

Cellular Automata and their Applications to Molecular Fluids

Organised by the Statistical and Thermodynamics Group of the Royal Society of Chemistry, held in Manchester, U.K. on 19 - 20 July, 1994.

In recent years there has been growing interest in the application of cellular automata and lattice gas models to simulate complex systems. The basic operation of these methods is to move particles from one lattice node to another according to well-defined collision and propagation rules. By choosing these rules appropriately, a wide variety of phenomena can be modelled. Furthermore the methodology lends itself ideally to efficient parallelization on a computer, and thus complex systems may be studied which could not be investigated using more conventional techniques.

Approximately 50 people attended the conference, coming from this country and abroad and coming from both industry and universities. As intended, this was not a conference simply for the active practitioners of lattice gas simulations - throughout links were made with theoretical, experimental and conventional simulation work done on the systems of interest and the choice of invited speakers reflected this.

The talks covered a very wide range of topics and the abstracts, given below, illustrate quite how versatile cellular automata can be!

Finally, we would like to thank the sponsors of this conference (Schlumberger, Unilever, Shell and Courtaulds), and also CCP5 for their role in enabling Professor Tony Ladd to visit this country.

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Numerical Simulations of Particle Suspensions

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Recent research has led to the development of a promising new technique for simulating hydrodynamically interacting particles [1-3]. The method combines Newtonian dynamics of the solid particles with a discretized Boltzmann model for the fluid; its most important feature is that the computational cost scales linearly with the number of particles. Suspensions containing up to 1000 spheres can be simulated on a desktop workstation; many more particles can be tracked on a massively parallel computer.

In this talk I will outline the basic ideas of the method and discuss two current applications; sedimentation of non-Brownian particles, and the development of hydrodynamic interactions in colloidal suspensions. Comparisons with experiment will also be discussed.

- [1] A.J.C. Ladd, Phys. Rev. Lett., 70, 1339 (1993)
- [2] A.J.C. Ladd, J. Fluid Mech., 271, 285 (1994)
- [3] A.J.C. Ladd, J. Fluid Mech., 271, 311 (1994)

Numerical simulations of Colloidal suspensions via the Lattice Boltzmann method

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In this talk, several methods of treating the solid-fluid boundaries in Lattice Boltzmann simulations of solid particle suspensions will be discussed. A novel and very simple method of adding fluctuations to the fluid will be presented as well as results from simulations of long-time diffusion in concentrated suspensions.

Dynamics of Colloidal Suspensions

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At high concentrations, the dynamics of suspended colloidal particles are complicated by hydrodynamic couplings transmitted through the liquid suspension medium. Formally the hydrodynamic problem can be stated simply. In practical situations, due to the long range and intrinsic many-body nature of hydrodynamic interactions, its analytic solution has proved largely intractable. For these reasons, simulations, including both lattice gas and lattice Boltzmann modelling of the suspension medium, are proving valuable.

The talk will attempt a largely qualitative summary of the basic physics of colloidal suspensions. The forces (Brownian, direct and hydrodynamic) acting on the particles will be identified, and the relevant time scales discussed. Both quiescent suspensions, in which the particles undergo interactive Brownian motions, and suspensions out of equilibrium, due to the application of external forces, will be considered.

Computer Simulation of Complex Fluid Flows

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Research on complex fluids has great significance for a large number of industrial applications as well as for newly emerging technologies. A novel computer simulation technique, the Lagrangian/Eulerian method, for modelling complex fluid flows will be presented. The essential feature of our method is to explicitly track the motion of fluid elements in a Lagrangian fashion and to solve for the fluid fields on a co-moving grid in an Eulerian manner at each physical time step. The method is able to track the details of the fluid motion, e.g. deformation and stream lines, and it also can advance free surfaces in a physical way. The method has been applied to model continuum viscoelastic fluids, concentrated suspensions, foams and concentrated emulsions. The results of extrusion flows as well as simple shear and Poiseuille flows will be reported.

A Lattice-Boltzmann study of Dispersion in Porous Media

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The value of the dispersion coefficient for a tracer particle present in a fluid flowing through a porous medium can differ substantially from its value in the static fluid. Essentially the fact that the diffusing particle experiences variations in the fluid velocity during its travels causes additional “hydrodynamic” dispersion. For this effect to be significant the ratio of the typical time taken

by the particle to diffuse over some characteristic length scale must be large compared to the typical time taken to convect an equivalent distance. Otherwise the motion of the tracer becomes uncorrelated before it has time to sense the non-uniformity of the velocity field. The ratio of the two timescales defines the Peclet number. Here we describe a numerical study of this phenomenon.

The lattice-Boltzmann approach is well suited to the simulation of flows in complex geometries. There also exists a computational trick which allows one to calculate the velocity autocorrelation function (VACF) for tagged particle motion in the system, averaged over all the trajectories that a particle could possibly follow. The dispersion coefficient is then the integral over all time of this correlation function. By employing this approach we have been able to calculate dispersion coefficients as a function of Peclet number for flow through various three-dimensional structures. We describe results obtained for a periodic array of spheres, for which we can compare with the experimental results of Gunn and Pryce, and for random packings of spheres. The latter serves as a first approximation to a simple sandstone. By studying the timescale for the decay of the VACF we are able to explain the Peclet number dependence of the dispersion coefficient in the two systems.

Polymer Melts as Complex Fluids - current theoretical approaches

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Polymer melts are not immediately candidates for molecular simulation as the range of timescales between molecular motion and viscoelastic relaxation is prohibitively wide. Instead a coarse-grained molecular model has proved to be the most fruitful approach, based on the topological interaction of the molecular segments. This “tube model” has been applied now to a wide range of molecular architectures - recent results on comb polymers will be presented. The challenge is the application to non-linear flows, which demands an interface with the type of approach presented at this meeting by X-F Yuan. A set of calculational rules adaptable to numerical processing, and based on the molecular dynamics of branched polymers, is proposed.

Molecular Level Simulations of Polymers

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There is a growing current interest in the atomistic dynamical modelling of dense amorphous

polymers in order to provide understanding of bulk properties such as stress/strain behaviour, the glass transformation and penetrant diffusion in terms of chain structure, chain motions and interchain forces. One of the major advantages of computer simulation is the control that one has over the parameters defining the problem and the external conditions that can be imposed. One of the major challenges is to relate the behaviour observed in computer simulations, which is obtained on small samples on ultrashort time scales, to behaviour of macroscopic samples obtained in the laboratory on time scales ten orders of magnitude longer.

In this talk we shall review what has been achieved to date and what is feasible for the near future using molecular dynamics and Monte Carlo simulations, with either coarse grain and “atomistic” force fields. In addition we shall discuss recent progress [1,2] in the use of parallel processing computers to extend the system size, simulation times and precision of data.

- [1] D.Brown, J.H.R.Clarke, M.Okuda and T.Yamazaki, J.Chem.Phys., 1994, 100, 1684
- [2] D.Brown, J.H.R.Clarke, M.Okuda and T.Yamazake, J.Chem.Phys., 1994, 100, 6011

Long Range Correlations and Non-Gibbsian States in LGA's

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Non-Gibbsian stationary states occur in dissipative non-equilibrium systems. They are closely connected with the lack of detailed balance, and the absence of a fluctuation-dissipation theorem. These states exhibit spatial and temporal correlations that are long ranged under generic conditions, even in systems with short range interactions, provided the system has slow modes, and there is some anisotropy either in configuration space or in state space. The anisotropy may come from imposed fields (driven diffusive systems, temperature gradients). In the statistical mechanics of dissipative systems, such as stochastic cellular automata, the asymmetry is only in state space. Here the equilibrium states are non-Gibbsian, they may be spatially uniform with long range pair correlations, $g(r) \sim r^{-d}$ (with dimensionality d), and may even exhibit instabilities, and lead to the formation of clusters or patterns.

Topological Defects, Porod Tails and Growth Laws for Phase Ordering Kinetics

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The concept of topological defects provides a unifying framework for the study of phase ordering kinetics. For n -component vector fields, we show that the structure factor exhibits the power-law tail, $S(k) \sim A\rho/k^{d+n}$, provided $n \leq d$, where d is the spatial dimension, ρ is the density of defect core (e.g. the area of domain wall per unit volume for $n = 1$) and A is a calculable constant. Combined with the dynamic scaling hypothesis, this result can be used in a calculation of the energy dissipation rate to derive growth laws for general n and d , for both conserved and non-conserved dissipative dynamics.

**Correlations and Renormalization in Lattice Gas Automata:
Applications to a Model of a Chemically Reacting System**

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A method is described for calculating corrections to the usual Chapman-Enskog analysis of lattice gases due to the buildup of correlations. It is shown that these corrections can be expressed as a diagrammatic series, and closed-form expressions are given for the vertices in these diagrams. Subsets of these diagrams can be identified that correspond to the kinetic ring approximation or to truncations of the BBGKY hierarchy, and the relationship of this method to the more familiar Green-Kubo method is discussed. The method is then applied to a lattice gas for the Schloegl model chemical reaction. Corrections to the equilibrium densities and diffusivities are calculated and compared with numerical experiments. The impact of the correlations on the stoichiometry is elucidated.

**Materials Science Applications: Cellular Automaton Simulations
of Cement Hydration and Microstructure Development**

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Cellular automaton algorithms, which operate on a starting digital image of a water - cement suspension, are described. The algorithms simulate the microstructure development process due to hydration reactions that occur between cement and water, converting the viscous water - cement suspension into a rigid porous solid. The processes modelled include dissolution, diffusion, reaction and aggregation.

Methods are presented for generating two and three dimensional images representing suspension

initial conditions; these are derived both from micrographs of real cements and computer based algorithms. The 2D initial images are based on the processing of backscattered electron and X-ray images of real cement suspensions. The 3D images employ either spheres to represent cement particles, or, more realistic randomly shaped particles via an algorithm which smooths and thresholds a 3D lattice whose sites are initially populated with random white noise.

A convenient measure of the point at which the initial paste turns into a solid material is the percolation threshold of the solids. Consideration of these models led to the prediction and subsequent experimental observation of a sharply-defined onset of shear wave propagation, from ultrasonic measurements through hydrating cement slurries. The amount of hydration needed to reach the percolation threshold can be determined in the present simulation, and our results are compared with the time of shear wave onset in actual cement slurries.

Variants of the basic model provide insight into both early time behaviour that is of primary interest to oil-well cementing and the later time microstructural properties that are of interest in the construction industry.

It is hoped that this example will serve to highlight the potential range of applications of cellular automata within material science.

**Boundary conditions and interfaces
in lattice gases and lattice Boltzmann equations**

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Lattice gases have been introduced by Frisch, Hasslacher and Pomeau [1] as an efficient way to simulate fluid flows with a simplified molecular dynamics. These models consider point particles moving synchronously on a regular lattice and obeying conservation laws during their collisions on the nodes of the lattice. The field as matured in the lattice Boltzmann equation [2], in which the particles are replaced by their occupation numbers. This latter step is the fundamental ingredient for the theoretical derivation of the Navier-Stokes equations and, at the same time, it has been proven an effective tool for the direct simulation of flows at low to moderate Reynolds numbers (up to several thousands).

Although the theory of these models is now well understood in the context of periodic boundary condition, the implementation of solid walls and of external flows relies on empirical or semi-empirical recipes such as the well known “bounce-back condition”. An attempt to clarify this point will be presented in the context of a Couette flow for general orientations of the boundary with respect to the lattice. These results will be extended to Poiseuille flow oriented along one of the axis of symmetry of the lattice.

Some results will also be presented for systems with interfaces such as the two-color model introduced by Rothman et al. [3] or the liquid-gas model introduced by Appert and Zaleski [4].

- [1] U. Frisch, B. Hasslacher, and Y. Pomeau, “Lattice-gas automata for the Navier-Stokes equation”, *Phys. Rev. Lett.* **56**,1505–1508 (1986).
- [2] F. J. Higuera, and J. Jimenez. “Boltzmann Approach to Lattice Gas Simulations.” *Europhys. Lett.* **9**, 663–668 (1989).
- [3] A. K. Gustensen, D. H. Rothman, S. Zaleski, and G. Zanetti, “Lattice Boltzmann model of immiscible fluids”, *Phys. Rev. A* **43**, 107–114 (1991).
- [4] C. Appert and S. Zaleski, “Dynamical liquid-gas phase transition”, *J. de Phys. – France II* **3**, 309–337 (1993).

A Cellular Automaton Model of Microbial Growth.

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A stochastic, cellular automaton model of diffusion controlled, two compartment, population growth is introduced. The behaviour of the model system has many features in common with the growth of bacterial populations. Automaton variables can be interpreted in terms of macroscopic properties of structured growth media and measured growth parameters for particular microorganisms. The automaton illustrates different growth regimes and structural properties of colony growth.

Two dimensional Lattice Boltzmann simulation of immiscible fluid droplets in simple shear fields.

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In ([1]) Rothmann and Keller (RK) devised a simple rule which, when used to augment a Lattice Gas Cellular Automaton populated by particles of different species (colour) , segregates those particles and maintains an interface between them. Results from static simulations on Immiscible Lattice Gas Automata (ILG’s) demonstrated that the Laplace Law governing fluid-fluid interfaces was recovered.

Subsequent work at Sheffield Hallam University on the original algorithm of [1] implemented in parallel on considerably larger lattices revealed certain disadvantages the method. However these were largely overcome when RK’s idea was extended to Lattice Boltzmann simulations([2]) - results showed that interfaces with correct static properties might be produced and that these interfaces had (qualitatively at least) appropriate time-evolution behaviour.

This talk will review the various techniques used by workers over recent years to generate interfaces in LGAs, discuss their limitations, the parameters which influence their behaviour and the problems to be overcome in efficient implementation. It will present recent results from static simulations and from assessments of the dynamical, steady state properties of RK and RK-like interfaces in the context of fluid droplets under shear.

Possible modifications to the rule currently under development will be discussed.

- [1] Rothmann and Keller, J Stat Phys 52 (1988) pp1119
- [2] Gustensen, Rothmann, Zaleski, Zanetti, Phys Rev A 43 (1991) pp4320