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Development of a Genetic Algorithm for Molecular Scale Catalyst Design

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A genetic algorithm has been developed to determine the optimal design of a two-component catalyst for the diffusion-limited $A + B \rightarrow AB\uparrow$ reaction in which each species is adsorbed specifically on one of two types of sites. Optimisation of the distribution of catalytic sites on the surface is achieved by means of an evolutionary algorithm which repeatedly selects the more active surfaces from a population of possible solutions leading to a gradual improvement in the activity of the catalyst surface. A Monte Carlo simulation is used to determine the activity of each of the catalyst surfaces. It is found that for a reacting mixture composed of equal amounts of each component the optimal active site distribution is that of a checkerboard, this solution being approximately 25% more active than a random site distribution. Study of a range of reactant compositions has shown the optimal distribution of catalytically active sites to be dependent on the composition of the ratio of *A* to *B* in the reacting mixture. The potential for application of the optimisation method introduced here to other catalysts systems is discussed.

INTRODUCTION

There has been much recent interest in the concept of catalyst design at the molecular scale, utilising developments in surface science technology to manipulate the composition of the surface layer of catalytic materials in order to optimise activity or selectivity with respect to a particular reaction (1). Examples include the epitaxial deposition of ordered metallic monolayers on metallic or metallic oxide supports (2) and the application of microelectronic circuitry fabrication technologies to deposit metal particles of around 100 Å in regular arrays on silica and alumina supports (3, 4). Given, therefore, that the technology exists in principle to construct surfaces at the molecular level we must begin to develop methodologies for designing such surfaces for particular reactions.

In this paper, the optimisation of a surface containing two catalytically active sites is considered. Possible application of the method described might therefore lie in the optimisation of bimetallic or metal–metallic oxide composites, where the use of such two-component catalysts in preference to single-component materials has led to significant improvements in catalytic activity and product selectivity (5). In particular the use of supported Pt–Rh and noble metal/reducible oxide catalysts for CO oxidation at low temperatures has been the subject of considerable interest (6) due to the importance of this reaction in the automotive industry as a pollution control measure. Bimetallic catalysts have also been utilised in more complex reactions such as the cracking of hexane over a metallic catalyst composed of an alloyed overlayer of Au on Pt(111) and Pt(110) (2), a reaction established in the petrochemical processing industries. In this latter reaction the selectivity with respect to the various cracking products was found to be highly dependent upon the surface coverage of Au. This variation was partly attributed to the formation of active sites of differing coordination number to the pure platinum crystals, illustrating the potential for catalyst design at the molecular level.

Genetic algorithms (GAs) are general purpose optimisation methods which are finding increasing application in a wide range of disciplines, particularly in engineering and the physical sciences. Although they are based on the mechanisms of Darwinian evolution, and extensive use of biological terminology is made in their description, their use is by no means restricted to biological problems. GAs are particularly useful in situations where no analytic optimisation techniques are available. Recent examples of their application to optimisation problems include the determination of the minimum energy configuration of metallic clusters (7), pipeline design (8), and the design of nuclear magnetic resonance imaging coils (9). In this paper we consider the use of a GA to determine the optimal design of a two-component catalyst for the simple monomer–monomer reaction between two identical but distinguishable species *A* and *B*. In this hypothetical reaction each component is considered to adsorb preferentially on to one of two types of surface sites. Once adsorbed the reactants may undergo a Langmuir–Hinshelwood type reaction to form the reaction product, *AB*, which immediately desorbs. To maximise the activity of the catalytic surface, we must optimally determine both the number and the geometric configuration of

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the two types of sites. Since we have available no analytic optimisation technique appropriate for searching among all possible site configurations, we must employ a nonanalytic method, such as the GA.

The method adopted is outlined below to give an overview of the general principles underlying evolutionary optimisation. The method of combining the optimisation routine with the Monte Carlo simulation is then considered in detail followed by a discussion of the results obtained. In the final section we draw conclusions and consider the possibility for extension of the GA-based approach to more realistic catalytic systems.

METHOD

General Approach

GAs (10, 11) utilise a biological metaphor in an attempt to solve an optimisation problem by the propagation of some desirable characteristic of the solutions through the selective breeding of a set of possible solutions. Such an approach has proven to be a robust method of locating solutions to complex problems with many local maxima. In addressing the optimisation of a catalyst surface comprising two types of site a set of such surfaces is first generated at random to represent an initial set of estimates to the solution, each of these possible solutions representing a particular arrangement of the two site types on a planar surface. The activity of the catalyst surfaces is then determined by a Monte Carlo simulation of the reaction conducted on each of the surfaces. Each of the catalyst surfaces and its associated activity are then passed to the GA to determine the next set of solutions, and the process is repeated until a single best solution emerges. This procedure is illustrated schematically in Fig. 1. While the GA provides no guarantee of optimality, its ability to provide good solutions to a wide variety of optimisation problems has well-established theoretical foundations (11).

A genetic algorithm, in its simplest form, works by breeding successive sets of binary strings, which are encoded versions of possible solutions to an optimisation problem. The actual solution corresponding to a particular set member is known as its phenotype, while the binary string to which it corresponds is known as its genotype or chromosome. In the present case the phenotype is the representation of the two-dimensional catalyst surface, as shown in Fig. 2. The set of solutions being processed by the GA at any one time is known as the population. The population size remains constant throughout the optimisation process.

A number of operations analogous to biological reproduction may then act on this population to produce subsequent generations of solutions, the driving force for evolutionary improvement being the more frequent selection of the more successful individuals in the population for breeding into subsequent generations. The production of successive populations is achieved by using two fundamental genetic operators, crossover and mutation, shown in Fig. 3. The crossover operation can be considered the means by which desirable characteristics of the successful solutions are spread in the population by exchange of genetic information. Each of the individuals in the population is selected for breeding with a frequency related to its fitness, which in this case is related to the activity of the corresponding catalyst surface as determined by a Monte Carlo simulation. Two individuals are selected and a crossover attempted with a specified probability of success. If the mating is successful a section of each of the two parent chromosomes is selected and swapped between the parents to produce a pair

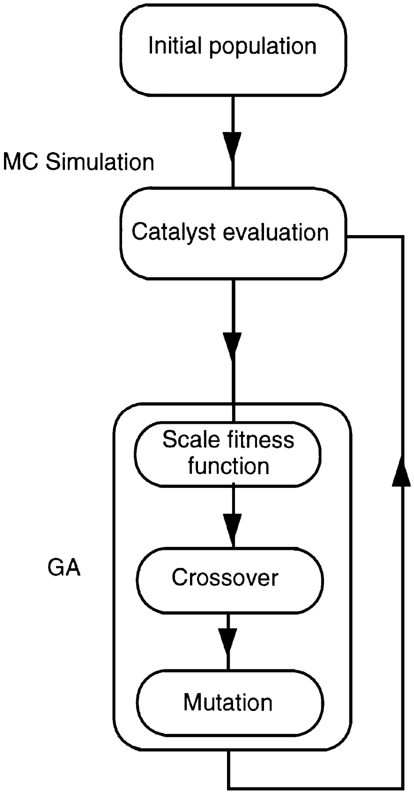

FIG. 1. Flow diagram illustrating the optimisation procedure. A Monte Carlo simulation provides the GA with a measure of the catalytic activity of the surface, r, known as the fitness function. This activity is then scaled to determine $F(r)$, the scaled fitness function, which is proportional to the probability of selecting the surface for breeding into the next generation.

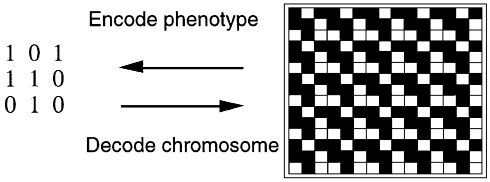

FIG. 2. A 2-D binary chromosome and a section of the corresponding phenotype representing a catalyst surface.

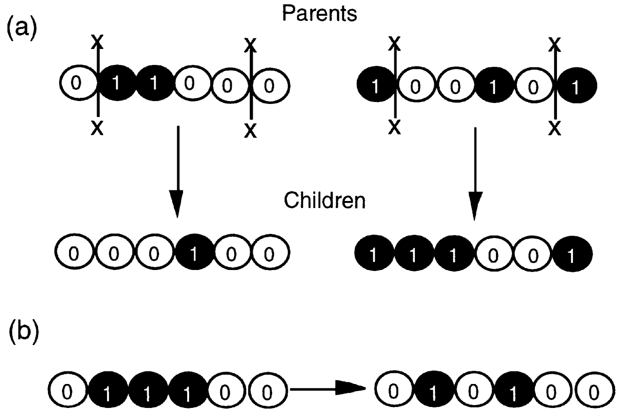

FIG. 3. The basic genetic operators in a genetic algorithm acting on a one-dimensional chromosome. In the crossover operation (a) all genes between either a single point and the end of the chromosome (one-point crossover) or between two points (two-point crossover) are swapped with the corresponding genes in the other parent chromosome; thus the next generation of chromosomes retains some of the characteristics of the previous generation. In mutation (b) an attempt is made to alter each individual gene with a specified probability of success. In a binary chromosome a successful mutation leads to the mutated gene assuming the alternative binary digit.

of chromosomes as children. The parents are then discarded and the offspring are carried forward into the next generation maintaining a constant population size. If the mating is unsuccessful the parents are carried over into the next generation in an unmodified form. To maintain diversity within the population and to introduce a random search mechanism an additional operation, mutation, is used. After the crossover operations have led to the next population, each gene of each individual may mutate with a particular probability (often around 0.1–3%) to assume the value of the alternative binary digit. The analogous process in biological evolution is the selection of genetic material (chromosomes) for recombination and reproduction, based upon the performance or “fitness” of the organism (phenotype) comprising this genetic material.

Optimisation of the Catalyst Surface

In summary there are two stages in the optimisation procedure; first there is the determination of the activity of a member of the population and, second, the use of this activity in guiding the evolution of the population towards an optimal solution by use of the GA.

Determination of Catalytic Activity by Monte Carlo Simulation

To test the fitness of a solution and hence optimise the catalyst surface using the GA, each member of the population of solutions must be characterised using objective criteria. In this case the appropriate criterion is the catalytic activity of that member of the population. In this work a Monte Carlo simulation (12, 13) has been used because it overcomes the limitations of deterministic models by making no prior assumptions regarding either surface homogeneity or the distribution of reactants on the surface. In contrast, a classical approach to modelling surface reactions uses simple algebraic equations to describe the rate of the surface reaction assuming a random distribution of each of the reacting species on a homogeneous surface. Such models assume a spatially averaged description of the adsorbed species. This assumption will be inaccurate for a reaction on a disordered surface as information regarding the geometric distribution of active sites cannot be incorporated into a single algebraic expression for the reaction rate. Furthermore, for diffusion-limited reactions of the type considered in this example it has been demonstrated that, even on a homogeneous surface, the particles are not randomly distributed and segregation of the reactants is observed as a consequence of fluctuations in the local particle density (14). Clearly, selection of a Monte Carlo algorithm to quantify the fitness of a population member in terms of its catalytic activity is appropriate here.

In this work we use the Monte Carlo simulation to calculate the catalytic activity of each surface with respect to the diffusion-limited $A + B \rightarrow AB\uparrow$ reaction (15). The simulation is performed on a regular lattice of 120×120 sites with periodic boundaries. Each site, labelled as S_1 or S_2, may adsorb specifically only one of the two species. The reaction is assumed to follow Langmuir–Hinshelwood type kinetics and can be considered to consist of three irreversible steps: two adsorption steps and a reaction step to produce the product which immediately desorbs. The rate constants for each stage are taken to be infinite; the initial adsorption step is therefore the rate-limiting process.

$$\begin{aligned} A + S_1 &\rightarrow AS_1 \\ B + S_2 &\rightarrow BS_2 \\ AS_1 + BS_2 &\rightarrow S_1 + S_2 + AB\uparrow. \end{aligned} \qquad [1]$$

The above mechanism closely resembles that for the oxidation of CO by two-component catalysts. In these catalysts the rate of CO oxidation is enhanced relative to a single component noble metal catalyst by the addition of a second component, typically either another metal or a metallic oxide (16). In the case of the pure noble metal catalyst the CO oxidation rate is inhibited by the saturation of the metal surface by CO at all but very low CO/O_2 ratios. The addition of a second component which specifically adsorbs only oxygen would, therefore, greatly increase the rate of CO oxidation. Examples of such systems include Rh/Cu and Sn/Pd bimetallic catalysts (17, 18). In these systems the Cu and Sn partially oxidise to form adsorption sites for oxygen only, the rest of the surface being almost fully covered with CO. These two types of adsorption site can be considered equivalent to the S_1 and S_2 sites in the simplified model considered in this paper. The design of such a bimetallic material was considered qualitatively by Herz *et al.* (19) for the related $A + (1/2)B_2 \rightarrow AB\uparrow$ reaction.

The Monte Carlo method employed here follows directly from that described in previous work on both homogeneous surfaces (15, 20) and heterogeneous surfaces (21). The reacting system is considered to consist of an infinite bulk phase composed of the reacting species which adsorb, at random, onto a catalytic surface represented as a regular array of adsorption sites. The reaction is simulated by a series of discrete events:

(i) A particle is selected from the gas phase for adsorption with a probability proportional to the abundance of its species in the gas phase.

(ii) A site on the lattice is selected at random and the particle adsorbs and occupies the site if the site is of the correct type and is vacant (i.e., reaction by the Eley–Rideal mechanism is not permitted).

(iii) Each of the neighbouring lattice locations is checked in a random order. If a neighbouring AB pair is formed then both desorb, vacating both adsorption sites.

Steps (i)–(iii) are repeated until a steady value is obtained for the rate of production of the AB pairs. This value is taken to represent the activity of the catalytic surface and is passed to the GA to determine the probability of selection of the surface for breeding into the next generation.

Application of a Genetic Algorithm to Catalyst Design

The optimisation procedure was implemented in C++ on a Silicon Graphics Indigo2 workstation using prewritten software to provide the basic genetic operators (22). Each surface was encoded as a two-dimensional binary array, where 0 and 1 represented sites S_1 and S_2, respectively. A 6×6 array was typically used, although arrays up to 8×8 sites were used to confirm the results obtained. The catalyst surface was constructed by tiling this base array over a larger grid of 120×120 sites to minimise errors resulting from the use of a finite lattice. For the simple reactions of the type considered in this work we would not expect the repeating unit to be larger in size than a few lattice spacings as the size of the reacting molecule is equivalent to only a single lattice location. The Monte Carlo simulation was then conducted by making a large number of repeated adsorption attempts and the activity estimated by counting the number of AB product molecules produced. As the simulation is a stochastic process the catalyst activity returned is a random variable, so the simulation on each surface was repeated a number of times to obtain a reliable measure of the surface activity. In the present case the simulation output was taken to be reliable if it could, with 95% confidence, correctly distinguish two surfaces differing in only one site on the basis of their activities. Typically around 10^6 Monte Carlo steps (MCS) and five repeated trials were required to obtain data accurate enough to differentiate between such a pair of similar surfaces.

Given that R is the total number of product molecules produced in a given simulation run the activity of the catalyst surface can be defined in terms of the turnover frequency, r, the number of reactions occurring per unit time on a lattice composed on N lattice sites. Time, t, was measured in terms of Monte Carlo steps, where 1 MCS is defined as N attempted adsorption events. The measure of catalytic activity can therefore be written as

$$r = \frac{R}{t \cdot N}. \qquad [2]$$

As the difference in activity between the optimal and average catalytic surfaces was typically only 20–30%, the probability of selecting the more successful surfaces in a given generation for breeding would not be much greater than that for a randomly selected surface. A scaled fitness function, $F(r)$, was defined in order to increase the bias towards the preferential selection of the more catalytically active surfaces. This function was defined as a simple power law of the form

$$F(r) = r^{\gamma}. \qquad [3]$$

Let r' denote the activity of the most active surface in the population and $\bar{r}$ the corresponding average activity of the population. The exponent, γ, was adaptively chosen so as to maintain the ratio $r'/\bar{r}$ at the constant value, n, giving

$$\gamma = \frac{\ln(n)}{\ln(r'/\bar{r})}. \qquad [4]$$

Typically n was taken to have a value of 3, leading to the best surface from each population being selected for breeding three times as often as a surface with the mean activity.

The parameters required by the GA are the population size, probability of a successful crossover occurring and the mutation rate. There are no established quantitative ways of obtaining an optimal set of parameters for a GA and the parameters used in this work were obtained by a process of trial and error. Typically the probability of a successful crossover was taken as 0.85 and the probability of mutating an individual gene as 0.02. There is also some freedom in the specification of the details of the crossover operation. Here it was found useful to take into account the geometry of the two-dimensional catalyst surface by implementing a two-point crossover, similar to that shown in Fig. 3 but acting directly on the two-dimensional arrays used to represent the surface rather than on the commonly used one-dimensional string. Without this modified crossover operation it proved to be impossible to get the algorithm to converge. The details of the crossover operation are given in the Appendix.

RESULTS AND DISCUSSION

Figures 4 and 5 illustrate the results obtained for the $A + B \rightarrow AB\uparrow$ reaction for the case where the reacting

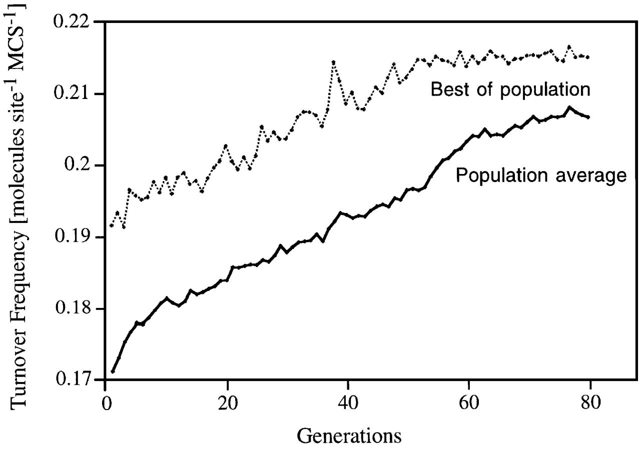

FIG. 4. Optimisation of a two-component catalyst for the $A + B \rightarrow AB\uparrow$ reaction with equal quantities of each reactant in the bulk phase. The turnover frequency, calculated by the Monte Carlo simulation, of the most catalytically active member of the population is compared with that calculated as the population average. The first 80 generations of evolution are shown.

mixture consists of equal quantities of A and B. These results show the algorithm locating the optimal solution after approximately 50 generations. A good indication that the algorithm has arrived at an optimal, or locally optimal, solution is the convergence of the population towards a single solution. This was indicated by the narrowing of the difference between the activity of the best surface of the population of each generation and the average activity of the whole population, as shown in Fig. 4. Figure 5 illustrates the evolution of the optimal solution for this problem, demonstrating the spreading of the desirable $\left[\begin{smallmatrix}0 & 1\\ 1 & 0\end{smallmatrix}\right]$ group forming the checkerboard pattern, where 0 and 1 each represent one of the two possible surface sites. These results agree with the heuristic arguments put forward regarding the design of a similar catalyst for the $A + (1/2)B_2 \rightarrow AB\uparrow$ reaction (19) in that the checkerboard pattern allows for the maximum number of active boundaries between differing adsorption sites. From Fig. 5 we also see that the density of S_1 sites in the best solutions immediately approaches and remains close to the value 0.5, which is indeed its value in the optimal solution. Closer inspection of the whole population shows that the majority of surfaces have approximately this density after only a few generations. We interpret this as showing that the GA manages to find the optimal site density well before it solves the more difficult problem of finding the optimal geometric configuration of sites. Evaluation of surfaces with a range of site densities confirms that the activity depends very strongly on this parameter. Thus the GA could help us identify significant design parameters even in situations where it is unable to identify an optimal solution.

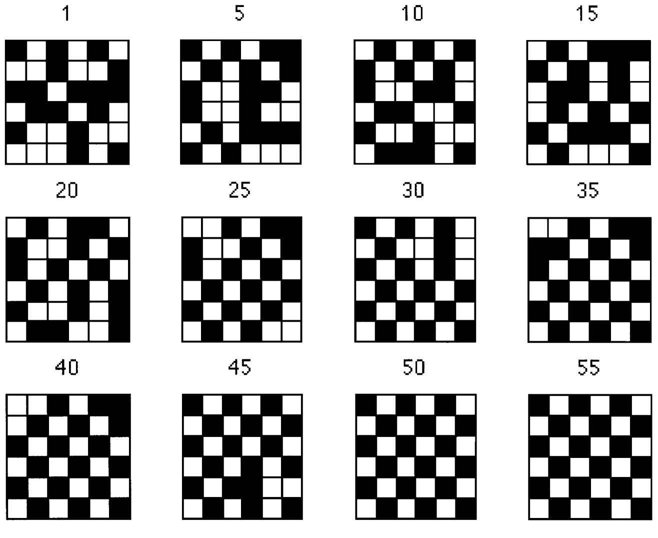

FIG. 5. Evolution of the solution to the $A + B \rightarrow AB\uparrow$ reaction with equal quantities of each reactant in the bulk phase as illustrated by the best solution from each generation. The generation from which each solution was selected is indicated above each grid.

Although it might have been considered possible to predict that the checkerboard pattern would have been the optimal site distribution for the case when the reactants are present in the bulk phase in equal quantities, it is not so clear what the optimal distribution should be for other bulk phase compositions. As a further test of our methodology we considered the design of catalyst surfaces for a range of bulk phase compositions. Altering the bulk phase composition such that one of the species becomes twice as likely as the other to attempt in adsorption even leads to noticeable changes in both the site densities and their configuration. The most active solution found in this case is shown in Fig. 6, with the repeating unit being

$$\begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

This result is significant as it demonstrates that altering the gas phase composition leads to a compensating effect in the site density. In this example doubling the adsorption probability halves the number of adsorption sites for that particular component, with the consequence that the surface coverages of the two species remain approximately equal.

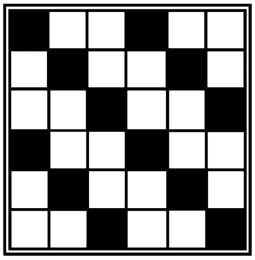

FIG. 6. Optimal solution with unequal quantities of each reactant; in this case the ratio of A to B is 2 : 1. Species A and B adsorb on the black and white sites, respectively.

Again these results show that the surface arranges itself such that the number of reactive sites at the interface between the two adsorption sites is maximised. This diagonal arrangement was maintained for the higher $A : B$ ratios of 3 : 1 and 4 : 1, there being no further tendency to reduce the number of adsorption sites for the species predominant in the bulk phase. This limiting behaviour is to be expected due to the formation of adsorption sites which only border sites of the same type if one of the surface sites becomes too predominant. These unreactive sites cannot lead to the formation of reactive pairs and so are avoided by the optimisation algorithm. The arrangement shown in Fig. 6 represents a compromise between the optimal number destiny of each type of site and the increasing tendency to form unreactive sites with several neighbouring sites of the same type.

CONCLUSIONS

We have introduced a rational methodology for the optimal design of simple catalyst surfaces at the molecular level using a genetic algorithm in combination with Monte Carlo simulations. This method has been applied to a simple $A + B \rightarrow AB\uparrow$ reaction occurring on a two-component catalyst. In the particular case where the bulk phase is a mixture of equal amounts of each component, the surface yielding the highest catalytic activity is found to be a checkerboard distribution of the two catalytically active sites. Deviations from this configuration are observed for cases in which the bulk phase is not composed of an equal mixture of the reactants. As the relative proportion of one of the species increases, the number of adsorption sites for that species tends to a ratio of 2 : 1 in favour of the site adsorbing the deficient component. Further increase in this ratio does not occur, as it would require the formation of sites on the catalyst surface which border only sites of the same type, and thus could not be involved in the formation of reactive pairs.

The purpose of the present paper is to introduce the use of the GA as a practical tool in catalyst design problems and to demonstrate that, in the case of a model system, the catalyst designs obtained are sensible. Thus we are now in a position to consider the extension of this approach to more complex systems. Two possible extensions are considered here. First, the incorporation into the design process of effects such as sintering, adsorbate-induced surface reconstruction, and poisoning. Second, the use of GAs to identify key catalyst design parameters even in situations where the determination of optimal catalyst structure may be unfeasible.

To generate catalyst designs which take account of the deactivation processes mentioned above, we need only replace the simple Monte Carlo model used here with one which incorporates the required physical phenomena. Processes such as sintering (23), surface reconstruction (24, 25), and catalyst poisoning (26) have been investigated extensively using Monte Carlo methods, so their incorporation would be a simple extension of the work presented here. Furthermore, the use of Monte Carlo simulation for the evaluation of fitness is not essential. A GA-based process could be used in conjunction with other evaluation techniques, for example, molecular dynamics simulations of analytical models. The use of Monte Carlo methods is very demanding on computing resources, so the use of more efficient evaluation techniques, where appropriate, would be desirable.

In the application of GA-based design to a highly complex system, such as a silica-supported metal catalyst, it might prove impossible to obtain a unique, optimal catalyst design within an acceptable simulation time. The results of this paper suggest that the use of the GA might still provide us with valuable indications of which design parameters are important and with estimates of their optimal values. Let us take the example of a Pt/silica catalyst. On a first iteration we might identify particle size and the ratio of isolated to clustered hydroxyls on the silica surface as the important influences on catalyst performance. The identification of these key characteristics would reduce an intractable design problem to one in which we need to optimise over a small set of variables. A further GA could then be used to address this restricted problem.

APPENDIX

The GA will result in the exponential spread of compact clusters of genes which produce above average fitness, according to the schema theorem introduced by Holland (11, 27). A schemata is a template describing a subset of chromosomes with some common characteristic at particular positions in each chromosome. This generalisation is facilitated by extending the set of characters used to encode each chromosome by the inclusion of a wild card character, here denoted as an asterisk, which permits the gene to assume any value, that is either 0 or 1 in the case of a binary encoding. For optimal performance it is required that the known elements of any schemata be located as close to each other as possible to avoid fragmentation of the schemata in the crossover operation. It is for this reason that two-dimensional binary strings were used to encode the surface as opposed to the conventional one-dimensional string encoding. For example, the schemata

$$\begin{bmatrix} 1 & 0 & * & * \\ 0 & 1 & * & * \\ * & * & * & * \\ * & * & * & * \end{bmatrix}$$

may be identified as "good" as each 1 has a 0 neighbour but in the 1-D representation the desirable $\left[\begin{smallmatrix} 1 & 0 \\ 0 & 1 \end{smallmatrix}\right]$ group is unlikely to be maintained as the [10**01..] string is quite likely to be fragmented in the crossover operation. To overcome this limitation we implemented a two-point, two-

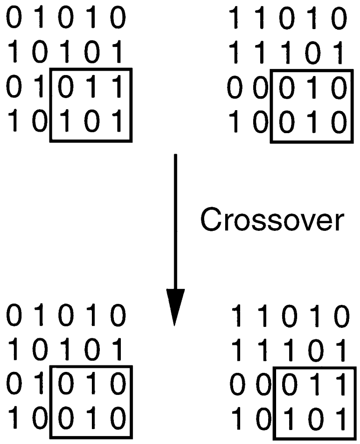

FIG. A1. The two-dimensional crossover operation.

dimensional crossover by crossing over rectangular sections of the binary string as illustrated in Fig. A1. Two points were selected at random in one of the parent chromosomes and the resulting section was swapped with the corresponding section of the other parent chromosome, producing the two children.

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