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Diffusion model for the treeing process of electrodeposition experiments

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Abstract

We simulate a model that captures all the features of the silver electrodeposition experiment in a rectangular cell. We study the bulk of the aggregates on the basis of a treeing process (M. Matsushita, P. Meakin, Cluster size distribution of self-affine fractals, Phys. Rev. A 37 (1988) 3645; F. Romá, C.M. Horowitz, E.V. Albano, Numerical study of the development of bulk scale-free structures upon growth of self-affine aggregates, Phys. Rev. E 66 (2002) 066115). The model proposed is a diffusion limited process in $1+1$ dimension, where one dimension is the linear size L and the other the height. In our model the particles are dropped from the top of a rectangular lattice and are allowed to diffuse. The diffusion upwards is forbidden, whereas in the other directions the particles are allowed to diffuse with probability $1-p$ to the lateral nearest neighbors positions and with probability p downwards. Here p takes into account the strength of the electric field. When a newly deposit particle has a nearest neighbor which belongs to only one tree, it sticks to that tree. If the particle has more than one nearest neighbor that belongs to different trees one of them is selected at random and the particle sticks to the chosen tree. We compute the r.m.s height h_s, the r.m.s width w_s and the size distribution of the trees N_s as function of the mass s of the ''frozen'' trees for different values of p. We found that the scaling behavior with s of h_s and N_s depends on p, while w_s does not depend on p. In the limit $p \to 1$, the values obtained for the exponents, that characterize the scaling behavior of the magnitudes studied here, are close between the error bars, with the one found in the experiment of silver electrodeposition (C.M. Horowitz, M.A. Pasquale, E.V. Albano, A.J. Arvia, Experimental evidence of the development of scale invariance in the internal structure of self-affine aggregates, Phys. Rev. B 70 (2004) 033406 ; E.V. Albano, R.C. Salvarezza, L.Vázquez, A.J. Arvia, Validity of the Kardar–Parisi–Zhang equation in the asymptotic limit of metal electrodeposition, Phys. Rev. B 59 (1999) 7354), that was suggested to belong to the Kardar–Parisi–Zhang (KPZ). For finite p, the results suggest that our model may belong to another universality class. We also studied the finite size effect in the values obtained for the exponents and found that the p dependence is not due to finite size effects.

Keywords: Irreversible growth processes; Dynamic scaling; Interfaces and surfaces

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1. Introduction

The study of evolving interfaces or surfaces has recently attracted attention due to their potential technological applications. These interfaces can be found in many physical, chemical and biological processes. Examples include film growth either by vapor deposition or chemical deposition [1], bacterial colony growth [2] and propagation of forest fire [2]. The interface of these systems exhibits self-affine properties over a large range of spatial and temporal scales. Therefore, almost all the studies focuses on the scaling properties of the interface that are mainly characterized by the scaling behavior with the linear size L and the time t of the roughness $W(L,t)$, defined as the r.m.s (mean square root) at a given time t of the height fluctuations,

$$W(L,t) = (\langle h^2(t)\rangle - \langle h\rangle^2)^{1/2},$$

where $h(t)$ is the height of the interface at time t, and $\langle\cdots\rangle$ denotes averages over configurations and over the d-dimensional surface of typical size L. The r.m.s. roughness W of an interface is characterized by the Family–Vicsek scaling [3] with respect to t and L given by

$$W(L,t) \sim L^{\alpha} f(t/L^{z}),$$

where $z = \alpha/\beta$ is known as the dynamic exponent, β and α are the growth and roughness exponent, respectively. The scaling function $f(u) \sim u^{\beta}$ for $u \ll 1$ and $f(u) \sim$ constant for $u \gg 1$.

The study of the properties of the interfaces has been widely studied, in contrast there are only few studies on the bulk properties of the growing system [1,4]. It was shown that the measurement of the bulk properties of the process of growing is a useful experimental technique for the evaluation of the self-affine properties of some aggregates. Experimental evidence on electrochemically formed [1,5], shows that an alternative approach for the characterization of the system into universality classes can be achieved analyzing the growing branched patterns of frozen structures, that arise due to competitive processes among neighbor growing structures. These branched structures or ''trees'' can be characterized by their r.m.s height h_s and r.m.s width ω_s that exhibit scaling invariance with the mass s characterized by exponents that, within the error bars, are close to those predicted by the Kardar–Parisi–Zhang equation in $d+1$ dimension, where d is the euclidean dimension of the surface of linear size and the other is the height of the interface. For $1+1d$, $\beta = 1/3$ and $z = \frac{3}{2}$.

Also numerical evidence, presented by Romá et al. [4], on the ballistic deposition (BD) model shows that the bulk of the aggregates formed can be broken into a set of infinite scale invariant structures called ''trees''. These trees present spatial and temporal scale invariance and its exponents can be related to the classical exponents of the KPZ universality class. The exponents obtained were compared to the ones obtained in the experiment of the electrochemically formed presented in Refs. [1,5], where the intensity of the electrical field and the electromigration does not play any role. The results suggest that the experiment fall into the KPZ universality class.

In this paper, we present a model for the electrochemically aggregate where the electromigration and the intensity of the electrical field is taken into account. We analyze the bulk's properties of the growing patterns on the basis of a treeing process; i.e any growing can be thought as the superposition of individual trees.

2. Treeing and self-affinity

Suppose an aggregate that grows over a d-dimensional substrate. The aggregate is formed by trees that compete between them to grow and give rise to the total pattern. The structural properties of the individual trees and the entire aggregate are determined by the growing mechanism. The scaling behavior of h_s and w_s with s of the trees is given by [6]

$$h_s \sim s^{\nu_{\parallel}}; \tag{1}$$

$$\omega_s \sim s^{\nu_{\perp}}, \tag{2}$$

where $\nu_{\parallel}$ and $\nu_{\perp}$ are the correlation lengths exponents parallel and perpendicular to the main growth direction of the aggregate respectively [6]. When $\nu_{\parallel} = \nu_{\perp}$ the aggregates are self-similar but when they are different the aggregates are self-affine. If N_s is the number of trees with s particles, the number of particles per unit of

volume is given by

$$N = \sum_s \frac{sN_s}{L^d} = \sum_s sn_s, \tag{3}$$

where n_s is the tree size distribution that scales with s as

$$n_s \sim s^{-\tau} f\left(\frac{s^\sigma}{N}\right), \tag{4}$$

where $f(u)$ is a scaling function that dominates the scaling behavior for big values of s.

Substituting Eq. (4) into Eq. (3) and taking the main term of the sum, the scaling relation between the exponents $\sigma = 2 - \tau$ is obtained. Using the fact that $\sigma = v_\parallel(D - d)$ [6] for self-affine aggregates with geometric dimension D, τ is given by

$$\tau = 2 - v_\parallel(D - d). \tag{5}$$

This equation relates the exponent τ of the size distribution of the trees with the exponent $v_\parallel$, the geometric dimension D and the spatial dimension d. For the experimental conditions of Ref [3], $D = 2$ and $d = 1$ then

$$\tau = 2 - v_\parallel. \tag{6}$$

Taking into account that the bulk of a tree V_s scale with s as $V_s \sim h_s\ \omega_s^d \sim s^\pi$ [See Eqs.(1) and (2)],

$$\pi = v_\parallel + \mathrm{d}v_\perp, \tag{7}$$

when $\pi = 1$ the trees are compact, while for $\pi > 1$ they are not.

Identifying the parallel length correlation $\xi_\parallel \sim t^{1/z}$ with ω_s and h_s with t [6], and using Eqs. (1) and (2), we obtain

$$z = \frac{v_\parallel}{v_\perp}. \tag{8}$$

Therefore, the dynamical exponent z that is an exponent that characterize the interface is related with the exponents that characterize the bulk's properties.

3. Model and simulation results

Our model take place in a $1 + 1$ dimensional square lattice, where one dimension is the surface of linear size L and the other is the height of the interface. The interface is represented by the set $\{h_i\}$, $i = 1, \ldots, L$, where h_i is the height of the interface at column i. Periodic boundary conditions in the lateral dimension are assumed. Particles are dropped at a random column from a height five times the height of the maximum value of the interface and are allowed to diffuse with probability p downward and $1 - p$ in the lateral directions. Upwards diffusion is forbidden. Here p represents the strength of the electric field and $1 - p$ takes into account the mass transport or "diffusion" due to the thermal energy transferred by the bulk current to the solution where the particles travel. At the beginning the interface is flat, without loss of generality we assume that $h_i = 0$, $i = 1, \ldots, L$, as the initial condition of the interface, therefore the particles deposited at the flat interface are seeds for the treeing process. If a particle reaches a position nearest neighbor of a tree, it is considered as a new deposited particle that belongs to the same tree as the one of the nearest neighbor particle. If there is more than one nearest neighbor belonging to different trees one of them is selected at random and the particle is incorporated to that tree. The process stops when exactly M particles are deposited. In Fig. 1 we show a typical configuration of the trees after M particles were deposited. We analyze the bulk properties of the frozen trees.

As in our problem we have a tuning parameter p we expect that the exponents $v_\parallel$ and $v_\perp$ will depend on p. Notice that in our problem $p = 1$ is not a random deposition [2] process because the process of selection of the tree, where a new particle will stick, generate correlations between nearest neighbors. Therefore, it is expected that for $p \to 1$ the process will belong to the same universality class as the BD growth model i.e. the KPZ universality class. On the other hand, when diffusion is the main driving force we expect $v_\parallel(p) > v_\parallel(p = 1)$

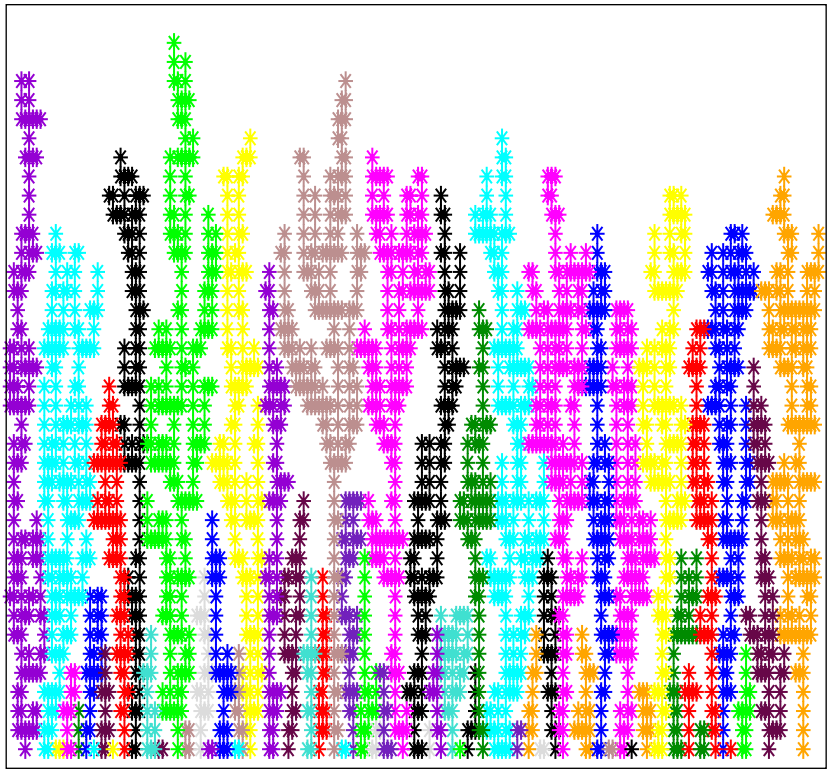

Fig. 1. Plot of one configuration of the trees formed with our model for $p = 0.64$ and $L = 250$. Different colors indicate different trees. We can see some trees that cannot grow anymore because they are overlapped by other trees. Those trees are frozen structures were our analysis was done.

because as p decreases the particles are allowed to diffuse more in the lateral direction reducing the screening of the trees.

In order to prove our ansatz we perform extensive numerical simulations of our model to study the magnitudes of the bulk properties of the growing process such as h_s and ω_s as function of s, given by

$$h_s^2 = \frac{1}{s}\sum_{i \in s} h_i^2; \tag{9}$$

$$\omega_s^2 = \frac{1}{s}\sum_{i \in s} \omega_i^2, \tag{10}$$

where h_i and ω_i are the height and the width of the column i. As the main direction of growth is perpendicular to the surface we analyze only the cases $p \geqslant 0.32$. In Fig. 2(a) we show the log–log plot of h_s as a function of s for different values of p. We can see that as p decreases the slope of h_s increases, thus $\nu_\parallel \equiv \nu_\parallel(p)$. The exponent $\nu_\parallel(p)$ was obtained by a least square fitting of the data minimizing the χ^2 function [7] with a power law in the region where it is valid, using as the standard deviation the errors of the simulations. The values obtained are plotted with their errors in the inset of Fig. 2(b) and shown in Table 1. We can see that the values of $\nu_\parallel(p)$ depend on p. In Fig. 2(b) we plot h_s as a function of $s^{\nu_\parallel(p)}$, it is easy to see the linear relation. In order to obtain $\tau(p)$ we compute $N_s(s)$. The values of $\tau(p)$ were obtained by the same method than $\nu_\parallel(p)$ fitting N_s as function of s with a power law. In Fig. 3 we show the plot of $N_s/s^{-\tau(p)}$ as function of s. The flat behavior of the curves for big values of s, where the power law holds, shows the agreement with the computed values of $\tau(p)$. In the inset of Fig. 3 we plot the values obtained for this exponent as function of p with the error bars. Also this exponent depend on p. Performing the same analysis for $\omega_s(s)$, we find that $\nu_\perp \simeq 0.4109 \pm 0.0003$ independent of p. In order to confirm this result, we compute the conditional distribution of ω_s given s and p, $P(\omega_s/s, p)$.

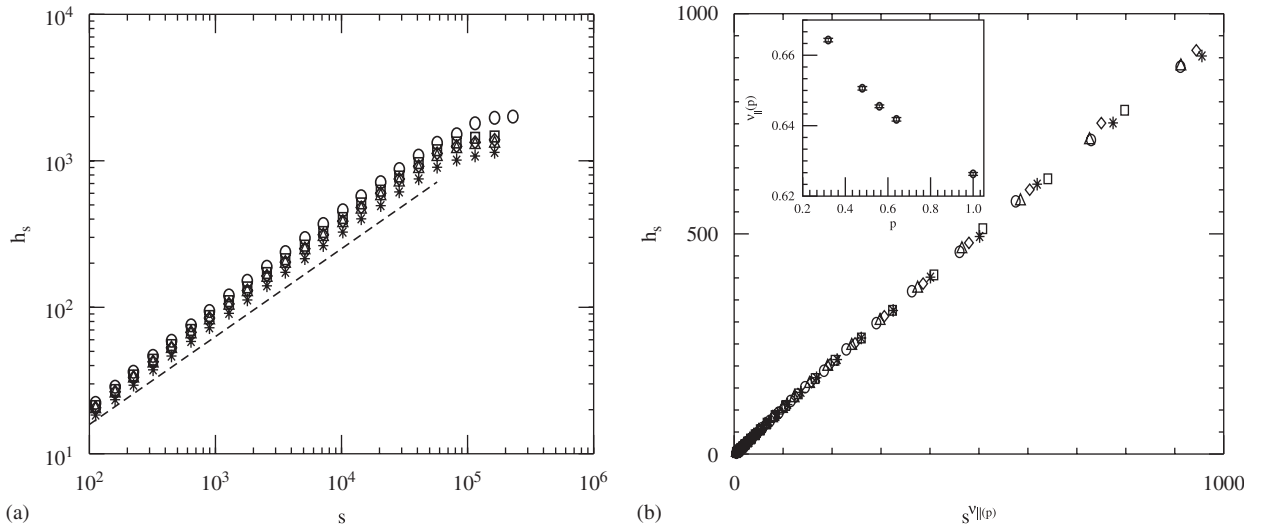

Fig. 2. (a) Log–Log plot of h_s as function of s for different values of p, $p = 1$ (∗), $p = 0.64$ (△), $p = 0.56$ (⋄), $p = 0.48$ (□) and $p = 0.32$ (◯). The dashed line is used as a guide to show the KPZ behavior of h_s ($h_s \sim s^{0.6}$). (b) Plot of h_s as function of $s^{\nu_\parallel(p)}$ for different values of p. The symbols represent the same as in (a) with the values of $\nu_\parallel(p)$ from Table 1. The collapse of the curves indicates that $h_s \sim s^{\nu_\parallel(p)}$. In all these simulations $L = 10^3$, $M = 10^6$ and the averages were done over 10^6 realizations. In the inset plot, we show $\nu_\parallel(p)$ as function of p with the error bars using the method of least squares with a power law as explained in the text.

Table 1
Values of the exponent $\nu_\parallel(p)$, $\pi(p)$, $\tau(p)$ and $z(p)$ as function of p from the simulations compared with the values of the experiment of silver electrodeposition [4], the BD model and the actual values of the exponents of the KPZ and Edward Wilkinson (EW) [8] universality classes.

p	$\nu_\parallel(p)$	$\pi(p)$	$\tau(p)$	$z(p)$
0.32	0.6648(2)	1.0757(5)	1.333(3)	1.618(2)
0.48	0.6506(5)	1.0615(8)	1.350(1)	1.583(2)
0.56	0.6455(4)	1.0564(7)	1.361(7)	1.571(2)
0.64	0.6418(5)	1.0527(8)	1.382(4)	1.562(2)
1	0.6263(4)	1.0372(7)	1.399(3)	1.524(2)
EXP	0.63(3)	1.09(7)[1]	1.37(4)[1]	—
BD	0.6	—	1.4[4]	—
KPZ	0.6	1	1.4	1.5
EW	0.65	1	1.35	2

We find that this distribution can be approximated very well with a Gaussian function,

$$P(\omega_s/s) \sim \exp\left[-\frac{(\omega_s - \langle\omega_s\rangle)^2}{2\sigma^2}\right], \tag{11}$$

where $<\omega_s> = \omega^* \sim s^{\nu_\perp}$ and ω^* is the most probably value of the distribution, that in a Gaussian coincides with the mean value. In Fig. 4 we plot $P(\omega_s/s)s^{\nu_\perp}$ as function of $\omega_s/s^{\nu_\perp}$. The collapse of the curves indicates clearly that $\nu_\perp$ depends only on s and does not depend on p. From the values obtained for $\nu_\parallel(p)$ and $\nu_\perp$, we compute $\pi(p)$ using Eq. (7) (see Table 1). We can see that as p decreases $\pi(p)$ increases, so the trees are slightly less compact. Replacing the values of $\nu_\parallel(p)$ and $\nu_\perp$ in Eq. (8) we obtain $z(p)$. The values are shown in Table 1 and plotted with the errors in Fig. 5. From the results we can see that as p decreases, z increases. All our results suggest that for $p < 1$ our model does not belong to the KPZ universality class. In order to see whether our results are or not due to finite size effects, we compute the exponents for different system sizes. In Figs. 6 and 7

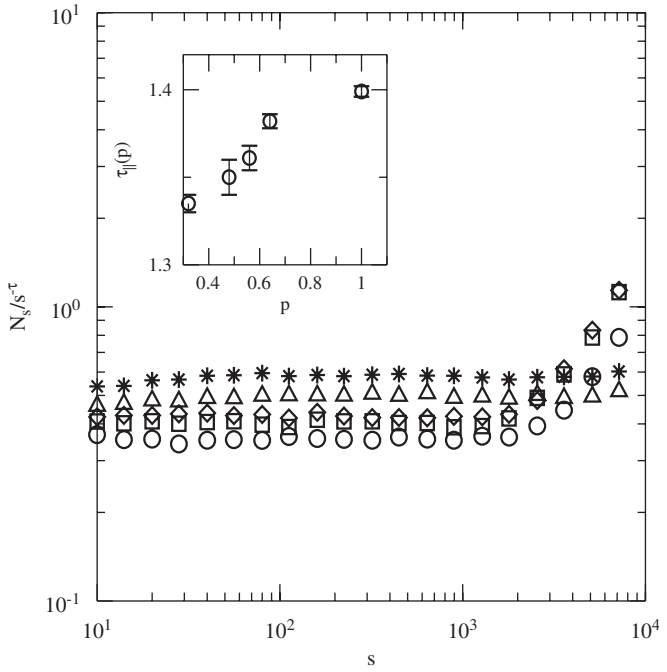

Fig. 3. Plot of $N_s/s^{-\tau}$ in log–log scale only to display better the fact that for big s, $N_s/s^{-\tau}$ is constant. The symbols represent the same values of p than in Fig. 2. In the inset figure, we show the values obtained for $\tau(p)$ as function of p with the error bars using the method of least squares with a power law as explained in the text.

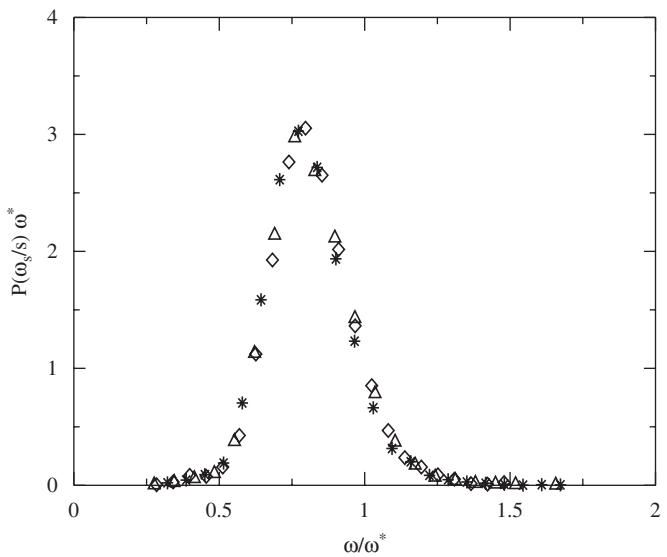

Fig. 4. Plot of $P(\omega_s/s)$ as function of s for different values of p. The collapse of the curves shows that the ω_s does not depend on p. The symbols represent the same values of p as in Fig. 2.

we show the results obtained for $v_{\parallel}(p)$ and $\tau(p)$ for $p = 0.32$ and different system sizes. We can see from the figures that after some characteristic system size close to $L = 1000$ the values obtained, between the error bars, does not depend on L. Thus, in our model, the correction to scaling due to finite size effects are not necessary for the values displayed in Table 1. We checked that this is also the case for other values of p.

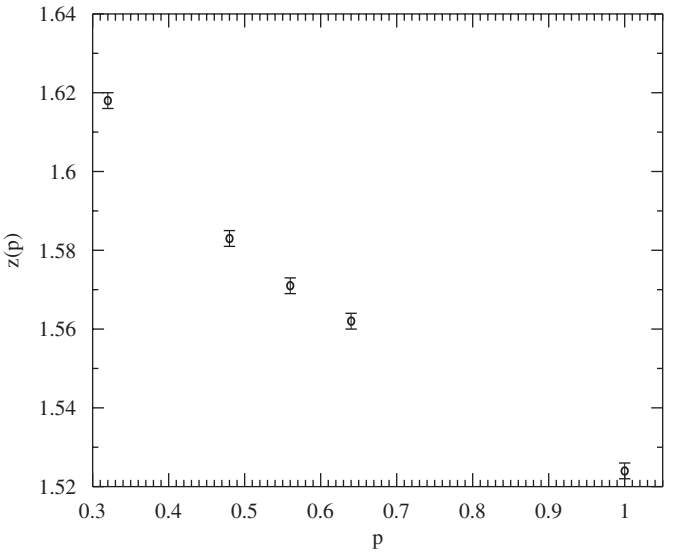

Fig. 5. Plot of $z(p)$ as function of p with the error bars, for $L = 1000$ and 10^6 realizations. Our results shows that as p increases z decreases to a value very close to the one of the KPZ universality class.

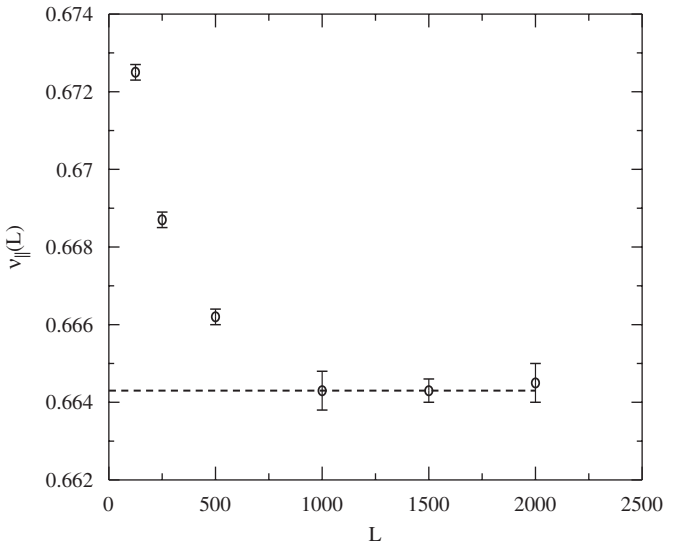

Fig. 6. Plot of $v_{\parallel}(p)$ as function of L for $p = 0.32$ with $L = 125, 250, 500, 1000, 1500$ and 2000. The dashed line is used as a guide to show the value reported in Table 1. Our results shows that after some characteristic size around $L = 1000$ the exponent does not depend on L. Thus the finite size effects of our model are weak.

4. Conclusions

In this work we present a model where the strength of the electric field and the diffusion are taking into account through a parameter p. Our model represents well the electrochemical aggregate experiment in the limit $p \to 1$. In this limit we find exponents close to the one of the $1 + 1d$ KPZ model. The finite size effects are very weak and after a characteristic value of $L{\sim}1000$, $v_{\parallel}(p)$ does not depend on L. The exponent $\tau(p)$, does not depend on L between our error bars. We can conclude that, except $v_{\perp}$, all the other exponents depend on p. Our results could be explained by a crossover from a KPZ regime for $p = 1$ to another universality class as p decreases.

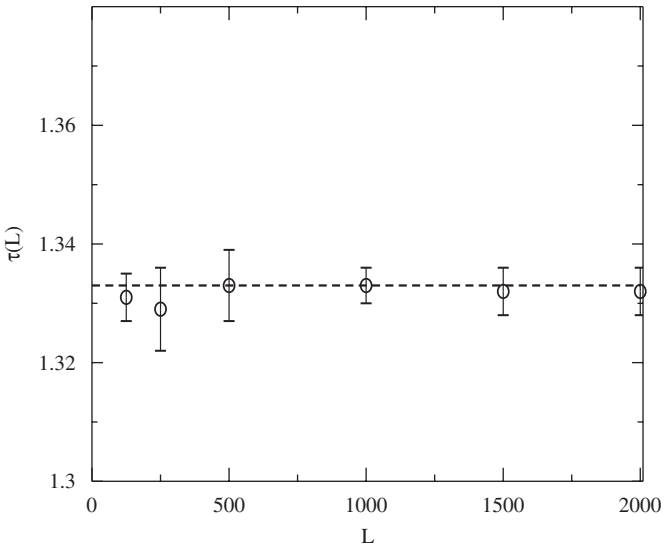

Fig. 7. Plot of $\tau(p)$ as function of L for $p = 0.32$ with $L = 125, 250, 500, 1000, 1500$ and 2000. The dashed line is used as a guide to show the value reported in Table 1. Our results shows that between our error bars this exponent does not depend on L.

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