MOLECULAR DYNAMICS CALCULATIONS OF FEEDBACK PROCESSES IN AN OPEN SYSTEM (CSTR)

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Molecular dynamics (MD) calculations were done on the following feed back (autocatalytic) processes occuring in a continuous flow stirred tank reactor (CSTR):

$$A + B \rightarrow C; \qquad B + C \rightarrow D + B; \qquad D \rightarrow 2B$$

where A and B are continuously supplied by inflow and all species flow out of the reactor. We have recently given the conditions under which a second order kinetic phase transition is(particularly)observed in the CSTR using deterministic kinetics[1]. This kinetic phase transition occurs at a point of marginal stability and manifests itself in the phenomenon of "critical slowing down". Its possible biological significance may be that of a very efficient kinetic buffer and a low pass filter for periodic perturbations imposed on the open system. In this work we report MD calculations of the above system consisting of ~500 molecules. All molecules are considered to be hard spheres. The state of the system is computed exactly by the solution of the equations of motion[2] after a number of collisions (about 10,000) have occured. Periodic boundary conditions are imposed and elastic and reactive collisions - as specified by the above mechanism - are possible. The CSTR conditions are simulated by a spatially random addition and removal of labelled spheres. Stationary state concentrations from the MD simulations are different (< 10%) from the normalized stationary concentrations of the deterministic calculations. Fluctuations in the number of particles are enhanced in the neighborhood of the critical flow rate where the kinetics of the stochastic system shows a drastic slowing down effect in analogy to the phenomenon of critical slowing down in the deterministic CSTR calculations. We show with the aid of Fourier analysis that the fluctuation spectra is shifted towards smaller frequencies. Fluctuations are only weakly damped and relax slowly. Deviations from the mean of the distribution function of particles are observed: the Poisson distribution broadens and separates into two to three maxima. We believe MD to be a potent tool in the simulation of chemical reactions occuring on surfaces or in compartments and involving small numbers of molecules (a few to several thousand). Thus kinetic processes in biological cells may be simulated.

1. M.Heinrichs and F.W.Schneider, J.Phys.Chem., July 1981
2. P.Ortoleva and S.Yip, J.Chem.Phys., 65, 2045 (1976)