A Course in . . .

TOPICS IN TRANSPORT AND REACTION IN MULTIPHASE SYSTEMS

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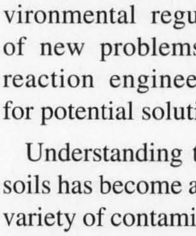
Chemical reaction engineering is a subject that requires a combination of thermodynamics, chemical kinetics, transport phenomena, and computational and applied mathematics in order to be fully understood. Furthermore, the physical systems of interest to the subject usually involve two or more phases, several components, and a strong coupling between mass, momentum, and heat transport and the chemical kinetics. Moreover, current industrial applications and government demands for environmental regulations have brought about a plethora of new problems where the fundamentals of chemical reaction engineering play a crucial role in searching for potential solutions.

Understanding the processes of transport and reaction in soils has become a crucial aspect of cleanup efforts in a wide variety of contaminated sites. The knowledge of how chemical reactions interplay with processes of mass, momentum, and energy transport is a helpful tool in identifying new strategies for air and water pollution control and for achieving better quality in microelectronics processes, coating, and cure techniques in material synthesis and processing. These few examples illustrate dramatically the importance of having a solid training in the subject of chemical reaction engineering. The applications, of course, do not diminish the importance of the subject in perhaps more traditional applications such as catalytic reaction engineering where the search for better yield and an improved selectivity still continues.

Based on the framework given above, it seems logical and timely to devote some effort to put together a graduate-level course that focuses on teaching topics that integrate transport phenomena with chemical reactions. The need for such integration has been pointed out in workshops related to new demands in chemical engineering education[1] and in international seminars on modeling chemical reactors.[2] Early efforts in trying to teach transport phenomena coupled with reactions from "first principles" can be found in Whitaker and Carbonell[3] and in Slattery,[4] and some integration can be found in the text by Rosner.[5] The contents of Table 1 in that text deviate considerably from those discussed by this author, however.



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The lack of textbooks on the subject, the rich variety of phenomena found within the domain of chemical reactions and diffusion,[6-8] and the widespread use of simplified reactor models in chemical engineering,[9] among other things, have kept the realization of this course from reaching full development. In addition to the important technological applications, the framework previously described identifies a rich learning environment for chemical engineering graduate students.

This paper describes a course on topics in "Transport and Reaction in Multiphase Systems." In the following sections the reader will find some thoughts about the ideas behind the course and the teaching technique, a description of the outline and how it is implemented, the course requirements and its supporting materials, and some concluding remarks.

IDEAS BEHIND THE COURSE AND TEACHING TECHNIQUES

A general outline of the course is given in Table 1. The course covers topics ranging from basic concepts in fluid mechanics and kinetics to concepts in boundary layers, convective mixing, transport and reaction in porous media, and applications in fluid interfaces. One of the course goals is to introduce students to the various aspects of the subject rather than producing a specialist, and in this sense the course has more breadth than depth. The key problems in each unit, however, are discussed in detail, and homework and exercises are designed to give the student an opportunity to work on the (physical and algebraic) details. Furthermore, a term paper requires additional work. It is chosen with strong input from the student and provides an excellent opportunity for the student to become knowledgeable in a particular aspect of the course.

The course is largely based on literature published in scientific journals, thus giving students an opportunity to read a paper critically and to propose alternative methods of attacking a problem. Justifying steps and discussing the validity of an author's hypothesis provide a good vehicle for students to evaluate an author's work. Another goal of the course is to foster attacking a given problem from a "research point of view" and, therefore, increasing the student's skills in investigating new problems. At the end of the course, and if this goal is achieved, the students will be better prepared for their own research.

The course is taught in an active-learning environment called "The Colloquial Approach."[10,11] In this mode of instruction, the student is the center of the learning process and the professor becomes a vehicle for organizing and providing material for the discussions. The professor conducts the discussions in a way that everyone participates fully in the process.—"the lecturer" is replaced by "the coach." In general, students regard the technique as a powerful and effective way for learning new material, for building confidence to attack problems, and for providing an environment where the many aspects of a problem are exposed to critical analysis.

COURSE DESCRIPTION

The course begins with a discussion of the vectorial and/or tensorial formulation of fundamental quantities such as divergence, gradient, and basic integral theorems such as the Green and Stokes theorems. The geometrical interpretation of these concepts and their potential application to the analysis of different systems is introduced. Different types of coordinate systems are reviewed and their relevance to fluid mechanics and transport phenomena is brought into perspective. The Leibnitz rule is used to motivate introduction to the Reynolds and general transport theorems, and the students are challenged to identify potential uses of such a tool.

Next, introduction to the concept of a continuum is addressed from a particle dynamic point of view.[12] Students

TABLE 1

Course Synopsis

Rigorous analysis of transport phenomena at the micro- and macro-scale levels in systems with mixtures of several components and featuring more than one phase. Topics include, for example: **1**-Boundary layer flows with surface reactions; **2**-Analysis of the mixing effect from a (fundamental) mechanical point-of-view, and with and without chemical reactions; **3**-Analysis of the transport in porous and structure media; use of the surface and volume averaging techniques; **4**-Analysis of the transport process at interfaces with and without chemical reactions; rigid and flexible fluid-fluid interfaces; **5**-Special applications.

Course Outline

1. Fluid Mechanics, Transport Phenomena, and Kinetics

Review of vector and tensor algebra; index notation; review of fundamental concepts in fluid mechanics; Eulerian and Langragian coordinate systems; constitutive equations; stress tensor; Reynolds transport theorem; general transport theorem; conservation equations from an axiomatic point of view; connection between conservation equations and kinetics; concept of a continuum and the relation to multicomponent mixtures; Cauchy equation of motion; Navier-Stokes equation; continuity and energy equations

2. Boundary Layer Theory with Reaction

Hydrodynamics boundary layer model; Prandtl's differential model; Von Karman's integral approximation; integral formulation for the case with reaction on the surface (Cambre and Acrivos analysis); different types of reactions; first-order and Langmuir-Hinshelwood kinetics; Rosner's analysis of diffusional falsification of activation energy and reaction order; extension to include non-isothermal systems; equations for multicomponent systems

3. Mixing Processes and Reactions

Laminar flow systems; convective-diffusive transport, Taylor-Aris problems, and the area averaging procedure; effective diffusivity and effective convective velocity; effect of reaction on the wall; single component formulation and extension to multiphase reaction systems; derivation of averaged equations and closure procedures; applications to network of reactions; introduction to the theory of moments; introduction to lamellar mixing models; fluid mechanics of mixing in single extruders; macro- and micro-mixing and the problem of averages; concept of material surfaces and description of mixing coupled with diffusion and reaction; mixing in premixed reactors and the effect on conversion and selectivity; mixing and polymeric reactgions; introduction to chaotic mixing.

4. Heterogeneous Catalytic Systems

Introduction to the method of spatial (volume) averaging—definitions, concepts, and procedures; connections with the area-averaging procedure; diffusion and reaction problems in a pellet; derivation of averaged equations; equation of motion in porous media; different geometrical scales; Darcy's law; permeability tensor; extensions to analyze isothermal packed-bed reactors; introduction to averaging procedures; homogenization method—Stokes flow in periodic structures; multiple scale analysis of effective transport; averaging methods using tools from molecular hydrodynamics and linear filtering theory (Cushman's analysis); periodic porous structures (Brenner's analysis)

5. Introduction to Interfacial Transport and Reaction

Surface coordinates; algebra of surface tensors; differential operators in the surface; Green and Stokes' theorems in the surface; Reynolds transport theorem for surfaces; kinematics of the surface; surface stress tensor; equations of motion in the surface; boundary conditions and the relation to surroundings; effect of reaction in the formulation of transport (i,.e., momentum, energy, and mass) equations for the surface; application of surface-averaging techniques to derive surface macrotransport equations; introduction to the rheological aspects on the interface; introduction to theory of mixtures for the analysis of transport and reaction on the surface.

are generally familiar with analysis of the dynamics of single body or collection of bodies, and these ideas are used along with concepts of summation, limits, and Riemann's integrals to build up the ideas of a continuum. Then concepts such as linear momentum, angular momentum, and torque are reinterpreted in terms of their new concept of the continuum. At this point, the molecular description of transport phenomena is discussed as an alternative to the continuum description. An organization introduced by Rosner[13] and a paper by Peters[14] on molecular engineering are useful references. Students are asked to compare both alternatives and their potential advantages and disadvantages and to think how they are related. The idea of "average" is briefly discussed.

Conservation principles of mass are introduced from an axiomatic point of view[15-18] and integral balances are derived. Conservation of total mass of a system and conservation of mass of one component are discussed and the role of chemical reaction is analyzed in detail. Students are questioned about their views for the cases when the reaction is homogeneous or when it is at the boundaries of the system, and whether or not each case requires a separate conservation axiom. Microscopic equations are derived and the relation between "balance equation" and "chemical reaction" is discussed. A paper by Cassano[19] is helpful in bringing students on track for the analysis. Most of the discussion is centered on isothermal systems of one component, but generalization to non-isothermal and multi-component systems is addressed. Also, organization of the transport processes used here follow the ideas of Cerro[17] (see Figure 1). Applications of the different (integral and differential) models to the various kinds of reactors is performed in homework, exercises, and additional reading.

After the fundamentals have been introduced in the first unit, various applications where convective-diffusive transport is coupled with reaction are analyzed in the second unit. I believe that from the students' point of view, it is simpler to analyze cases where the reaction occurs at the boundaries of the system rather than in the bulk. In this regard, boundary layer flows offer an excellent choice for learning prototypes.[10] These types of systems have been analyzed by Chambre and Acrivos,[20-24] among others, and their analysis stresses understanding of the phenomenon by using approximate methods. This type of approach is not only useful to gain a deeper understanding of the behavior of the system, but it also provides the student with a very rich environment for developing research abilities.

After the students have mastered the basic ideas of a problem, they can always extend them to include cases where, perhaps, numerical computation is the only choice for the solution of velocity, concentration, and temperature field equations. The basic concepts play an important role in guiding the calculations— this aspect seems trivial to an experienced professor, but the new generation of researchers needs to learn how useful it is. Boundary layer flows also offer a possibility of being studied by using integral balances—a concept that was introduced in Unit 1. These integral balances play a key role in Von Karman type approximations for momentum transfer and, by extension, mass and energy transfer. Integral equations offer a useful way to analyze and model sophisticated reactor models in heterogeneous media.[25-26]

The analysis of convective-diffusive transport and reaction following the work of Acrivos and Chambre is complemented by discussing the efforts of Rosner[27-28] on studying the effect of transport on the falsification of activation energy and reaction order. The work by Rosner also uses approximate solution methods for a variety of external and internal flows and for flat plate geometry. Students are asked to compare the different analyses (*i.e.*, Chambre and Acrivos vs. Rosner's work) and to identify general and specific aspects in the study that could be useful in other physical situations and geometry. For example, homework sets and exercises are designed to apply the concepts to boundary layer flows in cylindrical catalytic surfaces and the students are encouraged to propose simplifications that could lead to approximate solutions. At the end of the unit several situations are addressed to motivate the extension to include multicomponent and multiphase systems. Among these examples, chemical vapor deposition[29] and heterogeneous combustion[5] are useful systems.

The next unit focuses on processes of mixing with chemical reactions. Here, the idea of studying mixing relies on kinematic and/or mechanistic approaches rather than on empirical concepts. The book by Ottino,[30] and some of his papers as well as papers by Ranz,[31] offer interesting points of view and a framework useful for student discussion. In the course, the study of this perspective is preceded by analysis of dispersive mixing in tubular Poiseuille flows following a Taylor-Aris ap-

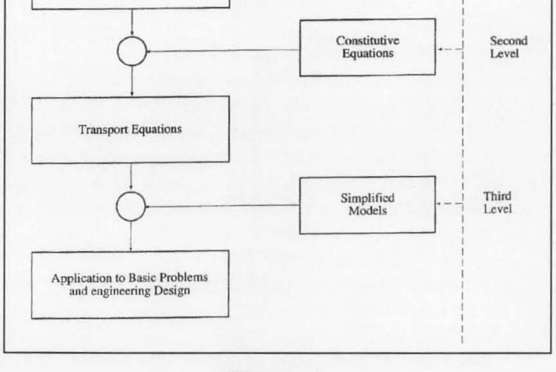

Figure 1

proach.[32-36] The effect of different types of convection (*e.g.*, Couette, squeezing, and plug flows) and the roles played by surface and bulk reactions are discussed.

A process of area averaging to derive simplified or effective transport equations is used and compared with the Taylor-Aris approach. Students are asked to compare this type of analysis with the ideas behind the methodology proposed by Ranz and Ottino and to think about new aspects of the technique. Provoking questions such as, "Is it true that everything that the Ottino approach is able to explain in convective-mixing can also be achieved by a Taylor-Aris technique?" help to keep the students busy thinking and exploring. As in the other unit, papers are assigned to read, and homework and exercises are designed to cover important details.

After the class has been exposed to area averaging approaches and has derived effective or macrotransport equations, the students are ready to move on to analyzing transport and reaction in heterogeneous catalytic systems (see Unit 4 in Table 1). Students are first introduced to the different types of media (*e.g.,* disordered and ordered) and to the different mathematical and geometrical descriptions that researchers use to describe transport and reaction in these media. Chapters from Adler's book[37] and the review paper by Sahimi, *et al.,*[38] are useful in giving students an introduction and perspective of the amount of work found on this topic in the literature.

Once the introduction and preliminary ideas have been finished, the class focuses on the volume-averaging approach following Whitaker,[39,40] Carbonell,[41] and Slattery.[42,43] The volume averaging theorem is derived following arguments borrowed from differential geometry[44-45] and then applied to a catalytic pellet with transport and reaction.[46] Reading material from the papers referred to above is assigned, as well as from Whitaker.[2] Other methodologies for averaging purposes that are available in the literature are briefly mentioned, and their differences are compared with the volume-averaging technique. For example, the homogenization method described by Sanchez-Palencia[47] and the averaging technique based on molecular hydrodynamics and the linear filtering theory[48] are introduced.

Finally, the ideas of deriving macrotransport equations and the relation with Green functions following Brenner's procedures[49] is brought into perspective. The relative amount of time spent in the different techniques depends on the interest of the students in the class and varies from year to year.

The final unit of the course is devoted to analysis of transport and reaction in fluid-fluid interfaces (see Unit 5, Table 1). These topics are offered on an optional basis. An appropriate background for the mathematics and mechanics required to understand transport phenomena in systems with fluid-fluid interfaces can be found in Chapters 8 and 9 of Aris,[50] and the study of fluid mechanics of surfaces in units follows closely the exposition in Chapter 10 of that text. This presentation is largely based on the analysis by Scriven.[51] The author believes that if the students master this material, they will have an excellent introduction to a variety of interesting applications where surface transport phenomena and reaction are important.[52-53]

TABLE 2
Sample Term Papers

- Isothermal Squeezing Flows with Chemical Reaction
- Convective-Diffusive Transport and Reaction in Couette Flows
- Basic Kinetics in Modeling a Gas-Liquid Phase Pulsed Corona Batch Reactor
- Effect of a Surfactant on the Mass Transfer with Reaction in an Ascending Bubble
- Convective-Diffusive Transport and Reaction in a Membrane Reactor: An Integral-Spectral Approach
- Effectiveness Factors in Boundary Layer Flows with a Chemical Reaction
- Modeling of a Single Pellet Diffusion Reactor
- The Spatial Volume Average Theorem Applied to Transport and Reaction in a Catalytic Pellet

COURSE REQUIREMENTS

The coursework includes one midterm exam, one final exam, several quizzes, and a term paper (see Table 2 for sample term papers). It also requires submission of homework sets and exercises as well as reading specific assigned material from the various literature sources. These reading assignments may be "formal" and replace the homework set in a particular week, and in that case, the assignment may also include questions to be answered and problems to be solved. The assignment is not graded, but students are strongly encouraged to write notes and to prepare material for submission.

After the regular class time, the instructor is usually available for individual discussions related to the formal assignments. Following the philosophy of the "Colloquial Approach," students are not given an answer to a particular question, but are motivated to look at the problem from different angles and to propose their own conclusions. Discussion among classmates is strongly encouraged and should be conducted in a professional manner.

An important aspect of this course is that every student must submit a complete folder for evaluation. This is a *portfolio* type of evaluation[54] that is very effective in keeping the student highly motivated to perform activities that are not graded. All of them must be included in the folder, where they are reviewed by the instructor.

The term paper (or project) is usually selected by follow ing one of two alternative routes: the students may choose a particular topic that interests them, but one that is suitable for the integration of concepts from transport and reaction along the lines of the course, or the instructor proposes topics to the students. In either case, the student is given a few weeks to prepare and submit a prospectus with a relatively well-defined scope that includes main references. During this period the student receives feedback from the instructor in order to identify a problem that looks reasonable for the purpose and extension of the course.

Students are encouraged to start writing a progress report in parallel with the analysis of the project problem. A preliminary version of this report is due a few weeks before the final submission at the end of the semester. The instructor meets with the student to discuss the status of the project and to suggest alternatives. A final fifteen-minute presentation in front of the class is peer-evaluated, and the comments and remarks of classmates are considered as part of the grade—this motivates students to be as professional in their presentations as possible. Faculty members and other students are invited to these report presentations. Some of the projects have been successfully continued and expanded into Master of Science theses with the submission of papers to refereed journals.

SUPPORTING MATERIALS

Students are admitted to this course only after they have been exposed to an advanced course in fluid mechanics. Typically, students have taken a fluid mechanics course in the applied mathematics program at FSU (using the text *Introduction to Fluid Dynamics* by Batchelor[55]) or a course on transport phenomena in the chemical engineering department (using the text *Energy, Momentum, and Mass Transfer in Continua*). Chemical engineering students have also taken the applied mathematics and chemical thermodynamics graduate-level courses. In addition to the texts mentioned above, *Introduction to Fluid Mechanics,* by Whitaker,[15] is helpful in several aspects of this course. Many of the students have copies of these textbooks from previous graduate-level courses or from their undergraduate studies. All of the papers cited in the previous sections are also used as references by the students of the course and, therefore, they end up with a body of references that can be useful in future work.

As the reader may conclude, there is currently no textbook that includes all the aspects of this course, but we suggest that the students use the texts by Brenner and Edwards[49] and by Levich[56] as references.

CONCLUDING REMARKS

The course has been successful in introducing students to advanced concepts and fundamentals of transport and reaction in a variety of physical systems that are relevant for current chemical reaction engineering applications. Feedback from the students indicates that the intense level of involvement in the various aspects of the different units has helped them considerably in understanding the behavior of the systems that have been studied. They have also pointed out that they feel more confident in their ability to attack research problems in their own projects. Students who have the proper background in fluid mechanics and in chemical reactor design and kinetics at the undergraduate level are the ones that benefit the most, and they are also the strongest supporters of the course.

Although the contents of this course cover a wide variety of topics, the author has been able to develop a coherent and systematic way of delivering the material by focusing on fundamental principles and theories. There is a smooth flow of ideas and applications to which the student is constantly exposed during discussions and exercises. Some students have also pointed out that the course has been useful in helping them to acquire an idea of "structure" or "framework" and a level of hierarchy among the different topics covered during the semester.

The author believes that the course is a helpful introduction for students who need (or desire) to be involved in further studies of multiphase problems such as dispersions[57] and other disordered[58] or ordered systems with the interplay of more than one phase. A good complement to this course would be a course focussing on the fundamentals of catalysis and kinetics from a theoretical and experimental point of view.

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