UNCERTAINTY IN MATERIALS MODELING, SIMULATION, AND DEVELOPMENT FOR ICME

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Abstract

The importance of integrated computational materials engineering (ICME) has been acknowledged in the past decade. Yet, the reliability and robustness of simulation prediction under uncertainty have not been well recognized, given that all simulation models have errors involved. Particularly, discrepancy between models and physical world and numerical approximation for ease of computation are the major sources of model-form uncertainty. Data fitting and model calibration processes introduce additional input uncertainty because of systematic and random errors inherited in experimental data. Various uncertainty quantification (UQ) methods can be applied in materials simulation to assess the impacts of uncertainty. However, given the high cost of simulation mechanisms themselves, efficiency is a challenging issue for the application of UQ methods. In this paper, an overview of major UQ methods is given. The sources of uncertainty in materials modeling and simulation as well as technical challenges are discussed. The application of UQ methods in materials design and simulation is reviewed.

Introduction

The importance of integrated computational materials engineering (ICME) has been acknowledged in the past decade. The major paradigm shift is to provide data-enhanced and simulation-based tools so that sound decisions can be made during materials design. One of the core factors in decision making is uncertainty. Uncertainty exists universally in a real-world setting, and most of the decisions are made under uncertainty. It has been part of science development particularly in the domains of philosophy, mathematics, and physics for centuries. The quantification of uncertainty led to a new branch of mathematics, known as probability. Although different interpretations of probability co-exist and debates between scholars from these different schools last for centuries, it has been generally accepted by all that the source of uncertainty is our lack of knowledge about future. In the domains of physical sciences, two sources of uncertainty are differentiated. One is the lack of perfect knowledge, and the other is the random fluctuation of nature. The former is referred to as *epistemic* by many nowadays, whereas the latter is *aleatory*. Any uncertainty phenomenon we observe is the conflated effect of these two components. The differentiation of these two components is pragmatic and mainly for decision making practitioners. Epistemic uncertainty often appears as bias or systematic error in data or simulation results and is regarded as reducible. Increasing our level of knowledge can reduce the epistemic component of uncertainty. In contrast, aleatory uncertainty appears as random error and is irreducible. Random fluctuation inherently exists in physical matters such as atoms and electrons above absolute zero temperature. When decision makers can differentiate the sources of uncertainty, the risk of go/no-go decision is easier to be managed. Gaining more

knowledge to reduce the epistemic component of uncertainty will generally lead to more precise estimation of the risk, which is the combination of chances and consequences.

Widely used modeling and simulation (M&S) software packages for ICME, such as density functional theory (DFT), molecular dynamics (MD), kinetic Monte Carlo (kMC), dislocation dynamics (DD), phase field (PF), and finite-element analysis (FEA), predict physical phenomena at different scales and provide decision support for engineers and scientists. The sources of uncertainty associated with these tools should be identified if we would like to make robust decisions based on the results of these computational tools. All models require certain levels of abstraction thus approximation error is inevitable. The causes of these errors are the major sources of epistemic uncertainties in M&S tools. The forms of models we choose in the approximation lead to *model-form uncertainty*. In addition, empirical models are derived from experiments via data fitting procedure. All measurement has systematic error or bias from device or human. This leads to bias in parameters of the fitted model, known as *parameter uncertainty*. Model form uncertainty and parameter uncertainty are the major factors of epistemic uncertainty. Needless to say, bias also exists in the data archived in ICME databases which are collected through either experiment or simulation.

Uncertainty propagates through modeling and simulation procedures. M&S tools with imprecise and inaccurate inputs will produce uncertain output. *Uncertainty quantification* (UQ) is the process of representing uncertainty and predicting its propagation quantitatively in data and models. The predicted uncertainty is used for risk analysis and decision making. Given the enormous amount of work that has been done in UQ methods, the purpose of this paper is not to provide a comprehensive literature review of UQ methods themselves. Rather, an overview of the available methods is provided to assist materials scientists in the ICME community in selecting UQ methods for specific problems.

Sources of Epistemic Uncertainty in Multiscale Modeling and Simulation

In DFT simulation, the major source of model-form uncertainty is the exchange-correlation potential functionals, where many-particle interaction is approximated and simplified in data fitting procedure. In addition, the pseudopotentials are typically used to replace the Coulomb potential near each nucleus in the calculation, which also introduces approximation error. Error is also introduced in the tradeoffs between long-range and short-range dispersions and between efficiency and accuracy during the approximation. In the self-consistency calculation of ground state energy, the chosen threshold for convergence also introduces numerical error.

In MD simulation, the major source of model-form and parameter uncertainties is the uncertainty associated with the interatomic potential function. As the input of MD, the approximation error is naturally propagated to the output prediction through the simulation process. Other sources of uncertainties include the cut-off distance in simulation for ease of computation, the imposed boundary conditions that may introduce artificial effects, simulation acceleration through modified potentials or the application of physically unrealistic high strain rates which intends to overcome the time scale limitation of MD, the computational error with different computer architecture because of round-offs in floating-point numbers or task distribution and sequencing in parallel computation, the systematic error in measurement data that are used in model calibration, as well as other unknown biases introduced during the model construction process.

In kMC simulation, the major sources of epistemic uncertainties are the incomplete event catalog and imprecise rates or propensities. The accuracy of kMC simulation depends on the validity of

complete knowledge of all possible events. Further more, the true rates in physical world can vary along time. They are also dependent on the state of the system. For instance, external loads can change the diffusion of defects. Crowding effect exits in reactions where molecules easily block reaction channels. In kMC, events are also assumed to be independent for the convenience of computation. In reality, they may be correlated. The unknown correlation between events is another source of model-form uncertainty.

In DD simulation models, the major approximations include the empirical modeling of stress field, phenomenological rules for dislocation interactions, linear approximations of dislocation curves during discretization, and numerical solutions of ordinary differential equations.

In PF simulation, the major sources of model-form uncertainty are the empirical models of energy functionals, and numerical treatment in solving partial differential equations.

In summary, unknown bias and numerical treatment of any model in simulation introduce epistemic uncertainty into the model. As a result, the prediction as the simulation output is inherently imprecise. When macroscopic quantities as statistical ensembles are of our interest, the output is also inaccurate. Therefore, the simulation output contains both epistemic and aleatory uncertainties. Quantification of these uncertainties is necessary to estimate the possible extent of inaccuracy and imprecision, establish the confidence in model verification and validation, and improve the robustness of simulation prediction.

The uniqueness of UQ for ICME is the need of considering uncertainty propagation between multiple length and time scales in material systems. Uncertainty observed at a larger scale is the manifestation of the uncertainty exhibited at a smaller scale. For instance, the non-deterministic strengths of material specimens are due to the statistical distributions of grain boundaries and defects. The randomness of molecular movement known as Brownian motion is from the stochasticity of physical forces and interactions among electrons at quantum level. The ability of modeling the propagation of uncertainty between scales will allow us to have a holistic view of material systems without the artificial separation of scales. Existing M&S tools simulate material systems with the range from nanometers to micrometers. The major challenge of UQ in these tools is the information exchange between different models, where assumptions of scales and boundaries are made a priori. For model validation, not all physical quantities predicted in simulation can be directly observed, especially those at small length and short time scales. Measurable quantities at larger scales are typically used to validate models. The assumed or derived correlation between the measureable and unobservable also introduces uncertainty, in addition to sensing errors.

UQ Methods

Various UQ methods have been developed in the past five decades. Most of the UQ approaches are developed based on probability theory. Alternative approaches [1] such as evidence theory, possibility, interval analysis, and interval probability have also been developed, particularly with the differentiation between aleatory and epistemic uncertainty. With respect to the application of M&S, the methods can be categorized as intrusive and non-intrusive. Non-intrusive UQ methods do not require an internal representation of uncertainty in the M&S tools. The original tools are treated as black boxes, and the UQ methods are implemented as parent processes to call M&S tools for necessary evaluations. In contrast, intrusive UQ methods require the modification of the original M&S software tools so that uncertainty can be represented internally.

The commonly used non-intrusive UQ methods include Monte Carlo simulation, global sensitivity analysis, surrogate model, polynomial chaos, and stochastic collocation. The common intrusive UQ methods include local sensitivity analysis and interval-based approaches.

Monte Carlo simulation. Monte Carlo (MC) simulation is the earliest attempt to quantify uncertainty inherent in physical models, where pseudo random numbers are generated by computers and used in evaluating models, although the technique was originally devised to numerically calculate deterministic integrals in quantum mechanics. The effectiveness of MC relies on how "random" the numbers generated by the pseudo random number generators (PRNGs) are. Common implementations of PRNGs as the core of MC to generate uniformly distributed numbers include linear congruential method [2, 3] and its various extensions [4, 5, 6, 7, 8] for longer periods and better uniformity, feedback shift register generators [9, 10], and Mersenne twister [11]. Based on the uniformly distributed numbers, random variates that follow other distributions can be generated via computational methods such as inverse transform, composition, convolution, and acceptance-rejection. When MC is applied to assess the uncertainty associated with the inputs of a model, inputs with predetermined distributions are randomly generated. They are used to evaluate the model or run the simulation many times. The distribution of the resulting outputs is then used to assess the effect of uncertainty, which is quantified with statistical moments of different orders.

The major issue of MC for UQ in ICME is its computational cost. The quality of uncertainty assessment depends on how many runs of simulation can be conducted in order to generate statistical distributions of outputs and draw meaningful conclusions from the results. If each run of simulation is expensive such as DFT and MD, the cost of UQ will be high in the pure sampling based approach. In addition, MC also requires the predetermined input distributions from which the samples are drawn. If there is a lack of prior knowledge about the distribution types or lack of data, the effect of model-form uncertainty needs to be assessed. Second-order Monte Carlo (SOMC) [12] is a natural extension of MC to study the uncertainty associated with the distributions. In the outer loop, the parameters or types of statistical distributions are randomly sampled. In the inner loop, classical MC is applied with each of the sampled distributions to study the variability. The SOMC results can provide an overall picture of the combined effects from both epistemic and aleatory uncertainty.

Global Sensitivity Analysis. Sensitivity analysis is a general concept of studying how uncertainty in the output of a model is attributed to different portions of uncertainty associated with model inputs. Global sensitivity analysis (GSA) [13] quantifies the effect of each individual input as well as their joint effects by conducting the analysis of variance. For a given model or simulation input-output relation $Y=f(X_1,\ldots,X_n)$, by fixing input variable X_i to be x_i^* one at a time, the conditional variance of the output $Var(Y|X_i=x_i^*)$ is typically less than the original total variance $Var(Y)$. The difference between the two is an indicator of the contribution of variance from input variable X_i, i.e. the sensitivity of output uncertainty with respect to input X_i. Given all possible value of x_i^*, $Var(Y|X_i=x_i^*)$ itself is a random variable. Thus, it is easier to use its expectation $E(Var(Y|X_i))$. The deterministic value $Var(Y)-E(Var(Y|X_i))$ therefore is a metric to quantify the importance of input X_i. Similarly, the conditional expectation $E(Y|X_i=x_i^*)$ is a random variable. Equivalently, the value $Var(Y)-Var(E(Y|X_i))$ is an indicator of the contribution of variance from input variable X_i, since $Var(Y)=E(Var(Y|X_i))+Var(E(Y|X_i))$. The first-order Sobol' sensitivity index [14, 15] $S_i=Var(E(Y|X_i))/Var(Y)$ is commonly used to quantify the sensitivity of output with respect to the uncertainty associated with the i^{th} input variable as the main effect. The

interactions among input variables are estimated by second- or higher-order sensitivity indices. By fixing two variables X_i and X_j simultaneously at a time, $Var(E(Y|X_i,X_j))$ measures the joint effect of the two variables. The second-order index $S_{ij}=Var(E(Y|X_i,X_j))/Var(Y)-S_j-S_j$ shows the interaction effect of the two input variables. Higher-order indices can be defined in a similar way. Instead of variance, other moment-independent sensitivity indices [16, 17] were also proposed to quantify the importance of input uncertainty based on cumulative distributions or density functions directly.

It should be noted that GSA does not require a known or closed-form mathematical function $Y=f(X_1,\ldots,X_n)$ for the input-output relation. For black-box simulation, if enough runs of simulation are conducted with proper design of experiments to generate statistically meaningful pairs of input and output, GSA can be performed by the analysis of variance.

The limitation of GSA for ICME is similar to the one in MC, since MC sampling is typically needed to estimate the variances. Notice that traditional MC only provides the information of overall output distributions, whereas GSA can provide fine-grained information of individual and compound effects of input variables. Instead of variance, other measure of uncertainty such as interval range can also be applied for GSA [18]. This can reduce some computational cost of sampling.

Surrogate models. When the input-output relations in simulation models are too complex or unknown, surrogate models can be constructed to approximate the response between inputs and outputs. The simplified surrogate models, typically in the form of polynomials or exponentials, can improve the efficiency of model evaluations and predictions. For UQ, surrogate models are used for sensitivity analysis. The input-output responses can be generated by experimental designs such as factorial, fractional factorial, central composite, and orthogonal designs [19, 20, 21]. The resulting models are generally called response surfaces. They are constructed by interpolation or regression analysis of the results from simulations with combinations of input variable values. Particularly, to improve the efficiency of sampling from inputs under uncertainty, Latin hypercube sampling and Kriging were developed. With the constructed response surfaces, performance of new input values and the sensitivity can be predicted without running the actual simulation itself.

To construct response surfaces, the input variable values can be generated via Latin hypercube sampling (LHS) [22, 23, 24]. LHS is a stratified sampling strategy for variance reduction where samples are taken from the predefined input subspaces. For each subspace, there is an equal probability that the input samples are drawn from. The sampling is then performed within each of these subspaces so that all subspaces can be covered by much fewer samples than classical MC sampling. Thus, the number of samples can be significantly reduced while results are still statistically representative. LHS is also extended to dividing subspaces with unequal probabilities, and the estimates are weighted by the corresponding probability values [25]. LHS is a versatile tool and can be applied to study statistical properties of responses directly without constructing response surfaces. The limitation of LHS is that the efficiency of the sampling strategy depends on the prior knowledge of probability distributions associated with input variables. When there is a lack of knowledge about the types and parameters, the variance reduction technique may introduce bias.

Kriging [26, 27], also known as Gaussian process regression, is an interpolation method (although regression appears in the name) to find approximated response models between input's

and outputs. It predicts new functional value from some existing ones by modeling the underlying but unknown true function as a Gaussian process. The prediction is the weighted average of the existing values plus an additive noise as a stationary covariance process with zero mean. With the estimated covariance from existing data via maximum likelihood estimation (MLE), the new value is predicted. More specifically, the prediction of new functional value $y(\mathbf{x})$ at any $\mathbf{x}$ is $y(\mathbf{x})=\mathbf{f}(\mathbf{x})^{\mathrm{T}}\boldsymbol{\beta}+\varepsilon(\mathbf{x})$ where $\mathbf{f}(\mathbf{x})=[f_1(\mathbf{x}),\ldots,f_p(\mathbf{x})]$ is a vector of p polynomial basis functions in the so-called universal Kriging. With m existing inputs $\mathbf{x}^*=(\mathbf{x}_1,\ldots,\mathbf{x}_m)$, the corresponding output vector $\mathbf{Y}=[y_1,\ldots,y_m]^{\mathrm{T}}$, and an $m\times p$ matrix of polynomial values $\mathbf{F}=[\mathbf{f}_1(\mathbf{x}^*),\ldots,\mathbf{f}_m(\mathbf{x}^*)]^{\mathrm{T}}$, the estimation of functional value at the new position $\mathbf{x}$ is $\underline{y}(\mathbf{x})=\mathbf{f}(\mathbf{x})^{\mathrm{T}}\underline{\boldsymbol{\beta}}+\underline{\varepsilon}(\mathbf{x})$, where $\underline{\boldsymbol{\beta}}=(\mathbf{F}^{\mathrm{T}}\mathbf{R}^{-1}\mathbf{F})^{-1}\mathbf{F}^{\mathrm{T}}\mathbf{R}^{-1}\mathbf{Y}$ is the generalized least square estimators of the coefficient $\boldsymbol{\beta}$, $\mathbf{R}$ is the $m\times m$ correlation matrix for m data points, and $\underline{\varepsilon}(\mathbf{x})=\mathbf{r}(\mathbf{x})^{\mathrm{T}}\mathbf{R}^{-1}(\mathbf{Y}-\mathbf{F}\underline{\boldsymbol{\beta}})$ is the estimator of covariance with $\mathbf{r}(\mathbf{x})$ being the correlation function at the new position $\mathbf{x}$. Classical Kriging as the interpolation method has been used in surrogate model construction from deterministic simulations [28, 29,] as well as its application in design optimization [30, 31].

One issue of universal Kriging for UQ is that the functional response with polynomials as bases is assumed. Therefore model-form uncertainty is introduced. To improve the accuracy of prediction, Bayesian model update approach with multiple models has been proposed [32]. Similarly, blind Kriging modifies polynomials with Bayesian variable selection by incorporating experimental data [33]. Another issue is that the covariance for the underlying true function is unknown. Some correlation functions have to be assumed or fitted from existing data with MLE. An important model-form uncertainty thus is introduced. Stochastic Kriging or composite Gaussian process [34, 35] were introduced to decompose the covariance into two components. One covariance process is from the underlying true function, whereas the other is from experimental data. This approach allows for the decomposition of model-form uncertain. Nevertheless, the assumption of the unknown covariance of the two Gaussian processes still has to be made.

Polynomial chaos expansion. Polynomial chaos expansion (PCE) [36, 37, 38] approximates random variables in functional or reciprocal space with orthogonal polynomials as bases. Different polynomials are available for different probability distributions. Orthogonality ensures the efficiency of computation and ease of truncation error quantification. However, efficiency is still a challenge for PCE. For instance, solving a stochastic differential equation is reduced to solving many deterministic differential equations. The efficiency of computation is directly related to the truncation, which also depends on types of distributions and the corresponding polynomials. Some distributions such as those with long and heavy tails cannot be efficiently modeled using PCE approximation.

Stochastic collocation. When the number of input parameters for simulation models is very high, such as hundreds, the direct construction of the high dimensional response surface will become inefficient. Stochastic collocation [39, 40] is an approach to alleviate the curse of dimensionality. The main idea is to choose the sampling positions of input wisely for functional evaluations in conjunction with the orthogonal polynomials in the problem solving process (i.e. partial differential equation with random inputs) so that a much sparser grid can be used.

Local sensitivity analysis. Different from GSA, local sensitivity analysis (LSA) studies the effect of input uncertainty locally. A straight-forward way is to estimate derivatives of models with the finite-difference approach where the difference between two responses is divided by the perturbation of inputs. The finite-difference approach is a non-intrusive approach to assess

sensitivity. In addition to these direct (forward) methods, adjoint (backward) SA approaches were also developed for deterministic simulation based on differential equations [41, 42]. A more efficient LSA method that is specific for stochastic simulation is to estimate the derivatives of the expected values of output performance, i.e. the expected values of stochastic derivatives or gradients, from simulation directly. This can be achieved by either varying output performance w.r.t. input parameters as the infinitesimal perturbation analysis [43, 44], or by varying the probability measures w.r.t. inputs as in the likelihood ratio method [45, 46, 47]. These approaches are intrusive in order to promote efficiency of computation.

Interval analysis. Interval analysis [48] is a generalization in which interval numbers replace real numbers, interval arithmetic replaces real arithmetic, and interval analysis replaces real analysis. Interval arithmetic was originally developed to solve the issue of numerical errors in digital computation due to the floating-point representation of numbers. Intervals provide a distribution-neutral form to represent uncertainties and errors in measurement or digital computation. Interval probability is an extension of probability theory to include epistemic uncertainty. It is also closely related to Dempster-Shafer evidence theory and possibility theory. One drawback of the intrusive methods based on intervals is the range of variations can be overestimated and grow pessimistically wide based on classical interval arithmetic during propagation.

UQ in Materials Design and Simulation

In this section, the application of UQ methods in materials design and simulation is reviewed. Chernatynskiy et al. [49] provided a review related to UQ in multiscale simulation. Here, the review is focused on the research efforts related to materials simulation, especially those that emerged in the most recent years, indicating the rapidly growing interest of UQ for ICME.

Uncertainty in materials simulation was initially recognized with sensitivity of inter-atomic potential selection in MD simulation of solid materials [50, 51], water molecule [52, 53], irradiation damage [54], and others, as well as the effect of cut-off radius. Only recently, formal UQ methods have been applied. To study the sensitivity of macroscopic properties w.r.t. interatomic potential parameters, Jacobson et al. [55] constructed response surfaces with Lagrange interpolation. To construct better interatomic potentials in MD, Frederiksen et al. [56] applied Bayesian update with experimental data sets. Cailliez and Pernot [57] as well as Rizzi et al. [58] applied Bayesian model calibration to calibrate interatomic potentials parameters. Angelikopoulos et al. [59, 60] showed the applicability of Bayesian model calibration with water molecule models. Rizzi et al. [61] applied PCE to study the effect of input uncertainty in MD. Cailliez et al. [62] applied Kriging in water molecule MD model calibration. As an intrusive approach, Tsourtis et al. [63] developed Langevin dynamics in MD so that the effect of input uncertainty is assessed. As another intrusive UQ method, Tran and Wang [64] developed an interval based MD mechanism via Kaucher interval arithmetic to assess uncertainty propagation.

The sensitivity of first-principles simulation was recognized more recently. Mortensen et al. [65, 66] estimated the distribution of exchange-correlation functional errors from experimental data via the Bayesian update approach. Hanke [67] performs local sensitivity analysis of DFT with respect to input parameters with finite difference estimation of the first derivative of binding energy. Pernot et al. [68] predicted systematic errors associated with the exchange-correlation functionals of different crystal structures using regression analysis. He [69] used the stochastic Kriging approach and developed efficient and robust first-principles saddle point search algorithms where model-form uncertainties of DFT and surrogate are quantified simultaneously.

Choi et al. [70] applied response surface model for simulation based robust design of material compositions. Koslowski and Strachan [71] studied the local sensitivity of MD, PF, and DD in crystal elasticity via perturbation.

Concluding Remarks

To facilitate the wide adoption of ICME by academic researchers and industry practitioners in materials design, the reliability of simulation should be assessed so that it can provide better confidence to the 'consumers' of such information about how credible the predictions are. Even though it is well recognized that simulation results are very sensitive to model inputs, for instance, interatomic potentials and cut-off radius for MD, existing practice of not providing uncertainty information gives the perception to the information consumers that modelers are not willing to reveal the details and model development tends to be ad hoc. This obviously hinders the adoption of ICME methods. On the other hand, simulation practitioners' unwillingness to perform UQ for their models is a reflection of the lack of understanding of uncertainty in ICME research community. It is most likely because of their perception that uncertainty is equivalent to error-prone and unscientific. Therefore, researchers of UQ methodology must reach out to educate and fill this knowledge gap. Much more work still needs to be done to explore the potential of UQ to improve ICME methods and tools.

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