – Adjust for feedstock etc. online rather than offline adjustments with huge safety margins
• Simulation science can and has to play a catalytic and important role in bringing innovation to the energy market place
  – Reinvigorate the economic machine
## Collaborators

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