To the Editor:

In "Direct and Split Operator Approaches with ELLAM for Reactive Transport Equations," (August 2007, pp. 2161–2169, DOI 10.1002/aic.11234), Younes and Fahs[4] consider the simulation of reactive transport by a standard split operator approach. This topic is important because of the widespread application of reactive transport problems. These problems are often simulated using split operator methods. We will show, however, that the key conclusion of this article is not true in general and applies only to a limited range of test problems.

Standard split operator algorithms decouple reaction and transport by integrating each process separately. Reaction and transport processes are approximately coupled by using the result from the first integration (e.g., transport) as the initial condition for the second integration (e.g., reaction). This introduces a splitting error that is discussed by Younes and Fahs,[4] and others.[1] A general reactive transport problem with m species is written as

$$\frac{\partial u_i}{\partial t} = \underbrace{L_i(u_i)}_{\substack{\text{Arbitrary n}-\text{dimensional}\\ \text{linear transport operator}}} + \underbrace{F_i(u_1, u_2 \ldots u_m)}_{\substack{\text{Arbitrary time dependent (kinetic)}\\ \text{reaction operator}}} \quad \forall i = 1, 2 \ldots m, \qquad (1)$$

where u_i is the concentration of the i^{th} species. Recently, we have shown that for transport then reaction splitting, the splitting error for small Δt is given by

$$E_i = \underbrace{\frac{\Delta t}{2}\left[\sum_{j=1}^{m}\frac{\partial F_i}{\partial u_j}\mathbf{b}_j\right]}_{\textit{Boundary Error}} + \underbrace{\frac{\Delta t}{2}\left[\sum_{j=1}^{m}\frac{\partial F_i}{\partial u_j}\mathbf{A}_j\mathbf{u}_j - \mathbf{A}_i F_i\right]}_{\textit{Internal Error}} + O\left(\left[\Delta t\right]^2\right) \quad \forall i = 1, 2 \ldots m. \qquad (2)$$

To arrive at Eq. 2, a general form of the spatially discretized transport operator is considered[2] ($L_i(\mathbf{u}_i) = \mathbf{A}_i\mathbf{u}_i + \mathbf{b}_i$). For single species reactive transport ($m = 1$) subject to one-dimensional advection-diffusion ($L_1 = D\partial^2/\partial x^2 - v\partial/\partial x$) the splitting error simplifies to

$$E_1 = \frac{\Delta t}{2}\left[\underbrace{\frac{dF_1}{du}\mathbf{b}_1}_{\textit{Boundary Error}} \quad \underbrace{-\frac{d^2F_1}{du^2}D\left(\frac{\partial u}{\partial x}\right)^2}_{\textit{Internal Error}}\right] + O\left(\left[\Delta t\right]^2\right). \qquad (3)$$

Equation 3 shows: (1) the internal error is independent of advection, (2) the internal error is proportional to the diffusivity, (3) both the internal and boundary error are proportional to the timescale of reaction, and (4) the net splitting error is proportional to Δt. The internal error is also independent of advection for multiple species reactive transport when all species are subject to the same rate of advection.[2–3] Equation 2 shows that a simple way to reduce the splitting error is to keep Δt small since $E_i \to 0$ as $\Delta t \to 0$. Other conditions also lead to small splitting errors, such as considering problems with small diffusivities or small reaction rates. Since the internal splitting error is independent of advection, a standard splitting algorithm will perform very well when advection dominates diffusion and reactions are slow. This is true provided that suitable numerical methods are used to approximate the reaction and transport operators.

This analysis highlights a serious flaw in Younes and Fahs conclusions. Younes and Fahs[4] focus exclusively on advection dominant problems, where the timescale of advection is large compared to the timescale of diffusion. Results in Figures 2–6[4] involve very small diffusivities with $D/(v\Delta x) \ll 1$, meaning that the terms governing the internal error in Eq. 2 are small. The reaction rates in all the kinetic problems are also small. For example in Figures 2–3[4] $K\Delta t \ll 1$ and in Figures 5–6[4] $\alpha\Delta t \ll 1$ meaning that all terms in Eq. 2 are negligible. All results for time-dependent reactions (Figures 2–6[4]) involve parameters that give small splitting errors. This explains the conclusion, "numerical experiments have shown SNIA_ELLAM to be highly accurate and efficient..." (pp 2167).[4]

These conclusions are not true in general. Eq. 2 (and supporting simulations[2–3]) show that split operator algorithms can be inaccurate for diffusion dominant conditions or where reaction rates are sufficiently large. These difficulties arise because standard split operator algorithms are first-order accurate in time, which can make them inefficient compared to any higher order method. Since Eq. 2 is independent of the spatial approximation of the transport operator, the algorithm proposed by Younes and Fahs[4] suffers from the same limitations. In general, these limitations cannot be overcome by (1) using specialized approximations for advection dominant conditions, (2) using a large number of subtime steps, or (3) by artificially adjusting to reduce the boundary error near inlet boundaries.[3] It is more appropriate to conclude that the numerical experiments in Younes and Fahs[4] show that the SNIA_ELLAM algorithm can be accurate and efficient for a limited range of problems only, and that the algorithm suffers from the same limitations that affect all standard splitting algorithms. Had Younes and Fahs chosen different parameters (i.e., larger D, K or α), these limitations would have been obvious and the conclusions different. Further details regarding methods to overcome the splitting error are given elsewhere.[3]

Literature Cited

1. Barry DA, Miller CT, Culligan-Hensley PJ. Temporal discretisation errors in non-iterative split-operator approaches to solving chemical reaction/groundwater transport models. *J Contam Hydrol*. 1996;22:1–17.
2. Simpson MJ, Landman KA. Theoretical analysis and physical interpretation of temporal truncation errors in operator split algorithms. *Math Comput Simulat*. 2007a. doi: 10.1016/j.matcom.2007.01.001. Available online.
3. Simpson MJ, Landman KA. Analysis of split operator methods applied to reactive transport with Monod kinetics. *Adv Water Resour*. 2007b;30:2026–2033
4. Younes A, Fahs M. Direct and split operator approaches with ELLAM for reactive transport equations. *AIChE J*. 2007;53:2161–2169.

Matthew J Simpson
Dept. of Mathematics and Statistics,
University of Melbourne,
Victoria 3010 Australia
E-mail: M.Simpson@ms.unimelb.edu.au

AIChE Journal, Vol. 54, 588, (2008)

DOI 10.1002/aic.11383
Published online December 17, 2007 in Wiley InterScience (www.interscience.wiley.com).