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Preface

Many physical and chemical processes display chaotic behaviour. Furthermore, the morphology of porous solids such as adsorbents and catalysts can be described by the use of fractal concepts. The objective of this special issue of *The Chemical Engineering Journal* is to focus on the applications of theories of chaos and fractals in chemical engineering. A brief introduction to the various papers is discussed under several subheadings.

1. Use of fractal concepts in porous structures

Punčochář et al. analyse the modelling of fractal structures by means of partially scaled fractals and suggest a practical application of this technique to the modelling of gas–solid reactions. Adrover and Giona develop a predictive model for the permeability of porous matrices, starting from twodimensional images of thin sections. In this model a new length scale parameter, related to the decay of pore–pore correlation function is introduced. The model does not contain any adjustable parameter, but is limited to twodimensional structures to date. The extension of this approach to three-dimensional structures is considered to be feasible.

Transport and kinetics on fractal model structures are topics of major interest for their application to design and optimisation of catalytic performance. The article by Sahimi reviews the scaling theory and the effective medium approximation results for transport and vibration on disordered and fractal structures.

The analysis of reaction/diffusion schemes on fractal structures is developed in four articles, by applying different approaches.

Giona et al. develop a renormalization method to solve linear transport schemes on fractal lattices with arbitrary accuracy. The method is based on Green function renormalization on finitely ramified fractals. The main advantage of this approach is that the experimental observable quantities can be readily obtained by iterating a low = dimensional system of recursive relations in the pivotal Green functions. The inclusion of boundary conditions is treated by means of boundary condition transformations. This method is applied to sorption properties of fractals and to the determination of the effectiveness factor for firstorder reactions. The extension of this approach to non-linear schemes, to infinitely ramified fractals and to the influence of boundary conditions is also discussed.

First-order reactions on a fractal structure are investigated by Mougin et al. The authors consider the devil's comb, a two-dimensional space-filling loopless structure, as a model pore network. Coppens and Froment analyse the influence of surface fractal dimension and of surface fractal morphology in catalysis. By tuning the catalyst surface morphology, the selectivity of catalytic reforming for example, can be influenced. Borrelli et al. study the modelling of diffusion-limited gasification of carbon by means of a branching pore model.

Adsorption is probably one of the most consolidated fields of application of fractal concepts in heterogeneous chemistry. The work of Avnir, Farin and Pfeifer (1983, 1984) demonstrate that many surfaces of adsorbent materials can be characterised by means of a fractal dimension. The overwhelming majority of theoretical models proposed to predict the influence of fractality on adsorption equilibria are thus far restricted to the case of singlecomponent adsorption and are not suitable for mixtures.

In 1990 Keller proposed a class of adsorption isotherms, for mixtures of an arbitrary number of components satisfying the thermodynamic constraint induced by the Maxwell equations. The fractality of the adsorbent is considered in the Keller model, in the form of the Avnir–Pfeifer scaling for the monolayer coverage. The eventual energetic heterogeneity is described by means of the partial-pressure dependence of the adsorption energies. The Keller model proves to interpret and predict fairly accurately adsorption isotherms for microporous materials. The basic structure of the Keller isotherm consists of a generalised Langmuir–Freundlich equation. Rudziński et al. analyse this class of isotherms and show that the Langmuir–Freundlich model is related to the assumption of the lack of correlation amongst the adsorption energies of the various components.

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2. Rheology of suspensions

Lapasin et al. develop a rheological model to predict the viscosity of aggregated suspensions. A model which includes the fractal structures of the aggregates is proposed for the relative viscosity of dense suspensions. The model shows good agreement with experimental data from epoxy–acrylic systems and aluminium silicate for different concentration of the disperse phase.

3. Multifractal characterisation of flow

Zhong et al. study the multifractal characterisation of flow in circulating fluidized beds (CFBs). They show that CFBs exhibit multifractal properties and introduce a multifractal-based technique for characterising the intermittency of instantaneous voidage signals under different flow conditions. From the results the authors suggest that the cluster motion may be behave as a multiplicative process. Since multifractional behaviour can also be created by other processes, further study is required before the physical insight into turbulent systems can be applied.

4. Chaotic mixing

The kinematics of mixing plays an essential role in many forms of chemical production (e.g. pharmaceuticals). The study of mixing is important to prevent undesirable effects in reaction. The study of chaotic motion of tagged particles in periodic flows proves to be a useful route to understand the details of mixing. Such models, based either on simplified velocity patterns or on the solution of low-Reynolds number Navier–Stokes equation under periodic excitement (the cavity flow) show to be in good agreement with dye tracer experiments. Muzzio and Liu analyse chemical reactions in time-periodic flows. They consider simple bimolecular reactions and competitive–consecutive reactions by stressing the influence of chaotic mixing on reaction kinetics. Because for a bimolecular reaction the ability of both reactants to mix is a critical factor in determining the rate at which a product is formed, Sawyers et al. investigate the yield of two initially separated reactants of such a system in a tubular reactor of different coiling geometries at laminar, steady flow conditions with high mass Peclet number. They find that coiling and flow kinematics have a strong effect on the length of the interface, separating the two reactants. If the coiling axis is periodically changed in the flow direction, above a critical value of the switching length, an exponentially growing interface length can occur.

5. Chaotic dynamics

If the emphasis on the nonlinearity of our world is not focused on its (multi)fractality, but on its chaotic dynamical evolution in time, in principle two approaches are possible.

The first, which may be found in the literature of the last 10–20 years is based on the acceptance of the (often unexpected) chaotic dynamics; one tries to take this chaotic behaviour into account and to implement its consequences. Since the early nineties the Kolmogorov entropy and some version of the dimension (e.g. correlation, boxcounting) have been tried to characterise regimes and regime transitions in multiphase (reactor) systems. Some examples are shown in this issue. Only recently has one group tried to improve and to extend the existing nondimensional scaling rules to cover the restricted predictability of these systems as well; based on the Kolmogorov entropy a chaos based scale up methodology was proposed at the 1996 ISCRE Symposium (Schouten et al., *Chem. Eng. Sci., 51* (1996) 1991–2000), which takes care that the short term dynamics are similar in the scaled systems.

The second possible approach is that instead of accepting the chaotic behaviour of the system one can try to exploit it. The basic idea is that because a chaotic system is very sensitive to tiny changes in the conditions, relatively little energy is required to completely change the hydrodynamic regime of the system and to control it in the new form by applying a proper control algorithm. Ott, Grebogi and Yorke (OGY) have made a start to provide such an algorithm; however, so far no one has actually performed such a strategy in a chemical engineering setting; only suggestions are made to apply it, for instance to control any required hydrodynamic regime in multiphase flow systems (e.g. bubbleless fluidization to improve fluidized bed mass transfer). Research groups at Oak Ridge National Laboratory (TN, USA) and Delft University of Technology (The Netherlands) are known to work in this area.

Qammar et al. examine control of a chaotic polymerisation reaction employing conventional feedback control strategies.

6. Multiphase flow regime characterisation

Drahoš et al. use wall pressure fluctuations to characterise the intermittent flow patterns in a horizontal pipe. They investigated the transition from plug flow to slug flow (where a weak sign of deterministic chaos was observed) to annular flow (where the resulting transition patterns showed the intermittency route to chaos). Hay et al. use the crosssectional average correlation dimension to characterise the global bubble column hydrodynamics and the average bubble size in a gas–liquid contactor. Briens et al. study the liquid and gas maldistribution in bubble columns and in gas–liquid–solid fluidized beds. They use an approximate box counting dimension which can be calculated so rapidly that it can be used on line.

From the raw signal of a bubble probe the gas maldistribution can be calculated, from a local conductivity probe the liquid maldistribution can be derived.

Fuller, Flynn and Daw perform measurements in a 70 MW coalfired pressurized fluidized bed boiler, a cold simulated slice of this boiler and a 30 MW pulverized coal combustion facility. They show that multiple analytical tools, both linear (e.g. conventional statistics, power spectra, mutual information) and nonlinear (e.g. correlation dimension and Kolmogorov entropy), are needed to sufficiently describe and discriminate between the different boiler operation conditions; together they serve as a powerful tool for boiler diagnostics. Finally, Nguyen et al. show that regimes in multiphase reactors can also have a spatial component besides a temporal one, making the system more complicated. They argue that the dynamics of rising bubbles in a dense fluid can be best described as smallboxspatio-temporal chaos with a flow instability; they show that their experimental results are similar to the results of their earlier developed bubble-interaction model that takes drag forces and bubble coalescence into account.

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