6
Multiscale Process Modeling

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6.1
Introduction

This chapter covers multiscale modeling by discussing the origins of such phenomena in process and product engineering as well as discussing the approaches to the modeling of systems that seek to capture multiscale phenomena. The chapter discusses the development of the partial models that make up the multiscale model, particularly focusing on the characteristics of those models. The issue of partial model integration is also developed through the use of integrating frameworks. Those frameworks are analyzed to understand the important implications of model coupling and computational behavior.

Throughout this chapter, reference is made to granulation processing, which helps illustrate the concepts and challenges in this important area.

6.2
Multiscale Nature of Process and Product Engineering

6.2.1
The Origin and Nature of Multiscale Engineering Systems

Multiscale systems, and hence their models, exist due to the phenomena they contain or seek to represent. This is due to the fact that thermodynamic behavior and rate processes undergird our main view of the scientific and engineering world.

The underlying phenomena come into focus depending on the granularity of our perspective, which is influenced by the history of scientific investigation and, indeed, our own backgrounds. This perspective ultimately deals with length and time scales that can vary from atomic to global scales, or beyond. We can investigate time scales of nanoseconds to millennia, or length scales from nanometers to light-years, as seen in Villermaux (1996), who illustrated the typical scales dealt with in chemistry,

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physics, chemical engineering and astronomy. Hence, we are presented with a spectrum of scales depending on where we wish to view the system under study.

Our modeling efforts are simply a mapping of our understanding of these phenomena into a convenient mathematical or physical representation. The amount of scale-related information we incorporate into our models determines the multiscale degree of that representation. In some cases we can work on a single scale of time and/or length, or incorporate two, three or more scales within our models. This latter case is the area of multiscale modeling, which we address here.

As evidenced by the literature on the subject of multiscale engineering systems, there has been an explosion in interest since the mid 1990s (Li and Kwauk 2003). Papers on this topic at the start of the 1990s were very few. By 2000 a ten-fold increase in publications occurred and it continues to grow at a phenomenal rate. It is an area of intense research driven mostly by applications in science and engineering, especially materials science, mathematics and physics. Li and Kwauk (2003) and Glimm and Sharp (1997) provide examples from many disciplines. In fact, multiscale models are often multidisciplinary.

Within chemical engineering, the multiscale approach facilitates the discovery and manufacture of complex products. These may have *multiscale product specifications*, that is, desired properties specified at different scales. Biotechnology, nanotechnology and particulate technology – in fact, product engineering in general – are driving the interest in the multiscale approach (Charpentier 2002; Cussler and Wei 2003). In addition, despite the continuing increase in computing power, there are problems of practical interest that will remain intractable when tackled by direct, "brute force" methods. Multiscale techniques provide a way of making these problems feasible. There are a growing number of tools, methods and representations for engineering systems, yet little fundamental conceptual analysis leading to overall frameworks that help guide the modeling of multiscale systems.

6.2.2 Length, Time, Other Scales and their Representation

Because process engineering has its roots in physics and chemistry, the properties of models reflect the underlying time and length scales on which important phenomena occur. This can be from the quantum mechanical length scales of 10^{-13} m with time scales of 10^{-16} s to global scales of 10^{4} m and 10^{8} s and higher. At one extreme we are concerned about subatomic behavior, while the other extreme represents global processes that might have characteristic times of years or decades.

Small scales are of significant interest in determining product properties whereas large-scale processes can be of interest to process engineers involved in areas such as climate change, environmental impact and supply chain management.

Figure 6.1 shows a general scale map appropriate to process and product engineering. It is an adaptation of work by Grossmann and Westerberg (2000). It is noticeable that there is a general relationship between length and time scales that reflects the time constants over which phenomena occur at different lengths of behavior.

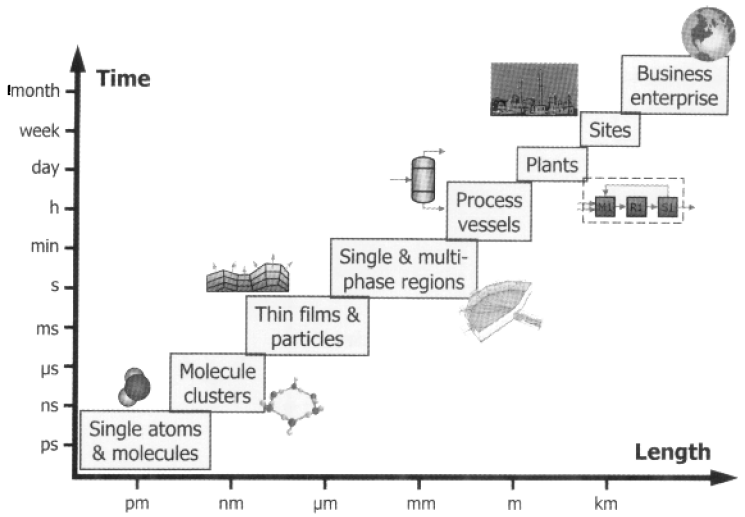

Figure 6.1 A general scale map.

It is now widely appreciated that product quality is often determined at scales well below the scales applicable at the processing or macrolevel. Hence there is intense interest in micro- and nanoscale behavior in product design.

For example, the development of granulation processes via drum or pan granulation is a multiscale operation, where final product quality is determined not only by the macroscale processing equipment level, but also at the microscale level of particle formation and interaction. A typical granulation circuit diagram is shown in Fig. 6.2, which highlights the principal operating equipment in the circuit. In this case, the circuit consists of the granulator where fine feed or recycle granules are contacted with a binder or reaction slurry. Growth occurs depending on a number of operational and property factors. Drying, product separation and treatment of recycle material then occurs. For this application, Fig. 6.3 shows a scale map from Ingram and Cameron (2002), which considers the key phenomena as represented by length and time scales within the processes. The scales represent individual particles through to agglomerates and then onto processing equipment and finally the complete circuit.

Besides the length and time scales, a *detail* scale could also be considered, which seeks to develop models with varying degrees of fidelity in relation to the real world phenomena. This form of scale can consider such issues as:

- The granularity of the system view in terms of number and types of balance volumes and the degree of aggregation that takes place.
- The number of key mechanisms related to flows, reaction, heat, mass and momentum transfers within the model and their inclusion or exclusion. Of particular interest is the complexity and fidelity of the constitutive relations. These issues can have a significant impact on the validity region of the resultant process

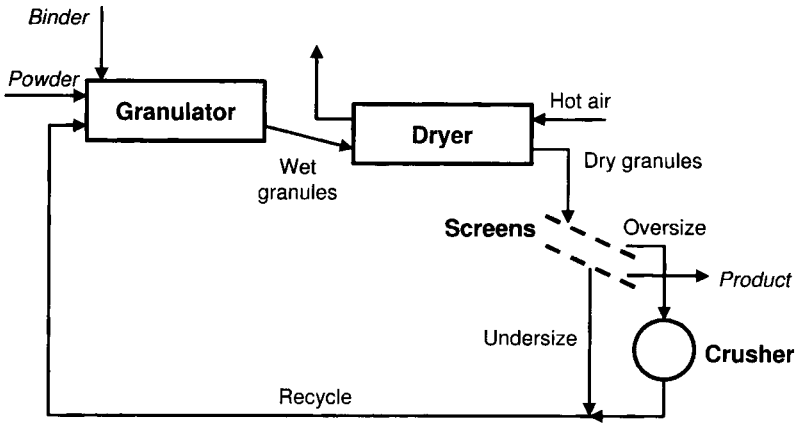

Figure 6.2 Typical continuous granulation plant.

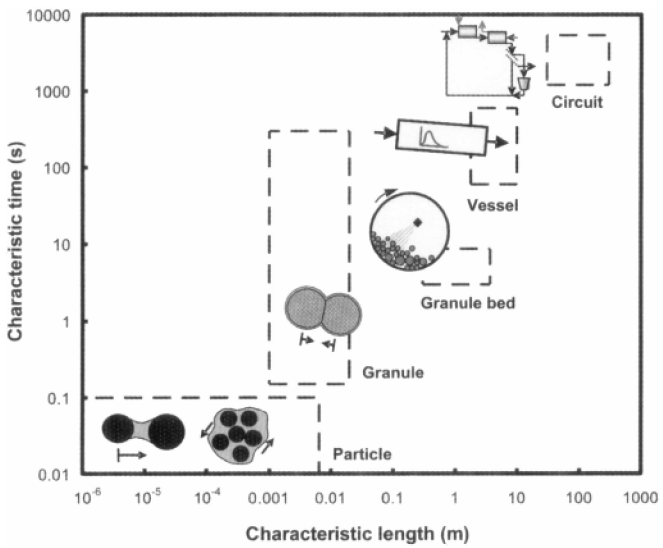

Figure 6.3 Scale map for granulation processes.

or product models, as different relations such as equations of state or property models are used.

- Species identification and representation using, for example, "lumped" representations common in pseudo-component approaches to petroleum fractions.

The detail scale complements the traditional time and length scales that are common in multiscale modeling.

6.2.3
Key Issues in Multiscale Process Modeling

We mention briefly some of the key issues in multiscale modeling before addressing some of those issues related to modeling and integration in Sections 6.3 and 6.4.

The principal issues relate to:

- Scale identification and selection for a specific modeling goal: For any modeling task the issue of identifying what scales are needed to be represented in the final model is an important consideration. As a first step, the literature contains a variety of scale maps and diagrams that show the hierarchical organization in specific application areas (for examples Alkire and Verhoff 1994; Maroudas 2000). There are several approaches that are more fully elaborated upon in Section 6.3. However, an understanding of the time scales of interest can often dictate the final scales of length and time that are needed in the model.
- Model representation: In what form does the model exist? This question is often answered by our understanding of the system under study and the phenomena we can identify. It is most often the case that grey-box models are used at several scales, because we often have some mechanistic understanding of the system phenomena. At the same time there are system parameters that are calculated via data fitting or, in some cases, are averaged values from another calculation at a lower scale. A spectrum of models exists from completely black box to mechanistic descriptions.
- Model integration: Model integration refers to linking the *partial models* that apply at a single scale into a composite, multiscale model. It still remains a challenging area and one where much is yet to be done to resolve this important issue. Section 6.4 discusses a number of these issues, with reference to several application areas. A number of integration frameworks exist, which possess distinct characteristics of information flow between scales and hence computational and other properties.
- Model solution: Solution of multiscale systems remains another major challenge, especially where distinct model forms are present within the composite or multiscale model. This is a huge area and beyond the scope of the present chapter. Some aspects are briefly discussed in Section 6.4.4.2.

6.3
Modeling in Multiscale Systems

The general approach for developing a process model for multiscale systems is an extension of that for conventional, nonmultiscale process systems (Hangos and Cameron 2001a). This approach, called the "seven-step modeling procedure", included the following stages:

1. model goal set definition (modeling problem specification);
2. model conceptualization (identifying controlling factors);
3. modeling data: needs and sources;

4. model building and model analysis;
5. model verification;
6. model solution;
7. model calibration and validation.

Thus, this section focuses on the special elements of the extended general approach that make it applicable for modeling multiscale systems as well.

Similar to the conventional case, the modeling problem specification consists of the definition of the process system with its boundaries, subsystems, components, mechanisms together with the description of the modeling goal. In the multiscale case the process system description as well as the modeling goal may call for a multiscale model when there are order of magnitude differences in either length or time behavior between the system elements. A separate subsection below deals with the specialties of the modeling goal in modeling multiscale systems.

An extended seven-step modeling procedure (Hangos and Cameron 2001a) can also be followed as a general approach in the multiscale case when some of the steps need special care and procedures that are described below.

6.3.1 Multiscale Modeling Strategies

Most often the need for developing a multiscale model arises in step 1 *(Problem definition)* or step 2 *(Identify controlling factors)* of the seven-step modeling procedure. Here, one usually identifies the necessary scales that become part of the problem definition. Thus, the first two steps should be repeated for each of the scales to develop individual modeling problem definitions and to identify the relevant controlling factors. This way a set of scale-driven related submodels is created.

Interest in system behavior over a long period of time (steady-state properties) often excludes phenomena, and hence scales, operating on a very fast timeframe. This eliminates the fast components of the system. In other cases such as modeling startup and shutdown performance of processes the intermediate time scales are of main interest. Here, some exclusion of slow and very fast components can be made (Robertson and Cameron 1997a,b).

In addition, modeling decisions should be made on how to organize the information flow between the partial models, that is, to determine the multiscale modeling framework (Ingram, Cameron and Hangos 2004) as discussed in Section 6.4.

Having formulated the modeling problems for each of the partial models, identified the controlling factors and reviewed the data available, we can turn to constructing the model in step 4 of the seven-step modeling procedure. There are two fundamentally different approaches to doing this: the *bottom-up* and the *top-down* approaches. As their name suggests, bottom-up approaches start with constructing the partial model in the finest resolution scale and proceed towards the coarser scales. Alternatively, top-down approaches start with the coarsest scale partial model

and refine its elements using finer scale submodels if necessary. Two other approaches have also been suggested in the literature. A *simultaneous* approach, which has been used industrially in the context of new product design (Lerou and Ng 1996), involves developing models at each scale of interest at the same time, and then linking them together. *Middle-out* modeling is the method of choice in some multiscale biological applications (Noble 2002). It refers to building up a model by starting with the scale that is best understood and has the most data, and then working "outwards" (to finer and coarser scales) from that.

In the following, the key elements of the extended modeling procedure, the development and role of the multiscale modeling goal set, and the specialties of the top-down and bottom-up strategies of model construction will be described in more detail.

6.3.1.1

The Role of the Modeling Goal

Any process model is developed for a specific use or possibly multiple uses. These uses influence the goals that the model must fulfill. It is, however, important to recognize that modeling goals normally change, are refined, deleted or added as the modeling cycle proceeds. This is clearly seen in the modeling process of multiscale systems, where the original modeling goal might indicate the use of a multiscale approach and then the modeling goal set of any partial model is established and refined incrementally as the modeling proceeds.

Overall Modeling Goal

The modeling goal is typically a statement with three major components:

- the need to develop a model in some relevant form;
- an application of the model for a given purpose;
- a reality that is being modeled.

These three aspects can in turn be decomposed into lower level goals that are applicable to the partial models. This can be seen in the overall modeling goals such as: "Develop a model for evaluating control options for chemical vapor deposition (CVD) of...".

The *overall modeling goal* will determine the number and hierarchy of the scales and their integration framework. Multiscale models are needed if the process system has controlling factors or mechanisms with very different scales covering several orders of magnitude. The modeling goal or the requested modeling accuracy might require partial models of finer granularity. A goal might include inputs and outputs at different scales, e.g., in CVD, where reactor operating conditions (macro) might be achieved such that film microstructure has acceptable smoothness (micro) or simply feasibility. Alternatively, the modeling feasibility can dictate partial model inclusion.

An example of such a case can be a dynamic modeling problem of an industrial granulator drum for fault detection and diagnosis purposes. Figure 6.4 shows some multiscale aspects of the granulation system and indicates some of the key informa-

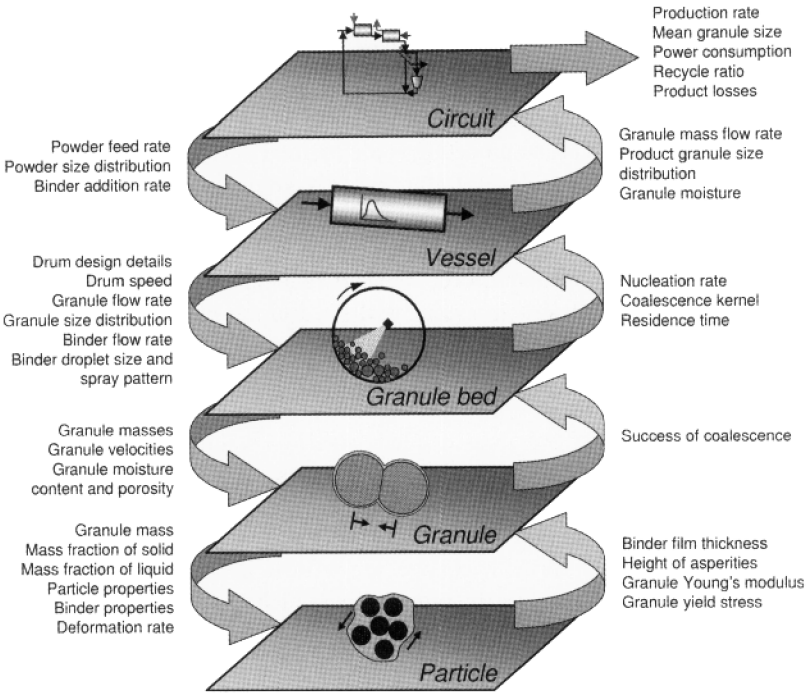

Figure 6.4 A multiscale view of linking granulation phenomena.

tion flows that exist. Because the malfunctions and faults in this equipment can be consequences of the granulation mechanisms, transport phenomena, fluid dynamics and operation procedures affecting the whole system, one needs to have several scales, a granulation particle scale and equipment scales integrated in a multiscale modeling framework.

Modeling Goals for the Partial Models

The systematic decomposition of the overall modeling goal to the individual scales is difficult and has not yet been studied well in the literature. If we consider the modeling goal to be a multifaceted statement then some of the facets originate in the original overall modeling goal, i.e., those goal elements that are relevant to the partial model related to a particular scale are simply inherited from higher level goals.

The other *integrating facets* in the modeling goal set of a partial model ensure its consistency and purposefulness from the viewpoint of the multiscale integrating framework applied to the modeling problem. This integrating part may contain variables in other partial models to be computed with a given accuracy, and may determine the data or other model ingredients to be used that are delivered by other partial models.

Continuing the granulator drum example, the granulation particle submodel will inherit the goal facets related to the malfunctions caused by the granulation process

itself. That is, it should be dynamic and should describe the formation, growth, and breakage of the particles. The integrating facets in the modeling goal set ensure that the model to be developed should produce the source and kernel functions in the granule population balance in every time instance with a given accuracy that is needed to complete the conservation balance equations on the equipment scale.

6.3.1.2

Gradual Model Enrichment or Iterative Deepening

The philosophy of the top-down approach for multiscale model development is very simple: start from the overall system model on the coarsest resolution scale (largest time or space scale) and develop a new partial model on a finer resolution scale if any facet in the overall goal set requires it. This approach can be regarded as *iterative model deepening*, which is directed by the modeling goal and its sensitivity with respect to the elements of the process model being developed. The top-down approach has been viewed as the best method for process engineering because time and cost pressures favor quick application of the results, with a minimum of detailed modeling. Later on in the lifecycle of the process, model refinement can be applied as required.

The iterative model deepening technique is applied in step 4 *(Model building and model analysis)* of the seven-step modeling procedure when the number of scales and the model integration framework have already been selected. We then start from the overall system model on the lowest resolution scale with the overall modeling goal and determine which facets of the modeling goal are not satisfied. By using sensitivity analysis, it is possible to determine which model elements influence the missing goal facets. The model is then enriched by a partial model on a finer resolution scale for the necessary model elements. Repeating the above deepening steps until the entire modeling goal set is satisfied constructs the final multiscale model.

It is worth mentioning that the iterative model deepening procedure is similar to the approach applied for empirical model building (Hangos and Cameron 2001a).

If we again consider the granulator drum example, we start constructing the overall multiscale model by developing the material and energy balances of the drum and find out that we need finer models for the convective and diffusive flows, together with the source terms describing the particle birth, growth, and breakage processes in the granule population balance.

6.3.1.3

Model Composition

The bottom-up approach of constructing multiscale process models is also applied in step 4 of the modeling procedure as an alternative method to the iterative model-deepening procedure. The overall model construction starts by building the partial models on the highest resolution scale. These models are then integrated according to the selected multiscale integration framework to prepare the modeling problem statements for the submodels on the next, coarser resolution scale.

Bottom-up model composition is a common way to build multiscale models, when the submodels originate from different sources and/or are based on different princi-

ples. The advantage of this approach is that substantially different models may be integrated if a suitably selected framework is found. The drawback is that the resulting model is often not homogeneous in its approach, purpose, and accuracy or it may happen that a partial model cannot be suitably integrated. Model composition ensures that the upper level models have a sound fundamental basis and may consequently be more reliable than those developed by model enrichment (Section 6.3.1.2). However there is the risk of starting the modeling process at a level that is too fundamental, which may lead to accurate but inefficient modeling.

6.3.2
Partial Models: Approaches and Classification

In this section we focus on the partial models or submodels of a multiscale process model. For this purpose, we assume a well-posed modeling problem for each partial model, i.e., a system description and modeling goal specification for any partial model. This implies that the mechanisms and data available can be determined individually.

The approaches of model building of the partial models, that is the development of the model equations, are essentially the same as in the classical case: we may apply mechanistic approaches based on first principles or a black- or grey-box model. Similarly, the classification of the resulting partial models goes along the same lines as in the general, nonmultiscale case.

6.3.2.1
Mechanistic Approaches for Process Models

The mechanistic approach of developing the model equations of a process model uses first principles to construct the ingredients of a partial model. The model-building subprocedure (Hangos and Cameron 2001a) is followed in this case with the following substeps:

1. system and subsystem boundary and balance volume definitions;
2. define the characterizing variables (inputs, outputs, and system states);
3. establish the balance equations for conserved quantities: mass, energy, momentum and number, etc.;
4. transfer rate specifications;
5. property relation specifications;
6. balance volume relation specifications;
7. equipment and control constraint specifications;
8. modeling assumptions.

In the multiscale case, however, steps 1 and 2 need special care, because these are partially determined by the other partial model(s) and by the integrating framework of the overall multiscale model.

As the result of a mechanistic approach to partial model construction, we obtain a process model with standard ingredients (see Section 6.3.3.2). This makes it relatively easy to integrate the resulting partial model into a multiscale framework (see Section 6.4).

Table 6.1 Classification of partial models.

Type of Model	Criterion of Classification
Mechanistic	Based on mechanisms/underlying phenomena
Empirical	Based on input-output data, trials or experiments
Stochastic	Contains model elements that are probabilistic in nature
Deterministic	Based on cause-effect analysis
Lumped parameter	Dependent variables not a function of spatial position
Distributed parameter	Dependent variables are functions of spatial position
Linear	Superposition principle applies
Nonlinear	Superposition principle does not apply
Continuous	Dependent variables defined over continuous space-time
Discrete	Only defined for discrete values of time and/or space
Hybrid	Containing continuous and discrete behavior

6.3.2.2

Black- and Grey-Box Modeling

Usually, either engineering knowledge or data are not available to construct a fully mechanistic so-called "white-box" model, but we obtain a partial model with unknown model parameters and/or structural elements. One can then use measured data from the real process to give an estimate of the unknown model parameters or to construct an empirical, so-called "black-box" model for the unknown model element. This way a fully determined model can be obtained in the *model calibration and validation* step of the modeling procedure.

It is important to note, however, that in most of the cases it is rather difficult, if not impossible, to calibrate a grey-box partial model, because one only has measured data from the overall process system and not from its subsystems corresponding to partial models (see Section 6.4.4.3 for more details).

In some cases a fully black-box model should be developed by using empirical model building (Hangos and Cameron 2001a) that is similar in its approach to gradual model enrichment. Typical of these models are Box-Jenkins and neural networks.

6.3.2.3

Model Classification

The classification of the resulting partial models is done similarly to the general case. As the characteristic of the different classes of models have a great impact on the solution techniques and on the application area we briefly recall the criterion of classification and the type of models in Table 6.1 (Hangos and Cameron 2001a).

It is important to observe that a classification criterion generates a pair or triplet of model types and all classification criteria can be applied to a particular partial model.

6.3.2.4 Particular Modeling Techniques for Different Scales

We make a brief digression here to list some of the techniques that have evolved to describe specific scales. In approximate order of increasing scale, they include:

- computational quantum chemistry and molecular mechanics to deduce basic chemical properties on the electronic/atomic scale;
- molecular dynamics, Monte Carlo and hybrid methods that predict the ensemble behavior of many molecules;
- assorted techniques for front tracking, interface modeling, particle interactions and so on, grouped roughly as "mesoscale" models;
- computational fluid dynamics for detailed flow prediction;
- unit operation modeling and process flowsheet simulation, most familiar to chemical engineers, for vessel and plant scale studies;
- environmental simulation and business enterprise modeling on the "megascale".

Each technique has many variations, both broad and subtle, and will likely require contribution from specialists in the field. This reinforces the cross-disciplinary nature of much multiscale modeling work.

6.3.3 Characteristics of Partial Models

This section deals with the characteristic properties and model elements of partial models in a multiscale process model with an emphasis on those model properties and ingredients that are important from the viewpoint of multiscale modeling.

6.3.3.1 Model Types

The classification of partial models and the resulting model types are already described in Table 6.1 in Section 6.3.2.3. There are some model types that often arise as partial models in multiscale systems and are therefore of special importance.

Lumped and distributed parameter models. Most often both of these types of partial model can be found together in a multiscale model. The models in the finest scale are typically distributed parameter dynamic models, while the models on the coarsest scale are often lumped parameter models. The integration of such mixed-type partial models into a multiscale integration framework needs special care (see Section 6.4).

Deterministic and stochastic models. There are some characteristic phenomena often found in multiscale systems, such as fluid dynamics, diffusion, heterogeneous

kinetics and the like that are often described by using stochastic models on a fine scale. The integration of such partial models into a framework of deterministic models is usually performed by using averages of different types (mean values for stochastic variables, time, and/or space averages).

6.3.3.2

Standard Ingredients of Partial Models

Partial models in a multiscale model have the same standard ingredients as usual process models. In this case, however, it is of crucial importance to have all the ingredients clearly specified in order to be able to integrate the partial models into a multiscale framework. The seven-step modeling procedure ensures that the resulting model possesses all ingredients in a consistent way. Therefore mechanistic partial models need no further effort to ensure this.

Balance volumes. Homogeneous or quasihomogeneous parts of process systems over which conservation balances are constructed are called balance volumes. They are fundamental elements of a partial model in a multiscale system. The union of all balance volumes in a partial model spans the entire domain of the partial model, while the balance volumes in different partial models are related in a way determined by the multiscale integration framework.

Model equations (differential and algebraic). The differential equations in a dynamic partial model usually originate from conservation balances and they are supplemented by constitutive algebraic equations that make the model complete from both the engineering and mathematical viewpoints (Hangos and Cameron 2001b). The algebraic equations are of mixed origin: they describe extensive-intensive relationships, transfer and reaction rate equations, equations of state, physicochemical property equations, balance volume relations, and equipment and control relations. In addition, conservation balance equations are also algebraic equations in static partial models.

Model variables and parameters. Variables in a dynamic partial model are time and possibly space-dependent quantities, which are either differential variables if their time-derivative appears in the model equations or algebraic variables otherwise. There are also model parameters present in a partial model; their value is regarded as constant. Some of the variables are given a value by using specification equations in order to make a model with zero degrees of freedom: specification equations are also part of the model. In dynamic partial models, some of the variables, the potential input variables, are also assigned a given "value," a time-dependent function.

Initial and boundary conditions. Similarly to any process model, partial models are sets of ordinary and/or partial differential (or integro-differential) and algebraic equations. In order to make the model well-posed from a mathematical sense, we need to give suitable initial and boundary conditions, which are also part of the model.

Modeling assumptions. Although not always stated explicitly, modeling assumptions are key ingredients of any process model, because they document any decision the modeler had taken while developing the model. This way modeling assumptions are artifacts of process modeling and serve as key elements in model documentation, verification and consistency checking (Hangos and Cameron 2001a).

6.3.3.3

Particular Considerations for Integrating Multiscale Process Models

Some of the above standard ingredients of partial models have particular significance when integrating partial models into a multiscale framework. These are briefly listed in this subsection, while the way they are used is described in Section 6.4.

Variables and parameters. Some of the characterizing variables of the partial models in a multiscale process model, such as the conserved extensive quantities (mass, component masses, and energy), the thermodynamic state variables (temperature, pressure, and concentrations), and the rate variables, are usually related in a way determined by the integration framework. In addition, some of the model parameters in a partial model of coarser scale may be determined by the modeling output of partial models of finer scales. Such interscale relationships appear in a partial model in the form of additional specifications or equalities for the model parameters.

A simple example of such an interscale relation can be the value of the porosity of an equipment scale model determined by another partial model on a particle scale.

Initial and boundary conditions. Phenomena occurring at an interphase boundary often call for a partial model describing them in a finer scale. The result of such small scale modeling enters into the equipment-scale model as an expression for the boundary condition of that particular interphase boundary.

Constitutive equations. Certain algebraic variables in partial models of coarser scales, such as reaction or transfer rates, serve typically as connection points between partial models of different scales. Their determining "connecting" constitutive equations contain variables of different scale partial models, where the variables of finer scales are usually averaged to obtain the connecting algebraic variable on the coarser scale.

A simple example of such a connection can be a reaction rate of a heterogeneous catalytic reaction, where the reaction rate equation serves as a connecting point between the equipment scale and the catalytic particle scale variables. As a result of the particle scale model, an algebraic relationship should be somehow determined that describes the average reaction rate in a point of the equipment as a function of the average concentrations and temperature at the same point in a time instance.

6.4
Multiscale Model Integration and Solution

Model integration is the process of linking partial models that exist at different time and length scales into a coherent, composite multiscale model. Integration is the essence of multiscale modeling. If information did not flow between the scales of interest, the model would not be multiscale! Two models sharing information at the same scale does not constitute a multiscale system.

In this section we will explore the challenges and status of model integration, compare alternative integration schemes, and address some implementation matters.

6.4.1
The Challenges and Status of Multiscale Integration

The multiscale modeler potentially faces several challenges in performing model integration because it may be necessary to link partial models that:

- *Span a vast range of length scales.* For example, in electrodepositing of metal onto printed circuit boards, electrical function is influenced by lattice defects of $O(10^{-10})$ m, local hydrodynamic and mass transfer processes affecting the deposition rate occur over $O(10^{-4})$ m, and current distribution must be controlled over the entire job, which is $O(1)$ m in size (Alkire and Verhoff 1994).
- *Operate on very different time scales.* In semiconductor fabrication using chemical vapor deposition (CVD) for instance, individual diffusion and reaction events occur at atomic time scales of $O(10^{-13})$ s and thin film growth takes place over minutes $O(10^{2})$ s, while the total processing time for a multilayer film may be hours or days $O(10^{4})$ s (Jensen 1998).
- *Have disparate natures.* The models may have different dominant phenomena, be continuous or discrete, have different dimensionality, be deterministic or stochastic, exist in concept only or already be implemented in commercial software, and be drawn from different scientific disciplines.

These challenges raise several types of issues that need to be tackled for successful integration (Pantelides 2001):

- *conceptual issues,* for example, deciding what information should flow between the scales and how, or investigating the benefits of reformulating the partial models to allow better integration;
- *mathematical issues,* regarding how well-posed the problem is, for instance;
- *numerical issues,* such as the choice of numerical method for efficient and robust solution of the composite model;
- *application issues,* including the software engineering work needed to link diverse software across different platforms.

Despite the difficulties, there are many successful examples of multiscale model integration from a wide range of disciplines. It is partly the diversity of the applica-

tion areas and the complexity of the implementation details that obscures the common features in these examples. There are the beginnings of a classification scheme for model integration methods, and there is some knowledge of their characteristics. However, we are not yet at the point of understanding, in a quantitative and general way, how the choice of integration method affects the resulting multiscale model nor how to select the best integration method for a particular modeling problem.

6.4.2 The Classification of Model Integration Methods

Multiscale models can be classified by the way the partial models at each scale are linked. The very act of classification is enlightening. It draws out the similarities and differences in the broad range of existing multiscale models. It also makes comparing alternatives easier. Several authors have proposed classification schemes, often just in passing. A widely accepted scheme has not emerged and there is a confusion of terms. We will summarize a few of these classification ideas then recommend a particular one in the next section.

Maroudas (2000) divides multiscale models in materials science into two categories: *serial* and *parallel*. Serial multiscale models result from sequentially scaling up the partial models: the outputs of the finer scale models become the inputs of the coarser scale ones. By way of contrast, in parallel multiscale models, the partial models exist side by side and are solved together.

Stefanović and Pantelides (2000) identified three ways in which molecular dynamics (MD) for property estimation could be integrated with traditional process models. First, the MD model is run to generate "pseudo-experimental data," which are then fitted to find macroscopic material parameters. The second possibility is sequential like the first, but instead the macroscopic parameters are found directly from the microscale simulations. In the third approach, the macroscopic model calls the microscopic one on demand to find a relationship between macroscopic variables. This technique eliminates the need for macroscopic parameters.

Phillips (2001), whose interest is materials science, distinguishes two broad classes of multiscale models: those that work in *information passage* mode, and *models with internal structure*. Information passage models are of two subtypes. In the first, the fine-scale model is used to generate macroscopic parameters, which are then used by the coarse scale model. In the second, the microscale model is *transformed into* a macroscale model, that is, a *new effective macroscale theory* is derived from a microscale one. Models with internal structure, which in Phillips' area are typically "mixed atomistic-continuum models," are where the microscopic model is adjacent to or embedded within the macroscopic model and they are solved together.

Guo and Li (2001) and Li and Kwauk (2003) are concerned with predicting the structure of dynamic, multiscale systems with competing mechanisms. They describe three modeling approaches of increasing complexity and illustrate them by referring to gas-solid fluidization. The first approach is *descriptive*: each scale of interest is modeled separately without attempting to link them together. The second is

correlative, where fine scale information is scaled up for use in coarser scale models. Volume-averaging of parameters is a possible technique, for example. In the third approach, *variational*, the partial models are solved together subject to the minimization of some quantity that reflects the strengths of the competing mechanisms that influence the system structure. The last approach can handle regime transformations.

These schemes represent different views of multiscale integration, which reflect their originators' backgrounds and modeling objectives. They all distinguish between sequential and simultaneous application of the partial models, and most discriminate between variations on the sequential method. Pantelides (2001) presented a classification scheme for multiscale process modeling with four categories, *serial, simultaneous, hierarchical* and *parallel*, that encompass most of the ideas above. Ingram and Cameron (2002) refined and extended this scheme with an additional class in order to distinguish between different methods of simultaneously combining partial models.

6.4.3
A Framework Classification for Process Modeling

We introduce the term *framework* to describe the way partial models, which apply at different scales, are linked, or integrated, to form a composite multiscale model. Figure 6.5 shows the extended version of Pantelides' classification scheme for multiscale frameworks that is proposed for process modeling. The main division is between *decoupling* and *interactive* frameworks. The serial and simultaneous frameworks decouple the solution of the partial models, so that one partial model is solved (in some sense) then the others are solved in turn. In contrast, the interactive frameworks-embedded, multidomain, and parallel-essentially involve simultaneous solution of the constituent partial models. Another view of the classification scheme,

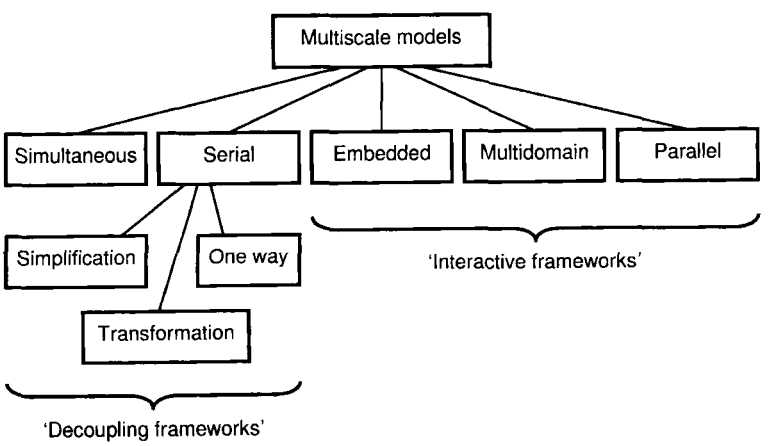

Figure 6.5 Classification scheme for multiscale models based on the framework used to link the partial models.

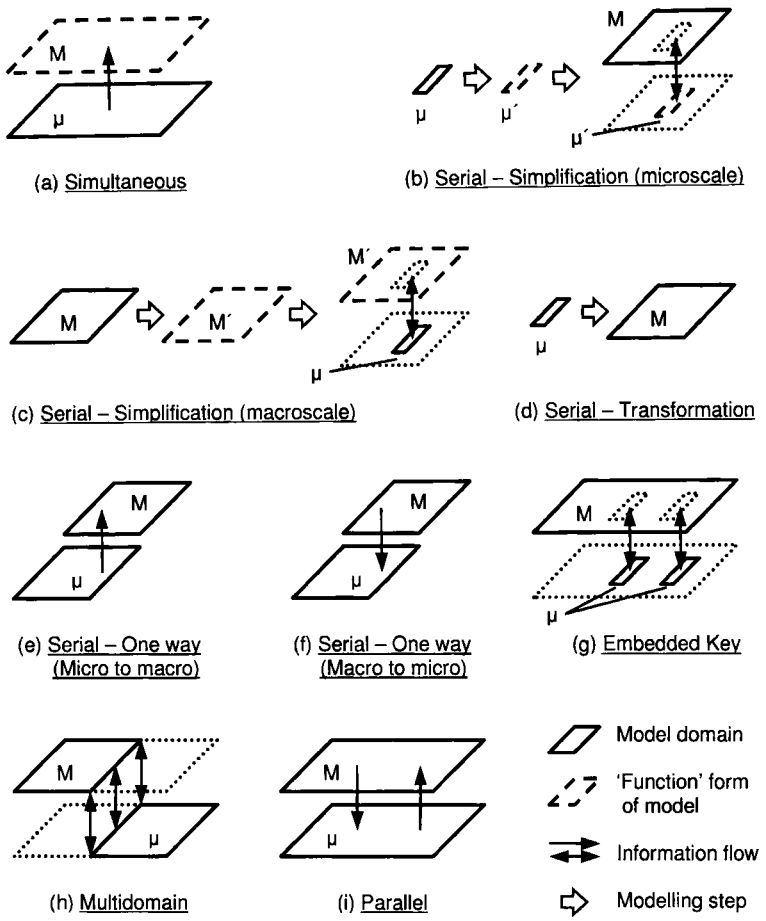

Figure 6.6 Domain relationships and information flows in models with microscale (μ) and macroscale (M) parts.

expressed in terms of the model domains and the information flows between them, appears in Fig. 6.6. The framework classification attempts to define the broad conceptual options for linking partial models. It is not intended to discriminate between the very specific techniques that are used in particular applications.

The following sections explore the frameworks in more detail, by giving a short description, common application situations, some advantages and disadvantages, and the main challenges associated with them. The integration of two partial models at a time is discussed. The fine and coarse scale models are referred to as microscale and macroscale models, respectively.

6.4.3.1
Simultaneous Integration Framework

Description

In simultaneous integration, the microscale model is used to describe the system in its entirety (Fig. 6.6a). This approach corresponds, for example, to using discrete element modeling to predict the trajectory of every particle (microscale level) in a fluidized bed (macroscale level), or the use of computational fluid dynamics (CFD) (microscale) to model the detailed fluid flow in a complex vessel (macroscale). The macroscale "model" simply summarizes or interprets the detailed microscale results, usually by totalizing, averaging or otherwise correlating the microscale data. This is why the macroscale model in simultaneous integration could be better called the "macroscale function." In the fluid bed example above, the macroscale function might estimate the average bed expansion or gross solids circulation rate, while in the CFD case, the macroscale function might calculate the residence time distribution. The microscale and macroscale models are decoupled in the sense that information is transferred in one direction only: from the microscale to the macroscale.

Application

Simultaneous integration is used when:

- it is not possible to model any part of the system with sufficient accuracy at the macroscale level;
- some part of the region can be successfully modeled at the macrolevel, but the micro- and macroregions cannot be meshed together well enough (this is multidomain integration; see section 6.4.3.4);
- it is desired to view the system entirely at the microlevel.

It also serves as a baseline integration method against which other frameworks can be tested because it has "zero integration error" (Solomon 1995; Werner 1999).

Advantages and Disadvantages

The advantages of simultaneous integration include the potentially high levels of detail, flexibility and accuracy. The main disadvantage is the very high computational burden, the highest of all the frameworks. This limits the size of the system and the length of time that can be simulated. Artificially reducing the system size risks failing to capture large-scale/long-time effects (McCarthy and Ottino 1998). Despite continuing growth in computer power, at least for the intermediate future, there will be problems of practical interest where microscale modeling of the entire system is impossible (Chan and Dill 1993). Simultaneous integration will also likely generate a large amount of detailed microscale data that is largely irrelevant to the modeling objective.

Challenges

These include:

- improving the accuracy of the microscale model, because any microscale error will flow to the macroscale;
- increasing solution efficiency via improved numerical algorithms and distributed computing;
- recognising when simultaneous integration is truly necessary.

6.4.3.2

Serial Integration Framework

Description

There are three broad, partially overlapping possibilities in serial integration: simplification, transformation and one-way coupling.

1. Simplification

   The microscale model covers a small part of the system domain. Usually, it is this model that we "simplify" (Fig. 6.6b). In this case, the microscale model is simplified by just "fitting a curve" to computed input-output data, by systematic order reduction methods, or by analytical solution if possible. The simplified or solved microscale model creates a relationship between macroscopic variables that is easier to evaluate than the complete solution of the original microscale model. We should perhaps refer to the microscale model as the "microscale function." The macroscale model spans the system domain and calls the microscale function on demand. This kind of integration is used, for example, to link local and global scales in climate modeling. Mechanistic city-scale calculations relating atmospheric and urban variables to pollutant fluxes can be approximated by polynomials, which are then used in a global atmospheric chemistry model to predict climate change under different development scenarios. A more familiar example may be the analytical solution of the reaction-diffusion equations in a porous catalyst to yield a Thiele modulus-effectiveness factor relationship that is then used in reactor scale modeling.

   In serial integration by simplification, information flows in both directions between the micro- and macroscales. However, the framework is decoupling in the sense that the solution process has two stages. First, the microscale model is first simplified to a "microscale function," and then the macroscale model is solved, which involves calling the microscale function.

   Often the microscale model is "simplified", but sometimes it is the macroscale one (Fig. 6.6c). For example, if the focus of a modeling exercise were a particular unit in a process, then a model of the process excluding that unit could be built and then simplified, to provide a computationally cheap approximation to the operational environment of the unit. In this case, there is a "macroscale function" (the rest of the plant) that is called as required by the microscale model (the particular unit).
2. Transformation

   Another possibility in the serial integration framework is transformation. The microscale model describes a small part of the system domain. It is "formally

transformed" into a macroscale model (Fig. 6.6d). This process is also called upscaling, coarse-graining, degree of freedom thinning, and constructing new effective theories or laws. Now, the microscale model is no longer needed and the system domain is described entirely by the new macroscale model. Many techniques are used for upscaling: volume averaging, renormalization and homogenization, among others. The three named methods have been used to upscale the equations for diffusion in porous media. No flow of information occurs between the microscale and macroscale models during solution because, in effect, the microscale model has been eliminated. Only the macroscale model must be solved. It is a decoupling framework in this sense.

3. One-way Coupling
   The third variation on the serial integration framework occurs when, because of the nature of the system, information flows naturally between the scales *in one direction only* (Fig. 6.6e,f). Or, that the approximation is close enough to satisfy the modeling goal. Physical vapor deposition (PVD) is an example. A vessel scale (macro) model predicts the average spatial distribution of metal sputtered onto a substrate in a PVD chamber. A "feature scale" (micro) model can then track the build up of the deposit layer on features, such as holes and trenches, on the substrate surface. Another example is the use of (microscale) molecular dynamics to calculate a diffusion coefficient, which is later used at the vessel (macro) scale via Fick's law. The information may flow either from the microscale to the macroscale, or vice versa. The independent model is solved first, and then the dependent one is solved. In this framework the solution of the models is decoupled in one direction.

Application

The use of serial integration depends on the broad strategy chosen:

- *Simplification.* Virtually any microscale (or macroscale) model can be simplified by order reduction or approximation techniques; some models may be solved analytically under appropriate simplifying assumptions.
- *Transformation.* The ability to use transformation depends on the upscaling method chosen and the nature of the system. For example, sufficient "scale separation" is needed for homogenization (Auriault 1991).
- *One-way coupling.* This method can be used when the nature of the system is such that one scale is dependent and the other is independent, or at least we can treat them as such. There is no feedback between the scales.

Many well-known relationships in science and engineering can be viewed as applications of serial multiscale integration by simplification or transformation. Examples include: equations of state derived from the kinetic theory of gases, the rheological equation for Newtonian fluids, Thiele modulus-effectiveness factor expressions for reaction-diffusion problems, and even, in some sense, Newton's law of universal gravitation (Phillips 2001).

Advantages and Disadvantages

The principal advantage of the serial approach is the elimination of the expensive, detailed microscale model. Consequently, serial integration can potentially produce the most efficient models among the five frameworks. Many powerful mathematical techniques for order reduction, analytical solution and upscaling can be applied to the microscale model. These techniques not only enhance calculation efficiency, but also, and perhaps more importantly, highlight the essential nature of the microscale model. They strip away unnecessary detail to reveal how (a few) key variables influence macroscale behavior. The disadvantages include the restricted accuracy and flexibility of the approach. Considerable effort may be needed to apply the mathematical techniques referred to above.

Challenges

The basic challenge is to find an appropriate balance between fidelity and computational cost through manipulation of the microscale model. This includes learning which mathematical techniques are best used to simplify or transform the microscale model, and knowing when one-way coupling is acceptable. A further challenge is to provide mechanisms for revising the microscale representation as required (Oran and Boris 2001, p. 439).

6.4.3.3 Embedded Integration Framework

Description

The microscale model is "formally embedded" within the macroscale model in this framework (Fig. 6.6g), which was termed hierarchical by Pantelides (2001). The macroscale model spans the system domain, while the microscale model is local, restricted to a relatively small part of the domain. The microscale model calculates, on demand, a relationship between macroscale quantities. Hence, while its domain is small, the microscale model may be called (instantiated) at many points through the system. Ab initio molecular dynamics is an example of the embedded framework. In this application, the macroscale model is an MD simulation that tracks the motion of each molecule based on the forces that act upon it. The microscale model is an electron-atom scale computational chemistry method, such as density functional theory, that calculates the intermolecular force (potential) function *on the fly* as the MD simulation proceeds. The embedded approach is a true, interactive multiscale method because information is passed between two models that are actively being solved.

Application

This framework is used when a suitable macroscale model exists but needs to be "informed" by localized microscale simulation, *and* the microscale model cannot be acceptably simplified. If a suitable simplification of the macroscale model were available, serial integration via simplification (Section 6.4.3.2) should be used to reduce computing demands.

Advantages and Disadvantages

Embedded integration has a natural appeal because of its orderly, hierarchical nature. It potentially has the flexibility and accuracy of simultaneous integration, but with a much lower computational load. The detailed, expensive microscale calculations are performed only where and when they are required. A disadvantage is the need to run the microscale model at all, because it may still consume the bulk of the computing resources. Because the micro-macro interface in embedded and serial (simplification) integration may be similar, it should be possible to swap these methods with little change needed in the macroscale model.

Challenges

The challenges include:

- finding the smallest domain and shortest time needed to simulate the microscale model to provide accurate results with a minimum amount of calculation;
- enhancing the micro-macro interface beyond that used in traditional serial (simplification) modeling. For example, Stefanović and Pantelides (2000) show a new approach to linking molecular dynamics information with unit operation modeling.

6.4.3.4

Multidomain Integration Framework

Description

In the multidomain framework, the microscale and macroscale models describe separate but adjoining parts of the whole system (Fig. 6.6h). It is sometimes called "hybrid modeling" because it is often used to create hybrid, discrete-continuous models. There are many multidomain models in materials science. In investigating the fracture of brittle solids, for example, the region around the growing crack could be modeled with a discrete, atomistic, microscale method: molecular dynamics. Relatively far from the crack, the solid could be modeled at the macroscale level with a continuum technique, such as the finite element method. The interface between the micro- and macroscale domains may be either a point, line or surface, or it may be a buffer zone with a volume that is nonzero, but small compared to the size of the micro- and macrosimulated regions. The models in the two regions interact across an interface. This multidomain framework is a true, interactive multiscale method, with a two-way flow of information between the partial models via the interface.

Application

Multidomain integration is used where some parts of the system can be adequately described at the macrolevel, while in other regions, only a microscale model will suffice. It is often used in models with heterogeneous media, for example, the gas and solid phases in chemical vapor deposition, or the catalyst and bulk phases in a packed bed reactor. In these applications, the microscale region is usually fixed in space. The other main application for multidomain models is materials science, par-

ticularly the field of fracture mechanics. There, the microscale model is applied when the macroscale model fails some error criterion; the microscale region may change as the simulation proceeds.

Advantages and Disadvantages

Like embedded integration, the multidomain method couples micro- and macroscale models to reduce the computational burden compared to simultaneous integration, while maintaining microscale realism where needed. The greatest disadvantage is the potential complexity of the micro-macro interface. It is important to guarantee the continuity of thermodynamic properties and transport fluxes across the interface, and to avoid unphysical wave reflections (Brenner and Ganesan 2000a,b; Curtin and Miller 2003).

Challenges

The main challenge is to formulate a seamless interface between the microscale and macroscale regions. In some applications, techniques are also needed to move, grow and shrink the microscale region as the simulation proceeds to minimize computational requirements.

6.4.3.5 Parallel Integration Framework

Description

Both microscale and macroscale models cover the entire system domain in parallel integration (Fig. 6.6i). However, the models are complementary in the detail with which they describe the important phenomena. There are currently few examples of parallel integration in chemical engineering. All combine a CFD model with a traditional unit operation model. For example, in a bubble column reactor, two phenomena are important: hydrodynamics and process chemistry. In a parallel framework, the microscale model might treat the fluid mechanics in detail via CFD, while the process chemistry could be represented in an abbreviated manner by a simple gas source (or sink) term. The macroscale model, on the other hand, could contain a comprehensive reaction scheme that was assumed to take place in simple fluid flow regime, such as a well-mixed or plug-flow region, or some combination of these. Current parallel models are solved by successive substitution. The macroscale model predicts some quantities that are inputs to the microscale model, while the micromodel outputs some variables that the macromodel requires. The models are run alternately until convergence. Parallel integration is an interactive multiscale method because both models are active and must be solved in concert.

Application

To date, parallel CFD-unit operation models have been developed for a bubble column reactor (Bauer and Eigenberger 1999, 2001), a batch stirred tank reactor (Bezzo, Macchietto and Pantelides 2000), an industrial crystallizer (Urban and Liberis 1999) and a high temperature electrochemical reactor (Gerogiorgis and Ydstie 2003). The

parallel method is suitable where the important mechanisms in the system are weakly coupled (Pantelides 2001; Urban 2001). However, it is likely that the strength of the coupling that can be successfully accommodated by the parallel method would increase as the macroscale model is more finely discretized. Very strongly coupled systems may require embedded integration.

Advantages and Disadvantages

The key advantage of parallel integration is the division of the system into two simpler problems. It is also a way to form a multiscale model from an existing software package that has limited interface options. The main disadvantage is the inherent approximation of the method and its consequent limitation to systems with weakly coupled active mechanisms.

Challenges

One challenge for parallel CFD-unit operation models is to determine efficiently, perhaps automatically, an acceptable combination of ideal flow regions needed to approximate the CFD flow pattern. Another is to replace successive substitution with a more efficient and stable solution method. However, the most open challenge is to expand the range of applications beyond the CFD-unit operation examples reported so far. A general question is how best to partition the controlling phenomena between the parallel models.

6.4.3.6

Discussion of the Scheme

The extended classification scheme proposed for process modeling groups multiscale models according to how the microscale and macroscale models are linked. It helps in understanding the structure of multiscale models, but there are some open issues:

- The classification scheme does not provide formal definitions for the frameworks. A given multiscale model could potentially fall into more than one category. Conversely, applying a framework to given partial models does not guarantee a unique multiscale model; many variations are possible.
- While some qualitative properties of the frameworks are outlined, we lack comprehensive comparative information. There is little guidance and there are no quantitative tools available to help select the integration method. These would be especially useful early in the modeling process.
- It is unclear how, or indeed if, all frameworks could be applied to a given modeling problem. Some problems do not naturally seem to suit certain integration methods and the underlying reasons need to be understood.
- The way the integration framework depends on the partial models to be integrated, and conversely, how the partial models affect the properties of the integrating framework, also needs careful further investigation.
- The classification scheme considers the integration of two scales at a time. However, it can be applied to multiscale models with more than two scales. For example, in a three-scale system, the micro- and mesomodels could be linked with one

framework, while the meso- and macroscales could be linked with another framework. This "pairwise" scheme may not be adequate to describe all multiscale models with more than two scales.

Some of these unresolved issues can be tackled through structural analysis techniques.

For systems that may exist in more than one state or regime, there is an additional element needed in model integration. It is to define the criteria used to identify the operating regime or prevailing physical structure of the system. Li and Kwauk (2003) have developed a method for multiscale models.

6.4.4 Application of the Frameworks

So far, we have looked at some issues of conceptual modeling for multiscale process systems. There are also mathematical and software issues involved in implementing the models: the use of existing software, solution methods, and model testing.

6.4.4.1 The Use of Existing Software

The chemical engineering community has already addressed many of the challenges in linking existing, previously incompatible process simulation software through the CAPE-OPEN project and its successors (Braunschweig, Pantelides, Britt, et al. 2000). CAPE-OPEN defines a set of open interface standards that allows parts of process simulators from different vendors to work together in a "plug-and-play" manner. The standard is maintained by the CAPE-OPEN Laboratories Network, CO-LaN (http: www.colan.org). Their Web site contains a list of current projects and discusses issues in linking process engineering software.

Work at the University of Aachen has been centered on the component-based hierarchical explorative open process simulator (CHEOPS) (http: www.lfpt.rwth-aachen.de/Research/cheops.html) as an integrative modeling and solution environment.

There are commercial examples of multiscale software integration. Some of these include linking a CFD package with another software type, for example, process modeling software (gPROMS with FLUENT and STAR-CD), a process simulator (ASPEN Plus with FLUENT), and a gas-phase/surface chemistry simulator (CHEMKIN with STAR-CD).

A related development is underway in the field of medical research as part of the IUPS Physiome Project (Hunter and Borg 2003; Hunter, Robbins and Noble 2002). A series of XML-based markup languages is being developed to capture and exchange information on human physiology – from the scale of cells to the whole body.

6.4.4.2

Solution Methods for Multiscale Models

Solution efficiency is important in multiscale modeling because the partial models may use computationally intensive techniques, such as quantum chemistry, molecular simulation and so on. A key feature of multiscale modeling is transforming a problem that is intractable when viewed at the finest scale into a manageable one when considered at multiple scales. The modeling techniques used at each scale have established specialized solution methods. The challenge is to meld the scale-specific solution techniques into an efficient strategy for solving the multiscale problem.

There is a broad and powerful class of numerical methods known as multiscale (multigrid, multilevel, multiresolution, etc.) schemes (Brandt 2002). They seek to solve problems that are defined by equations at one scale, but have a multiscale character. This numerical approach complements the concept we have advanced in this work, namely defining separate models at each scale and linking them through a framework. The multiscale numerical method consists of recursively constructing a sequence of solutions to a fine-scale problem at increasingly coarse scales. Large-scale behavior is effectively calculated on coarse grids, but it is based on information drawn from finer scales. Multigrid techniques have been applied to a vast range of problems. A few relevant to process engineering are: high efficiency methods for fluid dynamics, solution of partial differential equations in general, data assimilation and other inverse problems, optimal feedback control, efficient molecular dynamics and molecular modeling, and global optimization. See Brandt (2002) for a comprehensive review. Not included in that review is the recent "equation-free" (or "gap-tooth/projective integration") method (Kevrekidis, Gear and Hummer 2004) that has been developed in the context of process systems.

The solution of multiscale models constructed from partial models linked by a framework involves several issues, including:

- *The effect of the framework* – In some frameworks, for example, the simultaneous and some serial ones, the solution process is decoupled. The best method at each scale can then be employed in isolation. The nature of the mathematical problem is different for each of the interactive frameworks. In multidomain integration, the models only interact through boundary conditions, while in embedded integration, information may also flow through the state variables and their derivatives at any location, through transport coefficients and fluxes, and source terms, for example. Thus far, only successive substitution has been used to solve parallel multiscale models. There are also fundamental questions, such as how the properties – differential index, stability, computational complexity, and so forth – of a multiscale model are determined by the properties of the partial models and the chosen framework.
- *Time stepping* – The partial models will usually have quite different time scales. In interactive frameworks, this results in stiff problems. Different microscale and macroscale time steps are often used. The microscale time step is usually much smaller than the macroscale one, but not always. When the microscale model is

stochastic, temporal averaging is helpful in damping out microscale "noise," which would otherwise propagate into the macroscale model (Raimondeau, Aghalayam, Katsoulakis, et al. 2001).

- *Updating strategies* – Partial models that have been simplified may need to be updated by referring to the original model, either periodically or when their estimated error becomes too high (Oran and Boris 2001). The accuracy required for the simplified model can be judged through error propagation methods, similar to those used in design sensitivity studies as presented, for example, in Xin and Whiting (2000).
- *State transitions* – For systems that may exist in different regimes, identification of the prevailing state is required. Li and Kwauk (2003) solved the multiscale model together with an optimization problem: minimization of a function (stability condition) that reflects the competing mechanisms that determines the system structure.
- *Parallelization* – Parallel processing fits naturally into some multiscale codes (Laso, Picasso and Öttinger 1997; Broughton, Abraham, Bernstein, et al. 1999).

6.4.4.3

Model Verification and Validation

The verification and validation (V&V) of models in science and engineering is a discipline in its own right (Oberkampf, Trucano and Hirsch 2003). The goal is to lend confidence (Kister 2002; Best 2003) to the use of the model. Pantelides (2001) identified the construction of validated models as a major challenge in chemical engineering. Here we touch on some aspects related to multiscale models.

Verification is "the process of determining that a model implementation accurately represents the developer's conceptual description of the model and the solution to the model," that is, solving the equations correctly. It is a matter of software engineering and mathematics. There are two aspects: code verification and solution verification (Oberkampf, Trucano and Hirsch 2003). The former can be split into the verification of the numerical algorithm and software quality assurance. Solution verification involves checking for gross consistency (such as overall mass conservation), spatial and temporal convergence, and consistency with trusted solutions. Essentially, it is being confident that the *numerical error* in the predictions is acceptable and the qualitative behavior of the solution, for example the stability, corresponds to the developer's expectations.

In multiscale modeling, each partial model can be verified in isolation, using dummy functions or typical values for any variables that are shared between scales. The composite multiscale model should then be verified. This tests the integrating framework. Code and solution verification can be applied as before, but additional "trusted solutions" are potentially available. For each pair of linked scales, the chosen framework could be checked against simultaneous integration of those scales, since that has "zero integration error" (Solomon 1995; Werner 1999). The process is to compare predictions for the coarse scale variables using the chosen framework against the predictions of the same coarse variables derived from a complete simulation of the system at a finer scale. In practice, complete simulation of the system at

the microscale may not be feasible, but selected microscale simulation of regions with typical or extreme behavior may be. Another possibility is to check the integration of three scales at a time against a two-scale model for the special case where the intermediate scale should not alter the solution (Gobbert, Merchant, Borucki, et al. 1997). Of course, this is most convenient when a suitable existing two-scale model is available, which might occur as part of the iterative model building process when it is decided that another scale is needed.

Model validation is "the process of determining the degree to which a model is an accurate representation of the real world from the perspective of the intended uses of the model". It is a question of getting the actual physics of the system right. Ideally, validation consists of testing the model against experimental data drawn from a set of carefully targeted experiments (Pantelides 2001; Oberkampf, Trucano and Hirsch 2003). Both the data and the model will contain uncertainties. Two issues that are important for multiscale model validation are measuring data over a range of scales and estimating model parameters at different scales.

If a model contains parameters that are unknown, that is, too uncertain, experiments can be used to estimate them. For hierarchical models, which include multiscale models, parameter estimation can be applied simultaneously at all scales, sequentially to the scales in some order, or independently at each scale, or a mixture of these (Robinson and Ek 2000). The ideal situation is to determine each parameter independently. Conversely, the simultaneous approach is seen as "bad empiricism" (Randall and Wielicki 1997), to be used only for parameters with mechanisms that are poorly understood. In climate science and meteorology, there are complex multiscale models with many poorly known parameters. Brandt (2002) discusses the use of (multiple) multiscale computational approaches to assimilating data on the fly into dynamic models of the atmosphere.

Good data helps in both parameter estimation and model validation. For multiscale models, we would like data at each scale of interest. Different measurement techniques are used at different scales; see, for example, Balazs, Maxwell, deTeresa, et al. (2002); Gates and Hinkley (2003) for some techniques used in materials science. Like the various modeling techniques that describe the phenomena at different scales, we can locate the tools of measurement on the logarithmic time and length axes of a scale map (see Figs. 5 and 6 of Gates and Hinkley (2003)). However, there is a fundamental difference in the multiscale nature of models and measurements, at least at small scales. For models there is a rough positive correlation between time and space scales: small processes operate quickly and large processes slowly, in general. The opposite is true for measurement. There is an approximate negative correlation: it takes a long time to measure small things, and a short time to measure large ones. The consequence for multiscale modeling is that it is not possible to gather data to allow the *direct* validation or parameter estimation of some partial models. We have no choice but to view their effects through the filter of intermediate scales.

6.4.5
Summary of Multiscale Model Integration

Multiscale model integration refers to linking models at different scales together into a coherent whole. The constituent models may operate on vastly different characteristic time and length scales, and may be of disparate kinds. There are many reported examples of model integration and we are beginning to understand its principles. Classification of the types of integration is helpful. One classification scheme proposed for process modeling identifies five broad frameworks for multiscale integration: simultaneous, serial, multidomain, embedded and parallel. There is some qualitative information available on their properties and when they can be applied. Aside from conceptual modeling concerns, there are issues of integrating existing software, solution of the model, and model validation.

There is a good opportunity now, through using the large number of published models as examples, to improve our understanding of model integration. Key elements will be a clearer classification of integration methods and a suite of modeling tools to estimate the performance of a multiscale model – in terms of both dynamic and computational properties – at an early stage in the modeling process. As computing power increases and becomes more accessible, previously infeasible modeling techniques will become ripe for integration into multiscale models. New ideas will be needed to achieve tighter integration of different kinds of models.

6.5
Future Challenges

There is strong and increasing interest in the multiscale approach. Many examples of multiscale modeling are now scattered widely throughout the literature and there are the beginnings of a general "theory of multiscale modeling." However, like any rapidly evolving field, the development is uneven. In specific areas, we can choose between several sophisticated multiscale techniques, while in other fields multiscale thinking has barely made an impact. We highlight here some future challenges in the multiscale modeling of process systems.

Overall Strategy for Multiscale Modeling

In Section 6.3 we outlined a general model building strategy and discussed its extension to multiscale modeling. Solomon (1995), Li and Kwauk (2003) and others provide alternative viewpoints. Are these strategies sufficient for the *efficient development* of *parsimonious* multiscale models? One item that deserves more attention is the role of the modeling goal in multiscale systems. How can we use a statement of the modeling goal to drive model building towards:

- Identifying and selecting the scales to include in the model?
- Developing or choosing among alternative partial models at each scale?
- Guiding the integration of the partial models into a multiscale model?

We need a better understanding of the decomposition of goal sets in multiscale problems: relating goals to scales and appreciating the filtering effect of the integrating framework.

Rethinking the Partial Models

Pantelides (2001) expresses the point well: "it should not be taken for granted that techniques (e.g. for CFD or molecular modeling) that have evolved over long periods of time for 'stand-alone' use automatically possess the properties that are necessary for their integration within wider computational schemes. Indeed, our experience has been that the contrary is often true." We may need to reformulate the partial models to assist with mathematical aspects of "tighter" model integration: ensuring well-posedness, continuity, efficient Jacobian calculation, and so on. Stefanović and Pantelides (2000) provide an example.

Model Integration

Classification is the first step in generalizing our understanding of the options for linking multiscale models. Several classification schemes have been proposed (Sections 6.4.2 and 6.4.3), and there is some anecdotal information available on the properties of the classes. Are there more useful classification schemes? Attempting a formal definition of the classes, for example, Ingram, Cameron and Hangos (2004), may help improve upon current ideas. We also need to develop techniques to answer the question: how do the properties of the partial models and the nature of the integration method contribute to the properties of the resulting multiscale model? A suite of characterizing model *metrics* that can be applied at different stages of the modeling process will be of assistance here.

Numerical Methods

Solving the partial models from some scales may entail a very high computational load. Indeed, the possibility of the repeated solution of such models in a multiscale simulation highlights the need for efficient numerical methods. Multiscale models might run across different platforms and processors. The partial models may be of different types (Section 6.3.2.3) and will almost certainly have very different time scales. Specialist techniques, refined over time, are usually available for the models from different scales. We need to understand how the most suitable numerical methods for the partial models and the chosen integration scheme interact in order to develop efficient and robust solution methods for multiscale models.

Multiscale Modeling Tools

A multiscale perspective would enhance existing CAPE tools. To be effective aids for multiscale modeling such tools should:

- permit partial model development;
- allow various integration schemes to be applied;
- generate metrics that characterize the partial and multiscale models;
- provide specialist solvers for different scales;

- store validation data over the range of scales;
- archive the underlying assumptions of the partial models.

Extending the current work on open interface standards and heterogeneous simulation would facilitate these efforts.

On a final note, we need to maintain a watch on how other disciplines are approaching the challenge of multiscale modeling and its application. There are interesting developments in materials science, ecology, climate studies, medicine and many other fields (Glimm and Sharp 1997; Li and Kwauk 2003).

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