CCSI™

Carbon Capture Simulation Initiative

CCSI Validation and Uncertainty Quantification Hierarchy for CFD Models

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Acronym List

Acronym	Descriptive Name
ADA-ES	ADA Environmental Solutions
CCSI	Carbon Capture Simulation Initiative
CFD	Computational Fluid Dynamics
CO_2	Carbon Dioxide
DOE	Department of Energy
MFIX	Multiphase Flow with Interphase eXchanges
MWe	Megawatt Electric
NETL	National Energy Technology Laboratory
PSRI	Particulate Solid Research, Inc.
SA	Sensitivity Analysis
TGA	Thermogravimetric Analysis
UA	Uncertainty Analysis
UQ	Uncertainty Quantification
V	Validation

1. Introduction

In order to use the information and predictions generated by the Carbon Capture Simulation Initiative (CCSI) Toolset with confidence, it is critical that the physical and mathematical models are continually and methodically validated with experimental data at various scales, and uncertainties in the modeling process are characterized and quantified. This document outlines a plan for CCSI validation and Uncertainty Quantification (UQ) of the Element 2 models to identify and quantify the computational fluid dynamics (CFD) simulation errors measured in relation to experimental data and to provide a quantitative assessment of uncertainties.

Because of the infeasibility of designing and conducting validation experiments on the full-scale carbon capture systems, we will follow a systematic and hierarchical approach in designing physical experiments (and/or utilizing available measurement data sets) for comparison with predictive computational models. This approach will divide the complex utility scale solid sorbent carbon capture system into several progressively simpler tiers: pilot scale cases, laboratory scale cases, and unit problems, as illustrated in Figure 1.

This layered strategy will incorporate uncertainty in both parameters and models to assess how accurately the computational results compare with the experimental data for different physical phenomena coupling and at different geometrical scales. The validation and UQ efforts will be supported by experimental data from a variety of sources, including the U.S. Department of Energy (DOE), National Energy Technology Laboratory (NETL) facilities and NETL-supported industrial experiments at external locations (DOE-NETL Industrial Challenge Problems) as well as available literature data. The plan for the integrated validation and UQ activities for CCSI CFD modeling and simulations is based on the expertise and experience of the CCSI team members as well as their state-of-the art knowledge of basic concepts, principles, and procedures (Oberkampf 2010, Roy 2011, Roache 1998, Grace 2004, and AIAA 1998).

2. Overview

The CCSI CFD development work, which is focused on the specific multi-physics of the carbon capture systems of interest to CCSI and the capabilities of the models and codes, needs to be supported by a quantitative assessment of the confidence in the predictions. The contributing elements in the development of a computational predictive capability for the CFD models and simulations are verification, validation, and UQ. While many types of models (1-dimensional, reduced order, process models, etc.) are being developed for the CCSI Toolset, validation efforts will focus mainly on the CFD models. The other models can then be validated against (or calibrated to) those validated models. This will ultimately allow the fundamental, validated physics-based models to be used to improve the other CCSI models.

Verification addresses the correctness and functionality of the computations and denotes the process of establishing the precision of the numerical solution of the computational model in comparison to the accurate solutions. The verification process may be divided into two areas: code verification and numerical solution verification. The CCSI CFD researchers are developing their models and codes in an open-source multiphase flow computer code MFIX. MFIX has been developed by employing appropriate software engineering practices and has been subjected to software verification process in several studies. We can thus assume that the major features of the provided general-purpose MFIX computer code have been verified and are an accurate computational representation of the original mathematical models. Numerical solution verification starts after the particular model has been embodied in a verified code, the initial and boundary conditions have been specified, and all other auxiliary relations have been incorporated (verification of input data). The multi-physics models and sub-models, which are used to simulate the carbon capture system in MFIX, are well developed and widely used in industry and research. As such, we assume that the basic models and sub-models in MFIX are well verified and do not attempt to re-verify them as part of this work.

Model validation, the main objective of the validation (V)/UQ sub-team, is to develop the process for determining the degree to which a CFD model is an accurate representation of the real solid sorbent carbon capture processes from the perspective of the intended uses of the model (AIAA 1998). The execution of this process requires experimentally measured data to compare with the simulation results. We plan to obtain these data from a variety of sources including NETL facilities and NETL-supported industrial experiments at external locations (DOE-NETL Industrial Challenge Problems) as well as available literature data. It is important to obtain not only the experimental measurements for system response quantities (e.g., solid mass flux and density, pressure drop, particle velocity, capture efficiency, temperature, residence time, etc.) but also the estimation of the measurement uncertainties. Consequently, the model validation requires a statistical estimation of the difference between model predictions and experimental results, including experimental uncertainties.

Since many aspects of the CCSI CFD models are not exactly known, **UQ** is an important aspect of the modeling process. The CCSI UQ team (Element 6) is focused on modeling and investigating the uncertainty of the various models developed in the CCSI program. The CFD team (Element 2) will work closely with Element 6 to provide an integrated process for validating models and performing UQ analysis. UQ analysis will address specific uncertainty objectives, such as identifying important drivers, quantifying input distributions, understanding departure of the model from the underlying behavior, and determining what additional data to collect, through a sequential process designed to identify and characterize sources of uncertainty which can then be propagated, analyzed, and ultimately reduced through improved models or additional data collection.

The subsequent sections provide more details on the CCSI CFD model V/UQ plan and its execution.

3. Objectives and Expected Outcomes

The objective of the proposed validation and UQ work is to quantify the accuracy of the CFD modeling of the carbon capture system and to quantify our confidence in the model results. As will be discussed in the following sections, this work aims to validate the CCSI CFD models of the pilot (1 MWe), intermediate (25 MWe, 100 MWe) and full (650 MWe) scale carbon capture systems using a hierarchical validation approach. In this approach we use smaller scale models and simplified physics to investigate the accuracy of the various components of the CFD models and upscaling approaches used in CCSI's Particle and Device Scale Modeling activities (Element 2). The validation approach that we use is specific to the CCSI program and can be considered as "application focused validation." By "application focused validation," we mean that we will perform a series of logical and relevant model validation studies specific to the CFD problems within CCSI, which will provide a quantitative confidence range on the CFD predictions for the full scale system. Our goal here is *not* to develop and follow a general and fully hierarchical validation plan that can be applied ubiquitously to any coupled physical phenomena or system.

The outcome of this validation process will be estimates of the confidence range for system performance metrics for the developed CCSI CFD models and codes when used to simulate the carbon capture technologies of interest to the CCSI program. These estimates will be based on the confidence levels developed in the various decoupled unit problems as well as the upscaling procedure outlined in Figure 1. By considering smaller unit problems that isolate particular aspects of the multi-physics of the full scale system, we should be able to evaluate/isolate the contributions of different aspects of the model to the predictive errors, such as the effects of coupling different physics or the geometric upscaling of the coupled physics.

This task has the following specific major objectives:

- Quantify the accuracy of CFD modeling of laboratory and pilot scale solid sorbent carbon capture system(s).
- Estimate the accuracy of CFD modeling of the full scale system through hierarchical validation, UQ, and upscaling.
- Quantify the team's confidence in the CFD model results.

4. Validation Hierarchy

4.1 Overview

This section gives an overview of the CCSI Validation Hierarchy. Further details of each level will be given in following sections.

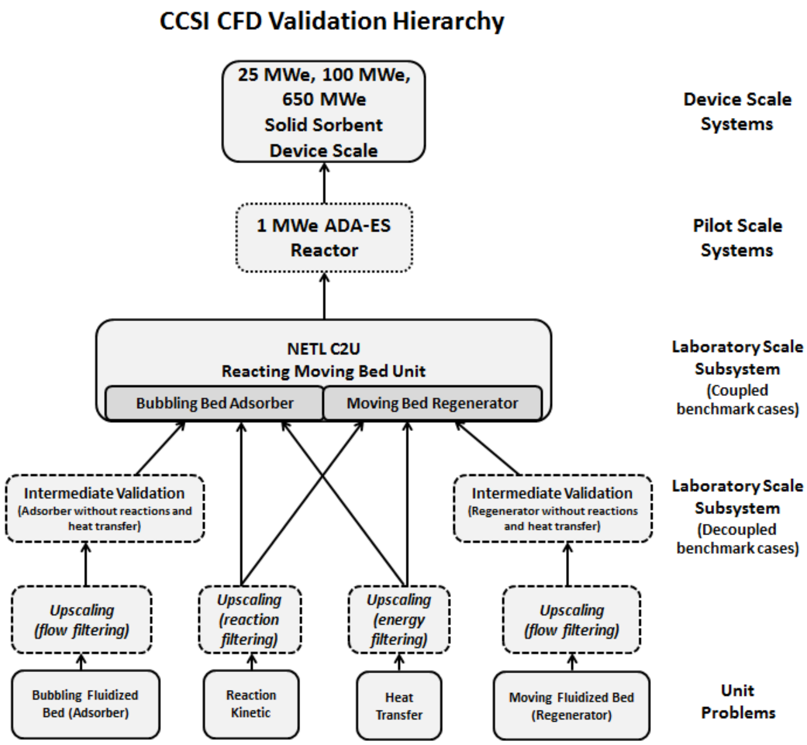


Figure 1: CCSI CFD Validation Hierarchy, Illustrating the Various Unit Problems of Interest, and the Levels of Validation that Lead up to Quantitatively Reliable Predictions for the Full Scale Device Systems

Solid sorbent-based carbon capture devices are complex engineering systems involving complicated multi-phase and multi-scale phenomena as well as state-of-the art industrial technology. To deal with this complex engineering system, we introduce a hierarchical structure (AIAA 1998, Oberkampf 2010) to estimate the confidence range of the CCSI CFD models' predictions (Figure 1). The complex solid sorbent carbon capture system is divided into several tiers of progressively increasing complexity.

The lowest level of the validation hierarchy is comprised of the unit problems encountered in the adsorber and regenerator, where each unit problem represents an important physical phenomenon. For the current system, the important physical phenomena, i.e., unit problems, are identified as (1) flow in a bubbling fluidized bed, occurring in the adsorber; (2) flow in a moving fluidized bed, prevalent in the regenerator; (3) reaction kinetics; and (4) heat transfer, which are expected to be significant in both the regenerator and adsorber. From a modeling perspective, each unit problem can be viewed as a set of models, and suitable choices for each model and its parameters will be based on existing knowledge or, if necessary, CCSI validation activities. For example, in the bubbling fluidized bed unit problem, several options are available for modeling the gas drag force experienced by the fluidized particles. As some model choices are expected to perform better than others, the identification of the best available models would be part of the CCSI validation activity.

The second level of the validation hierarchy addresses upscaling of the chosen models for simulating large geometries. It has been shown that in order to obtain quantitatively accurate results for multiphase systems, the grid size used in CFD simulations must be on the order of a few particle diameters (Agrawal 2001, Andrews 2005). When simulating a device which is tens of meters tall, it is not possible to have grid sizes in the range of 4-10 times the particle diameters (1 mm - 1 cm). However, the effects of fine scale structures can be accounted for in faster, coarse grid simulations through the development of "filtered equations" for gas-particle drag, effective gas viscosity, effective reaction rates, effective heat transfer coefficients, and other important model variables. Identification of existing filtered equations and/or development of unavailable filtered relationships will be part of the upscaling activity.

In the third level of the validation hierarchy, decoupled laboratory scale cases (flow without reactions or heat transfer) will be compared against predictions from the upscaled flow models. The NETL Carbon Capture Unit (C2U) is expected to provide useful validation data at approximately this scale despite some differences from the CCSI design. Experimental data from the NETL C2U system will be used for validation along with literature data. This step will help build confidence regarding the choice of models in the first level and the upscaling (filtering) procedure developed/implemented in the second level. It should be noted that it may not be possible to conduct decoupled experiments to test the performance of upscaled reaction or energy models. Quantitative assessment of the reliability of the model predictions can also be made for each unit problem, which will help in understanding the compounding of errors in the next level.

The fourth level of the hierarchy couples the unit problems (flow, reactions, heat transfer) together with the appropriate upscaling relationships for modeling a laboratory scale adsorber and regenerator such as the NETL C2U reactor or the 1 kWe ADA-ES system. This is the scale where fully coupled, and decoupled (cold flow), device scale measurement data will be available. The predictions of the coupled system will be validated against experimental measurements, including attempts to gain some understanding and/or functional relationships due to the compounding/coupling of errors of the unit problems.

The next tier of the validation plan is for pilot scale cases such as a 1 MWe system. At this time there are no 1 MWe systems in operation. However, ADA-ES is designing a 1 MWe system that, if available, could be used to validate a pilot scale CFD simulation.

The topmost level of the hierarchy is to obtain quantitative confidence on the predictions for the larger device scale systems (25 MWe, 100 MWe and 650 MWe). As of the writing of this validation plan, no full scale carbon capture systems are in operation nor are there any plans to construct such a system in the CCSI project life cycle. Therefore, the validation of the larger scale device models will be based on the validation activities in the lower tiers. The larger scale device simulations will rely on the information gained through the smaller scale validation tiers and the expertise of the Element 2 team. In developing these models the validated unit problems and upscaling methods will be employed. In addition, the expertise of Element 2 personnel will be used to determine the appropriate baseline operating conditions and parameters for the device scale models. The validation work at this larger device scale will also provide estimates of the error in the device scale models and the sources of those errors, which will come from the knowledge gained through the lower tier validation studies and propagation of uncertainty.

Our work at the larger device scale tier will lead to the development of baseline device scale models based on the validation work and our knowledge of the system. The development and use of these baseline models will be coordinated with other elements in the CCSI program to enable the development of reduced order models and for UQ analysis. It is expected that knowledge based calibration of the model parameters will be critical in obtaining better quantitative predictions.

4.2 Validation Tiers

This section gives details of each tier of the CCSI Validation and UQ Hierarchy.

4.2.1 Unit Problems

The unit problems for the CCSI CFD validation isolate the complex multi-physics of the CCSI reactor into simpler single-physics units. This includes the multi-phase flow in the bubbling fluidized bed of the adsorber, the reactions of carbon dioxide (CO_2) with the solid sorbent particles, the heat transfer in the reactor, and the multi-phase flow of the moving fluidized bed of the regenerator. The multi-phase flow of the adsorber and regenerator are considered in separate unit problems due to the different flow regimes present in the designs of the adsorber and regenerator that will affect the modeling methods used to simulate the two sub-systems. The operating conditions of the unit problem will be determined by the design of the full scale carbon capture systems. To determine the appropriate operating conditions for the unit problems, Element 2 is working closely with the Process Synthesis and Design team (Element 3).

The validation of the unit problems focuses on previously published experimental and code validation research. The unit problems based on the full scale system are common configurations and physical phenomena arising in many different multi-phase reactors used in numerous mechanical and chemical engineering industries. We will take advantage of the published literature on these unit problems to determine the most accurate modeling methods and model parameters.

A detailed literature search for each of the unit problems shown in Figure 1 has been done, and available literature has been compiled and evaluated by the CCSI Element 2 team and V/UQ sub-team members. The most appropriate literature cases have been identified and will be used for the validation and UQ analysis of the unit problems for the CCSI systems. The available literature for each of the unit problems is discussed below.

Bubbling Bed: Several validation studies of the hydrodynamics of bubbling beds are available in the literature (Hulme 2005, Li 2011, Herzog 2012, Lindborg 2007, Esmaili 2011, and Asegehegn 2011). From the available literature, the experimental work of (Kim 2003) has been selected as the most appropriate case for the CCSI validation studies of the bubbling bed unit problem.

(Kim 2003) completed an experimental study on the hydrodynamics and heat transfer in a bubbling bed with immersed tubes. The experimental setup measured 0.34 x 0.48 x 0.6 m with a staggered tube bank of 25.4 mm diameter tubes. The solid phase was sand with a mean diameter of 240 μm, and a density of 2582 kg/m³. Experiments were carried out at five different gas velocities with measurements taken around a central tube at various angular locations as discussed in Section 6. The hydrodynamic data published includes bubble and emulsion phase fractions, bubble frequency, and emulsion contacting time.

This paper was chosen for the bubbling bed validation case due to its similarities to the CCSI designs. The experimental setup replicates the desired geometry, uses similarly sized particles, tests a variety of gas velocities, and reports several variables that can be easily compared to numerical simulations. Additionally, initial validation studies using MFIX have been previously done at NETL (Li 2010).

Moving Bed: Very few experimental studies on the hydrodynamics of a moving fluidized bed are available in the literature. The most appropriate case for the CCSI validation work is that of (Yoon and Kunii 1970), who studied the gas flow and pressure drop through moving beds using two bed setups: a 7.0 cm diameter, 90 cm high acrylic pipe, and a 4.1 cm diameter, 45 cm high iron pipe. The particle size of the solid phase was varied throughout the experiments. The mean diameters were measured to be 133, 261, 430, and 1130 μm with densities of 2470, 2480, 2480, and 2480 kg/ m³, respectively. The experimental results include the solids velocity, pressure gradients, relationships between slip velocity and pressure gradients, and relationships between gas flow rate and pressure gradients.

This paper examines a moving bed with no perforated plates. Although the geometry and particle properties are slightly different than the current CCSI full scale design, the results from (Yoon and Kunii 1970) represent the best moving bed validation studies available in the literature. Despite the particle material being different, the range of diameters should allow good comparison by analyzing the particle size nearest to that being considered in CCSI (solid sorbent material 32D).

(Lapidus 1957) give theoretical results of the mechanics of vertical moving beds. Their research focuses mainly on the derivation of empirical relationships; however, they do present and compare the experimental data of (Struve 1955) and (Price 1951). (Struve 1955) used glass spheres of diameter 100 μm and water as the fluid phase; while Price used glass spheres with diameters of 8240 μm. The published data includes the holdup vs. slip velocity and holdup vs. fluid velocity. The experimental data presented in (Lapidus 1957) is sparse but should give a good starting point for validation. Further examination of the work of (Struve 1955) will be needed to apprehend the full experimental details. Due to the large solid phase diameter used by (Price 1951), we will not pursue this dataset at this time.

Heat Transfer: Many studies consider heat transfer in a fluidized bed; however it has been shown that the particle size has a significant effect on the results and accuracy of the simulation. Based on the full scale CCSI design, we conclude that the heat transfer data from (Kim 2003) is best suited for the CCSI V/UQ studies. The second half of (Kim 2003) reports experimental heat transfer results. The central tube in their setup included a resistance heater and an angular array of thermocouples. Using a variable power supply, this heat transfer probe was held at a constant temperature. The variables calculated and reported include the surface temperature and heat transfer coefficients (time-averaged and instantaneous) that should provide sufficient data for validation. Additionally, integrating heat transfer into the previously validated bubbling bed unit problem will save time by eliminating the need for a redundant hydrodynamics validation case.

Another possible heat transfer validation study is that of (Yusuf 2011), where a numerical study on the heat transfer in a bubbling bed with immersed tubes was completed. The setup was based on (Olsson and Almstedt 1995)'s experimental work. The bed setup measured 0.2 x 0.3 x 2.1 m and contained forty-six 20 mm diameter horizontal tubes in a staggered pattern. The solid phase was silica with a mean diameter of 700 μm and density of 2600 kg/m³. Similarly to (Kim 2003)'s experiment, one tube was set as the heat transfer probe and measurements were recorded around the perimeter. The void fraction and heat transfer coefficients (local and time averaged) are reported and can be used in conjunction with the original data for validation.

This paper has several favorable features: experimental data is available for the same setup, the simulation uses an Eulerian-Eulerian framework, the effect of bed pressure is reported, and the geometry is similar to the geometry in the adsorber. The unfavorable features include: large particle diameter, the poor representation of data, and the approximation of round tubes as square tubes. The tube-geometry approximation will undoubtedly cause some discrepancies when compared with the experimental data. For these reasons our initial heat transfer validation studies will focus on (Kim 2003), and if necessary, we will use the studies presented by (Yusuf 2011).

The particle size is an important parameter in heat transfer which should also be considered in our validation studies of the heat transfer unit problem. At this time we have not identified any appropriate validation studies or experimental data investigating the effects of particle size; however we will continue to look for appropriate cases and will consider any identified cases in our validation studies.

Chemical Reactions: The CCSI full scale carbon capture system is being designed for use with an NETL-developed sorbent material known as 32D. This material is new, so there is little data available on its behavior in a carbon capture system. Currently Element 1 is developing reaction models for 32D based on experimental data. Additionally, Element 1 is working with Element 6 on validation and uncertainty quantification studies of the recently developed chemical kinetic models. These models have been implemented into the Element 2 CFD models. The results of the UQ analysis will be used to propagate the uncertainties of the reaction models to the full scale model. Additionally, experimental operation of 32D in the C2U system is also planned and will be used to validate the CFD chemistry model under operating conditions similar to the design conditions of the full scale system.

MFIX has been used previously to model a number of reactive systems. (Das 2004a,b) completed a two-part study on dilute phase gas-solid riser reactors. Part two details the simultaneous adsorption of SO_2-NO_X from flue gases. One- and three-dimensional simulations were compared to the experimental data. The main reaction variable investigated was the SO_2 and NO_x removal and SO_2 and NO_x concentration. The one-dimensional simulation results were in good agreement with the experimental data. The three-dimensional simulation tended to under predict the removal values.

(Syamlal and O'Brien 2003) studied the decomposition of O_3 in a bubbling fluidized bed using MFIX. The simulation was based on the experimental work of (Fryer and Potter 1976). (Syamlal and O'Brien 2003) successfully simulated the hydrodynamics of the process; however, the reactive species concentration profiles were not in as good agreement. The O_3 concentration profile reached a minimum significantly closer to the base of the bed than (Fryer and Potter 1976) observed.

Both (Das 2004a,b) and (Syamlal and O'Brien 2003) investigated the accuracy of modeling reacting multiphase flow with MFIX. Although they consider different chemical systems than those considered in CCSI, they provide insight into the ability of MFIX to accurately model reacting multiphase flows. These papers show that MFIX is able to predict the appropriate trends, but may not be able to predict the exact quantitative results. This will be investigated further as CCSI implements the Element 1 reaction models and works with NETL's C2U system on CCSI specific laboratory scale experiments.

CO_2 Reactions with Amine: (Mebane 2012, Lee 2012) developed a reaction model for the reactions between CO_2 and an amine sorbent (32D) particle over a range of temperatures from adsorption data obtained by thermogravimetric analysis (TGA). Their model is a function of gas and solid species concentrations and local temperature. They validated the model with TGA data at various temperatures and discuss the applicability and shortcomings of the simple model. Additionally they have worked with Element 6 to investigate the uncertainties of their model.

4.2.2 Upscaling

It has been shown that local instabilities in fluidized systems affect the global flow behavior (Agrawal 2001, Igci 2012a). These instabilities are in the form of dynamic particle clusters that form and disintegrate with time (Figure 2). Since these clusters strongly affect gas-particle drag, effective viscosity, effective pressure drop, and effective reaction rates, among other properties, CFD simulations need to capture clustering behavior accurately to obtain quantitatively reliable predictions. One strategy is to use a grid size comparable to the cluster size, which is approximately ten times the particle diameter (Agrawal 2001, Holloway 2012). However, for simulations of large devices, such as those considered in the CCSI program that are tens of meters in size, using a mesh size ten times the particle diameter (1 mm - 1 cm) is computationally infeasible.

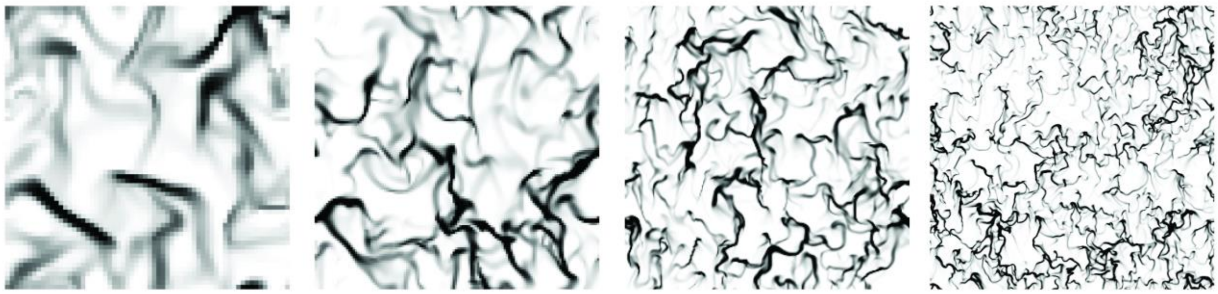


Figure 2: Snapshot of Instantaneous Particle Clusters, from (Igci 2012a) with Resolutions (left to right) 64×64, 128×128, 256×256, and 512×512 Grids. Local Instabilities, i.e., Particle Clusters, are Resolved Better at Higher Resolutions.

An alternate strategy to incorporate the influence of these small-scale clusters in coarse grid simulations is to use correction factors obtained by performing highly resolved simulations of a smaller representative system, then 'filtering' out the necessary correction factors to be used in coarse grid simulations. It should be noted that the filtering operation is dependent on the property of interest being corrected. Hence, the filtering operation needs to be performed separately for each of the conservation equations (mass, momentum, and energy) and for each of the constitutive relationships used. For the CCSI system, four sets of fundamental physical behavior are identified for filtering/upscaling gas-particle flow in the adsorber, flow in the regenerator, reaction kinetics, and heat transfer. Filtered models will be developed and validated for each physical behavior as part of the V/UQ work in collaboration with Element 2. Below we describe some of the available filtered/upscaling relationships, the shortcomings of the available relationships with respect to CCSI devices, and the proposed future efforts by the CCSI team.

Flow Upscaling in the Adsorber and Regenerator: Although both devices operate in very different fluidization regimes, with larger fluidizing velocities in the adsorber compared to the regenerator, a single model encompassing all regimes may be developed. In a series of papers by the multiphase CFD group at Princeton University (Agrawal 2001, Andrews 2005, Igci 2008, Igci 2011, and Igci 2012b), a model for filtered gas-particle drag, effective viscosity, and effective pressure drop has been developed. For validation, (Igci 2012a) compared predictions from a 3D riser model with the available experimental data from the Particulate Solid Research, Inc. (PSRI), facility in Chicago. Given the inherent (and unquantified) uncertainty in the experimental data, the filtered models for flow were able to predict the pressure gradient, particle holdup, and solid mass flux with reasonable accuracy. Development of similar filtered equations and validation efforts are also underway at the Université de Toulouse (Parmentier 2011). The validation efforts for the filtered flow models are far from complete and need to be tested for a wider range of particle/gas properties and model parameters. As a comprehensive validation effort is beyond the means of the CCSI effort, the validation team will attempt to validate some of the upscaling cases but will also rely upon independently published validation of these models.

Validation studies of the flow upscaling methodologies are currently ongoing. As part of the CCSI program, collaborators at Princeton University are investigating the accuracy of their filtered models (Agrawal 2001, Andrews 2005, Igci 2008, Igci 2011a, Igci 2012b) in comparison to experimental fluidized bed data. They are currently investigating three cases based on two NETL challenge problems and experimental data from (Zhu 2008). The cases consider different flow regimes, including a circulating fluidized bed, a bubbling bed, and a turbulent bed. Initial comparisons to the experimental data show that the filtered models perform relatively well; however, they do show sensitivity to different model parameters. This can be seen in the case of the circulating fluidized bed where the length of the exit pipe greatly affects the accuracy of the results (Figure 3 and Figure 4).

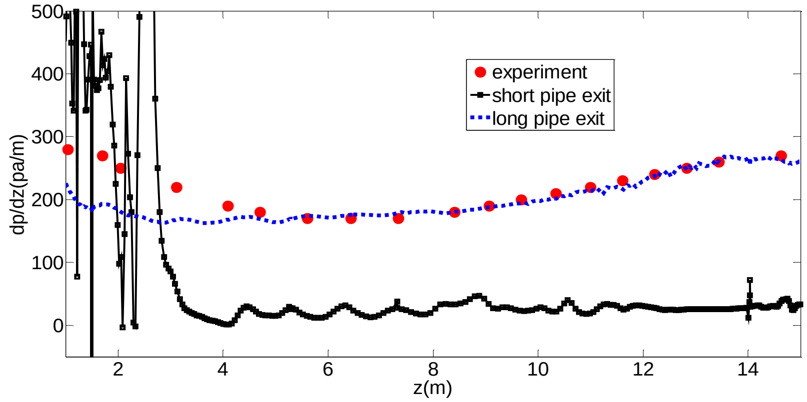


Figure 3: Distributions of Axial Pressure Gradient

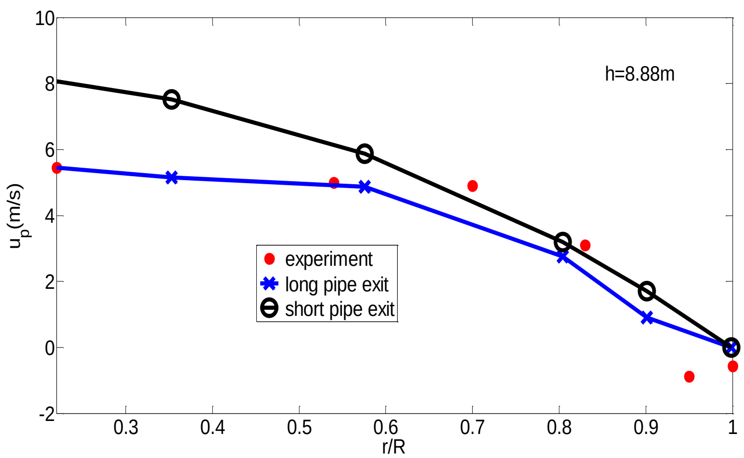


Figure 4: Distributions of Particle Velocities

The CCSI design also consists of an array of cooling rods in the adsorber and perforated porous plates in the regenerator. As the coarse grid full scale simulations cannot resolve every rod or hole, filtered models for upscaling the macroscopic influence of these internal geometry features will also be developed. Even though there have been a few validation studies on flow around a bundle of cylindrical tubes (for example (Holloway 2012)), there has been no progress towards development and validation of upscaling relationships. Element 2 has begun the development of a filtered model for the effects of heat transfer tubes in the adsorber. Validation of the filtered model will rely on the available literature data such as (Holloway 2012) and (Kim 2003).

Reaction Upscaling: Formation of clusters significantly affects the effective reaction rate in a unit volume of a fluidized bed (adsorber and regenerator). The available literature offers very preliminary information pertaining to upscaling of chemical kinetics (Holloway 2012). The reaction filtering method described in (Holloway 2012) is limited to a simple first order reaction catalyzed by the solid phase. In contrast, the particulate phase in the CO_2 capture reaction is a reactant and not a catalyst. A model for the chemical kinetics of CO_2 capture is described in a CCSI document authored by Element 1 (Mebane 2012), which is comprised of multiple reaction stages that include second and third order kinetics. Starting with the kinetics filtering approach described in (Holloway 2012), an upscaling approach for the more complex CO_2 capture reactions will be developed and validated. The validation may be obtained from experiments utilizing simpler geometries, as well as devices/materials directly related to CCSI efforts; either approach is suitable for validating the filtered reaction models. At this stage, the filtered flow equations described earlier may be included with the upscaled reaction models (as described in (Holloway 2012)).

Heat Transfer Upscaling: Although validation of the heat transfer unit problem has received some attention (see (Hamzehei 2010) for a recent example)), the CCSI validation team is unaware of any publication addressing the issue of upscaling heat transfer relationships in fluidized bed CFD simulations. Filtered heat transfer equations will be developed by the CCSI team in collaboration with Dr. Sankaran Sundaresan's multiphase group at Princeton. The first step in this process will be to assess whether filtered heat transfer equations are required, as small temperature variations need not necessarily be captured accurately; however, they may be compounded in an overall reactive system since the reaction rates and enthalpies of reaction are a function of temperature. The definition of 'small' with respect to the temperature range would depend on the sorbent and operating conditions. Once filtered heat transfer equations are developed, the CCSI team will focus on validating these results based on available experimental heat transfer data for standard test problems (available from literature) and heat transfer occurring in the CCSI devices. As the existing literature does not address this important CFD problem, this step will also represent a significant advancement in the fundamental understanding of heat transfer in energy systems.

As a comprehensive validation of all available and developed upscaling methodologies is not possible, the CCSI validation team will identify and focus on the individual problems that have a greater impact on carbon capture. For example, if temperature ranges are found to be small, heat transfer may be ignored. Moreover, the CCSI team will also rely on independently published studies and academic partners to help validate existing upscaling/filtered models.

4.2.3 Decoupled Laboratory Scale Validation

The decoupled laboratory scale validation will focus on larger scale solid sorbent systems that can be used to evaluate the simpler physics of the unit problems combined with upscaling. Validation data from experiments and literature will be used. In particular, the experimental C2U at NETL will be used to assess the accuracy of a simplified operation of a carbon capture reactor. The C2U system is designed for validation and operational testing of solid sorbent carbon capture systems. The C2U consists of bubbling bed reactors and is instrumented with numerous sensors for thorough data collection. Results from the experiments planned on the C2U system will be used to validate the upscaling of the unit problems in a carbon capture system. This will include modeling the experimental validation cases planned by NETL, such as non-reactive cold flow where solid sorbent particles are run through the system without reactions at isothermal conditions, and also working with NETL to plan CCSI specific validation experiments. At this time both two dimensional and three dimensional CFD models of the fully-coupled C2U circulating system are being developed for validation and design studies.

Decoupled laboratory scale validation will help us establish quantitative confidence in our choice of unit problem models and our upscaling filters. By isolating specific larger scale unit problems, we will gain a better understanding of how the various physics of the carbon capture reactor contribute to the compounding modeling error without blurring the source of error between the upscaling and the coupling of unit problems.

Currently there are a number of experimental laboratory scale fluidized bed systems that have been reported in the literature that include experimental data on decoupled experiments. These cases can be used as initial validation cases for the models developed in CCSI. An example of a literature case that may be used for validation is the experiments of (Igci 2011b). In the study, the authors have performed a set of 3D computational simulations (using MFIX) for validation of the filtered two-fluid model against the experimental data which was generated in a 14.2 m tall circulating fluidized bed riser with a 0.2 m internal diameter located at the PSRI experimental research facility in Chicago (Sun 1999). The experimental data set was specifically designed for benchmark modeling exercises. The authors present the physical properties of the particles and the gas used in the computational simulations and the simulation settings that are compared with the experimental case. The authors also consider the 3D filtered model equations with and without wall corrections. System response quantities (measured and predicted) include time-averaged radial profiles of particle volume fraction, axial particle phase mass flux, and axial superficial gas velocity as well as time-averaged gas pressure gradient profiles. The authors analyzed and compared the experimental data with the filtered model and considered the effects of the filter length, grid resolution, and boundary conditions on the accuracy of the model.

4.2.4 Coupled Laboratory Scale Validation

The C2U system and literature data will also be used to consider laboratory scale coupled validation. The coupled laboratory scale simulations will consider the effects of coupling the unit problems on the accuracy of the models and will allow us to quantify their contributions to the overall error of the models. By comparing the results of the decoupled and coupled validation problems, we will be able to investigate the effects of coupling versus the effects of upscaling on the accuracy of the simulations. This will help in the overall validation evaluation for the device scale systems. By isolating the individual contributions of error, we will be able to identify areas of improvement for future model development and experimental research to facilitate reduction of uncertainty through model refinement and collection of additional data.

The coupled laboratory scale validation will focus on data collected from the C2U system and data available in literature. A few examples of available literature data are listed below:

Coupled Heat Transfer and Hydrodynamics in a Fluidized Bed (Ebert 1993): In order to study the role of particles in augmenting heat transfer from the wall of a circulating fluidized bed, the authors carried out simultaneous heat and mass transfer experiments in a 20 cm diameter circulating bed operating at atmospheric conditions. The measured variables included heat transfer coefficients as functions of superficial gas velocity and suspension density, as well as the effects of particle density on heat transfer coefficients. The results for the overall mass and heat transfer parameters were also presented.

Carbon Capture Reactor (Seo 2009): The authors investigated a carbon capture reactor which uses a potassium-based solid sorbent in a bubbling fluidized bed reactor. The authors measured the physical properties of the sorbent, such as pore size, pore volume, and surface area after carbonation or regeneration, to investigate the extent of the carbon capture reaction. An experimental reactor with an inner diameter of 0.05 m and a height of 0.8 m was used and included thermocouple measurements throughout the reactor. The experimental setup also measured the CO_2 removal profiles as well as the amount of CO_2 capture. Although this case uses an alternative sorbent material from that considered in the CCSI program, the experimental data could still be useful for coupled simulations of the reactions, hydrodynamics, and heat transfer in a laboratory scale carbon capture reactor.

4.2.5 Pilot Scale Systems

If data should become available for pilot scale carbon capture systems, we will consider the validation of coupled and decoupled pilot scale problems in a manner similar to the laboratory scale cases. Pilot scale data would allow us to further validate the upscaling methodologies and coupling of unit problems.

At this time an experimental pilot scale carbon capture system does not exist; however, ADA-ES is currently designing a 1 MWe carbon capture reactor. We will work closely with ADA-ES to support them in this effort and to collaborate on validation experiments if ADA-ES is willing.

4.2.6 25 MWe, 100 MWe, and 650 MWe Device Scale Systems

Using computational tools to virtually develop larger to full scale carbon capture systems is the main focus of the CCSI program. At the time this document is prepared, there does not exist (nor are there any plans for) a full scale solid sorbent carbon capture system. As such we will most likely not have experimental data available for the validation of our full scale reactor models. Should the situation change and a physical system be built, we will work closely with the industry partner or laboratory to obtain data for validation of our larger scale models.

Based on the currently anticipated availability of experimental data, the validation plan for the 25 MWe, 100 MWe, and full scale system will rely on the validation and quantitative confidence gained at the lower tiers of Figure 1. For our larger (> 1 MWe or laboratory scale) to full scale models, we will assume that once the physics coupling of the unit problems and geometric upscaling of our models have been validated at the laboratory scale (and pilot scale if available), the validity of the modeling methodology will hold true for larger to full scale systems. This assumption relies on thorough validation at the unit scale, upscaling, and laboratory scales which will be the main focus of the CCSI validation work. The validation work at the lower tiers will provide us with a confidence range for the full scale system and will allow us to separate out the contributions of geometric upscaling and unit problems coupling to the overall predicted error of the full scale system. This will help to identify areas for improvement in model development and experimental property and parameter measurement, which in turn will enhance our ability to understand and manage the uncertainty in the CFD models being developed to model carbon capture systems. The CCSI validation team will attempt to find experimental data for larger scale systems, which are or similar to the CCSI design for validation; however, this will rely on the availability of data at these larger scales.

5. Uncertainty Quantification

Element 6 will work with Element 2 to incorporate UQ analysis at the various stages of the validation hierarchy. UQ analysis will start at the unit problem level and will entail close collaboration with Element 2.

5.1 Overview of Uncertainty Quantification

The UQ process begins with a Definition phase in which models are specified, and goals and objectives for the uncertainty analysis (UA) are developed that will guide the selection of UQ tools needed to obtain information about uncertainty that is needed to support the overall modeling and decision-making framework. Possible sources of uncertainty, including both model uncertainty and data uncertainty, are identified by working closely with Element 2 members using available data and knowledge to narrow down the set of variables of interest. Once the variables of interest have been defined, parametric distributions and/or model forms are developed in the Characterization step that capture the available information about uncertainty. Uncertainty may then be propagated either forward or backward through specified models in the system. Various types of analyses may be performed to study the impact of the uncertainties that are present. For example, sensitivity analysis (SA) can be conducted to learn about the relative impacts of different input variables and their importance in driving the uncertainty in model outcomes. Finally, the information gained from examining the existing uncertainty can be used to improve the models themselves or to inform the collection of additional data to ultimately reduce the uncertainty present in the system.

In this section overviews of three critical components to UQ, (i) calibration (ii) validation, and (iii) UA/SA are provided in the setting of a single model (e.g., a single unit problem). Finally, these concepts are used to provide a hierarchical UQ framework.

5.1.1 Calibration

UQ of the unit problems, for example, will proceed by first identifying the relevant input variables (e.g., pressure, temperature, etc.), model (i.e., calibration) parameters, and output variables (e.g., amount of CO_2 absorbed) of the unit problem under consideration. Determining the relevant parameters/variables can be accomplished by a combination of examining the findings of previous work in the literature and through conversations with modelers (Element 2). In conjunction with identifying relevant input variables and model parameters, appropriate distributions/ranges for the values of these quantities for the particular CFD application can be specified.

At this stage, an assessment will be made of how many model runs are computationally feasible for calibration and uncertainty characterization. If feasible, it would also be helpful to make some runs (e.g., from an LHS sample) of the model at this stage and perform SA (Helton 2007, Storlie 2009) to confirm and expand our understanding of how sensitive the outputs may be to the various model parameters. This approach is currently being taken with the first unit problem (bubbling bed), as discussed in Section 6.

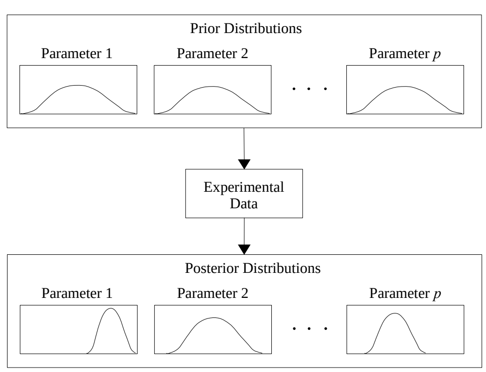


Figure 5: Overview of Bayesian Calibration

Once the relevant parameters are understood, an appropriate calibration scheme can be developed. A Bayesian-like calibration (Kennedy 2001) will be used, which will most likely need to explicitly deal with (1) emulation of the model output (Kennedy 2001, Higdon 2004), (2) treatment of functional output (Bayarri 2007, Higdon 2008), and (3) calibration parameter screening/selection (Bayarri 2007, Higdon 2008). A Bayesian calibration works by treating the model parameters as random variables and first defining a prior distribution for them (top of Figure 5). The computational model is linked to the experimental data through a statistical regression model, and the model parameters are then conditioned on the experimental data to provide a *posterior* distribution of likely parameter values that best describe the experimental data (bottom of Figure 5). In reality there is really one joint distribution of the model parameters. Figure 5 and subsequent figures of this type depict the entire joint distribution as several marginal distributions only for ease of presentation. The results of the calibration scheme then is (1) a posterior distribution characterizing the uncertainty of model parameters, and (2) an estimated model discrepancy to the observable process. A more detailed description of this process is provided in Section 6.2.

5.1.2 Validation

Model predictions will be compared with experimental data to check for any non-negligible bias or lack of fit. This can be accomplished by using the calibrated model to predict the data (we would predict data points that were **not** used in the calibration) and see how well we are doing at prediction of "out-of-sample" data points. In statistics this concept is called cross-validation and is a well-accepted means to "validate" a predictive model. The reason for using cross validation is that out-of-sample model predictions do not allow the predictions to “cheat” by tuning the model parameters to over-fit to the experimental data. These out-of-sample predictions will then be compared to the data in a model assessment framework such as those discussed in (Oberkampf 2006) and (Romero 2011). Essentially this boils down to the following scenarios represented in Figure 6 taken from (Romero 2011). In these cases a scalar output is assumed and there is uncertainty due to measurement error in the experimental data. There is also simulator output uncertainty due to the posterior uncertainty distribution of the calibrated parameters. These two forms of uncertainty are represented as confidence bands in the figure. It then comes down to expert judgment whether or not the model is performing well enough to be considered validated in these cases. If the uncertainty bands are two wide to make such a decision, then it is necessary to collect more experimental data (more replicates if the measurement error is making the experimental observation bands too wide or more space filling design points if the uncertainty in the calibration parameters is making the simulator output bands too wide).

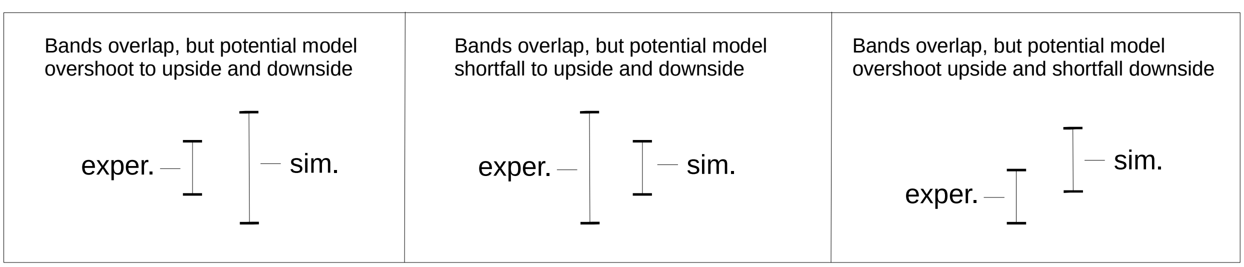


Figure 6: Generic categories of comparison of uncertainty intervals between experimental versus simulation results for a scalar output quantity.

If the simulator prediction bands and the experimental observation bands do not overlap at all, then there is some lack of fit. If the magnitude of the lack of fit is substantial enough to be deemed non-negligible by subject matter experts, than model improvements will need to be made. If necessary and/or feasible, an estimated model discrepancy can be used to guide model improvements until it is determined that the model is adequate, or that a model discrepancy needs to be incorporated somehow into the final characterization of the model.

5.1.3 Uncertainty Analysis and Sensitivity Analysis

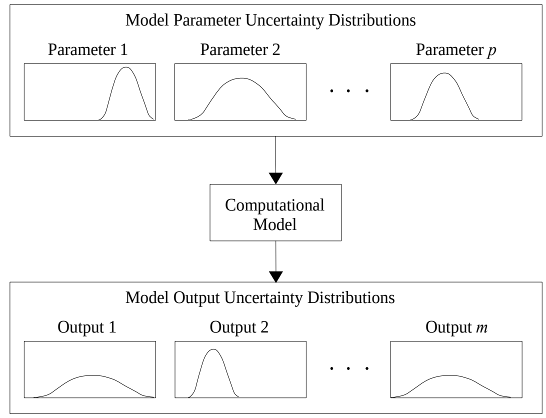


Figure 7: Propagation of Uncertainty in Model Parameters to Model Output

Whether or not experimental data are available, it is usually necessary to perform UA and SA on the model. UA is the process of propagating the uncertainty in the model parameters through to the outputs of interest (e.g., % CO_2 captured for the full scale system) to obtain a distribution of model output. If experimental data are available, the uncertainty in the model parameters refers to the posterior distribution resulting from the calibration. If experimental data are not available, the uncertainty in the model parameters refers to the prior distribution, either obtained from expert knowledge or a previous calibration, etc. In either case, propagating the uncertainty in model parameters to model output can be performed in the simplest case with Monte Carlo sampling as follows. First, sample a set of values for the model parameters from their uncertainty distribution (top of Figure 7), and then run the model with these values to obtain the relevant outputs. Repeat this process many times to obtain a sample from the resulting distribution of the outputs (bottom of Figure 7). A SA is also performed at this point to further understand the model (e.g., ascertain which model parameters contribute most to the uncertainty in the output, and ensure that the model behaves as expected when varying the model parameters, etc.).

It is very often the case that Monte Carlo sampling of the computer model is impractical due to limited computational resources. In this case, the UA and SA will need to be done using an emulator (i.e., a statistical approximation to the actual model) as in (Kennedy 2001, Higdon 2004, Storlie 2009, and Reich 2009) to perform the required Monte Carlo sampling. A final issue to consider is that we may need to distinguish aleatoric (e.g., associated degradation/attrition of sorbent material) and epistemic (e.g., intrinsic imprecision in the known values of model parameter) uncertainty sources as in (Helton 2000) when performing UA/SA.

5.1.4 Hierarchical UQ

UQ of the pilot and laboratory scale problems will proceed by again identifying the relevant input model parameters and output parameters that will likely be a subset of those parameters involved in the unit problem models. The result of UQ for the unit problems is a model form for each unit problem along with a posterior distribution of likely values for the corresponding model parameters. Therefore, the prior distributions of the model parameters at this stage will be their respective posterior distributions resulting from the previous calibration of the unit problems. A schematic providing an overview of the UQ hierarchical framework is provided in Figure 8. An assessment will be made of how many runs of the model are feasible for calibration and uncertainty characterization. Once again, a calibration scheme will be developed and applied to the experimental data to produce a posterior distribution characterizing the uncertainty of (1) model parameters, (2) model discrepancy to the observable process, and (3) the relevant outputs. Model predictions will be compared with experimental data (e.g., C2U experiments) as in the UQ of the unit problems to check for any non-negligible bias or lack of fit, and model improvements can be made if needed.

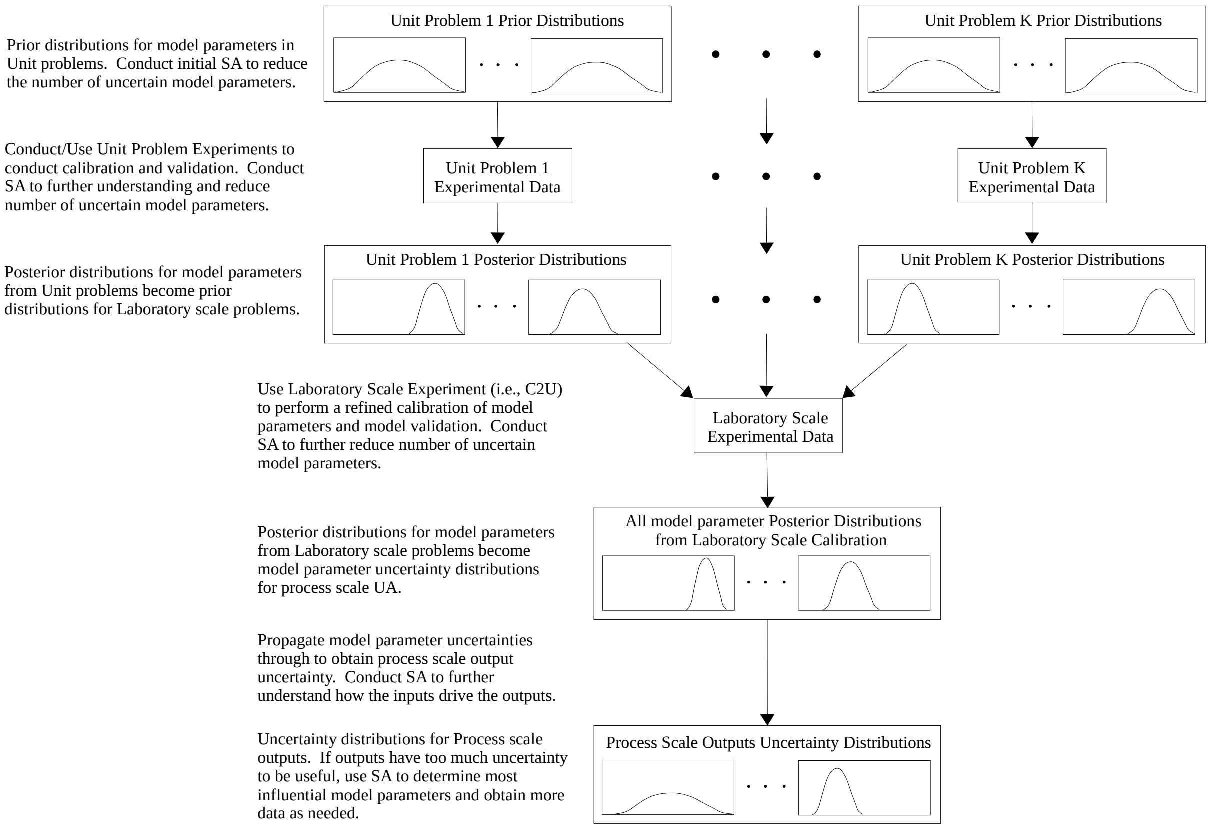


Figure 8: UQ Hierarchy Framework

In the hierarchy of moving from unit problems to laboratory scale problems, or laboratory scale to full scale, there are several models G_1, G_2, ..., G_K that are used to create a higher level model H where the term "used" does not mean that G_K is necessarily a sub-model of H. For example, suppose model G_1 is implemented via a CFD code (MFIX in this case) to model a case with only some of the physics present in the full scale case, e.g., flow with heat transfer, but not chemical reaction or isothermal chemical reaction. Model G_1 is run for cases where experimental data exists, and the model can be calibrated. These experimental conditions (geometry, temperature, etc.) will differ somewhat from the full scale conditions where we wish to run model H. Model H (up the hierarchy) uses MFIX again, but with all the physics coupled, and for a system for which we have no experimental data. However, again the model for the full scale system will have model parameters which are a subset of the model parameters involved in the laboratory scale problems. The Gs used to create H will depend on those model parameters. We can evaluate H at the model parameter values deemed appropriate by calibrating G_1,...G_K to generate full scale predictions. These predictions will be only as good as the assumption that the full scale model can be constructed with confidence and that it will have sufficiently accurate prediction capability based on the G_1, G_2,..., G_K, and the results of the laboratory scale problems. This assumption is virtually impossible to quantitatively verify without experimental data. However, we can get a qualitative measure of the plausibility of this assumption based on the results encountered during the upscaling from the unit problems to laboratory scale (and pilot scale if data is available) problems, for example.

Therefore to conduct UQ for the full scale model, the model parameters for the full scale model will be assigned their respective posterior distributions resulting from their last level of calibration data. These distributions will then be used to perform an UA by propagating the uncertainty in the model parameters through to the outputs of interest (e.g., % CO_2 captured) for the full scale system to obtain a distribution of model output. Propagating the uncertainty in model parameters to model output is performed in the simplest case with Monte Carlo sampling as follows. First, sample a set of values for the model parameters from their uncertainty distribution (top of Figure 7), and then run the model with these values to obtain the relevant outputs. Repeat this process many times to obtain a sample from the resulting distribution of the outputs (bottom of Figure 7). A SA will also be performed at this stage to obtain a distribution of model output.

6. Initial Validation and UQ Studies of the Bubbling Bed Unit Problem

A preliminary V/UQ study of the bubbling bed unit problem has begun. This initial study will be used to assess the validity of the MFIX implementation of a bubbling fluidized bed and to lay the foundation for the collaborations between the CFD modelers of Element 2 and the UQ team of Element 6. Work has already been completed on the mesh size and simulation time sensitivity of the unit problem. Ongoing work considering a SA of various model parameters will be used for calibration of the model.

The initial study of mesh size sensitivity allows us to reduce the total cell count of the simulation allowing for shorter simulation times. The sensitivity of the model to simulation time has also been investigated. Below we discuss the time needed for the bubbling bed to reach a statistically stationary state. The simulation results are averaged over time for comparison with the experimental data. The sensitivity of the simulation to the size and location of the time window for averaging was investigated for the bubbling bed unit problem. The details of the sensitivity analyses will be discussed later in this section.

6.1 Simulation Details

The following subsections describe the details of the MFIX simulation of the (Kim 2003) experiments.

Simulation Domain: The domain of the bubbling bed is divided into two sections, the bed and the freeboard as shown in Figure 9. The bed is defined as the lower section of the system where the solid phase primarily lies. The freeboard is the upper section of the system, occupied by gas and any solid particulate that is ejected upwards. The fluidization gas enters at the bottom of the bed and flows through the solid phase until it reaches the freeboard. The freeboard has a fixed-pressure outlet on the top where the gas flow exits the system. Within the bed region are twenty-five heat transfer tubes arranged in a staggered array following (Kim 2003), as shown in Figure 9a along with the dimensions of the tube array.

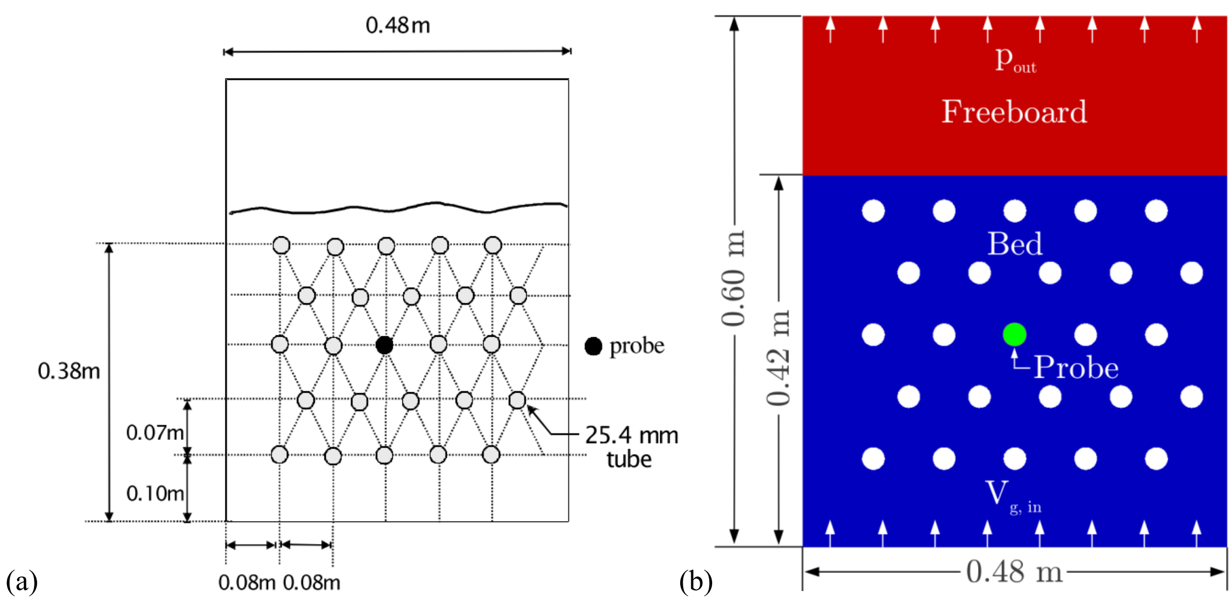
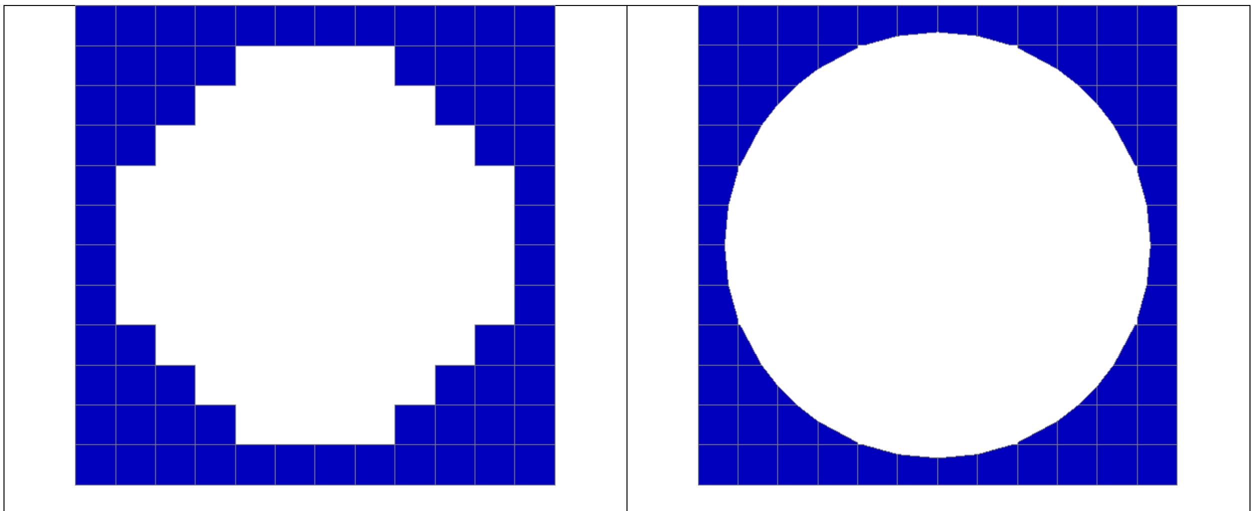


Figure 9: The Bubbling Bed Domain; the Experimental Setup of (Kim 2003) is Shown in (a) and the Simulation Domain is Shown in (b). In (b) Blue Represents the Solid Particle and Red the Gas.

Numerical Method: MFIX is used to model the bubbling bed unit problem. The unit problem focuses on validating the hydrodynamics of the bubbling bed, and as such, only the multiphase hydrodynamic equations are solved in MFIX.

MFIX offers two methods for defining geometry, traditional or cut-cell. The traditional method uses a Cartesian grid and approximates curved surfaces by defining entire cells as part of the geometry. The resulting geometry has "stair step" edges. At high grid resolution, the stair step approximation can be sufficiently accurate; however, at low grid resolution the approximation is poor. The cut-cell method approximates the geometry by dividing cells into quadrilaterals or pentagons. These new cut-cells provide significantly better approximations for geometry compared to the traditional method given the same grid resolution. A comparison of the traditional and cut-cell approximations for a tube can be seen in Figure 10. Simulations of both the cut cell and traditional grid methods were performed. The better resolution of the circular tubes in the cut cell method resulted in more accurate results when compared to the experimental data of (Kim 2003). As such, for the simulations discussed in this section only the cut cell mesh is considered.



(a) Traditional	(b) Cut-cell

Figure 10: Discretization and Approximation of Tubes for the 50K mesh resolution.

Grid Resolution: Several grid resolutions were studied to determine the lowest resolution that gives accurate results when compared to (Kim 2003)'s data. Reducing the grid size allows quicker run times, which will allow us to consider additional simulations for UQ analysis. Four grid resolutions were simulated with 20K, 30K, 40K, and 50K cells. These grid resolutions correspond to cell lengths of 3.8, 3.1, 2.7, and 2.4 mm, respectively. The simulation results for each of the cases were compared to the experimental data, and the sensitivity of the result was analyzed. The results of these analyses will be presented later in this section.

Experimental Data: The experiments of (Kim 2003) considered five different superficial gas velocities, 0.055, 0.07, 0.11, 0.126, and 0.161 m/s. The results for the superficial gas velocity of 0.126 m/s were reported in the most detail and therefore are used for comparison to the simulations.

Experimental data on the bubble frequency, bubble phase fraction, and emulsion phases contacting time were presented in (Kim 2003) at five points around one of the heat transfer tubes (shown in black in Figure 9a). As shown in Figure 11, the measurements were taken at +90, +45, 0, -45, and -90 degrees.

Data was collected for 28 seconds and was averaged over a time window of 14 seconds that ran from 14 to 28.

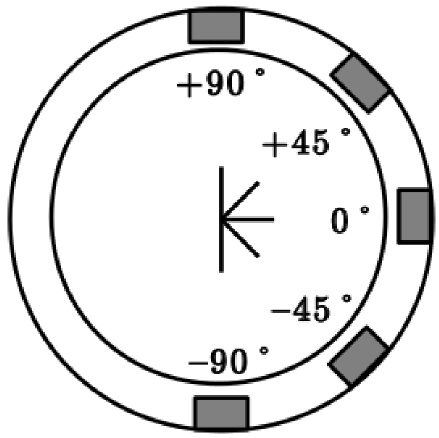


Figure 11: Measurement Probe of (Kim 2003)

Grid Resolution Study: The MFIX simulation domain shown in Figure 9b was run under the operating conditions of the experimental setup of (Kim 2003,), and the data for bubble frequency and phase fraction was calculated as in Equations 4 and 5 of (Kim 2003). The calculation is based on the predicted instantaneous void fraction at each time step *(*$\Delta t = 0.01\ s$). The results of the MFIX simulations are compared to the experiments in Figure 12 for a superficial velocity of 0.126 m/s and a fixed time window. Figure 12 compares the computed bubble frequency and bubble phase fraction for each of the MFIX simulations with the experimental data presented by (Kim 2003). These values are calculated from the instantaneous void fraction predicted by MFIX. To calculate the bubble frequency and phase fraction a cutoff threshold was defined to define a bubble in the bed (Li 2010). This calculation gives the impression that the lower resolution simulations can provide better predictions than the higher resolution simulations. However by analyzing the void fraction output directly as will be discussed in Section 6.2 it can be seen that the higher resolution simulations are less sensitive to the mesh resolution than the lower resolution simulations.

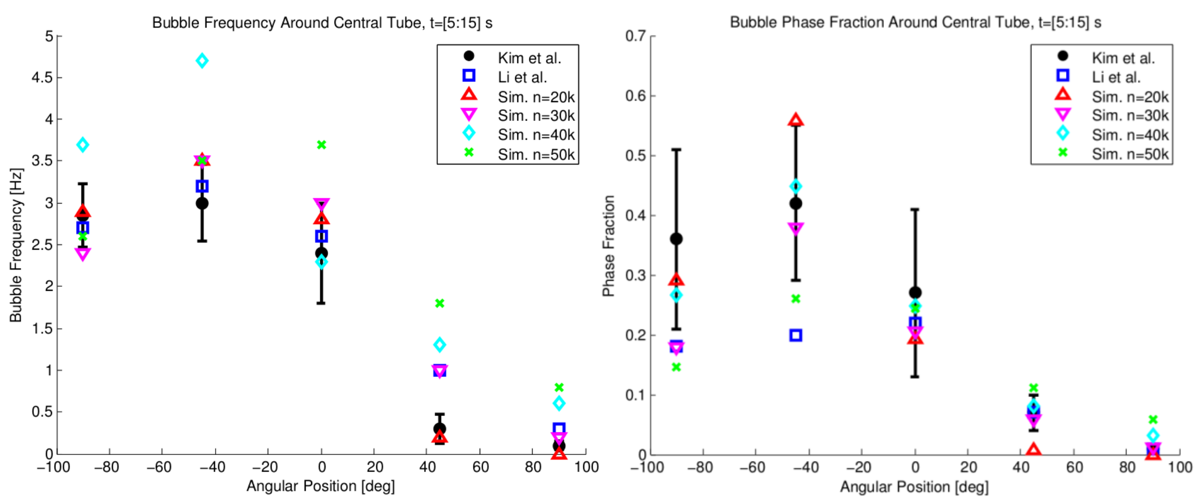


Figure 12: Plots of the Bubble Frequency and Bubble Phase Fraction for Each of the Mesh Resolutions Considered. The Black Circles are the Experimental Data and Uncertainty taken from (Kim 2003). Data is Shown for 0.126 m/s Superficial Velocity.

The 50K cell simulation was also run at a superficial velocity of 0.11 m/s. As seen in Figure 13, the simulation is able to more accurately predict the experimental data at 0.11 m/s than at 0.126 m/s.

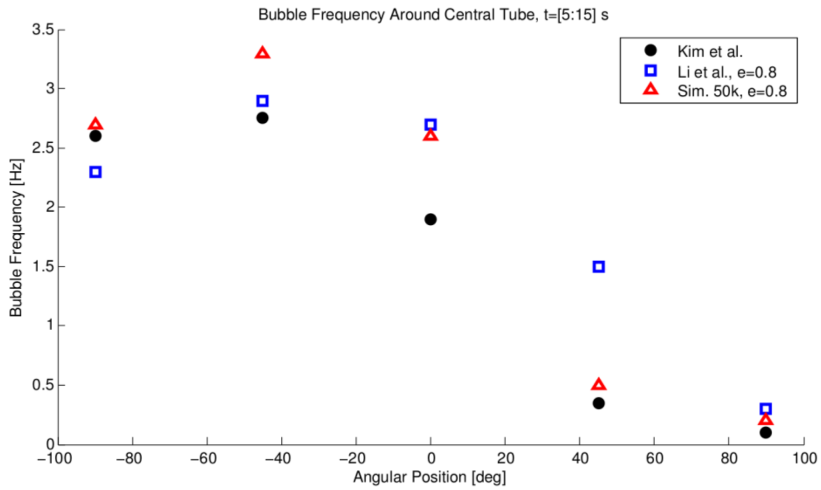


Figure 13: Plots of the Bubble Frequency and Bubble Phase Fraction for the 50K Cell Resolutions and a Superficial Velocity of 0.11 m/s. The Black Circles are the Experimental Data and Uncertainty taken from (Kim 2003).

Time Window Sensitivity: Additional simulations were run with the 50K cell mesh simulations to investigate the sensitivity to the time averaging window. The simulations were run for longer times and the simulation data was averaged over different time windows to investigate the sensitivity of the model to the simulation timeframe. For the 50K cell mesh simulations the data was analyzed up to 50 s and several averaging windows were considered. As seen in Figure 14, the size and position of the averaging window had little effect on the simulation results.

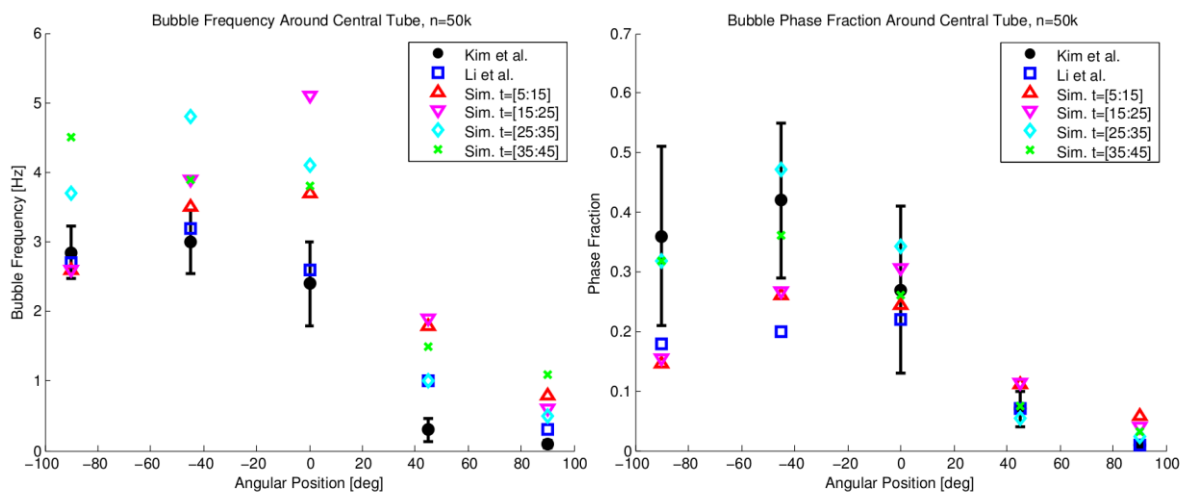


Figure 14: Plots of the Bubble Frequency and Bubble Phase Fraction for Different Time Windows Considered. The Black Circles are the Experimental Data with Uncertainty taken from (Kim 2003). Data is Shown for 12.6 cm/s Superficial Velocity.

Parameter Sensitivity: Ongoing work is underway to investigate the sensitivity of the bubbling bed unit problem simulations to MFIX model parameters. The model parameters of interest have been identified based on discussions between Elements 2 and 6 and the V/UQ sub-team. Initial simulations will focus on the model parameters shown in **Table 1**. The mesh and time sensitivity simulations were calculated using the initial values, while the remaining SA will consider the ranges and values shown in the table. The weighting of continuous variables is described by a range (R) and a mode (M).

Table 1: Parameters and ranges for UQ analysis.

Model Coefficients	Initial Values	Distribution of Values for UQ Analysis
θ_1 = Restitution (Particle-Particle)	0.9	R=[0.8,0.997], M=0.9
θ_2 = Restitution (Particle-Wall)	0.9	R=[0.8,0.997], M=0.9
θ_3 = Friction Angle (Particle-Particle)	30.0	R=[25,45], M=28.5
θ_4 = Friction Angle (Particle-Wall)	30.0	R=[25,45], M=28.5
θ_5 = Packed Bed Void Fraction	0.52	R=[0.4,0.6], M=0.5
θ_6 = Drag Model	Syamlal & O'Brien	
Syamlal & O'Brien		33.4%
Gidaspow		33.3%
Wen & Yu		33.3%

Initial simulations based on a simulation matrix developed by Element 6 have been started and will be used to calibrate the model as discussed in the UQ overview section (Section 5). As was done in the initial mesh and time sensitivity simulations, the void fraction, bubble frequency, and phase fraction will be compared between simulations, along with the instantaneous void fraction that is a direct output of MFIX. Initial plans call for 50 simulation runs; however, 25 cases will be run initially, and the results from the analysis of these cases will be used to adjust the number and focus of the simulations.

6.2 Uncertainty Analysis (UA) of Initial Unit Problem Simulations

UQ for the bubbling bed and any unit problem starts with the identification of the relevant model parameters along with their prior distributions, and the relevant model inputs and output(s). In this case, six model parameters $\boldsymbol{\theta} = [\theta_1,\dots\theta_6]$ were determined to be relevant, θ_1 = Restitution (Particle-Particle), θ_2 Restitution (Particle-Wall), θ_3 = Friction Angle (Particle-Particle), θ_4 = Friction Angle (Particle-Wall), θ_5 = Packed Bed Void Fraction, and θ_6 = Drag Model.

The experimental data for the hydrodynamics has two inputs (x_1 = placement angle of the sensor on the cooling tube, x_2 = superficial velocity) and three outputs (y_1 = bubble frequency, y_2 = bubble phase fraction, and y_3 = emulsion contact time). In our UQ studies, the lone model input of interest for calibration in this case is x_1. As mentioned previously, several superficial velocities were considered; however, the most complete set of data was taken at 0.126 m/s, so this is the velocity that will be considered in the UQ analysis. For the outputs, only y_1 and y_2 are considered, as y_3 is not reported for all cases or in as much detail as y_1 and y_2. The goal of the initial UQ analysis is to find good plausible values for θ that allow the model output y_m to match the experimental data y well at the observed input x_1 locations. In order to accomplish this, we must make several model runs at various $\boldsymbol{\theta}$ locations to assess how the model output varies over $\boldsymbol{\theta}$ space, and ultimately decide what part of $\boldsymbol{\theta}$ space is likely in the sense that it allows the model to match the data.

The model is somewhat computationally demanding, in that it takes approximately two days to make a run. Therefore we made some initial runs with $\boldsymbol{\theta}$ fixed at the values shown in **Table 1** to assess the impact of some of the model controls affecting run time, grid size, and simulated time. In particular, we ran the model at four different grid sizes of 20K, 30K, 40K, and 50K cells.

The 50K cell grid was considered the "gold standard" based on its use in previous work (Li 2010), so the aim here was to see how coarse of a grid we could use and still produce results very similar to the 50K cell grid. Another question to be answered is how long of a simulation time is needed for the process to arrive at a statistically stationary state? Figure 15 provides a graphical summary of the *void fraction* (the actual output of the MFIX model, which is then converted into bubble frequency and bubble phase fraction as discussed previously) averaged over four simulation cells around the 0 degree location on the probe tube for the four mesh sizes. These plots provide histograms of void fraction for the 0 degree location across all time steps of the model run. It is pretty clear that the distribution of void fraction (i.e., the likelihood of seeing a given void fraction value at a randomly chosen time step) is very similar for 40K and 50K cell mesh sizes, but possibly slightly different for the other two. The other angle locations presented similarly. We therefore decided on a 40K cell mesh size.

Figure 16 displays the distribution of void fraction averaged over the 0 degree cells for various time periods of the simulation. The earlier time periods appear to have distributions that are changing (through ~20 seconds).There does not appear to be any significant difference between any of the time periods after a time of 20 seconds. These findings were confirmed by comparing the distributions in the various time bins with Kolmogorov-Smirnov tests. The smallest *p*-value resulting from comparing the 20-25 second time period to the subsequent time periods was 0.27. We therefore decided to run the simulations out to 30 seconds (i.e., 10 seconds of steady state).

Since the model is expensive to run, the design of the $\boldsymbol{\theta}$ values at which runs are conducted is critical. For this, Latin Hypercube Sampling methodology (Helton 2003) is used to develop a design. This approach requires initial specification of distributions for the model parameters. Ultimately, this step would be necessary for the Bayesian calibration approach described below. The distributions of the model parameters were chosen based on previous knowledge and expert solicitation involving Element 2 team members, provided in Figure 17. Based on timeline and computational considerations, a Latin Hypercube Sample (LHS) of size 50 runs was chosen (split into two 25 run LHSs).Once these runs are available, they will be used in the Bayesian calibration scheme described below.

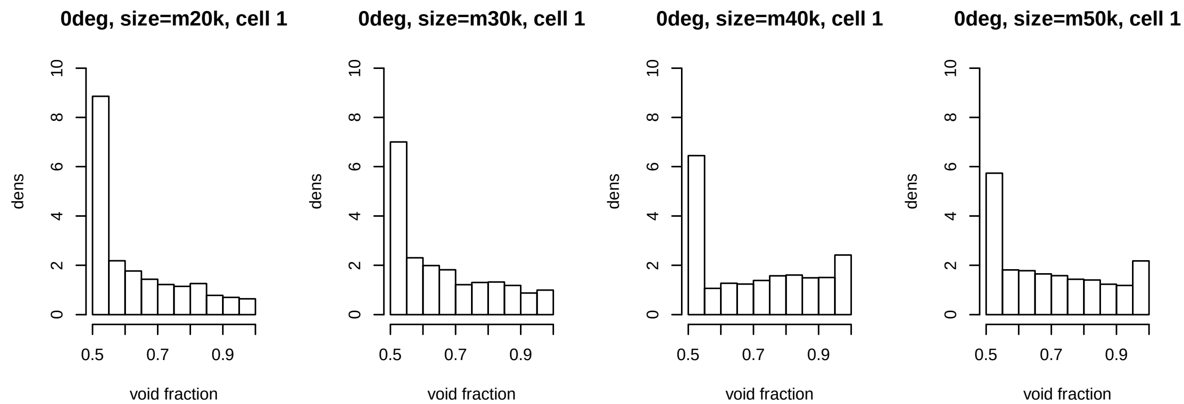


Figure 15: Distribution of Average Void Fraction for 0 Degree Cells for Various Mesh Sizes

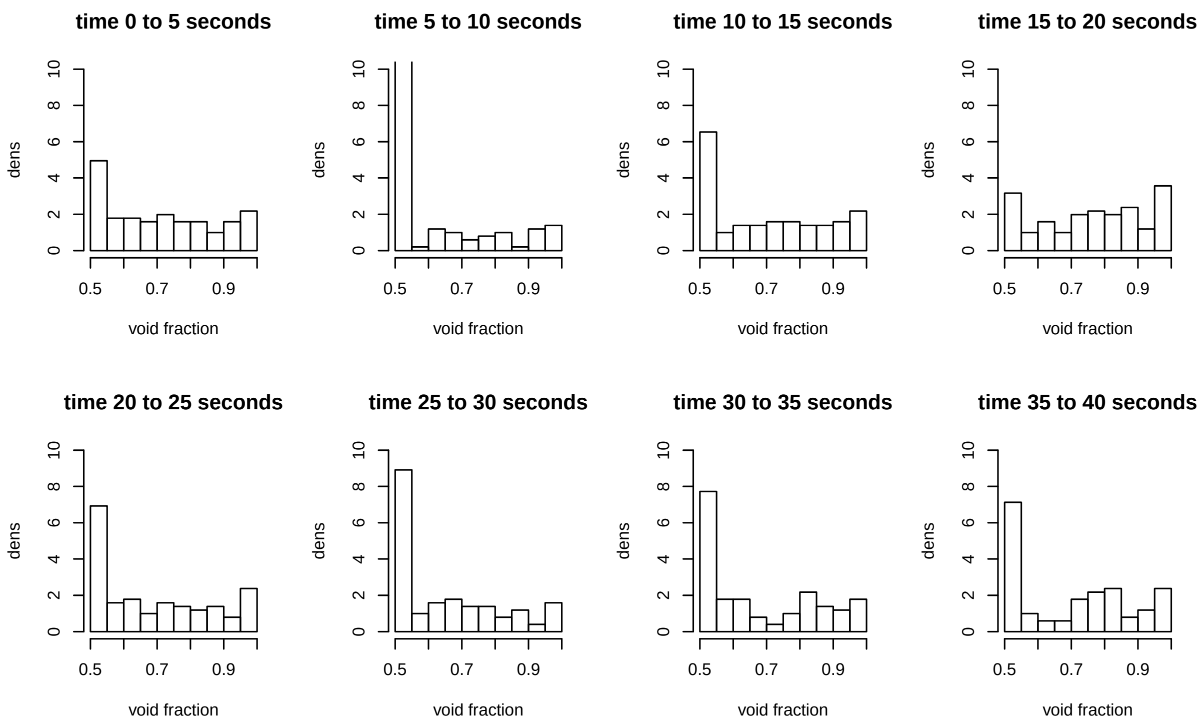


Figure 16: Distribution of Average Void Fraction of 0 Degree Cells for Various Time Periods of the Simulation

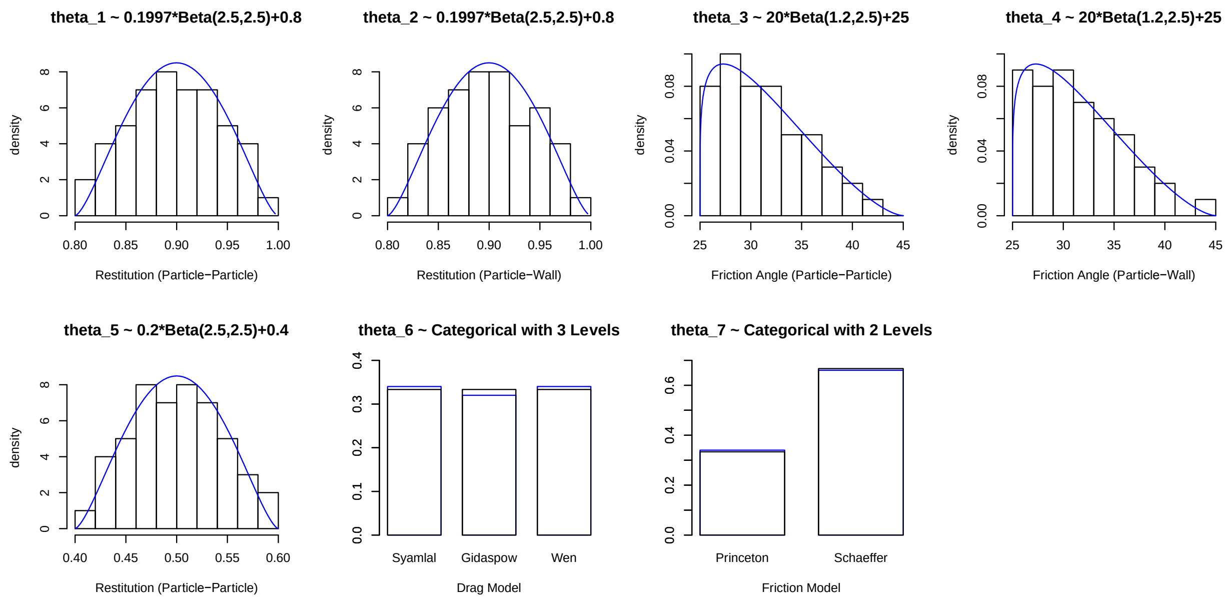


Figure 17: Prior Distributions of Uncertain Model Parameters for the Bubbling Bed Unit Problem. The Blue Lines Represent the Prior Distributions, and Histograms of the Resulting 50 Point LHS are Provided as Well

An overview of Bayesian calibration was provided in the previous section. Here the application of this approach as it applies to this specific unit problem is illustrated. Let $\boldsymbol{y}_i$ be a 2-vector of the observed bubble frequency and bubble phase fraction from the experimental data at angle locations x_i, $i = 1,\ldots,n$. We assume the following statistical model for y_i,

$$\boldsymbol{y}_i = f(x_i, \boldsymbol{\theta}) + \boldsymbol{\varepsilon}_i$$

where f is the MFIX calculation for bubble frequency and bubble phase fraction evaluated at $\boldsymbol{\theta}$, the "correct" or "true" value of the model parameters, and $\boldsymbol{\varepsilon}_i$ are measurement errors assumed to have a bivariate normal distribution $\boldsymbol{\varepsilon}_i \sim N(\boldsymbol{0}, \boldsymbol{\Sigma})$. Let $L(\boldsymbol{\theta}'; \boldsymbol{y})$ be the normal likelihood for a potential value of the model parameters $\boldsymbol{\theta}'$.The posterior distribution of $\boldsymbol{\theta}$ given the observed data $\boldsymbol{y}$, is $\pi(\boldsymbol{\theta}|y) \propto L(\boldsymbol{\theta}; \boldsymbol{y})\pi(\boldsymbol{\theta})$, where $\pi(\boldsymbol{\theta})$ is the prior distribution of theta which in this case is assumed to have the individual elements θ_i independent and distributed according to Figure 17. There is no closed form for $\pi(\boldsymbol{\theta}|y)$, as is most often the case, but we can sample from this posterior distribution with Markov Chain Monte Carlo (MCMC), which is a widely used approach in practice. The posterior sample can then be used to summarize the plausible values of $\boldsymbol{\theta}$, which could explain the experimental data, i.e., that would "calibrate" the model. One caveat to this procedure is that each evaluation of the likelihood L requires the evaluation of the MFIX model f, and in principle L will need to be evaluated thousands of times in the MCMC sampling routine, which is of course not practical when f takes ~2 days to run. The standard approach to deal with such situations is to use an emulator (or reduced order model) in place of f, which can allow for uncertainty in the actual value of f at unobserved locations. In this case, we would take the 50 evaluations of f from the LHS sample, and use those evaluations as additional observations in the likelihood function to fit an emulator (or surrogate or meta-model). This is described in detail in (Kennedy 2001) and (Higdon 2004).

Finally to "validate" the model, we generally intend to use "five-fold cross validation." This approach works by partitioning the experimental data into five disjoint subsets, then calibrating the model five times, each time holding out one of the subsets. The calibrated model is then used to predict the experimental data that were not used in the calibration (i.e., predict "out-of-sample"). If the model can predict the experimental data to within an acceptable tolerance (e.g., to within two standard deviations of the measurement error), then the model will be considered validated. If the prediction quality is too poor to be considered acceptable, then further refinements to the model can be considered, including the possibility of adding a discrepancy term to the MFIX model to allow it to make better predictions (Kennedy 2001, Higdon 2004, and Higdon 2008). Calibration and validation of all unit problems will proceed in a similar manner to that described here.

7. Limitations and Gaps

As discussed previously, the validation methodology laid out in this proposal can be considered as "application focused validation." This validation plan is meant to be specific to the carbon capture systems being considered as part of the CCSI program. As such, we have defined a very narrow scope for our validation activities that will allow us to complete the validation in the time frame available and with the funds available to us. This leads to certain limitations and gaps in the methodology that make this validation plan inappropriate for use on more generic multi-physics, multi-scale systems.

The first limitation is the unit problems selected and considered in the validation plan. Very specific unit problems are outlined in our validation plan and are based on the specific CCSI solid sorbent carbon capture system designs. Although some of the unit problems may apply to systems outside of the CCSI program, the overall analysis only applies to the CCSI specific systems and is not applicable to outside systems. Also, the unit problems that we have chosen to consider limit the validation plan to the currently considered CCSI designs. If extreme changes are made in the geometry, sorbent particles design, or flow regimes considered we may have to re-evaluate the validation plan.

Another limitation relating to the unit problems will be the availability of high quality experimental data and error estimates. We will be relying on previously published validation studies, and as such, will most likely not have direct access to the experimental data. We will only have access to the published information on the experiments and simulations. This may limit our ability to identify and quantify all of the errors in the unit problems. We will attempt to use the most complete validation studies available; however, we will most likely not be able to find high quality experimental data for all of the unit problems.

The second limitation is the proposed sequentially coupled approach for the overall validated uncertainty quantification goal. As mentioned above, this is proposed mainly due to the lack of experimental data with quantified measurement uncertainty at the unit problem scale. Hence the validation plan presented here aims first to provide quantitative confidence intervals on the CFD simulations for the baseline design. However, this analysis will be augmented by additional UQ studies where uncertain modeling parameters are varied within the bounds that are physically permissible by the problem, hence propagating uncertainties from input parameters to the various system performance metrics.

A third limitation is the inability to incorporate all parameters, and completely specify their associated distributions. In the initial definition phase of the UQ analysis, it is important to narrow down the set of parameters of interest to a modest size that is feasible for subsequent computations. The expensive function evaluations encountered in CFD studies necessitates focusing on a reduced parameter set. This results in a tradeoff between computational effort and the risk of excluding important parameters. The use of statistical experimental design techniques can ameliorate this situation by reducing the number of simulations required to examine high-dimensional parameter spaces. In addition, while expert knowledge can often be used to specify reasonable ranges for parameter studies, initial characterization of parameter distributions is generally approximate.

Another limitation of the validation plan is the gaps, i.e., omissions of the incrementally coupled cases, in the validation hierarchy laid out in this document. These include the consideration of the incremental coupling of the unit problems before upscaling, which would require a large amount of experimental data and numerous validation studies. The lack of data at the intermediate device scale (25 MWe, 100 MWe) is also a gap in available data that will most likely not be resolved during the validation process. Insufficient data at other scales may further limit the validation and UQ process.

Due to the gaps and limitations of the V/UQ studies proposed herein, we will rely heavily on the expertise and past experiences of the CCSI team members, especially those directly involved in Elements 2 and 6. Their understanding of the problems and insight from past experiences will help to guide the validation work to ensure that we are appropriately considering all of the physical and scaling effects that will affect the validity and errors associated with the CCSI modeling activities, and that we are minimizing the effects of these gaps and limitations on the validation of the CCSI device scale models.

8. Conclusions

A multi-tiered validation strategy has been laid out for the CCSI CFD modeling of the full scale carbon capture system. The goal of the proposed validation and UQ analysis is to validate the CFD predictions for a specific solid sorbent carbon capture system of interest to the CCSI program. We do not intend to lay out a general validation methodology that would be appropriate for other complex multi-physics systems. That is outside the scope of CCSI and the work proposed herein.

Instead, the proposed validation and UQ methodology aims to provide quantitative confidence on the CFD simulations of larger to full scale carbon capture systems by the coupling of simpler physical systems and upscaling to larger systems. As shown in Figure 1, a multi-tier methodology is proposed which divides the complexities of the full scale system into simpler sub problems. We will start by validating simple unit problems which represent pieces of the multi-physics of the entire carbon capture system. We will rely on previously published validation studies for the unit problems and will not attempt to re-validate the unit problems. We will next move on to considering the effects of upscaling and coarse graining methodologies which are used to deal with the geometrical issues associated with simulating a full scale carbon capture system. Validation of the upscaling will focus on the development of filtered models and their validation with experimental data and fine scale models. Next we will consider decoupled and coupled laboratory and pilot scale validation cases to investigate the effects of using the filtered models in larger scale systems and the combined effects of upscaling and unit problem coupling on the accuracy of the simulations.

The final tier of Figure 1 is the intermediate and full scale carbon capture systems. The overall goal of all of the validation activities is to quantify our confidence in the predictions of the full scale carbon capture simulations. Since there is no validation data available for a full scale carbon capture system we will use the information gained from the smaller scale validation problems to quantify our confidence in our full scale simulations. Based on the individual tiers of the validation plan, we will be able to separate the effects of coupling vs. upscaling vs. model selection on the overall error of the full scale simulations.

The validation methodology laid out in this proposal can be considered as "application focused validation." We have defined a very narrow scope for our validation activities that is specific to CCSI and will allow us to complete the validation in the time frame available and with the funds available to us.

As this effort progresses, the knowledge gained will be used to develop an integrated approach to model validation and UQ for the CFD models used in the CCSI program. The other models developed in the CCSI program (i.e., reduced order models, process and system level models, etc.) can then be validated against (or calibrated to) those validated CFD models. This will ultimately allow the fundamental, validated physics-based models to be used to improve the other CCSI models.

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