USING A WEB MODULE TO TEACH STOCHASTIC MODELING

MARKUS KRAFT, SEBASTIAN MOSBACH, WOLFGANG WAGNER*
University of Cambridge • Cambridge CB2 3RA, United Kingdom

Computational modeling in chemical engineering is becoming more and more a field in its own right, due largely to the rapidly increasing power of computers but also because of progress being made in developing numerical algorithms, which are necessary to solve sophisticated models.

The industry is highly interested because computer simulations have significantly lower costs compared to experimental studies. Some important ingredients for the field include accurate physical and chemical models in mathematical form, numerical values for the parameters that occur in these models (either taken from carefully selected experiments or from first-principles calculations), fast computers, efficient and powerful numerical methods, and—most importantly—competent engineers who are aware of the limitations of the models, parameters, and numerical methods.

Although important, the whole field of computational engineering is far too rich to be taught in a single course. In this article we discuss the teaching of stochastic (or "Monte Carlo") methods to students of chemical engineering. Monte Carlo methods have been shown to be highly efficient in many applications and can be found in various areas in the process and chemical industry, such as polymer synthesis, crystallization, liquid-liquid extraction, etc. They are also useful when it comes to simulating turbulent flames and their emissions as well as aerosol transport in the atmosphere. More generally, it has been demonstrated in some cases that stochastic models can account for effects that the corresponding deterministic models cannot. This is because fluctuations can sometimes significantly change the overall behavior of nonlinear physical models.

Another important aspect of Monte Carlo methods is, in our opinion, the connection to mathematics—which provides an appropriate language by means of the theory of stochastic processes. In the last decades a number of important mathematical results have been achieved that shift Monte Carlo methods from an intuitive, naive modeling level to the rigorous mathematical discipline of interacting stochastic-particle systems and their corresponding limit equations. A class of stochastic processes which is relevant for chemical engineering is Markov processes, in particular jump and Wiener processes (Brownian motion). To the best knowledge of the authors, so far in chemical engineering the subject has been taught from an intuitive point of view, focusing mainly on the physical motivation of the model. Examples can be found

Markus Kraft *obtained the academic degree "Diplom Technomathematiker" at the University of Kaiserslautern in1992 and completed his "Dr. rer. nat." in the Department of Chemistry at the same university in 1997. He has been a lecturer in the Department of Chemical Engineering at the University of Cambridge since 1999. His main research interests are in the field of computational chemical engineering.*

Sebastian Mosbach *studied physics and computer science at the University of Kaiserslautern, Germany, and obtained the equivalent of a masters (Part III of the Mathematical Tripos) in theoretical physics at the University of Cambridge, UK. He is currently studying for his Ph.D. in chemical engineering at Cambridge.*

Wolfgang Wagner *studied mathematics at the University of Leningrad (St.Petersburg) and received his Ph.D. in 1980. He is working at the Weierstrass Institute for Applied Analysis and Stochastics (Berlin) in the research group "Interacting random systems." His current fields of interest include Monte Carlo algorithms for nonlinear equations, and limit theorems for interacting particle systems.*

* *Address: Weierstrass Institute for Applied Analysis and Stochastics, D10117 Berlin, Germany*

in reference one as well as in the course CH 235 (AUG) 3:0 in the masters program taught at the IISc-Bangalore (<http://www.iisc.ernet.in/soi/ch.htm>), which is based in parts on the book by D.M. Himmelblau and K.B. Bischoff, *Process Analysis and Simulation*, first published by John Wiley in 1967.

With this in mind the authors felt a need to design a course to bridge the gap between the *physical*—say, direct simulation methods—and the more rigorous *mathematical* approach. First, we aim to enable students to understand current Monte Carlo methods on a more fundamental level and also to help them improve a given Monte Carlo method in terms of its numerical efficiency. Second, we teach students the connections between deterministic models and their stochastic counterparts given by a Monte Carlo algorithm.

A first result of the authors' activity is the 16-lecture course taught in the Department of Chemical Engineering at the University of Cambridge. The course, named Stochastic Modeling in Chemical Engineering, is given to students who are at an advanced undergraduate/beginning postgraduate level in the last (fourth) year of the undergraduate curriculum. At that stage, students have already been exposed to some computational techniques in process engineering, and they have a solid knowledge of models used in chemical engineering.

We demonstrate, while looking at a particular example, how a stochastic process can be obtained from its Master equation and discuss how Monte Carlo algorithms can be implemented on a computer.

The stochastic modeling course starts with examples in chemical engineering that lend themselves to a stochastic approach. After the introduction, we discuss how random numbers can be obtained using numerical algorithms. Then the notion of a Markov process is introduced, and the particular example of a jump process (death process) is examined.

Using this basis, we then develop a jump process that can be used to model a perfectly mixed gas in a tank reactor. For this system, a Direct Simulation Monte Carlo (DSMC) method is introduced that simulates how the physical quantities of interest change with time. The DSMC algorithm is based on the work of Gillespie[2, 3] published in the 1970s. We demonstrate, while looking at a particular example, how a stochastic process can be obtained from its Master equation and discuss how Monte Carlo algorithms can be implemented on a computer. For this algorithm, we present techniques for investigating numerical properties of Monte Carlo algorithms in general.

We then generalize the DSMC algorithm to arbitrary systems of ordinary differential equations (ODEs) and study coagulation of particles as described by the Smoluchowski equation.[4]

Finally, we introduce stochastic reactor models, which account for nonideally mixed chemical reactors. These models are based on the joint scalar probability density function transport equation, which is also frequently used for modeling turbulent reacting flow.

For all examples, we state a DSMC algorithm that can be easily implemented on a computer. The lectures are accompanied by example papers, which are discussed in small, supervised groups. These example papers are pencil and paper problems in a classical fashion; they do not contain any programming exercises, which would have to be carried out on a computer. This is partly because the students have not been taught a high-level computer language such as FORTRAN or C, and also because the implementation of algorithms as part of computer science does not provide any insight from the modeling perspective. The overall assessment of the students' learning progress is at the end of the academic year, when they have to complete four papers that cover all the courses taught in that year.

To introduce an element of continuous assessment and to give students the possibility of getting some working experience with stochastic algorithms, the stochastic modeling course is complemented by a Web module, which will be described in more detail later in this paper. The purpose of this Web module is to let students gain some experience on how to perform and investigate a Monte Carlo simulation algorithm without assuming any knowledge of a programming language.

The Web module can be accessed from every computer that runs Microsoft Internet Explorer or a similar Java-enabled browser. All students at Cambridge (and a large proportion of students worldwide) have access to such computers either at home or on campus.

The Web module, as it has been set up, also introduces an element of continuous assessment. As described in more detail below, it contains a set of tasks and exercises, which students have to complete either in small groups or on their own. They are asked to summarize their results and send a short report by mail to the research assistant or the lecturer who accompanied the students' progress. Some students even completed their reports at home during the vacation period and kept in touch via e-mail.

The content and design of the Web module are described in detail in the next section, which refers to one particular part of the stochastic modeling course—that dealing with the direct simulation of chemical reactions in a perfectly stirred batch reactor. Two reactions are studied: a simple chemical reaction for efficiency and convergence analysis, and the Belousov-Zhabotinsky (BZ) reaction as an example of a chaotic chemical system. The well-known BZ system has been

chosen to study the influence of fluctuations on chemical reactions. Furthermore, it presents an example of an oscillating reaction and aims to illuminate how such a system can be studied analytically using local stability analysis. In various exercises involving a number of numerical experiments with the Java applet, students have the opportunity to develop an understanding of the chemical reactions and see how the theoretical analysis is related to the actual behavior of the system.

We hope that by making this teaching resource available on the Internet we can encourage other university teachers to use it as an addition to their lecture courses. In our opinion, the module is not limited to chemical engineering courses but might also be useful in all physics, chemistry, or mathematics courses that contain elements of stochastic processes and/or nonlinear ordinary differential equations.

DESCRIPTION OF THE WEB SITE

The Web module can be accessed through the home page of the course "Stochastic Modeling in Chemical Engineering," < http://www.cheng.cam.ac.uk/c4e/StoMo>, or directly via <http://www.cheng.cam.ac.uk/c4e/WebModule>. The site is structured as follows: On the title page, the table of contents is shown in the form of links to all subsequent pages. Furthermore, at the top and bottom of every page there are navigational buttons, so that the user is led through the whole site step by step. The following pages can be found:

- *Introduction*
- *Theory*
  - *Some theory for a simple example*
  - *Some theory for the Belousov-Zhabotinsky system*
- *Algorithms*
  - *Algorithm for the simple example*
  - *Algorithm for the Belousov-Zhabotinsky system*
- *Numerical experiments*
- *Videos of actual experiments*
- *Questionnaire*
- *Web-based teaching—a survey*
- *Bibliography*

On the introductory page, we explain the subjects and the aims of the Web module and its connections to other teaching units. We focus on three areas: reaction engineering, Monte Carlo methods, and dynamical systems and chaos. We discuss how the Web module is related to these areas. And we specifically state the aims we want to achieve, which are:

1. *To provide a numerical tool, based on a Monte Carlo method, to simulate chemical reactions and understand the numerical properties of Monte Carlo methods for chemical reactions*
2. *To study a chemical reaction system analytically using linear stability analysis*
3. *To present an example for oscillating reactions and chemical feedback*

The Connection to Other Teaching units are specific to the chemical engineering course in Cambridge, but these courses are taught in similar fashion in other chemical engineering departments the world over. Curriculum containing materials that provide the basis for the modules' successful use and the problems' completion includes: Computer-Aided Process Engineering, Statistics, Mathematical Modeling of Chemical Reactors, Combustion, Bioprocess Engineering, Thermodynamics, and Kinetic Theory.

In the theory section, we introduce two example systems to be considered. The first consists of two very simple chemical reactions, allowing students to focus on investigating numerical properties of the algorithm rather than struggling with the complexity of the system itself. In the second, students are familiarized with the BZ reaction in some detail, but in order to avoid confusion, we restrict ourselves to a simplified oregonator mechanism due to Field, Körös, and Noyes.[5, 6]

$$A + Y \xrightarrow{k_3} X + P$$

$$X + Y \xrightarrow{k_2} 2P$$

$$A + X \xrightarrow{k_5} 2X + 2Z$$

$$2X \xrightarrow{k_4} A + P$$

$$B + Z \xrightarrow{k_c} \tfrac{1}{2} f Y$$

Here, X, Y, and Z are the species of interest in which the oscillations are to be observed, A and B are assumed to be constant, and f is an adjustable (not necessarily integer) parameter which arises due to the simplification of the model. By performing a transformation of variables[7] on the corresponding reaction-rate equations, we derive a system of three dimensionless ordinary differential equations:

$$\frac{dx}{d\tau} = \frac{1}{\varepsilon}\left[qy - xy + x(1-x)\right]$$

$$\frac{dy}{d\tau} = \frac{1}{\varepsilon'}\left[-qy - xy + fz\right]$$

$$\frac{dz}{d\tau} = x - z$$

The dimensionless parameters ε, ε', q, and f, which are functions of A, B, and the rate constants, determine the qualitative dynamical behavior of the system. Specifically, students are led through the calculation of the steady state and the so-called nullclines, which, using a number of graphs, provides an intuitive understanding of the time evolution in the phase space. Finally, we demonstrate how to perform a local stability analysis by means of linearization at the point of steady state including a classification of the eigenvalues of the Jacobi matrix.

On the algorithm pages, we write down explicitly the stochastic algorithms for both systems of chemical reactions. This is simply a specialization of the general method pre-

sented in the lectures, using the same notation, and is furthermore identical to Gillespie's method.[2, 3] As mentioned above, instead of referring to a particular programming language, we describe in words and formulae every step of the algorithm. In the lecture course, students have learned the concepts of reaction-rate functions (or simply, rates) $\tilde{K}_i$, waiting time parameter

$$\pi = \sum \tilde{K}_i$$

and reaction probabilities $p_i = \tilde{K}_i / \pi$. Equipped with this knowledge, students are well prepared to understand the algorithms. The one for the BZ system reads:

1. *Initialize the number of particles for each species, i.e., N_X, N_Y, and N_Z, set the time t equal to zero, and fix a stopping time t_{stop}.*
2. *Calculate the rates $\tilde{K}_i$, the waiting time parameter π, and the reaction probabilities p_i.*
3. *Generate an exponentially distributed waiting time τ, where the decay constant of the exponential is given by the waiting time parameter π. Generate a reaction index α according to the reaction probabilities p_i.*
4. *Perform the reaction α chosen in the previous step, i.e.:*
   - *If $\alpha = 1$ then increase N_X by 1 and decrease N_Y by 1*
   - *If $\alpha = 2$ then decrease N_X and N_Y each by 1*
   - *If $\alpha = 3$ then increase N_X by 1 and N_Z by 2*
   - *If $\alpha = 4$ then decrease N_X by 2*
   - *If $\alpha = 5$ then increase N_Y by f/2 and decrease N_Z by 1*
5. *Advance the current time t to $t + \tau$. If $t < t_{stop}$, go to step 2; otherwise stop.*

The description of the Java applet and the list of exercises are a central part of the Web site. The corresponding page first explains the purpose of the applet, then its elements and parameters, how to run a simulation, and how to obtain measurement data. The applet can be started by clicking on a link that opens a new window containing just the applet, so that students can run simulations and browse the Web site at the same time. Figure 1 shows the program with a typical output of the time evolution of the species, the phase space diagram, and numerical measurement data. It might be necessary to download the Java 2 Runtime Environment plug-in to view this page (the page has been tested using version 1.3.1_03).

Apart from experimenting with the applet in a rather unsystematic way, we expect students to complete a number of problems and exercises. The students who participated in the lecture course were asked to include answers to the problems in an essay to be handed in for grading.

The exercises focus on three areas: the numerical properties of the stochastic algorithm, the physical properties of the BZ system, and the characteristic features of dynamical systems. On the numerical side, students are asked to study systematic and statistical errors, their convergence, and some efficiency-related issues using the simple system of chemical reactions as a test case. Then, by investigating the BZ system, the theoretical knowledge on dynamical systems developed earlier is to be consolidated. Feedback on the reports was given in supervisions, which are tutorials for groups of two to three students (typical for the Cambridge system of education). In some cases students contacted research assistants through e-mail to ask specific questions.

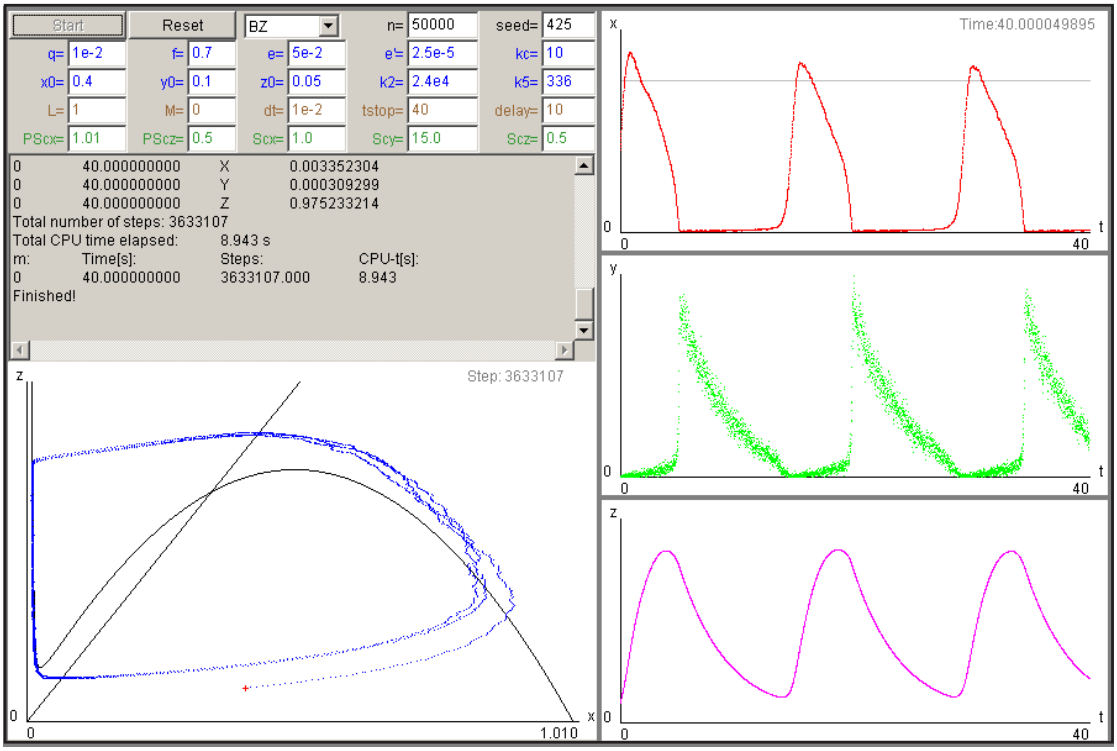

Figure 1. *Screenshot of the Java applet in action.*

For motivational purposes, we include an extra page with links to videos of BZ-reaction experiments. These videos were not produced by the authors, but they do complement the Web module, since running the Java applet can qualitatively reproduce the behavior of these experiments.

Lastly, the students can give feedback by completing an online evaluation form. We compiled a number of questions to gather information on how to improve the Web module for the next academic year. Different criteria are evaluated, including technical usability, organization of content, and quality of the problems and exercises. Students answer each question by choosing a number from 1 to 5. We also ask how long it took them to complete the problems, in order to estimate how to alter or add exercises in the future. In text boxes, we offer the opportunity to give more detailed feedback. Users can identify strengths and weaknesses of the Web module and comment on the Internet-based teaching approach in general.

On the page titled "Web-based teaching—a survey" we list a number of Web sites that also attempt to supplement conventional courses. We do this mainly because during the design phase of this Web module, we came across many examples that we thought deserved some advertising. We distinguish between different classes of teaching material and give short descriptions of a selection of Web pages. More details on various aspects of the material presented can be found in references one and seven through 13, which are all included in the Web module.

EVALUATION

The course Stochastic Modeling in Chemical Engineering is an elective in the fourth year. Typically up to 10 students sign up for it. At the end of each lecture course the students answer a questionnaire to assess the course's content and technical issues. Most students indicate they enjoyed completing the stochastic-modeling Web module, in particular the hands-on aspect of numerical experimentation. Also highly rated is the aspect of modeling real chemical reactions as shown in the videos. Furthermore, students remarked that the structure of the Web module allowed them to concentrate on better understanding the material without having to worry about the fine points of computer programming. With the activity presented as a Web module, they were able to progress through it at their own pace, wherever they had access to a computer. Most of the students, however, complained about the amount of material and the shortage of time they had to complete the tasks.

CONCLUDING REMARKS

In this paper, we described the course development on stochastic numerical methods in chemical engineering at Cambridge and presented a Web module, which is a central part of the fourth-year course Stochastic Modeling in Chemical Engineering. This Web module allows students at Cambridge to practice concepts taught in lectures, and it offers students worldwide a practical tool for studying stochastic methods and nonlinear chemical systems. Two chemical reactions in a perfectly mixed batch reactor can be studied using a DSMC algorithm implemented in a Java applet.

In working through the Web module, the users are supposed to write an essay that includes answers to a set of problems given in the module. To obtain these answers, students need to make extensive use of the Java applet. The Web module contains some additional material on the chemical and physical background of the reactions being studied. It also provides some basic material on linear-stability analysis.

Some videos and a survey on Web-based teaching complete the Web module. An online questionnaire gives users the opportunity to comment on various aspects and suggest improvements.

We view this course as a first step into Web-based teaching. We are planning to increase the number of Web modules for this particular course, but also hope to begin a virtual laboratory. Funding for this activity has already been made available by the Cambridge MIT Institute (CMI), and first results will be published in due course.

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