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Fractal growth in uv-irradiated DNA: Evidence of nonuniversal diffusion limited aggregation

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Branched structures and fractals occur widely in nature (trees, corals, sand dunes, etc.) showing its preference for nonintegral dimensionality characteristic of nonequilibrium growth phenomena [B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, New York, 1982)]. Neural networks, blood platelets, some cancerous growth patterns, etc. are also known examples of fractals in physiological systems. Many theoretical models have been invoked to explain different fractal and ramified structures [T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989)]. The diffusion limited aggregation (DLA) of particles in a random walk that was computer simulated by Witten and Sander [T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981)] is a more universal mechanism. Recently, a nonuniversal DLA has been theoretically proposed [P. Ossadnik, C. Lam, and L. M. Sander, Phys. Rev. E **49**, 1788 (1994)] where particles in a random walk are not of the same size. This paper reports the significant observation that uv-photolyzed DNA in an alkaline solution aggregates, on drying, in a fractal-like structure. Furthermore, this observation provides experimental confirmation of nonuniversal diffusion limited aggregation [P. Ossadnik, C. Lam, and L. M. Sander, Phys. Rev. E **49**, 1788 (1994)].

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Fractal growth based on the mechanism of diffusion limited aggregation [1,2] involves a random walk of particles and their subsequent sticking. This mechanism has been tested experimentally by creating a wide variety of growth conditions (see, for example, Ref. [1]). In this context, electrodeposition experiments where the aggregating particles move under the influence of an electric field stimuli are quite important [4–6]. Recently, in our laboratory [7–9] we have simulated experimental conditions for the growth of large size ($\sim 2-3$ cm) diffusion limited aggregates and other ramified structures in the ion-conducting polymer electrolytes in which the random walkers are of uniform radii (like I^- or SCN^-). However, one can visualize situations in which the radii (r) of all the aggregating particles (walkers) are not the same. Ossadnik *et al.* [3] have assumed the following power law distribution $P(r)$ for the aggregating particles:

$$P(r)=\begin{cases}\mu/r^{\mu+1} & \text{for } r\geqslant 1\\ 0 & \text{for } r<1,\end{cases} \tag{1}$$

and have shown that the resulting pattern is almost of the diffusion limited aggregation (DLA) type but different from the classical Witten and Sander pattern [2], depending upon the statistics of the particle size distribution. DNA has been chosen by us for the growth of the above nonuniversal DLA, apart from the importance of DNA in physiological processes, motivated by the facts given below.

Under uv irradiation, DNA is known to undergo various alterations, including fragmentation into molecules of different sizes. There are many well documented spectroscopic and other studies on the photolyzation products of DNA [10]. DNA, being a large molecule, is less likely to show random-walk-related diffusion limited aggregation but its fragments may. The motivation of the present study was to see whether (a) the natural and/or uv-photolyzed DNA aggregates from aqueous media as random clusters or shows some well defined fractal patterns, and (b) in the latter case, does it show experimentally nonuniversal DLA, in view of the likely particle size distribution.

Calf thymus DNA (Sigma) was used in the present study. The DNA was dissolved in triply distilled water. Aqueous NaOH solution was added to obtain high alkaline $p\text{H}\sim 11$. A part of this alkaline solution was put in a quartz cuvette and irradiated for 2 h using a 150 W xenon arc lamp (Applied Photophysics model 4060A) placed in the sample position of a (Applied Photophysics model SP 70B) fluorescence spectrometer. The irradiation wavelength was 260 nm with a bandwidth of ~ 20 nm. The irradiated solution was then poured into a glass Petri dish. In another Petri dish, the same amount of unirradiated solution was poured. After 5–6 days, as the solution dried up, fractal growth (of size $\sim 1-2$ mm) was observed in the Petri dish with uv-irradiated DNA solution [Fig. 1(b)] but no such growth was seen in the Petri dish with unirradiated DNA solution [Fig. 1(a)].

The experiment was repeated several times with different batch solutions. Similar results were obtained in different experimental runs. Furthermore, it may be noted that the fractal pattern of Fig. 1(b) was not an isolated observation, in a particular run, but such patterns were copiously distributed over the entire Petri-dish base. One such picture, with a larger field of view, is shown in Fig. 1(c). The fractal dimensions of these patterns were evaluated by the standard procedure described in Ref. [1] and it was found that $D\sim 1.7\pm 0.1$.

An interesting aspect of the fractal patterns shown in Figs. 1(b) and 1(c) is the occurrence of occasional dark "blobs" on the arms of these patterns. A normal computer-simulated Witten and Sander type DLA has clean branches and such was also our observation in iodine fractals grown by us [7,8]

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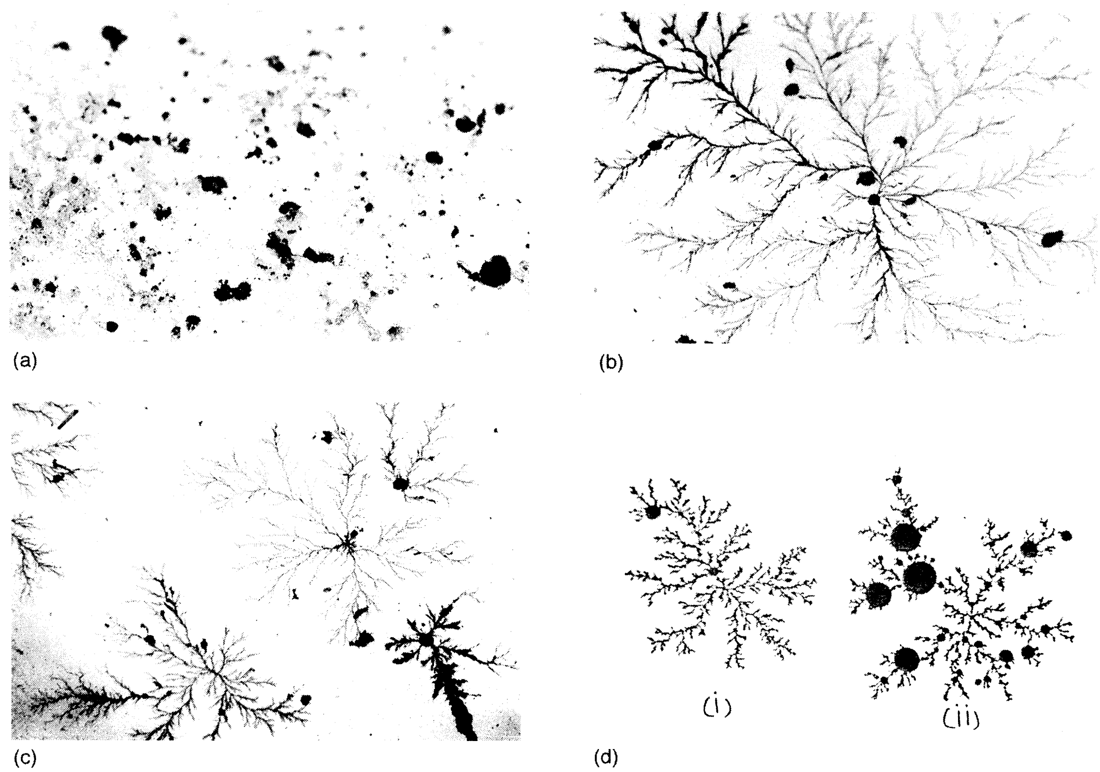

FIG. 1. Aggregation experimentally observed on drying of DNA solution (a) unirradiated; (b) uv-irradiated, magnification 100×, field of view 1 mm×0.75 mm; and (c) uv-irradiated, magnification 50×, field of view 2 mm×1.5 mm. (d) is the computer-simulated (Ref. [3]) nonuniversal diffusion limited aggregates: (i) small particles dominating the structure; (ii) transition point between the small particle domination and the large particle domination.

in ion-conducting polymers. We tried to avoid any unintentional dust particles falling and also doubly checked for the presence of any suspended impurity in the solution. Even after every precaution, the dark "blobs" on the fractal arms were always found to be present. So, these are real experimental observations. As mentioned earlier, Ossadnik *et al.* [3] studied the aggregation of particles with a size distribution, which would be the situation existing in the uv-photolyzed DNA. Two typical theoretical patterns from Ossadnik *et al.* [3] are reproduced in Fig. 1(d) for two relatively different dominations of particle size distribution. Our experimental observation given in Figs. 1(b) and 1(c) is astonishingly similar to the computer-simulated nonuniversal diffusion limited aggregate due to Ossadnik *et al.* [3].

From the above results, we conclude that uv-photolyzed DNA gives rise to nonuniversal diffusion limited aggregates or fractal patterns, which may have wide ranging consequences in radiation-damage-related phenomena in tissues. The above experiments also provide experimental confirmation of the nonuniversal diffusion limited aggregation model of Ossadnik *et al.* [3].

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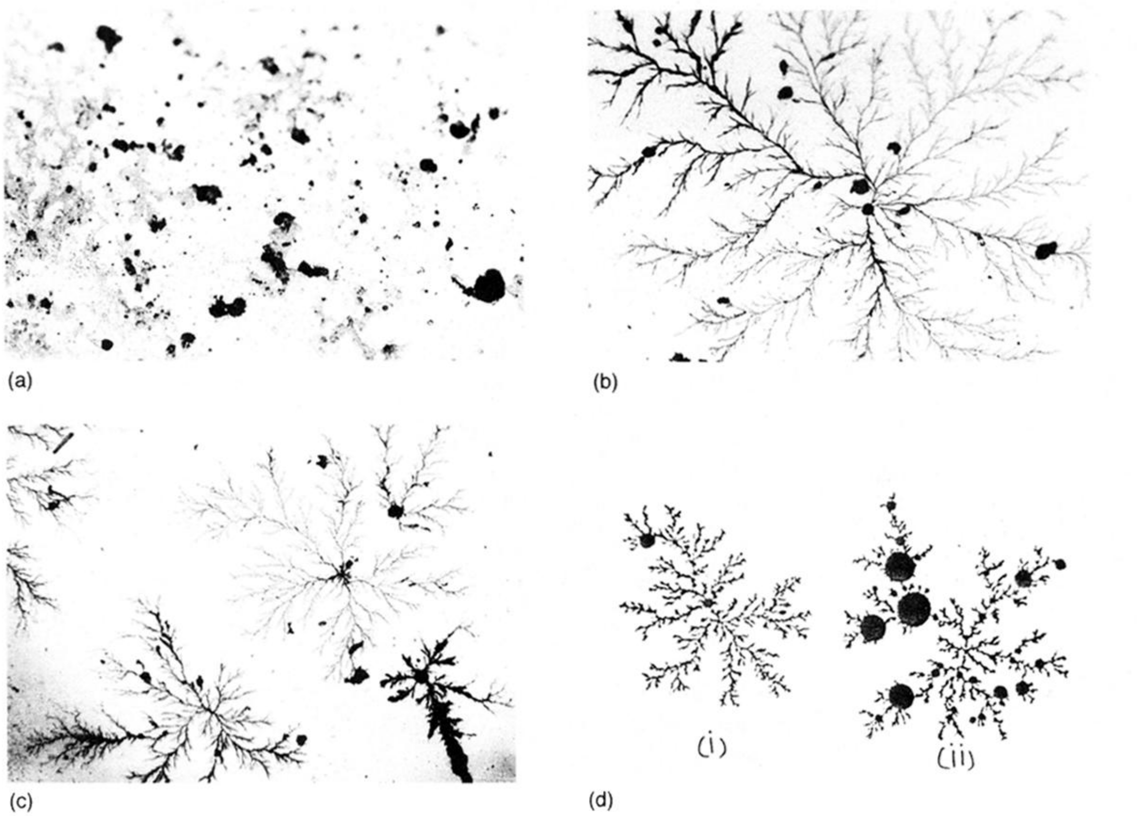

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