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A cellular automaton based model

for predicting intergranular corrosion in aerospace alloys

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SUMMARY

The cellular automata (CA) model is proposed for predicting intergranular corrosion. The appropriate CA rules are generated which can reproduce and predict the advance of a corrosion front The proposed model shows qualitative and quantitative agreement with experimental data on an aluminium Al2024 alloy in chloride solutions.

1. INTRODUCTION

Among the advantages of aluminium-based alloys for structural use is their high strength to weight ratio and their corrosion resistance [1]. However, while aluminium alloys are highly resistant to uniform (general) corrosion, they are much more susceptible to localised corrosion, especially intergranular corrosion, which is a localised attack along the grain boundaries which leaves the grains themselves largely unaffected. In order to estimate the progress of such corrosion in a given sample, it is possible to generate a numerical model of some sort. While there has been much effort spent in the development of electrochemistry-based models, the use of grey and black-box models remains largely unexplored. One exception to this is the use of cellular automata (CA) models which have recently been exploited to model the progression of uniform corrosion [2-16].

The object of the current paper is to apply the CA methodology to the case of intergranular corrosion. The probabilistic approach is chosen because it enables an easy simulation of the complex interactions between the chemical reactions in the electrolyte domain and the propagation of the corrosion path in the metal domain. The main focus is the simulation of preferential corrosion propagation along grain boundaries after the onset of local dissolution.

2. MODEL

The processes that are to be simulated are as follows: the metal is a set of grains separated by grain boundaries; the surface of a metal is separated from the electrolyte solution with a protective layer; when any of the three species - grain, grain boundaries or protective layer - is in contact with the solution, a reaction occurs which results in some corrosion product (thus a number of electrochemical reactions is replaced by the principal one); the corrosion product is able to diffuse into the solution.

We assume the rate of corrosion of the grains to be negligible compared to the corresponding rate for the grain boundaries, and the rate of corrosion of the grain boundaries to be dependent upon whether the boundary sites are adjacent to the grains, and if they are, to be different for each grain. Depending on the assumed diffusion rate, the corrosion product either dissolves within the solution immediately, or diffuses within the solution.

The system is modelled as a celullar automaton on a discretised lattice consisting of a number of cells. As we are dealing with a mesoscopic description, CA cells are not directly associated with the individual atoms, anions or cations, but to some limiting scale of resolution below which we consider the system to be unstructured, atomic size effects are thus not accounted for.

Each cell can assume N possible occupation states: E (empty), S (electrolyte), G (metal grain), B (grain boundary), P (corrosion product), and L (surface layer). The model has four parameters: P_{min}, P_{max}, P_{bound}, P_{layer}. Of those, P_{min} and P_{max} define the range of probabilities for the grain boundary (B) to corrode if it is in contact with a grain (G), P_{bound} is a probability for the grain boundary (B) not in contact with grain (G) to corrode, and P_{layer} is a probability for the corrosion of a surface layer (L).
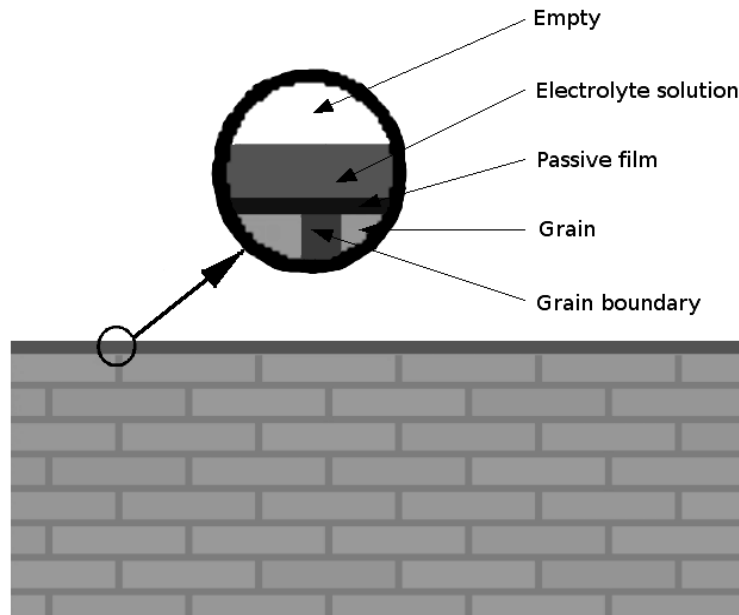
Figure 1: Initial state of the lattice.

Initially, the lattice is subdivided into the metal domain and the solution domain (see Figure 1). The metal domain consists of a number of grains (G) separated by boundaries (B). The metal domain is bounded by a thin surface layer (L) and a thin solution layer (S). Remaining sites in the solution domain are marked as empty (E). Each grain is initially assigned a random probability P_{grain} in the range between P_{min} and P_{max}. The value of P_{grain} is uniform for the whole grain.

At each discrete time step the electrochemical reactions are modelled by the events described by a set of CA rules aimed to mimic these reactions on a coarse-grained scale. The CA rules are local and determine the transition between different types of cells depending upon the state of the neighbourhood. At each discrete time step there are the following possibilities:

1. If B is in contact with at least one S and one G, it changes to S with probability P_{grain}, corresponding to the G site.
2. If B is in contact with at least one S but not in contact with G, it changes to S with probability P_{bound}.
3. If L is in contact with at least one S or P, it changes to S with probability P_{layer}.

After applying the CA rules, diffusion of the species into the electrolyte solution is modelled by a random walk process on a lattice. Potential walkers are the corrosion product sites (P), which are allowed to walk into neighbouring sites either occupied by electrolyte (S) or being empty (E). The sites are chosen in random order.

If the selected site is of type P, a random neighbour is selected; then the options are as follows:

1. If the selected neighbour is S, then it becomes P, and the current P site becomes S.
2. If the selected neighbour is E, it remains unaltered, and the current P site becomes S.
3. If the selected neighbour is neither S nor E, no changes are made.

The result mimics the diffusion of P species into the electrolyte E. The precise meaning of the concentration of P depends upon the parameters of the real system, for example, the concentration of any anion or cation.

3. SIMULATION RESULTS AND COMPARISON WITH EXPERIMENT

In this section we apply the CA model described above to simulate the experimental data obtained by Augustin et al [17]. In this regerence results are reported for the propagation kinetics of integranular corrosion in Al2024 alloy immersed in 1M and 3M chloride solutions. First we calibrate the parameters of our model to fit the experimental data. We then use the calibrated model to predict the time dependence of intergranular corrosion damage at larger times, and for different sample orientations.

The three-dimensional simulation is undertaken on a 240x280x240 lattice. Initially, there are 12x2x4=96 cuboid grains, each having size 20x140x160 cells, so that the aspect ratio of the grains is consistent with the experimental average grain size 100x700x300 µm. The width of the grain boundary is set to 4 cells.

The CA probabilities and time scale were adjusted using a least-squares method to obtain the best agreement of the predicted time dependence of the maximum corrosion depth with corresponding experimental data. It was assumed that $P_{min} = P_{max}$ (the same corrosion probability near all grains). The effect of different parameters upon the corrosion kinetics is presented in Figure 2, and the best agreement with experimental data is achieved for the following values of probabilities: $P_{min} = P_{max} = 1$, $P_{bound} = 10^{-3}$, $P_{layer} = 10^{-6}$ for 1M solution, and $P_{min} = P_{max} = 0.1$, $P_{bound} = 10^{-4}$, $P_{layer} = 10^{-4}$ for 3M solution. One time step corresponds to 0.04685 hours (1M solution) and 0.01187 hours (3M solution). Figure 3 presents the calculated maximal depth of corrosion as a function of time for 1M and

3M electrolytes, together with corresponding experimental points. It demonstrates satisfactory quantitative and qualitative agreement of experimental and simulation results, and the ability of the model to predict the advance of intergranular corrosion at longer times.
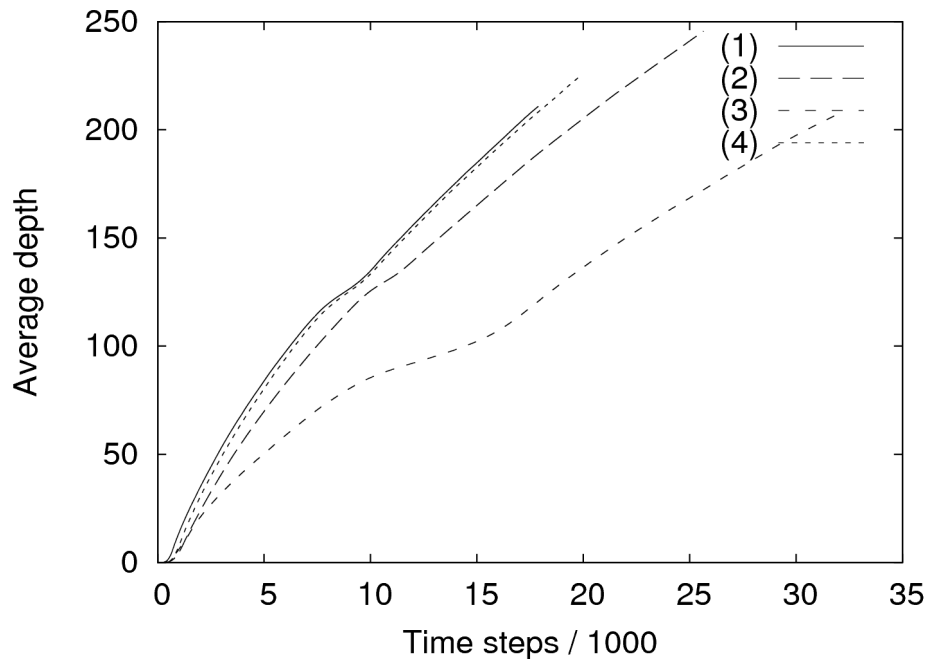

Figure 2: The effect of different parameters upon the time dependence of the average depth of intergranular corrosion damage. Different curves correspond to the following values of probabilities:

(1) $P_{min}=P_{max}=1.0$, $P_{bound}=10^{-3}$, $P_{layer}=10^{-6}$;
(2) $P_{min}=P_{max}=0.1$, $P_{bound}=10^{-3}$, $P_{layer}=10^{-6}$;
(3) $P_{min}=P_{max}=1.0$, $P_{bound}=10^{-4}$, $P_{layer}=10^{-6}$;
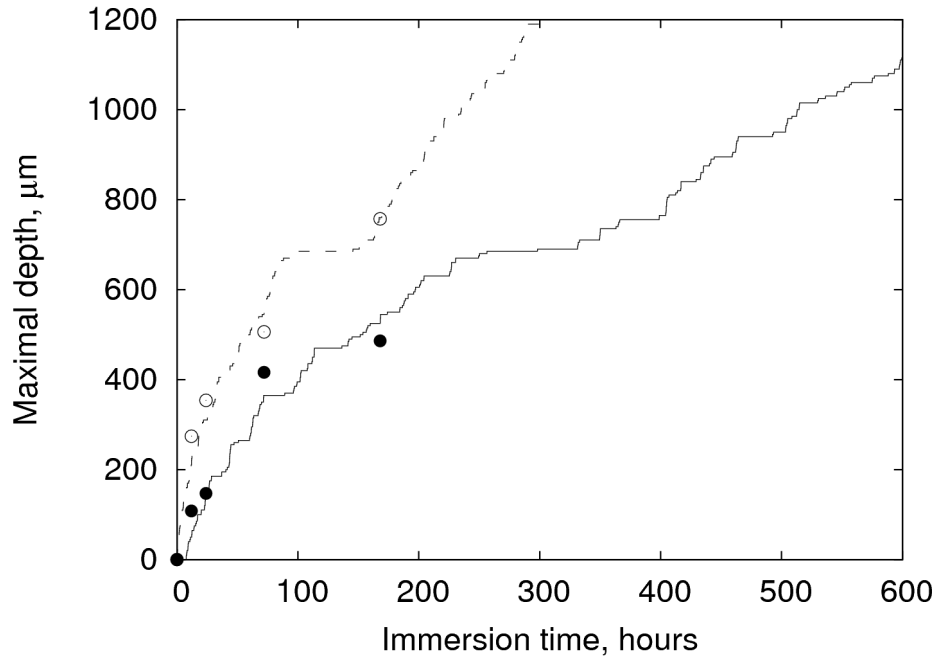*(4) $P_{min}=P_{max}=1.0$, $P_{bound}=10^{-3}$, $P_{layer}=10^{-7}$.*

Figure 3: Maximal depth of intergranular corrosion damage as a function of time. Circles represent the experimental data [17] for different electrolyte concentrations: 1M NaCl (filled circles) and 3M NaCl (empty circles). Solid line corresponds to simulation for 1M NaCl with Pmin=Pmax=1, Pbound=10^{-3}, Player=10^{-6}. Dashed line corresponds to simulation for 3M NaCl with Pmin=Pmax=0.1, Pbound=10^{-4}, Player=10^{-4}.

The model allows one to predict the corrosion propagation for different orientations of the sample. This is demonstrated in Figures 4 and 5, where the predicted maximal and average corrosion depth is depicted along longitudinal (L), long transverse (T) and short transverse (S) directions in the sample, using the values of the probabilities obtained by fitting the experimental data for 1M electrolyte solution.
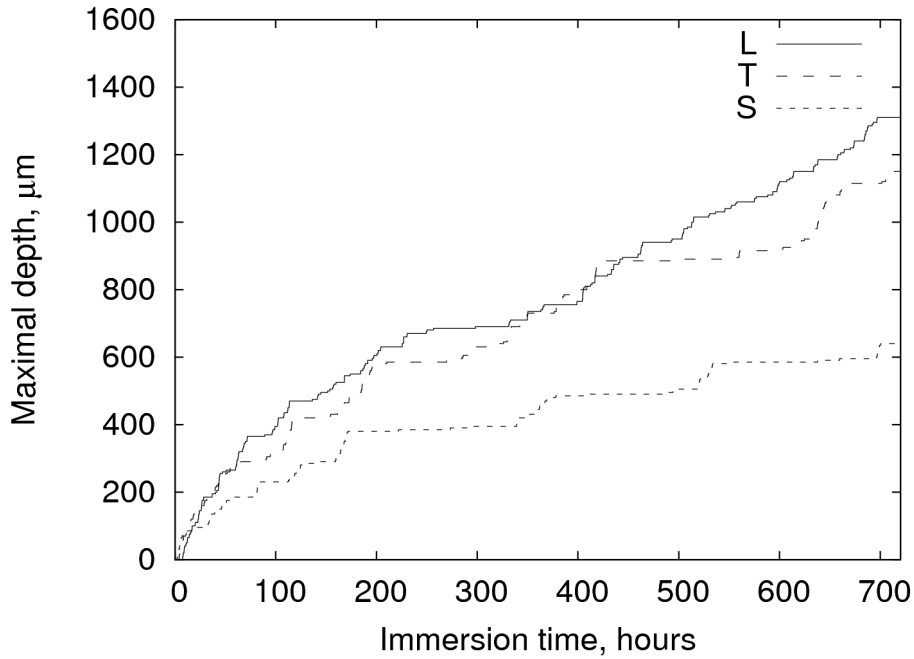
Figure 4: Predicted maximal depth of intergranular corrosion damage in 1M solution for different orientations of the sample.
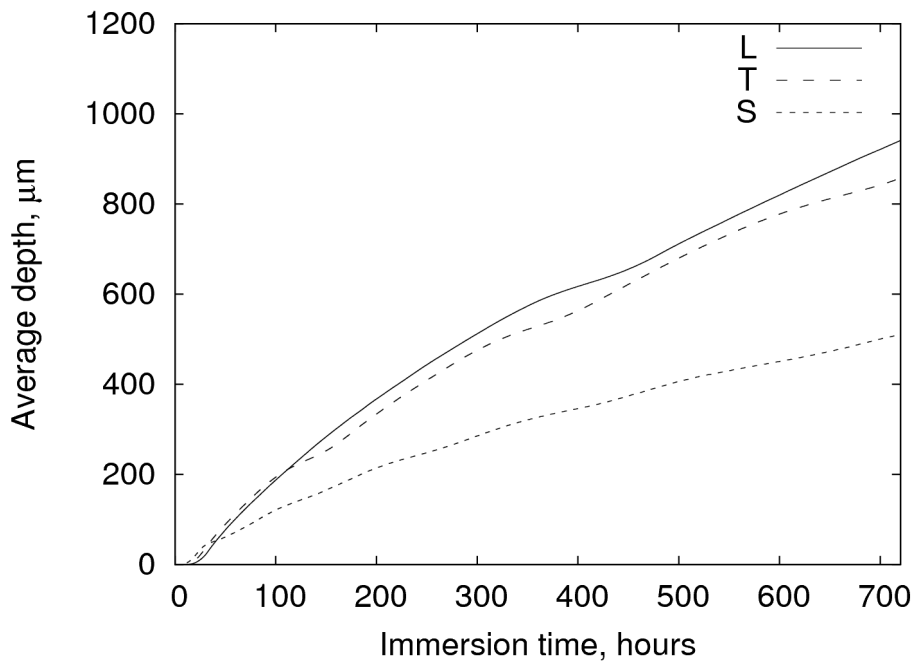

Figure 5: Predicted average depth of intergranular corrosion damage in 1M solution for different orientations of the sample.

5. CONCLUSIONS

In this paper we have developed a cellular automaton model for intergranular corrosion. The model is based on the set of CA rules in conjunction with a random

walk process, and has four adjustable parameters (probabilities). The adjustable parameters of the model have been calibrated to match the specific experiment, which has allowed a prediction of the propagation of the corrosion front for given conditions. 3D simulations show that the computations give good qualitative behaviour and can be computed in reasonable times. The simulation results show qualitative and quantitative agreement with experimental data on the advance of the corrosion front.

The set of CA rules can be modified to include a wider range of electrochemical phenomena occurring in the process of intergranular corrosion in given materials.

ACKNOWLEDGEMENTS

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