

vibrations occurring in a molecular system, are chosen. From molecular dynamics simulations, coordinates from equally spaced time steps along the trajectory are selected. These two approaches provide views of the changes that occur in electrostatic recognition as the system undergoes a number of concerted motions.

These computer graphics studies will be used to develop tools that provide insight into molecular recognition and interaction at a sufficiently detailed level to guide future site-directed mutagenesis experiments, which, in turn, will provide verification for these theoretical methods.

- 1 Sharp, K., Fine, R., and Honig, B. Computer simulations of the diffusion of a substrate to an active site of an enzyme. *Science* 1987, **236**, 1460
- 2 Allison, S. A., and McCammon, J. A. Dynamics of substrate binding to copper zinc superoxide dismutase. *J. Phys. Chem.* 1985, **89**, 1072
- 3 Getzoff, E. D., Tainer, J. A., Weiner, P. K., Kollman, P. A., Richardson, J. S. and Richardson, D. C. Electrostatic recognition between superoxide and copper, zinc superoxide dismutase. *Nature* 1983, **306**, 287

Molecular Modeling of Silicon Compounds

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Zeolites, both naturally occurring and synthetic, have found widespread application owing to their capacity for selective absorption and catalysis.¹ In recent years considerable effort has been directed toward the elucidation of zeolite structures and the mechanisms of their catalytic processes. This has included extensive diffraction studies, together with the recent application of high-resolution magic angle spinning NMR techniques. An alternative approach is to apply computational methods. To date these have mainly been carried out on small Si, O and Al clusters using *ab initio* or semi-empirical methods, and have been restricted as to the number of atoms under consideration.

A molecular mechanics model has been developed that can handle high silica zeolites. The first stage in this development was the extension of an existing empirical scheme for partial charge calculation to include silicon-containing molecules. During the parameterization it was shown that it is necessary to include both σ and π interactions between silicon and electronegative atoms.² Force constants to describe the vibrations of the Si-O fragments have been calculated from an extensive theoretical study, at various levels, carried out on a number of model compounds, including disiloxane, dimethoxy-dimethylsilane and silanol; the conformation of the cyclic molecule cyclotrisiloxane has also been investigated. Our calculations show the importance of including polarization functions in order to reproduce

unusual structural features of Si-O compounds.³ Finally, our molecular mechanics model has been tested against experimental geometries for silicon-oxygen containing molecules, and geometries and experimental energy differences for a number of alkyl silanes.⁴ In each case good agreement was obtained, validating our modifications to the MM2 force field, the application of our charges to molecular mechanics calculations and the ability of the calculated force constants to reproduce experimental features of silicon compounds.

To conclude, a discussion of applications of this model to zeolite structure will be presented. This includes optimization of substantial zeolite fragments with full relaxation.

- 1 Meier, W. M. *Pure Appl. Chem.* 1986, **58**, 1323
- 2 Abraham, R. J., and Grant, G. H. *J. Comp. Chem.* (in press)
- 3 Abraham, R. J., and Grant, G. H. (manuscript in preparation)
- 4 Abraham, R. J., and Grant, G. H. *J. Comp. Chem.* (submitted)

Spatial Density Distributions for Illustrating the Base Sequence Dependent Features of Double Helical DNA: Computer Graphic Visualization of Monte Carlo Chain Simulations

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Computer graphics is proving to be an essential tool in understanding the conformations and properties of biopolymers. The bulk of molecular applications to date have been static representations of crystallographically observed or energy minimized three-dimensional (3D) structures. A growing number of procedures, however, are being developed to study chain dynamics. Particularly popular is the animation of Newtonian molecular dynamics simulations, providing a visual description of the small-scale fluctuations of specific low-energy structures. Detailed study of large-scale molecular motions by such methods, however, is not yet computationally practical. Large-scale flexibility is generally described in terms of simplified artificial models or through computational short cuts that force the system over preselected energy barriers. Little, if any, attention has been given to visualization of the large-scale changes of macromolecular structure (involving the crossing of multiple torsional barriers) expected on the basis of local conformational flexibility.

Large-scale macromolecular motions are best deduced from direct Monte Carlo simulations. Such methods are particularly appropriate to studying polymers with properties determined by the chemical constitution of individual side groups. Specific chain conformations are generated from randomly chosen combinations of backbone structural parameters. Sequence-dependent features of structure are included in terms of the statistical