The Catalyst Pellet: A Very Rich POK in Progressive Learning Approaches for Transport and Reaction

Pedro E. Arce and Mario Oyanader

I. INTRODUCTION AND MOTIVATION

The concept of "Principal Object of Knowledge" or POK's was introduced in the "Colloquial Approach Environments" (Arce, 1994) to enhance the student learning and to promote a more efficient habit in engineering students to master difficult concepts. The tool was then extended to include a variety of subjects (Arce, 2000) in fluid mechanics, mass and energy balances, and continuum theory just to name a few examples. In this article, we discuss the role of the catalyst particle or pellet as a rich example of POK for students interested in learning about transport in porous media and heterogeneous reactions. The pellet is a multiscale-domain environment where diffusion process and heterogeneous reaction take place. The mathematical description of this transport and reaction system is complex and it exemplifies many multiphase and multicomponent systems (see, for example, Arce, 1994) very relevant to many engineering majors including Chemical, Biomedical, and Environmental Engineering.

The analysis of the literature shows that heterogeneous reactions and catalysis is a very populated subject. There are classical textbooks (Levenspiel, 1981) that introduces the students using somewhat simplistic models that quickly convey the information to the student and reach a mathematical description capable of obtaining concentration profiles and effectiveness factors. Others textbooks (see, for example, Fogler, 1992) presents a more "rigorous" introduction to the formulation of models and makes, also, connection with applied aspects such as the effectiveness factor determination and calculation. Textbooks with more sophisticated mathematics (Aris, 1969; Aris, 1979) takes a more pragmatic point of view and directly concentrates on a beautiful mathematical analysis with implications to practical aspects. Aris (1976) also has reported useful techniques to obtain information of a reaction-diffusion equation without actually solving the equation.

Based on the brief description presented above, it is a trained reader's choice to select one of these textbooks to read about the subject. It will, probably, be a matter of taste for this class of readers to chose one of these texts and enjoy the journey. Those more interested in the "back of the envelope calculations" will most likely be very comfortable with the simple and "global" type of analysis, i.e., Levenspiel's view. Others, with a more mathematical oriented taste will feel at home reading Aris's masterful treatise in diffusion and reaction. The spectrum of contributions could accommodate all levels that places in between these two limits.

The dilemma, however, seems not to be the same for the untrained reader—most likely the students! This class of readers will wonder how a complicated problem in two phases with reaction on the walls may be modeled as a domain with homogeneous reactions. Others may adventure a trial in a more sophisticate description but, perhaps, will not survive the mathematical machinery. Therefore, in spite of the fact that the subject of transport and reaction in heterogeneous media is populated with contributions, there is a need for a systematic approach of learning, based on first principles and that concludes with the overall or macro transport equations. This approach must follow a complete and logical sequence of steps.

In this contribution the authors will present an overall view of the status regarding problem where diffusion and reaction are involved from the point of view of the student learning and then present an "effective" and *progressive* approach to learn (sequentially) fundamental concepts. These will highlight the Catalytic Pellet as a very rich environment to learn about multiples (transport) scales and the role of multiphase process of a current relevancy in Chemical, Biomedical, and Environmental applications.

II. MORE PROGRESSIVE LEARNING APPROACHES

For systems with more than one phase and the presence of transport and reaction, it is usually a non-trivial task to introduce students to their physical as well as mathematical description. This is exactly the case of a catalytic pellet. It has two main phases, one the gas and the other the solid support with the active material. Instructors tend to use a view that renders the system to a simple case of homogeneous domain with just one phase and bulk reaction (see, for example, Levenspiel, 1981). While this approach brings the engineering equations, the inexpert reader, i.e., the students are left with a number of answered questions and a very confusing picture of the system. There is, however, a very logical and progressive approach that helps students to understand deeply the nature of systems such as the catalytic pellet and that follows a building block of knowledge (Arce, 1994). It includes the following main steps:

[Realistic physical picture→ rigorous mathematical description→process of homogenization→ engineering model→ solution→interpretation]

This is a very logical and a sequential series of steps that promote understanding of the system, offer an opportunity to the students to review concepts in previous courses, and give the chance to apply mathematical concepts learned in engineering math courses. In addition, the method promotes the overall student confidence in "engineering" an equation to describe the behavior of a complex situation. Several steps related to this progressive approach are detailed below.

1. A Sound Pedagogical Environment:

A systematic and progressive approach (Arce and Arce-Trigattti, 2000) to derive engineering equations in a catalyst pellet would be a more efficient and far less confusive exercise than those currently introducing the students by a "story telling" about "homogeneous reactions" in a true heterogeneous media. In general, students in engineering majors are quite comfortable with learning basic physical aspects of a problem and, then identifying a mathematical description that mimics closely the physics that they have "visualized". For example, it is quite rational to introduce students to a pore domain, within a catalytic pellet, where diffusion and (heterogeneous catalytic) reaction take place. Diffusion is present as the only transport mechanism inside the pore cavity so that reactants can travel from the bulk to the surface of the pore domain. Since the reaction is catalytic, students based on kinetics or physicochemical concepts have no problem in recognizing that it is located at the walls of the pore domain and, therefore, no reaction is present in the bulk of such pore. Furthermore, students that are familiar with heat surface sources can trivially associate this situation with a process at the boundary of the domain where diffusive fluxes and sources (i.e., reaction) must be involved. It is the equivalent situation to that of the heat conduction and heat generation with heterogeneous sources, a concept already introduced in the heat transfer course.

The physical description offered above is very realistic and strait forward to comprehend. There is no approximation, no "mysterious" concepts involved and no story-telling in the presentation. It is, therefore, a sound pedagogical and progressive description of a very complex phenomenon in a non-trivial domain. This description is very appealing to derive a mathematical description of the diffusion-reaction process inside the pore cavity. In fact, students that have already taken heat transfer and mass transfer usually find this situation a simple variation of the examples that they already encountered in the previous courses. What is needed next is the derivation of the differential model that involves the equation and the associated boundary conditions. This description constitutes the basis for the derivation of an engineering model useful for the calculation of the reactant concentration profiles. This is accomplished below.

2. A Robust and Progressive Approach

Based on the physical description given in section 1, above, the student here recognizes that the reaction in a catalyst pellet takes place at the surface (as it should be) and reactant must diffuse from the porous mouth towards the catalytic surface. Clearly, there is <u>no</u> homogeneous reaction within the domain of the pore cavity. Therefore in the general species continuity equation (Bird et al., 2001):

$$\frac{\partial C_A}{\partial t} + \vec{\nabla} \bullet \vec{N}_A = R_A(C_A) \tag{1}$$

the reaction term must be dropped; furthermore, if the pellet is under steady-state condition, then the time-derivative is also dropped. Therefore, equation (1) reduces simply to

$$\vec{\nabla} \bullet \vec{N}_A = 0 \tag{2}$$

If the students have been properly introduced to incompressible flows (Bird, et al, 2001), equation (2) is of the similar nature to that known as the incompressibility condition. To this point only transport concepts have been used by the student to derive the conservation or engineering equation. At this stage of the analysis, the geometry could be brought to the picture. If the rectangular geometry is the choice, then equation (2) reduces to

$$\frac{\partial N_{Ax}}{\partial x} + \frac{\partial N_{Ay}}{\partial y} = 0 \tag{3}$$

Now if, as it was stated before only diffusion is present, by using the Fick's law, equation (3) reduces to:

$$\frac{\partial^2 C_A}{\partial x^2} + \frac{\partial^2 C_A}{\partial y^2} = 0 \tag{4}$$

where the x-direction is the axial-direction in the porous cavity and the y-direction is the transversal-direction. Now, the student easily can identify boundary conditions for equation (4). This equation is identical to the Laplacian of the temperature where students focus on conduction heat transfer in a 2-D domain. By recognizing that the only transport mechanism is diffusion, by continuity of fluxes at the wall, and by remembering that the reaction takes place at the wall, the following boundary condition is easily written.

$$-D\left.\frac{\partial C_A}{\partial y}\right|_{wall} = R_A(C_{Aw}) \tag{5}$$

At this stage of the analysis, chemical engineering reaction concepts may be brought into the analysis. For an undergraduate student, R_A (C_{Aw}) is usually assumed of first order to become

$$R_A(C_{Aw}) = k(T)C_{Aw} \tag{6}$$

By using the same guidelines as before, the following boundary conditions are easily identified.

$$\left.\frac{\partial C_A}{\partial y}\right|_{center} = 0(symmetry) \tag{7a}$$

$$-D\left.\frac{\partial C_A}{\partial x}\right|_{porousmouth} = kg\left(C_A|_{p.m.} - C_A^{\infty}\right) \tag{7b}$$

$$\left.\frac{\partial C_A}{\partial x}\right|_{bottomofpore} = 0(impermeable) \tag{7c}$$

As mentioned before, the model above is very similar to those derived in heat transfer where now the source term is explicitly identified as a chemical reaction. This similarity enhances the student understanding of the system and reinforces the concept already described. The differential model (4-7) is a straightforward description of the physics present in the pore. Since the mathematical description follows very closely the physics description of the system under analysis, the students find it very appealing and very attractive leading to no confusion in the concepts and promoting an excellent understanding of the system. No storytelling is needed to write down such a model in this approach! Just a sound pedagogical environment and a very rational and very rigorous mathematical description will do the job.

3. Process of Homogenization.

The question is now, how can the student simplify the model and have a more “homogeneous view” of the problem? This is a typical question in transport phenomena problems where a more “global view” is desired to achieve meaningful results form either the measurement point of view and or the solution approach to the model stated in section 2, above. Two approaches will be discussed during the presentation:

<u>A- Simple Averaging Procedure.</u> By applying all area-averaging (i.e. please see definition of bulk concentration in Bird et al, 2001) to equation (4), the result will yield an ordinary differential equation in two types of concentration variables. One is the “local” concentration, C_A, evaluated at the pore wall and the other is the area-averaged concentration variable, $<C_A>$. This aspect is not trivial and it requires a very intensive effort from the students to realize that this situation needs a resolution before we can proceed to generate an engineering equation. One of such conditions and, in order to solve the model, the student must recognize that the following approximation is a possible “closure” to the problem:

$$\left.C_A\right|_{wall} \approx < C_A > \tag{8}$$

Which implies a very specific type of situation within the pore. This aspect will promote an interesting discussion among the students to identify what are these physical situations. Equation (8) is, in fact, one of the simplest “closure” procedures to have an engineering equation of the type.

$$\frac{d^2 < C_A >}{dx^2} + K_G < C_A > = 0 \tag{9}$$

Which is a "homogeneous" model for the pore domain where K_G now becomes a "global" constant with basic parameters of the model. Students most likely will not have any confusion from where equation (9) was derived. The students will recognize here that equation (9) is a "homogeneous" conservation equation for the pore but in the area-averaged variables, $\langle C_A \rangle$ rather than the local concentration, C_A, features by equation (4) and with modified or "effective" coefficients (K_G) rather than local coefficients such as the one identified in equation (4). Equation (9) is the basis now for additional engineering concepts such as the Effectiveness Factors (see, for example, Aris, 1974). Within the framework presented here, this concept could be viewed as a further step in the homogenization process in the catalytic pellet.

B – A Rigorous Averaging Procedure. By applying the procedure suggested, for example, by Whitaker (1983,2000), a more involved area-averaging procedure can be applied. This procedure yields an "effective" or "global" reaction rate with a constitutive equation that features various parameter involved in the problem. In short, this procedure will identify a more rigorous equation for the K_G parameter identified in equation (9). By studying this equation, the student is able to identify the various physical conditions capable of being represented by the "homogeneous" or area-averaged model. Details about this approach may be found in Whitaker (2000) and a discussion on them will be conducted during the session. A procedure such as this is perhaps more likely suited for a graduate level course.

III. ASSESSMENT

The assessment of the implementation of this approach in two different courses at the FAMU-FSU College of Engineering has shown a very promising trend. The students have been able to clearly perform better in exercises that involved conceptually the identification of quantities related to "global" parameters such as averaged concentration and "effective diffusion" as oppose to "local" concentration values and molecular diffusion. Students interviews at the end of the course have confirmed the mastering of the concepts and that they have achieved, in general, a deeper understanding of the different aspects in a heterogeneous system with diffusion and reaction. Furthermore, the platform of knowledge developed seems to be a very good tool to attack other more sophisticated systems such as a collection of pores in a catalytic particle. In addition, students have expressed their satisfaction in using concepts of engineering mathematics to develop "applied" models that are efficient in handling complex situations in transport and reaction.

IV. CONCLUSION

The article presents an analysis of the importance of a pellet as an environment where multi-scale transport process take place and introduces a systematic and progressive approach to derive differential models of the homogeneous types in a catalyst pellet. The approach avoids the storytelling methods followed in many classical textbooks. The same approach can be extended to include engineering equations valid for the entire pellet.

Once this approach has been introduced, the student in a rational fashion can extend the analysis from one-single porous cavity to a complete pellet. The procedure has shown that enhances the chances of the students to understand how a "homogeneous" type of description can be used as a useful approximation for describing the process of diffusion and reaction in a heterogeneous domain.

Some of the key benefits introduced by the approach presented here from the students point of view include: a- A realistic description of the physics of the situation, b- A clear identification of the role of the molecular diffusion and surface reaction, c- A chance to reinforce concepts already learned in previous courses, d- The opportunity for the students to apply math concepts learned in the engineering math courses, e-A clear chance of building blocks of knowledge in a sequential approach and f- Avoiding the use of story-telling arguments to derive engineering equations.

This approach also allows the students to enjoy the activities to "find things out" as Feynman used to say (Feynman, 1999). In fact, based on what we saw in our courses, the process of connecting the basic physics with the mathematical description creates a learning environment that will help the students to become a confident and alert individual. In many instances, the mathematical level required does not go beyond the one reached by the student in an undergraduate engineering math course. This approach differs in a remarkable way to that of "believe me this is the way that you must analyze this problem." Within this framework, the instructor tells the student a story about considering a "homogeneous" reaction with an effective diffusion coefficient and proceeding to write equations of change by using (usually) a shell balance for a "homogeneous" system! In contrast, the approach shown here follows a systematic procedure to derive conservation (engineering) equations in heterogeneous media.

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Contact Information for the Authors:

A- Pedro E. Arce, Department of Chemical Engineering, Tennessee Technological University, Box 5013, Cookeville, TN 38505; E-mail: parce@tntech.edu.
B- Mario Oyanader, Department of Chemical Engineering, Universidad Catolical del Norte, Antofagasta, Chile.