STOCHASTIC MODELING OF CHEMICAL PROCESS SYSTEMS

Part 2: The Master Equation

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THE GENERAL PHILOSOPHY of stochastic modeling was discussed in the first part of this series on stochastic modeling of chemical process systems. Moreover, a particular class of stochastic models was outlined, based on the master equation. In Part 2, the master equation will be discussed and an approximate solution technique known as the System Size Expansion will be presented. The formal apparatus developed here will be utilized in the third part of the series to model a chemically-reacting system.

In developing stochastic models based on the master equation, it is assumed: (1) that a population of discrete entities exists and evolves through interaction between the entities; (2) that the entities possess certain characteristics such as size, temperature, and chemical makeup, which distinguish groups of entities from other groups; and (3) that the entities exist in Euclidian space of zero or higher order. A stochastic model for this population can be derived based on the concepts of probability theory. The resultant expression for the joint probability of the random variables designating the distinct groups of entities in the population is known as the master equation [1,2]. The master equation arises directly from the assumption that the interactions between entities possess the Markov property; changes in the system depend solely on the present state of the population and not on its past states.

In what follows, the random variable N denotes the number of entities in a specific group in the population. Subscript j signifies the number of entities possessing feature j; each feature is assigned a positive integer. Similarly, multiple subscripts will designate distinct groups of characteristics, *e.g.*,

$$\left\{N_{i,j}: j \in \{1,2,3,\ldots\}, i \in \{-\infty,\ldots,-2,-1,0,1,2,\ldots,+\infty\}\right\}$$

can denote the number of entities with feature j, located at point i on a discretized number line. The joint probability of the random variables {N} will be denoted as P({n},t) or simply P, when $\{\{n\}: n \in (0,1,2,3,\ldots)\}$, *i.e.*, when the state space of N consists of the positive integers. However, for convenience of mathematical manipulation it will be desirable to approximate n as a positive real number *i.e.*, $\{\{n\}: n \in (0, +\infty)\}$, when performing the System Size Expansion introduced in the following section; the joint probability p becomes a joint probability density function denoted as p({n},t), or simply p. In both expressions, t refers to time since the model describes a process evolving in time. P({n},t) is interpreted as

$$P\left(\{N_1 = n_1, N_2 = n_2, \ldots\}, t\right)$$

which is the joint probability that the random variable N_1 has a value of n_1, the random variable N_2 a value of n_2, and so on at time t. It is also necessary to define a conditional probability, $P(\{n\}_1, t_1 \mid \{n\}_0, t_0)$, which is the probability that the random variable N_1 has a value of n_{11}, the random variable N_2 a value of n_{21}, and so on at time t_1, given that the random variable N_1 has a value of n_{10}, and so on at time t_0.

THE MASTER EQUATION

Letting $t_0 = t$ and $t_1 = t + \tau$, where τ is a small time interval tending toward zero, the conditional probability $P(\{n\}_1, t+\tau \mid \{n\}_0, t)$ can be expanded in a Taylor series

$$P\left(\{n\}_1, t+\tau \middle| \{n\}_0, t\right) = \left[1 - \tau \sum_{\{n\}} W_t\left(\{n\}_0, \{n\}\right)\right] \delta^k\left(\{n\}_1 - \{n\}_0\right) + \tau W_t\left(\{n\}_0, \{n\}_1\right) + 0\left(\tau^2\right) \quad (1)$$

The quantity $W_t(\{n\}_0, \{n\}_1)$ is the transition probability per unit time that the population changes from state $\{n\}_0$ to state $\{n\}_1$ in the time interval between t and $t + \tau$. The quantity

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$$\tau\sum_{\{n\}} W_t\left(\{n\}_0,\{n\}\right)-\tau W_t\left(\{n\}_0,\{n\}_0\right)$$

is the total probability of transition from state $\{n\}_0$ to any other state during the time interval between t and $t+\tau$. Thus, $\tau\ W_t(\{n\}_0,\{n\}_1)$ is the probability of a transition from $\{n\}_0$ to $\{n\}_1$ during the time interval between t and $t+\tau$, and

$$\left[1-\tau\sum_{\{n\}} W_t\left(\{n\}_0,\{n\}\right)\right]\delta^k\left(\{n\}_1-\{n\}_0\right)+\tau W_t\left(\{n\}_0,\{n\}_0\right)$$

is the probability that no transitions occur during the time interval between t and $t+\tau$.

Assuming that the states of the population possess the Markov property, $P(\{n\}_1,t+\tau)$ can be expressed as

$$P\left(\{n\}_1,t+\tau\right)=\sum_{\{n\}_0} P\left(\{n\}_1,t+\tau\,\middle|\,\{n\}_0,t\right)P\left(\{n\}_0,t\right) \tag{2}$$

Taking the limit of this expression as $\tau \to 0$ yields the master equation:

$$\frac{dP\left(\{n\}_1,t\right)}{dt}=\sum_{\{n\}} W_t\left(\{n\},\{n\}_1\right)P\left(\{n\},t\right) \tag{3}$$

where $W_t(\{n\}_1,\{n\}_1)$ is defined as

$$W_t\left(\{n\}_1,\{n\}_1\right) = -\sum_{\substack{\{n\}\\ \{n\}\neq\{n\}_1}} W_t\left(\{n\}_1,\{n\}\right) \tag{4}$$

MASTER EQUATION EXPANSION

The master equation, as given in Eq. (3), is in the form of an ordinary differential equation. Since $P(\{n\},t)$ appears only to the first power, the equation is linear. However, if the state space is large, Eq. (3) is a large system of coupled equations—one for each possible state. For example, for the set of two random variables

$$\left\{\{n\}_i=\{n_1,n_2\}_i : n_j\in\{0,1\}, i\in\{1,2,3,4\}, j\in\{1,2\}\right\}$$

there are four possible events

$$\{n\}_1=\{0,0\},\{n\}_2=\{1,0\},\{n\}_3=\{0,1\},\{n\}_4=\{1,1\}$$

Note that the state space of either of the two random variables, N_1 and N_2, consists of two events, *i.e.*, {0,1}. The resultant system of differential equations could comprise four coupled equations. In general, if k(j) is the number of events in the state space of random variable N, then the number of coupled differential equations could be equal to

$$\prod_j k(j)$$

Even if j is equal to 1, this could still result in a very large system of equations. For example, if the state space of random variable 1 consists of all the integers [k(1) equal to $+\infty$], the number of equations will be infinite. It is necessary, therefore, to develop an approximation procedure for the solution of such equations.

The use of the System Size Expansion is predicated upon the fact that often, for a system involving interactions between entities in the population, the magnitude of the change in the number of entities in the system following a transition is an extensive variable, *e.g.*, the number of molecules, but the dependence of the rate of transition on the number of entities is expressed as an intensive variable, *i.e.*, the concentration of molecules. As an example, consider a system consisting of two populations A and B, undergoing second order interactions between them in a volume Ω. Suppose that q members are in population A and r members are in population B, and that a transition takes place when a member of population A meets a member of population B. In most cases, the rate of such a transition will not only be proportional to q times r, but also inversely proportional to the volume squared. This follows intuitively from the image of the entities moving freely in the volume Ω. Decreasing Ω will increase the number of collisions between members of populations A and B. The rate of transition is thus dependent on the density or concentration of entities in the system.

Under the assumption that the System Size Expansion is valid, the term representing the rate of transition $W_t(\{n\},\{n\}_1)$ in the master equation, Eq. (3), is first rewritten as $W_t(\{n\};\{n\}_1 - \{n\})$, where $\{n\}_1 - \{n\}$ is the magnitude of the change in the random variables {N} during a transition. Letting $\{\xi\} = \{n\}_1 - \{n\}$, the rate of transition can be expressed as $W_t(\{n\};\{\xi\})$. It can further be rewritten as

$$W_t\left(\{n\};\{\xi\}\right)=\Omega\Psi_t\left(\left\{\frac{n}{\Omega}\right\};\{\xi\}\right) \tag{5}$$

if it is assumed to be a homogeneous function of the random variable. The rate of transition is now a function of the intensive random variables $\{N/\Omega\}$, and the

System Size Expansion can be introduced.

Making a change of variables and introducing the new random variables {Z} and the deterministic variables {ϕ} such that

$$\{N\} = \Omega\{\phi(t)\} + \Omega^{\frac{1}{2}}\{Z\} \quad (6)$$

the rate of transition is rewritten as

$$W_t(\{n\};\{\xi\}) = \Omega\Psi_t\left(\left\{\phi(t) + \Omega^{\frac{1}{2}} z\right\};\{\xi\}\right) \quad (7)$$

It will be seen later that the deterministic variables {ϕ} correspond to the macroscopic behavior of the system. The master equation, Eq. (3), is then of the form

$$\frac{dP\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}_1, t\right)}{dt} = \sum_{\{\xi\}} \Omega\Psi_t\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\};\{\xi\}\right) P\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}, t\right) \quad (8)$$

To proceed with the expansion, it is useful to define the first and second jump moments, A_i and $B_{i,j}$, respectively, and $\tilde{A}_i$ and $\tilde{B}_{i,j}$ as follows:

$$A_i(\{n\}) = \sum_{n_{il}} (n_{il} - n_i) W_t(\{n\},\{n\}_l)$$

$$= \sum_{\xi_i} \xi_i W_t(\{n\};\xi_i) = \Omega \sum_{\xi_i} \xi_i \psi_T\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\};\xi_i\right) \quad (9)$$

$$\tilde{A}_i\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) = \Omega^{-1} A_i(\{n\}) \quad (10)$$

and

$$B_{i,j}(\{n\}) = \sum_{n_{il}} \sum_{n_{jl}} (n_{il} - n_i)(n_{jl} - n_j) W_t(\{n\},\{n\}_l)$$

$$= \sum_{\xi_i} \sum_{\xi_j} \xi_i \xi_j W_t(\{n\};\xi_i,\xi_j)$$

$$= \Omega \sum_{\xi_i} \sum_{\xi_j} \xi_i \xi_j \psi_t\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\};\xi_i,\xi_j\right) \quad (11)$$

$$\tilde{B}_{i,j}\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) = \Omega^{-1} B_{i,j}(\{n\}) \quad (12)$$

The expression $W_t(\{n\};\xi_i\xi_j)$ denotes the dependence of the rate of transition on both n_i and n_j. If no such dependency exists, either ξ_i or ξ_j is identically zero. The master equation, Eq. (8), can then be expanded in powers of Ω to yield

$$\frac{\partial p(\{z\},t)}{\partial t} - \Omega^{\frac{1}{2}} \sum_i \frac{d\phi_i}{dt} \frac{\partial p(\{z\},t)}{\partial z_i}$$

$$= -\Omega^{\frac{1}{2}} \sum_i \frac{\partial}{\partial z_i}\left[\tilde{A}_i\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) p(\{z\},t)\right]$$

$$+ \frac{1}{2} \sum_i \sum_j \frac{\partial^2}{\partial z_i \partial z_j}\left[\tilde{B}_{i,j}\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) p(\{z\},t)\right] + O\left(\Omega^{-\frac{1}{2}}\right)$$

(13)

where p({z},t) is the probability density function of the new random variables {Z}, and $O(\Omega^{-1/2})$ represents terms of order $\Omega^{-1/2}$ and smaller.

To proceed further, the expansions of $\tilde{A}_i$ and $\tilde{B}_{i,j}$ in powers of Ω must be performed; they yield

$$\tilde{A}_i\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) = \tilde{A}_i(\{\phi(t)\}) + \Omega^{-\frac{1}{2}} \sum_j z_j \tilde{A}_{i,j}(\{\phi(t)\}) + O(\Omega^{-1}) \quad (14)$$

$$\tilde{B}_{i,j}\left(\left\{\phi(t) + \Omega^{-\frac{1}{2}} z\right\}\right) = \tilde{B}_{i,j}(\{\phi(t)\}) + O\left(\Omega^{-\frac{1}{2}}\right) \quad (15)$$

These expressions define the expansion coefficients $\tilde{A}_i$, $\tilde{A}_{i,j}$, and $\tilde{B}_{i,j}$. The expanded master equation, Eq. (13), thus becomes

$$\frac{\partial p}{\partial t} - \Omega^{\frac{1}{2}} \sum_i \frac{d\phi_i}{dt} \frac{\partial p}{\partial z_i} = -\Omega^{\frac{1}{2}} \sum_i \tilde{A}_i \frac{\partial p}{\partial z_i} - \sum_i \sum_j \tilde{A}_{i,j} \frac{\partial}{\partial z_i}[z_i p]$$

$$+ \frac{1}{2} \sum_i \sum_j \tilde{B}_{i,j} \frac{\partial^2 p}{\partial z_i \partial z_j} + O\left(\Omega^{-\frac{1}{2}}\right) \quad (16)$$

The terms of order $\Omega^{1/2}$ on both sides of this expression cancel if ϕ_i obeys

$$\frac{d\phi_i}{dt} = \tilde{A}_i(\{\phi(t)\}) \quad (17)$$

Letting Ω approach infinity (thermodynamic limit), the last term on the right-hand side of Eq. (16) vanishes, thereby yielding

$$\frac{\partial p}{\partial t} = -\sum_i \sum_j \tilde{A}_{i,j} \frac{\partial}{\partial z_i}[z_j p] + \frac{1}{2} \sum_i \sum_j \tilde{B}_{i,j} \frac{\partial^2 p}{\partial z_i \partial z_j} \quad (18)$$

where $\tilde{A}_i$, $\tilde{A}_{i,j}$, and $\tilde{B}_{i,j}$ are given by Eqs. (14) and (15). quations (17) and (18) are the expressions resulting from the System Size Expansion.

Even in the form given by Eq. (18), the master equation for the system may still involve a large number of variables {Z}, since the number of random variables is equal to the number of distinct populations in the system, which may be large. Nevertheless, Eq. (18) is a linear Fokker-Planck equation whose solution yields a multivariate, normal distribution; the linearity is in reference to the coefficients $\widetilde{A}_{i,j}$ and $\widetilde{B}_{i,j}$. In general, a Fokker-Planck equation is said to be linear if it can be written in the form of Eq. (18) and the coefficients do not depend on the random variables {Z}. Although the coefficients, $\widetilde{A}_{i,j}$ and $\widetilde{B}_{i,j}$, are linear, they are time-dependent through the dependence on {ϕ}, obeying the system of coupled, possibly non-linear, differential equations given by Eq. (17). To solve the Fokker-Planck equation, Eq. (18), it is necessary to first solve Eq. (17) for {ϕ}.

Solving Eq. (17) for {ϕ} can itself be a highly arduous task, especially if the equations are non-linear. Methods for solving the Fokker-Planck equations with the constant coefficient matrices, $\widetilde{A}_{i,j}$ and $\widetilde{B}_{i,j}$, are available, but the addition of a time-dependence quickly increases the complexity of the problem. Such difficulties can be circumvented in cases where a complete expression for p({z},t) can be substituted by expressions for its moments in general, and for its means, $<Z_i>$, and the cross-moments $<Z_iZ_j>$ in particular. This is accomplished by multiplying both sides of Eq. (18) by z_i or z_iz_j, and integrating over all variables from $-\infty$ to $+\infty$; this yields

$$\frac{d}{dt}\langle Z_i\rangle = \sum_j \tilde{A}_{i,j}\langle Z_j\rangle \tag{19}$$

and

$$\frac{d}{dt}\langle Z_i Z_j\rangle = \sum_k \left[\tilde{A}_{i,k}\langle Z_k Z_j\rangle + \tilde{A}_{j,k}\langle Z_k Z_i\rangle\right] + \tilde{B}_{i,j} \tag{20}$$

These expressions give rise to the governing differential equation for the covariances of {Z} as

$$\frac{d}{dt}\mathrm{Cov}\left[Z_i,Z_j\right] = \sum_k \left\{\tilde{A}_{i,k}\,\mathrm{Cov}\left[Z_k,Z_j\right] + \tilde{A}_{j,k}\,\mathrm{Cov}\left[Z_k,Z_i\right]\right\} + \tilde{B}_{i,j} \tag{21}$$

Returning to the original random variables {N} and using their definitions in terms of {ϕ} and {z}, the expressions for their means and covariances can be obtained from Eqs. (19) and (21), respectively, as

$$\frac{d}{dt}\langle N_i\rangle = \Omega\frac{d\phi_i}{dt} + \Omega^{\frac{1}{2}}\frac{d}{dt}\langle Z_i\rangle = \Omega\tilde{A}_i + \Omega^{\frac{1}{2}}\sum_j \tilde{A}_{i,j}\langle Z_j\rangle \tag{22}$$

and

$$\frac{d}{dt}\mathrm{Cov}\left[N_i,N_j\right] = \Omega\frac{d}{dt}\mathrm{Cov}\left[Z_i,Z_j\right] = \sum\left\{\tilde{A}_{i,k}\,\mathrm{Cov}\left[N_k,N_j\right] + \tilde{A}_{j,k}\,\mathrm{Cov}\left[N_k,N_i\right]\right\} + \Omega\tilde{B}_{i,j} \tag{23}$$

Note that Eq. (19) is identical to the linear equation resulting from linear stability analysis of Eq. (22). Consequently, the real parts of the eigenvalues of the coefficient matrix, $\widetilde{A}_{i,j}$, will be negative if the macroscopic behavior of the system is stable with ~ respect to fluctuations. Since $<Z_i(0)>$ is equal to zero, $<Z_i(t)>$ will be zero as long as the system is macroscopically stable. When this is not the case, the System Size Expansion is no longer valid and must be replaced by an alternate technique. Macroscopically, such behavior may correspond to a bifurcation point where two or more solutions branch from the original stable state. Probabilistically, the density function for the system would then no longer be unimodal; it is this property which invalidates the System Size Expansion at such points.

DERIVATION OF CORRELATION FUNCTIONS

The foregoing derivations of the expressions for the means and covariances of the random variables have yielded little information about the dynamic characteristics of the fluctuations. The auto- and cross-correlation functions, however, can provide this information. These functions yield measures of the influence of the value of a random variable at time t on the values of the random variables at time $t+\tau$. Two processes with equal means and variances but different auto-correlation functions can behave differently. For a Markov process, the auto- and cross-correlation functions can be easily derived [1]; the governing equations for them are the same as that for $<Z_i>$, Eq. (19). Defining the correlation matrix as

$$K_{i,j}(\tau) = \langle Z_i(0)Z_j(\tau)\rangle \tag{24}$$

the following set of differential equations can be derived by relating K(t) to $\mathrm{Cov}[Z_i,Z_j]$;

$$\frac{d}{d\tau}K_{i,j}(\tau) = \sum_k \tilde{A}^{s}_{j,k}\,K_{i,k}(\tau); \qquad K_{i,j}(0) = \mathrm{Cov}\left[Z_i,Z_j\right]^s \tag{25}$$

where

$$\tilde{A}^{s}_{j,k} = \tilde{A}_{j,k}\left(\left\{\phi^s\right\}\right)$$

$$\left\{\phi^s\right\} = \text{steady-state values of } \left\{\phi(t)\right\}$$

$$\mathrm{Cov}\left[Z_i,Z_j\right]^s = \text{steady-state covariance of } Z_i \text{ and } Z_j$$

Equation (25) is a direct result of the linear nature of Eq. (19) and of the fact that the process is Markovian. It also follows from Eqs. (24) and (25) and the relationship between the random variables {Z} and the original random variables {N} that the correlation functions for the random variables {N} can be found by solving Eq. (25) subject to the initial conditions

$$K_{i,j}(0) = \mathrm{Cov}\left[N_i,N_j\right]^s$$

where

$$\mathrm{Cov}\left[N_i,N_j\right]^s = \text{steady-state covariance of } N_i \text{ and } N_j$$

In Part 3, the final part of this series, the master equation and the System Size Expansion are applied to modeling of a chemically-reacting system.

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NOTATION

A_i	first jump moment
$\tilde{A}_i$	A_i / Ω
$\tilde{A}_{i,j}$	coefficient in expansion of $\tilde{A}_i$
$B_{i,j}$	second jump moment
$\tilde{B}_{i,j}$	$B_{i,j} / \Omega$
$Cov[N_i, N_j]$	$\langle N_i N_j \rangle - \langle N_i \rangle \langle N_j \rangle$, covariance of N_i and N_j
$Cov[Z_i, Z_j]$	$\langle Z_i Z_j \rangle - \langle Z_i \rangle \langle Z_j \rangle$, covariance of Z_i and Z_j
$K_{i,j}(\tau)$	correlation matrix defined as $\langle Z_i(0) Z_j(\tau) \rangle$ for Z_i and Z_j, or as $\langle N_i(0) N_j(\tau) \rangle - \langle N_i(0) \rangle \langle N_j(\tau) \rangle$ for N_i and N_j
N_j	number of entities possessing feature j
$N_{i,j}$	number of entities possessing feature i and feature j
$\langle N_i \rangle$	expected value of random variable N_i
$p(\{n\},t)$	joint density function of continuous random variables $\{N\}$
$P(\{n\},t)$	joint probability of random variables $\{N\}$
$P(\{n\}_1, t \| \{n\}_0, t_0)$	conditional probability of random variables $\{N\}_1$ at time t given the value of random variables $\{N\}_0$ at time t_0
$W_t(\{n\}_0, \{n\}_1)$	rate of transition from state $\{n\}_0$ to state $\{n\}_1$
Z_i	fluctuating component of random variable N_i
$\langle Z_i \rangle$	expected value of random variable Z_i
$\langle Z_i Z_j \rangle$	expected value of product of random variables Z_i and Z_j

Greek Letters

$\delta^k(x)$	Kronecker delta where $\delta^k(0) = 1$ and $\delta^k(x) = 0$ for $x \neq 0$
ξ_i	magnitude of change in random variable N_i
τ	small time interval tending toward zero
ϕ_i	deterministic variable corresponding to macroscopic behavior of N_i
$\Psi_t\left(\left\{\frac{n}{\Omega}\right\};\{\xi\}\right)$	homogeneous intensity of transition function
Ω	system volume

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RANDOM THOUGHTS

Continued from page 71.

straight A's for the rest of eternity; 2) pocket their tuition; and 3) don't give them the beer.

CORPORATE EXECUTIVE MODEL

Demand a high six-figure salary when offered the position of chancellor. When you get it, use the interest on your university's $200 million endowment to buy your way into financial control of a small but productive college in another state. Fire all their deans and department heads and put your own people in those positions. Move their best professors to your university, fire the others who don't have tenure, take any of their laboratory equipment you can use and sell the rest. Then fold the college and use the losses to offset the profits from the equipment sale, leaving yourself with a net annual corporate tax liability of $3.27. Keep doing this. When you've ruined enough small productive colleges to get your salary up to seven figures, announce that it is in the university's best interests to teach all classes in Japanese. Sell controlling interest in the university to the Kyoto Institute of Technology, participate in the dedication of the sushi bar where the Burger & Brew used to be, and retire just in time to miss the cafeteria riot and the disgusting things those ungrateful student hooligans do with all that raw fish.

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And that's all there is to it. With these few simple techniques we can easily transform our images and start to enjoy the good life.

On the other hand, there may be something to say for the status quo. As things stand now, most of us do our jobs without exploiting anyone's vulnerability or innocence, enriching ourselves at their expense, or trampling on their dignity. We may have to forego the Swiss bank accounts this way, but it still seems like a good bargain. We just have to be sure that our success is measured by the quality of our teaching and research and by nothing else...but then we're educators and scholars by profession, so there's no problem.

And now if you'll excuse me, I've got to get my notes together for the meeting at 10:00 where we review Greg Furze and Roger Snavely for promotion and tenure. Furze gets great teaching reviews and he's written a couple of research papers that people think very highly of, but there's not much by way of grants. Snavely is another story. He brings in a mint in funding, but his teaching evaluations are grim and his graduate students complain that they hardly ever see him, even though he keeps them here for as long as seven years. Should be an interesting meeting. ❐