Journal of Statistical Physics, Vol. 67, Nos. 5/6, 1992

An Extension of a Result of Grannan and Swindle on the Poisoning of Catalytic Surfaces

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Received October 8, 1991

Grannan and Swindle considered a model in which molecules of several types of reactant land on a catalytic surface. When two different reactants find themselves adjacent, they diffuse from the surface and leave the sites vacant. They showed that if the rate of bonding onto the surface of one reactant is sufficiently close to one, then the surface becomes poisoned by that type. In this paper we show that a sufficient condition for poisoning is that one reactant should bond at rate greater than that of the other reactants combined.

KEY WORDS: Catalytic surfaces; submartingale.

INTRODUCTION

Grannan and Swindle[3] introduced a collection of interacting particle systems to model catalytic surfaces. In these models n types of chemical, represented by states $\{1, 2,..., n\}$, fell on sites of the integer lattice Z^d. A vacant site was represented by state 0. Molecules of chemical type i fell upon vacant sites at rate p_i, where $\sum_i p_i = 1$. The model specified that no two different chemicals could occupy adjacent sites and so if a molecule of type i fell upon a vacant site adjacent to a site occupied by chemical j $(i \neq j)$, then the two chemicals would instantaneously react, leaving their respective sites vacant. If a molecule lands at a vacant site which has a plurality of adjacent sites occupied by different chemicals, then one of these is selected at random to react with the new particle.

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0022-4715/92/0600-1219$06.50/0

Grannan and Swindle[3] showed that "poisoning" occurred in two senses.

Theorem 1 (Grannan and Swindle). If $p_1 > 1/2$, then the only invariant measures are δ_i, $i = 1, 2, \ldots, n$. Here δ_i is the point mass at the configuration which is identically i.

Theorem 2 (Grannan and Swindle). There exists an ε so that if $p_1 > 1 - \varepsilon$ and η_0 has infinitely many stes in state 1 or state 0, then $\eta_t \to \delta_1$ a.s.

We will use ideas from the first result to strengthen the second. We show the following.

Theorem 3. If $p_1 > 1/2$ and η_0 has infinitely many 1's or infinitely many 0's, then $\eta_t \to \delta_1$ a.s.

In fact, from our results leading up to the proof of the theorem it will be plain that η_t can avoid a.s. convergence to δ_1 only if for some $i > 1$,

$$\sum_x I_{\{\eta_0(x) \neq i\}} < \infty$$

Here, given an event A, I_A denotes the function on sample space which is one if A occurs and zero if A does not occur.

Proof of Theorem 3. Before proving Theorem 3, we will assemble a few lemmas. The key fact we use is taken from Grannan and Swindle.[3] If $q(1) = 1$, $q(0) = 0$, and for $i > 1$, $q(i) = -1$, $1 > \lambda > (1 - p_1)/p_1$, and $|\cdot|$ denotes the L^1 norm, then

$$f_\lambda(\eta) = \sum_{Z^d} \lambda^{|x|/2} q(\eta(x))$$

satisfies

$$\Omega f_\lambda(\eta) > 0 \qquad \text{unless} \quad \eta = \delta_i \text{ for some } i$$

In the latter cases, $\Omega f_\lambda(\eta)$ is zero. The idea of using such functions was exploited by Durrett and Steiff.[2]

The positivity of Ωf_λ implies the following lemma.

Lemma 1.1. For $\lambda \in ((1 - p_1)/p_1, 1)$, $f_\lambda(\eta_t)$ is a submartingale.

Lemma 1.1 yields the following result.

Lemma 1.2. With probability one, η_t converges to a configuration in $\bigcup_{i=1}^n \delta_i$. The probability that $\eta_t \to \delta_1$ is at least

$$\frac{f_\lambda(\eta_0) - f_\lambda(\delta_2)}{f_\lambda(\delta_1) - f_\lambda(\delta_2)}$$

Proof. First we note that $f_\lambda(\eta_t)$ is a bounded submartingale. Therefore it must converge a.s. (see, e.g., Durrett[1]). It is elementary to see that η_t converges to a configuration in $\bigcup \delta_i$ if and only if

$$\text{for each} \quad x \in Z^d, \qquad \sup\{t: \eta_t(x) = 0\} < \infty$$

But if a site x has $\eta_t(x) = 0$, then a.s. at some later time s a particle must land or attempt to land on x, resulting in a jump of size at least $\lambda^{|x|+1}$ in $f_\lambda(\eta_s)$. Thus

$$\bigcup_x \{\omega: \sup\{t: \eta_t(x, \omega) = 0\} = \infty\} \subset \{\omega: f_\lambda(\eta_s) \text{ does not converge}\}$$

Given our first observation, the latter set has probability zero and we have proven the first part of our lemma.

For the second part, we simply observe that

$$E[\lim_{t \to \infty} f_\lambda(\eta_t)] \geq f_\lambda(\eta_0)$$

since $f_\lambda(\eta_t)$ is a bounded submartingale,[1] and that $f_\lambda(\eta_t) \to f_\lambda(\delta_1)$ on $\{\omega: \eta_t(\omega) \to \delta_1\}$ and $f_\lambda(\eta_t) \to f_\lambda(\delta_2)$ $[= -f_\lambda(\delta_1)]$ on $\{\omega: \eta_t(\omega) \to \delta_i$ for $i \neq 1\}$. ∎

Remark. Consider the case $n = 2$. For notational simplicity let the two chemicals be represented by 1's and -1's, with $p_1 > p_{-1} = 1 - p_1$. It follows from simple random walk considerations that

$$P[\eta_t \text{ hits the trap state } \delta_{-1} \text{ for some } t] = \left(\frac{p_{-1}}{p_1}\right)^{\sum_{x \in Z^d} (\eta_0(x) + 1)}$$

From the ideas which follow it will be clear that this probability is also the probability that η_t converges to δ_{-1}, which, in turn, is equal to 1 minus the probability that the system converges to δ_1.

Corollary 1. For each $\varepsilon > 0$, there exists an n so that for any x and any η with $\eta \equiv 1$ on the L^1 ball $B(x, n)$

$$P^\eta[\eta_t \to \delta_1] > 1 - \varepsilon$$

Proof. First take $x = 0$, observe that for n large enough, $\eta \equiv 1$ on $B(0, n)$ implies that

$$\frac{f_\lambda(\eta) - f_\lambda(\delta_2)}{f_\lambda(\delta_1) - f_\lambda(\delta_2)} > 1 - \varepsilon$$

The desired conclusion now follows from the second part of Lemma 1.2. For general x, we merely observe that $P^{\eta}[\eta_t \to \delta_1]$ is a function of η which is invariant under translation. ∎

Proof of Theorem 3. It is obviously sufficient to show that the probability that $\eta_t \to \delta_1$ is greater than $1-\varepsilon$ for arbitrary positive ε. Fix $\varepsilon > 0$. Choose n so large that the conclusions of Corollary 1.3 hold. For $x \in Z^d$, let $\sigma(x, n+2)$ denote the sigma field generated by the arrival processes at sites in $B(x, n+2)$. We consider an event in which each empty site in $B(x, n+2)$ is first invaded by $2d$ 1's to eliminate any non-1 neighbors, and then by a further 1 to ensure becoming a 1 itself. This then happens to each site in turn. Select an enumeration $0 = v_1, v_2,..., v_N$ of $B(0, n+1)$ so that for every $j > 1$, v_j is adjacent to v_i for some $i < j$. Let $A(x)$ be the event that in time interval (0, 1), only 1's attempt to fall on sites in $B(x, n+2)$ and that there are times $0 < t_1 < t_2 < \cdots < t_{N(2d+1)} < 1$ such that at t_i, $i \in [(2d+1)(k-1), (2d+1)k]$, a 1 attempts to land at site $x + v_i$. Then:

1. For each x in Z^d, $P[A(x)] = c > 0$.
2. If x_i are such that the sets $B(x_i, n+2)$ are pairwise disjoint, then the events $A(x_i)$ are independent.
3. If $\eta_0(x) = 1$ or 0 and $A(x)$ occurs, then $\eta_1 \equiv 1$ on $B(x, n)$.

Now if there are initially infinitely many 0's and 1's, then we may find an infinite sequence $\{x_i\}$ so that for each i, $\eta_0(x_1) = 0$ or 1 and so that the sets $B(x_i, n+2)$ are pairwise disjoint. Given facts 1 and 2, it must be the case that $A(x_i)$ occurs for some i. Given fact 3, this must imply that $\eta_1 \equiv 1$ on $B(x_i, n)$ for some i. The proof is completed by invoking Corollary 1.3. ∎

ACKNOWLEDGMENT

The research of T.S.M. was partially supported by NSF grant DMS-9157461.

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Communicated by J. L. Lebowitz