The journey from atoms to assessed engineering

**with quantitative confidence:**

Purpose, principles, and (some) practice

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With thanks to too many to list, but a few are musts:
Carter Edwards, Randy Summers, Jim Stewart, John Aidun,
Julie Bouchard, Laura Swiler, Allen Robinson,
Dave Sassani, Louise Criscenti, Peter Zapol (ANL), Carl Steefel (LBNL)

IMA Workshop: Uncertainty Quantification in Materials Modeling
IMA, University of Minnesota, Dec. 16-17 2013
Why Uncertainty Quantification (UQ)?

What’s the point of multiscale materials simulation anyway?

To be **insightful**
   identify, characterize, and assess crucial phenomena

To be **predictive**
   move beyond forensics and pretty pictures (and papers)

To be **meaningful**
   inform decisions (technical and programmatic)

**Goal:** establish credibility, quantitative confidence, in an evidence-based approach used to inform high-consequence decisions
What kind of decisions?

- Satisfy regulatory requirements in qualified assessment
- Support design choices for safety and performance
- Identify weakness\&strengths, gaps in system analysis
- Inform resource allocation and investment strategy
- Inform technical program priorities

Credible quantitative measures are necessary

Meaningful UQ is means to establish quantitative confidence
UQ alone is not enough

- UQ is only meaningful within Verification and Validation (V&V) regime
  Verification – are you solving what you think you are solving?
  Validation – how well does what you are solving represent reality?
  Uncertainties quantifies these questions
  This also applies to experimental data!

- V&V and UQ only make sense in the context of an application
  What are you solving for, precisely?

- Objective: acceptance of M&S “product”
  (1) Internal: Process to identify and mitigate weaknesses in models
    Make decisions – set priorities, allocate efforts
  (2) External: Establish quantitative confidence in results
    Make decisions informed by credible evidence

More questions than answers – defining a process, no clear answers
Process: Perfection vs. provably good (enough)

Goal: Establishing credibility, quantitative confidence

PCMM – Predictive Capability Maturity Model (Sandia)
Graded approach, greater risks => greater rigor

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Other, similar protocols: PMI (LANL), CAS (NASA) …

Process: approach that emphasizes every component in an assessment in a way balanced to maximize confidence in final output result
Multiscale materials modeling UQ challenges

Numerical UQ methods (relatively) well developed, but …

• Each application has unique multiscale sequence
  - custom UQ system every time?

• Physics/chemistry causalities not known \textit{a priori}
  - define network of UQ assessment without causality?

• Upscaling challenge
  - electrons to atoms to chemistry to thermodynamics …
  - how to propagate UQ through scales? After calibration?
  - with undetermined roadmap to ultimate (assessed) quantities?

• Cultural
  - materials disciplines do not embrace/express UQ principles?
Goal: assess waste form and long-term waste storage system for long-term disposal of nuclear waste subject to regulatory requirements.
Follow the radionuclides

What happens at barriers?
- corrosion, cracking, sorption

Cladding/canisters/package

What happens away from surface?
- transport, colloids, 2ndary phases
  aqueous solution pH, redox, cations, anions

Glass, ceramic, metal

Barrier layers, complexes, gels

What happens at the surface?
- leaching, transport, dissolution, …

Entire scenario might need to be modeled
Lots of coupled processes, unknowns
A System with well-defined process flows

Requirements-driven (PIRT)

Qualified applications (Verified)

VV/UQ’d data flow

Traceability, Reproducibility

Performance Assessment (PA) Codes

Surrogate Models

Hi-Fi THCM Codes

Constitutive Models

Subgrid Scale:
- Phenom.
- Experiment - UQ
- ModSim - Ver.

Discovery/Exploratory studies Methods Development

痕迹, Reproducibility

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Anatomy of a model - two parts

1. a physics abstraction (e.g. coarse-grained model)
2. parameter values that populate the model

- Phenomenological models - heuristic, irreducible
  - fit and calibrated to specific range
  - interpolative, not extrapolative

- Subgrid-physics-aware models - hand-off multiscale
  - potentially extrapolative (predictive)
  - experimental or mod-sim data
  - there is still fitting (calibration)

This duality a formal (and unsolved) challenge for UQ
Upscaling - “A miracle happens”

Two challenges:
1) accuracy of parameters
2) adequacy of physics abstractions (model form)

Model parameters usually refit (calibrated) for validation of application
  - Internal consistency (e.g. thermodynamic)
  - Model incompleteness

Physics abstraction mixes parameters (dependencies)
and may not be (quantitatively) reducible to sub-scale processes

Bridging this divide is unsolved challenge (both scientifically and UQ)
... more failures than successes (science), unclear process (UQ)
Requirements triage

UQ encapsulates:
- state of knowledge
- needs

UQ will determine:
- requirements
- priorities

Triage will determine flow of requirements
Constitutive Models

- Phenom.
- Experiment - UQ
- ModSim - Ver.

Req.,SA

UQ, Val.

Subgrid Scale:

Into high-consequence decisions: All data/process flows must be …

Traceable - provenance recorded
Reproducible - by others
With quantified confidence - VV/UQ

Required elements from all simulations feeding the system:

1) Necessary - Line-of-sight into system requirements
2) Verified - codes/solutions/models - (confidence in methods)
3) Validated - quantified errors and uncertainties (numerical and model)
4) Documented - sufficient to be reproducible (by others, 10 year later)
Materials Problem: Glass dissolution

A – rate of alkali surface release
B – rate of Si, Al surface detachment (tends to be limiting)
C – rate of $H_2O$ diffusion into glass
D – rate of silica diffusion through the gel (± SP) layer (at least partially limiting). Other constituents mass transport rates may also be important.
E – the aqueous composition at the RI is not the same as the Solution (aqueous silica concentration especially) and the surface area of the RI may be reduced by glass-gel contact area.
F – the gel and secondary phase layers may be acting as a mantle, in part isolating the fresh glass from the Solution.
Materials bulk phenomena

- Structure and chemistry of radionuclide-bearing phases and corrosion products
  - structure and phase stability as function of composition
  - volumetric and conductivity changes
  - cascade simulations, amorphization and recrystallization
  - point defects formation energies (v, i, anti-site, Frenkel, etc.)
  - point defect migration energies
  - nucleation and growth of gas bubbles, secondary phases, and cracks
  - volume diffusion of chemical constituents (esp. radios) to surfaces
  - grain boundaries, grain structure, microstructural evolution
  - diffusion along grain boundaries
  - surface free energies - exposed surfaces and cracks
  - microcracks, surface charges
  - indiffusion of water, reactions, and mechanical effects
Interface phenomena

- Chemistry of solid-aqueous interactions: corrosion, dissolution, reprecipitation, (as function of composition, oxidizing or reducing, pH, temperature, …)
  - surface/interface free energies
  - surface site energies and densities
  - energies of surface chemical processes, elemental attacks
    (WF degradation products, H\textsubscript{2}O, O\textsubscript{2}, H\textsuperscript{+},OH\textsuperscript{-}, H\textsubscript{2}O\textsubscript{2}, …)
  - migration/diffusion of chemical species along surfaces
  - out-/in-diffusion energies and rates of chemical species
    (water, bulk vacancies, ion exchange, radionuclide redox)
  - surface layer precipitation or dissolution: formation energies and rates
  - microbial effects
  - electrochemical corrosion
  - radiolysis effect on the chemistry
  - fugacities (partial pressures) of species in gas phase
  - equilibrium AND kinetic treatments of each of these
Upscaling within subcontinuum

- Phenom., Req., UQ, Val.
- Param., SA

Meso scale

Formulation of physics, Thermo., large structural data

Force fields, chemical networks, reaction/diffusion energies …

Dynamical/statistical (MD/MC)

Atomic chemistry:
- Phenom.
- Experiment
- QM/DFT.
Assembly into upscaling: Need for piecewise V&V, UQ

What simulation are you really doing?

Line-of-sight

What are the inputs (looking down)?
- model form/assumptions?
- parameter dependencies?
- how good are they?
- how good do they need to be?

What are the outputs (looking up)?
- what is the output quantity?
- what upscale needs this?
- how well does it need it?
- how good is the output?

Every scale must be up/down-scale-aware
VV-UQ are measure of awareness
Mapping V&V and UQ onto subcontinuum

• Presumptive line-of-sight
• Subcontinuum-adapted PCMM

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• Traceability, reproducibility, assessability
• Verification – code, numerics, models
• Validation
• UQ & SA
• Deliberate awareness of numerical vs. physical uncertainties

A start of a conversation, not a solution
Challenges

- **Tyranny of engineering timescale** (see: Becker 3:55pm)
  - Most projects are 3-5 years (at most) – “physics freeze” even shorter
  - Need for model development/validation data (not just engineering data)

- **Upscaling challenge** (e.g. several MD talks here)
  - How to propagate UQ through opaque barrier (calibration, model v. numerics)
  - How to define meaningful UQ with undefined physical causality networks
  - How to deal with every-UQ-is-a-custom-UQ analysis?

- **Avoiding the drunk-(mathematician)-under-a-streetlamp UQ approach**
  - Errors (uncertainties) in materials more model form and unknowable
  - Often too easy to do “simple” numerical analysis and ignore real risks

- **Science v. engineering culture** - Establish principles of VV&UQ in science
  - Incremental approach that enables credible piecewise VV&UQ
  - Presumptive line-of-sight as organizing principle
  - Articulate/enforce protocols for quantitatively meaningful simulations
  - Graded approach – rigor reflects sensitivity/importance
  - Promote culture of **quantitative-confidence** science for engineering
    - deliberate and explicit recounting of due diligence