Ecosystem of clusters made of self-propelled droplet surfers

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Introduction

Active matter can exhibit inherently complex behavior based on chemical and/or physical interactions among constituents. Among many types of active matter, liquid droplet surfers have attracted attention for their stable yet complex self-propulsion [Nagai et al. (2005); Chen et al. (2009); Čejková et al. (2014); Löffler et al. (2018); Tanaka et al. (2021)]. It has also been reported that such surfers can express complex collective behavior reminiscent of colony-forming single-celled organisms or the self-ordering within multicellular organisms [Tanaka et al. (2017); Cejkova et al. (2019); Kim et al. (2021)].

Alkyl salicylate droplet surfers [Tanaka et al. (2015)] are an ideal model system to study such a hierarchical structure constructed by active components, since several tens of droplets can be stably placed on the surface of an aqueous surfactant solution, usually contained within a Petri dish. This system has been studied so far for its ability to form ring-shaped clusters which periodically appear and disappear for more than a day [Tanaka et al. (2017)].

In this preliminary analysis of such systems we add another layer to the hierarchy. When the number density of the droplets is at a certain level (i.e., a less crowded system than in previously reported examples), we observe the formation of smaller, isolated clusters that coexist with each other and increase in size and complexity over time. Interestingly, the types and shapes of clusters observed are the same over repeated experiments, resulting in a reproducible "ecosystem" of clusters. The hierarchy is built from active droplets, their clusters, and finally the ecosystem of clusters. Here we report on a first attempt at characterizing this system over long timescales. We particularly focus on characteristic open clusters, called "gliders" due to their resemblance to gliders in Conway's Game of Life [Gardner (1970)]. These clusters seem to play an important role in perpetuating the ecosystem.

Methods

Droplets were made from 80 wt% of ethyl salicylate and 20 wt% of liquid paraffin. A small amount (0.0025 wt%) of dye, oil red O, was added for visualization. In total, 60 droplets with a volume of 20 μl were placed on the surface of aqueous 10 g/L sodium dodecyl sulfate solution in a glass Petri dish of 140 mm in diameter at 25°C. Images of droplets were taken using a high-speed CMOS camera (Baumer VCXU-04C, Frauenfeld, Switzerland) as time-stamped images of 512 × 512 pixels. The frame rate was 30 Hz. The elapsed time, t, was measured from the moment when the glass cover was placed on the Petri dish. The position of droplets was extracted from the images using ImageJ [Schneider et al. (2012)]. The lifetime, τ, of a cluster was calculated as a period for which they maintain the same cluster, measured and averaged within each 600 s time span.

Results and discussion

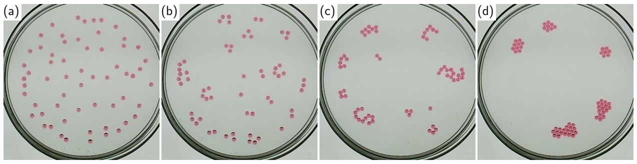

Figure 1: Snapshots of droplet cluster formation. (a) $t = 1$ h, (b) $t = 2$ h, (c) $t = 4$ h, and (d) $t = 19$ h. Diameter of the dish is 14 cm.

Figure 1 shows the evolution of clusters made of active droplets. At first droplets were moving around independently and randomly [Fig. 1(a)]. Then they started forming clusters [Fig. 1(b)]. The cluster size was increasing with time [Fig. 1(c)], and eventually they stopped moving [Fig. 1(d)]. The final stage of the evolution after ca. 20 h, was a single large crystalline cluster at rest.

We observe in general a relaxation process from a gas phase [Fig. 1(a)] to a solid phase [Fig. 1(d)]. This relaxation can also be seen in the lifetime τ of single droplets (monomers) [Fig. 2(a)], where τ decreased exponentially with t for at least the first 2 hours. However, the relaxation process was not a simple transition from gas to solid, as seen in Fig. 2 at $t = 2$ h and later, where several types of clusters coexisted with roughly the same lifetime. These clusters

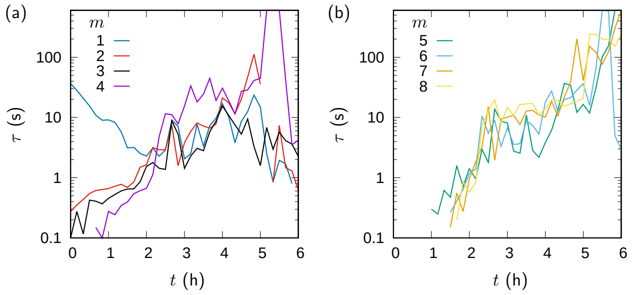

Figure 2: The lifetime τ of clusters of size m. (a) From monomers ($m = 1$) to tetramers ($m = 4$). (b) From pentamers ($m = 5$) to octamers ($m = 8$).

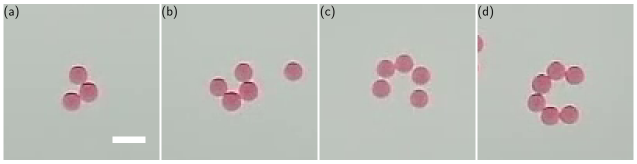

Figure 3: "Gliders": an open structure of clusters. The "gliders" move straight and much faster than other clusters. Each droplet is 20 μl in volume. The scale bar is 6 mm.

exchanged their components frequently with clusters disappearing to produce other clusters, exemplifying an ecosystem of clusters. Especially, both Fig. 2(a) and Fig. 2(b) show that there was a period ($t = [3, 5]$ h) when this ecosystem was stabilized as a dynamically stationary state. After this period, the lifetime of larger clusters increased towards the final equilibrium state.

The basic driving force of cluster formation is the attractive lateral capillary force acting among droplets on the surface [Tanaka et al. (2021); Kim et al. (2021)]. For $t \simeq [3, 5]$ h, clusters resist this attraction and small clusters coexist with larger ones because of their self-propulsion. A cluster collides with other clusters, breaks, and shuffles their components. As cluster size increased we observed that the overall translational velocity decreased. However, certain clusters were comparably active. They tend to have characteristic open, U-shaped structures as shown in Fig. 3, where clusters composed from chains of $3 - 6$ droplets are represented. They all moved in the direction of their convex "head" on a straight trajectory at higher speeds compared to other clusters.

Their motion continued until the U-shape was interfered with by other clusters or the the dish boundary. We call them "gliders" because of their resemblance to the gliders seen in the Game of Life [Gardner (1970)]. The formation of such "gliders" effectively maintained the cluster size distribution by their active randomization process, thus they were the key players to keep the dynamically stationary state of the ecosystem.

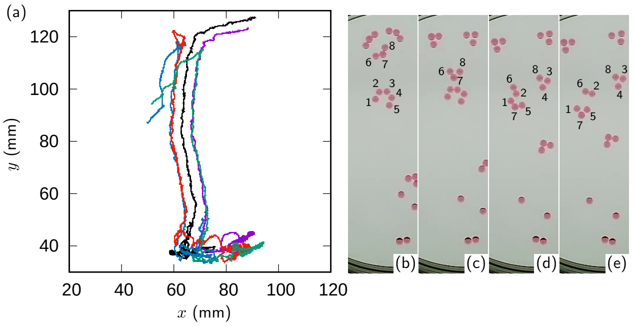

Figure 4: (a) A trajectory of droplets in a pentamer "glider" shown in Fig. 3(c). It traveled from bottom to top in the figure until it exchanged droplets with other structures. (b)-(e) Snapshots of the glider's collision. The numbers correspond to droplets in the pentamer (1–5) and in a trimer (6–8) that was also a glider. The intervals of the snapshots are, (b) 0 s, (c) 12 s, (d) 50 s, and (e) 60 s. Droplet volume: 20 μl

As an example of "gliders", Fig. 4 shows a trajectory of a pentamer moving from bottom to top. At first, these 5 droplets formed a random cluster. After some attempts, the pentamer rearranged into a U-shape and started translational motion. After traveling almost across the entire diameter of the Petri dish, it was stopped by collision with another "glider", in this case, a trimer. The collision reshuffled the components of clusters as shown in Fig. 4(b)–(e), and resulted in two trimers and a dimer. The "gliders" presented here have several properties similar to the gliders in the Game of Life. First, they need to form a specific shape to start moving. Second, the shape is easily destroyed by collisions. Third, there are many types of "gliders", mainly differing in their size. Finally, there is even something resembling glider guns in this system. When large clusters are formed, they tend to disintegrate soon and emit several small "gliders". More quantitative study is necessary to seek the origin of this resemblance.

Conclusions

As an ecosystem, clusters of droplets appear and disappear as they collide with each other driven by their self-propulsion. This dynamic overlays the general trend of decreasing individual driving forces over time. We found a peculiar type of cluster, named "gliders", that are especially effective in self-propulsion and reminiscent of gliders in the Game of Life. They have a characteristic open structure and move in a straight trajectory, thereby playing a significant role in the droplet exchange within the larger cluster ecosystem. To answer whether the resemblance between droplet clusters and gliders is just superficial, or whether there are deeper physical reasons for the similarity, rigorous modeling (for instance based on Kim et al. (2021)) will be necessary in future work.

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