The Branched Polymer Growth Model Revisited

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The branched polymer growth model (BPGM) has been employed to study the kinetic growth of ramified polymers in the presence of impurities. In this article, the BPGM is revisited on the square lattice and a subtle modification in its dynamics is proposed in order to adapt it to a scenario closer to reality and experimentation. This new version of the model is denominated the adapted branched polymer growth model (ABPGM). It is shown that the ABPGM preserves the functionalities of the monomers and so recovers the branching probability $b$ as an input parameter which effectively controls the relative incidence of bifurcations. The critical locus separating infinite from finite growth regimes of the ABPGM is obtained in the $(b, c)$ space (where $c$ is the impurity concentration). Unlike the original model, the phase diagram of the ABPGM exhibits a peculiar reentrance.

Keywords: Branched Polymer, Critical Transition, Reentrant Phase

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I. INTRODUCTION

The statistical mechanics of polymers has become a very prolific research area. During the last decades, many models have been proposed to describe the configurational properties of linear and branched polymers and a variety of methods have been employed to study these systems. The classic model for a linear polymer in dilute solution (or at high temperature) is the self-avoiding walk (SAW) [1,2]. On the other hand, connected clusters (lattice animals) provided a good model for branched polymers in the dilute limit [3].

The kinetic growth walk (KGW) was proposed as an alternative model to describe the irreversible growth of linear polymers [4-6]. Like the SAW, every KGW chain does not intercept itself. But whereas in a SAW the next step is randomly chosen from among all nearest-neighbor sites (excluding the previous one), in a KGW the choice is among the unvisited sites [4]. Therefore the KGW is less sensitive to attrition. Besides, although both models span the same set of configurations, the N-step SAW chains are all equally weighted while the statistical weights of the KGW chains can be different. Nevertheless, it was shown that both models present the same critical exponents [6].

Later, Lucena et al. [7] generalized the KGW in order to allow for branching as well as for impurities. This generalized model (which became known as the branched polymer growth model - BPGM) was found to exhibit an interesting phase transition (due to competition between hindrances and branching) separating infinite from finite growth regimes [8]. In the following years, several authors have studied the BPGM [8-15]. The topological and dynamical aspects of the model were investigated [8]. Besides, the system was shown to achieve the criticality through a self-organization growth mechanism [12] and to exhibit a transition from rough to faceted boundaries for large values of the branching probability [8,13]. The model was also studied through an exact enumeration of bond trees and ergodicity violation was discussed [14]. The question of the universality class of the BPGM was also investigated. In contrast to the common belief that branched polymers belong to the universality class of the Bethe lattice animals [16,17], the study of the growth process in chemical $l-$space with estimates of structural exponents led to the proposal that the BPGM belongs to the universality class of percolation [10,11]. On the Bethe lattice, this proposal is based on analytical results [11]. A further analysis using finite-size scaling techniques led to the conclusion that the BPGM is not in the same universality class of percolation in two dimensions [15].

In the present paper, we revisit the BPGM on the square lattice. We point out that, although the input parameter $b$ is named branching probability, it does not have effective control of the relative incidence of bifurcations (branches) in the simulated polymers. This seems to be an undesirable aspect of the model since the degree of branching is an important quantity that is usually measured in real ramified polymers. Besides, according to the growth rules of the BPGM, a monomer which is chosen to bifurcate (with probability $b$) but has only one empty nearest neighbor site is compelled to grow linearly thus changing its functionality from 3 to 2. In real experiments this change is not allowed since the concentrations of polymers with different functionalities are fixed. In order to preserve the functionalities of the monomers as well as to recover the meaning of the branching probability $b$, we propose a subtle modification in the dynamics of the BPGM. So this new version of the model aims to adapt it to a more realistic scenario and is called the...
adapted branched polymer growth model (ABPGM). In the following sections, we define the ABPGM and present the phase diagram in the \((b,c)\) space. This diagram is found to exhibit a peculiar reentrance. We introduce the concept of frustration and compare the effective branching rate \(b_{ef}\) with the input parameter \(b\) for both the original and adapted models. Our results concerning the ABPGM are shown to be much closer to the ideal behavior \(b_{ef} = b\). Finally, we present a discussion based on the clusters topologies of both models.

II. THE ADAPTED BRANCHED POLYMER GROWTH MODEL

First of all, let us review the BPGM. Consider a \(L \times L\) square lattice with a certain concentration \(c\) of sites randomly filled by impurities. At the initial time \(t = 0\), the polymer starts growing from a monomer seed located at the center of the lattice towards a random empty nearest-neighbor site. At time \(t = 1\), this chosen site is occupied by a monomer (growing tip) which now may bifurcate or follow in one direction (linear growth). The growth directions are randomly chosen among the available ones (those which lead to empty neighbors) in a clockwise way. At every time \(t\) \((t \geq 2)\), the process is repeated for all actual monomeric ends following the sequence of their appearances. If a polymer end has no empty nearest-neighbor site then it is trapped in a “cul de sac” and stops growing (it is then called a dead end). If only one adjacent site is available then the linear growth is obligatory. If at least two adjacent sites are empty then the growing end bifurcates with probability \(b\) or follows linearly with probability \(1 - b\). In a particular experiment of the BPGM, the polymer growth is simulated according to the above rules. Depending on the values of parameters \(b\) and \(c\), the polymer can either grow indefinitely or stop growing (die) at a finite time if all its current tips are dead ends. The experiment is finished either when the polymer touches the frontier of the lattice (infinite polymer) or when it dies (finite polymer). The ensemble over which averages are performed is constituted by a great number \(N_e\) of experiments.

Each polymer configuration of the BPGM can be identified with a self-avoiding loopless graph (bond tree) although the reciprocal is not always true. Thus the BPGM can be mapped into a particular subset of the ensemble of all possible bond trees. These tree graphs have been applied in models of branched polymers and can be classified by the number \(N\) of bonds and the number \(N_k\) of vertices with \(k\) bonds. In a typical finite polymer configuration of the BPGM, one may find twofold and threefold sites representing monomers of functionalities 2 and 3, respectively. The monomer seed and dead ends represent monofunctional monomers. Tetrafunctional units are impossible in this model once trifurcations are not allowed. For a tree graph generated by the BPGM one has the following constraints:

\[
\sum_{k=1}^{3} N_k = N + 1 \tag{1}
\]

and

\[
\sum_{k=1}^{3} kN_k = 2N . \tag{2}
\]

The occurrence of bifurcations in the model determines the relative incidence of trifunctional monomers (branches). This is basically the degree of branching of the polymer, an important quantity that is usually measured in real ramified polymers. However, although the parameter \(b\) is named branching probability, we shall see that it does not have effective control of the relative amount of branches (bifurcations) in the BPGM. Indeed, according to the rules of the BPGM, the parameter \(b\) is not the probability that any tip bifurcates but instead it is the conditional probability that a tip with two or more empty nearest neighbors bifurcates. Every tip with just one vacant nearest-neighbor site grows linearly with probability equal to one. From another viewpoint, it would also be desirable to consider that any monomer that is linking to the polymer has an effective probability \(b\) of being a trifunctional unit. Even if this were considered in the BPGM, a problem would become evident. Any growing end chosen (with probability \(b\)) as a trifunctional monomer would not be able to bifurcate if only one vacant site were available. In this case, the functionality of the monomer would not be respected since (according to the BPGM) the growing end would forcibly follow a linear growth like a bifunctional monomer.

In order to preserve the functionalities of the monomers we propose a modification in the dynamics of the BPGM. Besides to make the model a little more realistic, our proposal turns the branching probability \(b\) into an effective control parameter of the relative frequency of bifurcations. So it adapts the model to a scenario closer to reality and experimentation. We denominate it the adapted branched polymer growth model (ABPGM).

The ABPGM is defined just like the BPGM except for the following differences:

1. In the process of polymerization, let a free end (a growing tip with at least one empty nearest-neighbor site) be a trifunctional monomer with probability \(b\) or a bifunctional monomer with probability \(1 - b\). If the free end is a trifunctional monomer but there is only one empty nearest-neighbor site then it stops growing and becomes a dead end.

2. At every time unit, all current sites on the front of growth of the polymer are visited in a random sequence.
We remember that in the BPGM, a free end with just one empty nearest neighbor grows linear with probability one while in the ABPGM, since bifurcation is impossible, the free end stops and becomes a frustrated dead end. This frustration seems to be preferable and more realistic than forcing a trifunctional monomer to turn into bifunctional monomer as it occurs in the BPGM. So, in our proposal, the relative incidence of bifunctional and trifunctional monomers does not depend on the topology of the cluster anymore and is only controlled by the branching probability parameter. The second difference is also important: in the BPGM all tips are sequentially visited in a clockwise manner following the sequence of births whereas in the ABPGM they are visited in a random way. The sequential update of the growth front of the polymer (in the BPGM) simulates the formation of parallel chains in the infinite phase (for high $b$ and low $c$) like in a crystallization process. This mechanism leads to a faceted-to-rough transition that has been already studied [8, 13]. Figure 1a shows a typical BPGM configuration with faceted boundary generated with parameters $b = 0.5$ and $c = 0$ (for $L = 51$); the parallel ordering of chains is caused by the clockwise update of the growing tips. If we simulate the BPGM using the same set of parameters but with a random update instead of the clockwise update we get Figure 1b. In this case, two subsequent growing tips are probably located far apart so that they cannot produce parallel chains. The boundary is less faceted than before. Recently it has been shown that any deterministic growth order is non-ergodic in the sense that it spans only a subset of the space of all possible configurations [14]. Moreover, for the present purposes, this clockwise update mechanism is undesirable since the development of parallel chains corresponds to an increasing incidence of bifunctional monomers due mainly to geometrical effects rather than to the probability $1 - b$ itself. The relative incidence of bifurcations (which will be defined as the effective branching rate $b_{cf}$ in section 4) is much smaller than input parameter $b$. Indeed, we anticipate that $b_{cf} \approx 0.15$ for the cluster of Figure 1a whereas $b_{cf} \approx 0.30$ for the one of Figure 1b; so the last rate (corresponding to a BPGM cluster generated with random update) is closer to the input value $b = 0.50$. The effective branching rate gets still closer to the input value if we simulate the ABPGM. Figure 1c is a typical ABPGM configuration (generated with those same parameters $b = 0.5$ and $c = 0$) and presents $b_{cf} \approx 0.43$: the faceted front of growth seems to disappear and vacancies can be found inside the cluster.

The main feature of the BPGM is the phase transition separating finite from infinite regime. Lucena et al. [7] have defined a critical branching probability $b_c$ where the mean size of finite polymers diverges as $L \to \infty$. The critical value depends on the impurity concentration $c$ and separates the finite phase ($b < b_c(c)$) from the infinite one ($b > b_c(c)$). The probability that a polymer grows indefinitely is null for small values of $b$ and increases abruptly in the region of $b \approx b_c(c)$. This probability was estimated through the fraction $P_\infty$ of infinite polymers in the ensemble of configurations of the BPGM [7]. In Figure 2a, we reproduce typical plots of $P_\infty$ versus $b$ at different values of $c$ (with $N_e = 10^2$ experiments and $L = 1501$) for the BPGM. In Figure 2b, we present plots of $P_\infty$ versus $b$ at some values of $c$ corresponding to simulations of the ABPGM with $N_e = 10^4$ experiments and $L = 1501$. For $c = 0$, the behavior of $P_\infty(b)$ is analogous to that of the BPGM but now the threshold is little bit higher ($b \approx 0.06$ for the ABPGM while $b \approx 0.055$ for the BPGM). This difference increases...
with \( c \). The most interesting characteristic of the present
model may be observed when \( c = 0.18 \). For this value, the
curve raises at \( b \approx 0.32 \), then presents a plateau (where
\( P_\infty(b) \approx 0.7 \)) and finally falls! By a finite-size scaling
analysis we have verified that the height of the plateau
does not change significantly in the limit \( L \to \infty \). For
\( c = 0.19 \), the behavior of \( P_\infty(b) \) is gaussian shaped. This
means that, for certain impurity concentrations, as \( b \) in-
creases the system goes from a finite to an infinite phase
and then becomes finite again! Indeed, this reentrance is
confirmed in the next section when we determine the
phase diagram of the ABPGM through an analysis of the
correlation length.

\[
\xi \propto L \quad \text{for} \quad b < b_c,
\]

\[
\xi \propto \sqrt{L} \quad \text{for} \quad b > b_c.
\]

FIG. 2: Plots of the fraction of infinite polymers \( P_\infty \) versus \( b \)
for the BPGM (a) and ABPGM (b).

III. THE PHASE DIAGRAM

The mean size of finite polymers is a measure of the
correlation length \( \xi \) of the system. If a finite polymer
is generated during a simulation, the sizes \( l_x \) and \( l_y \) of
the smallest rectangle containing the cluster can be de-
determined. The correlation length can then be calculated
as \( \xi = \langle (l_x + l_y)^{1/2} \rangle \) where the average is performed over
all experiments with finite polymers \([7]\). We show typi-
cal plots of \( \xi \) versus \( b \) corresponding to simulations of
the BPGM (in Figure 3a) and ABPGM (in Figure 3b)
for different values of \( c \) and size \( L = 1501 \) (with \( 10^2 \) and
\( 10^4 \) experiments respectively). Each plot of \( \xi \) versus \( b \)
exhibits only one maximum except for plots of the ABPGM
with \( c > 0.175 \) where two peaks are detected! It can be
verified that all peaks of \( \xi \) do diverge when \( L \to \infty \).

FIG. 3: Plots of the correlation length \( \xi \) of finite polymers
versus the branching probability \( b \) for several impurity con-
centrations \( c \) in the cases: (a) BPGM and (b) ABPGM.

Let us first explain the behavior of \( \xi \) for the BPGM.
For a fixed value of \( c \) and as \( b \) increases, the polymer
is more likely to escape from steric hindrances and im-
purities so that the mean size grows to its highest value
at some \( b = b_0(L) \). Above this point, the system is de-
defined to be in the infinite growth regime (of course, the
truer critical point is obtained in the thermodynamic limit
\( b_c = \lim_{L \to \infty} b_0(L) \). As \( b \) continues to increase, the frac-
tion of infinite polymers grows and the finite polymers get
smaller so that \( \xi \) decreases. As higher impurity concen-
trations hinder the growth, \( b_0 \) increases with \( c \). The
critical line of the BPGM is the locus \((b_c, c)\) on which \( \xi \)
diverges \([7]\) and is shown in Figure 4 (dashed line).

Regarding the ABPGM, the same reasoning can ex-
plain the maximum of \( \xi \) (or the first maximum when
there are two peaks). But now this peak is located on
a higher branching probability \( b_1 > b_0 \) that compensates
the occurrence of frustrated dead ends. Just above \( b_1 \)
the system enters the infinite growth regime where it re-
mains unless a second peak appears (at \( b = b_2 > b_1 \) for \( c \n\) > 0.175). In the latter case the system returns to the fi-
nite phase! This reentrance from infinite to finite growth regime is a peculiar feature of the ABPGM. For this modified model, there is a certain range of values of $c$ where it is very probable that all free ends become frustrated (and stop growing) for a sufficiently large $b$ so that the polymer growth is finite again. The ABPGM phase diagram is also shown in the Figure 4. The reentrant phase only exists for $c$ in the small interval $[0.175, 0.195]$.

FIG. 4: The phase diagram of the models BPGM (dashed line) and ABPGM (solid line). Each critical line separates the infinite phase (at low values of $c$) from the finite one (at higher values of $c$).

We have also measured the fraction $P(N)$ of finite polymers with $N$ bonds. In Figure 5, we have a log-log plot of the polydispersity distribution $P(N)$ of the ABPGM on the critical line (running $10^5$ experiments in a $L = 1501$ lattice). The three sets of data correspond to the critical points: $b_1 \approx 0.06$ and $c = 0$ (black dots); $b_1 \approx 0.32$ and $c = 0.18$ (open circles); $b_1 \approx 0.93$ and $c = 0.18$ (squares). The data are fitted by a straight line with slope $-0.75$. So, on the critical line, $P(N)$ decays with $N$ as a power law. We have verified that outside the critical line, $P(N)$ decays exponentially with $N$. Lucena et al. have found a similar behavior in the BPGM.

FIG. 5: Log-log plot of the distribution $P(N)$ of finite polymers with $N$ bonds for simulations of the ABPGM (with $N_c = 10^4$ and $L = 1501$) at some points $(b, c)$ on the critical line.

IV. THE FRUSTRATION AND THE EFFECTIVE BRANCHING RATE

The frustration is a desirable event which distinguishes the ABPGM from the original model. It is defined as the interruption of the growth of any free end which was chosen (with probability $b$) as a trifunctional monomer but is unable to bifurcate since only one nearest neighbor site is available. It prevents such a free end to continue its growth linearly like a bifunctional monomer (as occurs in the BPGM) and consequently controls the relative incidence of branches.

Clearly, each polymer configuration of the ABPGM is also a bond tree which can be classified by the numbers $N$ (of bonds) and $N_k$ (of $k$-functional units). Besides, every dead end is a site which ceased to grow and so represents a monofunctional vertex; it can be subclassified as either a trapped site (if it is in a "cul de sac") or a frustrated dead end (otherwise). We denote by $\bar{N}_1$ the number of frustrated dead ends. For the BPGM, $\bar{N}_1 = 0$ since any free end never stops. In order to measure the relative incidence of frustrated ends among those free sites chosen as trifunctional monomers, we define the frustration rate as

$$f = \left( \frac{\bar{N}_1}{N_1 + N_3} \right)$$

where the average is performed over all experiments. For infinite polymers, the current sites on the front of growth should not be considered in the computation since their functionalities are undetermined.

According to the prior definition, $f = 0$ for any values of $b$ and $c$ in the BPGM. We remark that the null frustration of the BPGM does not mean that all free sites chosen
to bifurcate succeed but only that when they fail they are transformed in monomers with functionality two.

For the ABPGM, the frustration rate increases with $b$ and $c$ as it is shown in Figure 6. Indeed, both the excluded volume due to self-avoidance (which increases with $b$) and the impurities diminish the chance of success of any free end, so $f$ is a monotonically increasing function of $b$ and $c$.

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The plots of $b_{ef}$ versus $b$ at different values of $c$ for simulations of the BPGM and ABPGM (on a large $L = 1501$ square lattice) are shown in Figures 7a and 7b, respectively. The ideal behavior $b_{ef} = b$ is indicated as the dashed lines. Regarding the BPGM, there is evidently a large discrepancy between the input parameter $b$ and the output rate $b_{ef}$. For $c = 0$, $b_{ef}$ increases with $b$ up to a maximum (at $b \approx 0.2$) and then decreases to zero as $b \to 1$. This behavior is explained as follows: for small $b$, self-avoidance is reduced so that most sites trying to bifurcate succeed; but as $b$ increases, parallel linear chains (like those of Figure 1a) are forcibly generated due to both the increasing excluded volume and the clockwise update. For higher values of $c$, the presence of impurities hampers the formation of linear chains and thus $b_{ef}$ decreases slower (as a concave function). Anyway, for the BPGM, the discrepancy between $b$ and $b_{ef}$ gets more pronounced as $b \to 1$. On the contrary, for the ABPGM, $b_{ef}$ always increases with $b$ as a convex function (for any value of $c$) and the difference $b - b_{ef}$ is very small. In fact, for any $c$, the ratio $b_{ef}/b$ is approximately equal to 1 for small $b$, decreases until about 0.8 for intermediate $b$ and then returns to 1 as $b \to 1$. Such results corroborate the assertion that the input parameter $b$ controls the relative incidence of branches in the adapted model.

The effectiveness of the input parameter $b$ is evaluated by comparing it with the relative frequency of branches in a polymer configuration. For this purpose, we define the effective branching rate $b_{ef}$ as the ratio between $N_3$ and $N_2 + N_3$ averaged over all experiments:

$$b_{ef} = \left\langle \frac{N_3}{N_2 + N_3} \right\rangle$$

(4)

The plots of $b_{ef}$ versus $b$ at different values of $c$ for simulations of the BPGM and ABPGM (on a large $L = 1501$ square lattice) are shown in Figures 7a and 7b, respectively. The ideal behavior $b_{ef} = b$ is indicated as the dashed lines. Regarding the BPGM, there is evidently a large discrepancy between the input parameter $b$ and the output rate $b_{ef}$. For $c = 0$, $b_{ef}$ increases with $b$ up to a maximum (at $b \approx 0.2$) and then decreases to zero as $b \to 1$. This behavior is explained as follows: for small $b$, self-avoidance is reduced so that most sites trying to bifurcate succeed; but as $b$ increases, parallel linear chains (like those of Figure 1a) are forcibly generated due to both the increasing excluded volume and the clockwise update. For higher values of $c$, the presence of impurities hampers the formation of linear chains and thus $b_{ef}$ decreases slower (as a concave function). Anyway, for the BPGM, the discrepancy between $b$ and $b_{ef}$ gets more pronounced as $b \to 1$. On the contrary, for the ABPGM, $b_{ef}$ always increases with $b$ as a convex function (for any value of $c$) and the difference $b - b_{ef}$ is very small. In fact, for any $c$, the ratio $b_{ef}/b$ is approximately equal to 1 for small $b$, decreases until about 0.8 for intermediate $b$ and then returns to 1 as $b \to 1$. Such results corroborate the assertion that the input parameter $b$ controls the relative incidence of branches in the adapted model.

The branched polymer growth model (BPGM) was originally proposed [7] as a generalization of the kinetic
growth walk \cite{4} in order to include the possibility of ramification of the polymer as well as the presence of impurities in the medium. The model was found to exhibit a finite-infinite transition due to competition between branching and hindrances.

In this paper, we have proposed an alteration in the dynamics of the BPGM so as to adapt the model to an experimental realism. We have called it the adapted branched polymer growth model (ABPGM). The main difference between our proposal and the original model regards to the growth mechanism of a monomer which is chosen to bifurcate (with probability $b$) but has just one empty nearest neighbor site. In the BPGM, such a monomer is transformed into a bifunctional unit so that it grows linearly (with probability one). In our adapted model, that monomer stops and becomes a frustrated dead end. This frustration reveals as preferable and more realistic than changing the monomer functionality from 3 to 2 (as it occurs in the BPGM). This subtle change in the algorithm together with a random update of the growing ends lead to the formation of polymers with new topological patterns and adjusted degrees of branching. Indeed, we have shown that the effective branching rate $b_{ef}$ is very much closer from the input parameter $b$ in the ABPGM than in the original model.

We have found that the ABPGM presents a finite-infinite transition in the $(b, c)$ space with a peculiar reentrant phase in the small interval $0.175 \leq c \leq 0.195$. At this instance, we compare some typical graphs of both the BPGM and ABPGM at the fixed impurity concentration $c = 0.18$ for three increasing values of $b$: 0.2, 0.6 and 1 (Figures 8, 9 and 10, respectively). For the BPGM, all the three corresponding configurations are infinite clusters whose boundaries change from rough to faceted as $b$ increases. On the other hand, the typical ABPGM graph for $b = 0.2$ is a finite cluster (due to the occurrence of both trapped sites and frustrated dead ends); if $b = 0.6$ the cluster is infinite (since here higher branching overcomes dead ends) and if $b = 1$ the cluster is finite again (due to a high frustration rate).

We hope that the ideas presented here can add a new degree of physical understanding to the model as well as they can approximate it to the real ramified polymers.

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\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig9}
\caption{Typical graphs of both the BPGM (a) and ABPGM (b) for $b = 0.2$ and fixed $c = 0.18$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig10}
\caption{Typical graphs of both the BPGM (a) and ABPGM (b) for $b = 1$ and fixed $c = 0.18$.}
\end{figure}

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