Inferring the stability of concentrated emulsions from droplet configuration information

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Abstract When droplets are tightly packed in a 2D microchannel, coalescence of a pair of droplets can trigger an avalanche of coalescence events that propagate through the entire emulsion. This propagation is found to be stochastic, i.e., every coalescence event does not necessarily trigger another. To study how the local probabilistic propagation affects the dynamics of the avalanche, as a whole, a stochastic agent-based model is used. Taking as input, (i) how the droplets are packed (configuration) and (ii) a measure of local probabilistic propagation (experimentally derived; function of fluid and other system parameters), the model predicts the average size distribution of avalanches. In this article, we investigate how droplet configuration affects the avalanche dynamics. We find the mean size of these avalanches to depend non-trivially on how droplets are packed together. Large variations in the avalanche dynamics are observed when droplet packing are different, even when the other system properties (number of droplets, fluid properties, channel geometry, etc.) are kept constant. Bidisperse emulsions show less variation in the dynamics and they are surprisingly more stable than monodisperse emulsions. To get a systems-level understanding of how a given droplet-configuration either facilitates or impedes the propagation of an avalanche, we employ a graph-theoretic analysis, where emulsions are expressed as graphs. We find that the properties of the underlying graph, namely the mean degree and the algebraic connectivity, are well correlated with the observed avalanche dynamics. We exploit this dependence to derive a data-based model that predicts the mean avalanche sizes from the properties of the graph.

1 Introduction

Coalescence of a pair of droplets, in a concentrated emulsion flowing through a 2D microchannel, can trigger a cascade of similar events in their neighborhood. This gives rise to an avalanche that propagates through the emulsion [1, 2] (See Fig. 1 A, for snapshots of the propagating coalescence avalanche). Since the coalescence process depends sensitively on various parameters like the film thickness of the liquid phase between the droplets, instantaneous velocities, etc. which vary dynamically across the emulsion, the avalanche is observed to propagate stochastically. Bremond et. al. [1] even measured the probability associated with this local propagation as a function of the relative orientation of the droplets. Using this measure in a stochastic agent based model, we simulate the propagation of coalescence avalanches in concentrated 2D emulsions. We find that avalanches either propagate autocatalytically to destabilize the entire emulsion, or prematurely stop cascading leaving it relatively stable [3]. The avalanche dynamics depends on the size of the system, the packing aspect ratio, the local orientation of droplets [1], the number of avalanches triggered [4], and the fluid properties which in turn affect the overall propensity for coalescence propagation [3, 5].

In our previous investigations of the phenomenon, we studied coalescence propagation only on closely packed assemblies of monodisperse droplets (of the same size or volume). Note: often channel depths are smaller than the free droplet radius resulting in the droplet being squished between the top and bottom walls (like a ‘cylinder’). Hence, when droplet size is referred in terms of its radius \( R \), it refers to the observed circular radius of the cylindrical droplet. For these configurations, the total number of neighbors for each droplet was a constant except for those at the edge of the assembly. However, in real microfluidic applications, droplets self-organize to form different arrangements, which could be randomly close-packed, with differences in the neighbor configuration between droplets even in the bulk of the assembly. Depending on the application, droplets
may not always be monodisperse; i.e., they could be of different sizes. Polydisperse emulsions could significantly alter the neighborhood configuration of droplets. An important question then arises: how sensitive is the avalanche dynamics to the underlying droplet configuration, when material composition and all other properties are kept the same?

If the propagation is indeed sensitive to how the droplets are packed, it renders the stable operation of droplet-based devices non-intuitive. For instance, consider the dense flowing conditions in droplet-based incubators [6, 7]. Here, droplets reorganize dynamically during the flow as they move through the channel or are engineered to do so. Stability of the emulsions, i.e., the propensity to form large avalanches, becomes a function of time, making it hard to identify optimal operating conditions. Also, Bremond et al. [1] showed that cascades of coalescence events could result in the inversion of the droplet and continuous phases locally within the emulsion. They hypothesized that these phase inversion events had a higher chance of occurrence in polydisperse emulsions where propagation in small closed paths was more likely than in monodisperse emulsions: due to the anisotropic nature of the propagation.

Therefore, to understand these systems, to operate them stably, or to control the overall propagation of coalescence avalanches, we need to investigate the role of droplet configuration on the emulsion stability. In this study, we generate a variety of different droplet configurations and investigate the propagation of coalescence avalanches, using a stochastic agent-based model for coalescence propagation. Then, we bring graph theory to formally characterize the underlying droplet configuration—as a graph with nodes representing droplets and edges connecting droplets to their immediate neighbors—and investigate how the structural properties of the graph influences propagation. We then build a data-driven model that relates the topology of the droplet packing to emulsion stability.

2 Modeling the stochastic coalescence avalanches

Challenges with a first-principles approach
Coalescence of droplets is a multi-scale phenomenon [8, 9]. The continuous-phase film between two droplets has to drain and become thin enough to lead to its collapse that allows the droplet interfaces to make contact. The drainage process is rather complicated: a range of different interface configurations are formed as the thin film drains [10]. However, once these droplets touch, they form high curvature regions ($\gtrsim \frac{\pi}{1}$) that lead to large surface tension forces that pull the droplets together.

In the system we are interested in, droplets coalesce via a counter-intuitive mechanism: upon decompression. When droplets that are sufficiently close to each other get pulled away, a low-pressure region is formed in the interstitial region between the droplets, as the continuous phase fills in. This suction pressure pulls the interfaces together initiating contact between the interfaces [11]. Since these observations are reported in systems with large amounts of surfactant, there is reason to believe that coalescence is facilitated by the surfactant concentration gradients on the droplet interfaces. To completely resolve these structures and capture the different stages as droplets coalesce, one requires very-fine time ($\mathcal{O}(10^{-3})s$; [10]) and space ($\mathcal{O}(10^{-9})m$; [9]) resolutions in their simulations.

Furthermore, to simulate a coalescence avalanche, one has to capture, i) self-organization: the motion of droplets as they move through the microchannel, ii) nucleation events: coalescence between a pair of droplets which initiates a cascade of coalescence events, iii) dynamic processes in coalescence: interactions between dynamically growing coalesced clusters formed due to multiple coalescence events. These factors make any kind of first-principles approach to modeling coalescence avalanches computationally expensive, prohibiting the study at a system level.

Need for a simple model
Our goal in this study is to understand how propagation of avalanches depends on the way droplets are packed together. Hence, what we need is a model that will take the droplet configuration as input and simulate the stochastic propagation of an avalanche. The model should incorporate a measure of how coalescence events lead to newer events through the nearby droplets. For example, the probability associated with local propagation measured by Bremond and co-workers [1] can be incorporated into such a model. Also, since the avalanche propagates stochastically, it is important that we have a computationally simple model that allows us to generate independent realizations of the avalanche propagation (Monte Carlo study) to estimate the mean properties of the propagation phenomenon.

Stochastic agent-based model
We model coalescence propagation as a stochastic branching process on a group of $N$ droplets packed together in a tight configuration [3]; here, a branch emerges when two droplets coalesce and the branch grows (or propagates) stochastically via neighbors that are in close proximity to the recently coalesced droplets. The process continues till all the newly formed branches either stop propagating or there are no more droplets to coalesce. The droplets are assumed to be stationary during the entire propagation since, the speed associated with the propagation of the coalescence cascade ($\mathcal{O}(10^{-1})ms^{-1}$) is generally an order higher than the droplet movement speeds ($\mathcal{O}(10^{-1})ms^{-1}$). Propagation is carried out on a randomly packed droplet configuration that is assembled using the algorithm in [12, 13]. We used the code provided by the authors of [13] which can be found in [14], to produce dense monodisperse randomly packed droplet configurations (shown in Fig. 1 B). Note that the stochastic agent-based framework can be extended to account for the dynamic motion of the droplets and the coalesced clusters. However, in this article, we hold on to the simplifying assumption that droplets are static which aids
our current interest: to understand how the avalanche dynamics depends on the configuration, of how droplets are packed. For a detailed algorithm and implementation of the branching process, the readers are referred to [3].

**Local propagation rule** A pair of droplets is chosen randomly and allowed to coalesce. This initiates similar coalescence events in its neighborhood with a probability $\alpha \times G(\theta)$; here, $\theta$ is a measure of the local orientation of the droplets participating in the propagation and $\alpha$ is a parameter that varies with fluid properties such as viscosity, surface tension, etc. The form of $G(\theta)$ and the definition of $\theta$, are illustrated in Fig. 1 A ii. This measure was experimentally computed by ref [1] after analyzing over 2000 coalescence events in different parts of the 2D emulsion. The form resembles a cosine function ($G(\theta) = 0.3 \cos(1.6\theta) + 0.4$), which is the component of the pulling force experienced by a new droplet due to coalescence of a pair of droplets [3]. $G(\theta)$ favors propagation along the orientation of the coalescing pair ($\theta = 0$), which gives rise to avalanches that propagate as fingers through the 2D emulsion.

**Fluid properties and critical transitions** To understand how fluid properties affect the avalanche dynamics, one should explore the connections between the primary mechanism for coalescence between droplets (i.e., by decompression) and subsequent propagation, and the choice of fluids and operating conditions. It is known that coalescence by separation is prevalent when the interface mobility and the capillary numbers associated with the flow are low [15, 16]. And interface mobility is reduced when surfactant is added: due to the associated marangoni stresses. Most applications where droplets are packed in large numbers require addition of surfactant in sufficient quantities to stabilize the emulsions against coalescence due to compression [1, 6, 11]. Hence, as long as the viscosity ratio (droplet to continuous) is $> 0.1$ and the capillary numbers are within the range $< \mathcal{O}(10^{-2})$, which is typical in most microfluidic experiments, one can expect these systems to exhibit a propensity to coalescence upon decompression. While the presence of surfactants increases the chances associated with coalescence to propagation, it is important to note that increasing the wt% beyond ($> 10^2$ CMC) can reduce the propensity to propagate. Gunes et al [2] showed that the liquid bridges which are formed at the early stages of the propagation are destabilized at very high concentrations of the surfactant.

The exact choice of fluids will determine the overall propensity for propagation. We believe that this will linearly scale the local propagation probability, which is explained by the parameter $\alpha$. When $\alpha \approx 1$, every new coalescence event has the means to initiate more such events through their neighbors resulting in a cascade of coalescence events. $\alpha = 1$ corresponds to the conditions same as the experiments of Bremond et al [1]. When the capillary numbers associated with the system increase ($> \mathcal{O}(10^{-2})$) or when the viscosity ratio of the fluids used is lower than 0.1, the overall propensity to propagate reduces. We capture this by reducing the value of $\alpha$. When $\alpha$ is small ($< 0.7$), coalescence events do not propagate and the emulsion is stable.

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**Fig. 1** (A-i) - Snapshot from the experiments conducted by Bremond et al. [1] before the onset of a coalescence avalanche (courtesy: Nicholas Bremond, ESPCI France). Each droplet is around 22 $\mu$m in radius. (A-ii) - (In the box) Illustration of the local propensity for coalescence to propagate. When droplets 1 and 2 coalesce, a nearby droplet 3 coalesces with a probability based on its orientation $\theta$, as shown in the plot. (A-iii) to (A-v) - Images show the time lapse snapshots of the propagating avalanche. (B-i) to (B-iii) - Different droplet configurations which are input to the stochastic agent-based model along with the local propagation rule in (A-ii). (B-iv) to (B-vi) - Probability of an avalanche ($\mathbb{P}(A)$) as a function of its size $A$, computed via a Monte Carlo study of the stochastic model, for each of the input configurations (in (B i–iii)).
Therefore, a critical $\alpha_c$ exists which marks this transition from system-size spanning avalanches to a stable regime. Similar qualitative transitions based on surfactant concentration were reported by Baret and co-workers [5]. We find that the structure of the observed $G(\theta)$, which favors finger-like propagation events leads to a system size dependence of the critical transition $\alpha_c = f(N)$ [17].

**Monte-Carlo study** For every droplet configuration, we perform a Monte-Carlo study ($\sim 10^5$ simulations) of the stochastic agent-based model. Every run generates an independent realization of the stochastically propagating coalescence avalanche. From these independent runs, we compute the probability of occurrence of an avalanche $P(A)$, as a function of its size ($A$).

### 3 Results and discussion

**Stability of emulsions**

The structure of $P(A)$, the probability of an avalanche of size $A$, sheds light on the nature of propagation and the resultant stability of the emulsion. In our previous investigations [3, 4], where the propagation was studied on a hexagonally close packed arrangement of droplets, we observed $P(A)$ to have a non-monotonic shape (when $N \gtrsim 100$) with a maximum value at very small values of $A$ and a second peak at a large value of $A$ (red curves in Fig. 2i–iii). The second peak indicates that a significant fraction of the avalanches propagate through the entire emulsion, destabilizing it. One could call an emulsion ‘stable’ if the second peak can be avoided. One way to do this would be to make $\alpha < \alpha_c$, which reduces the propensity associated with local propagation giving rise to coalescence events that do not propagate. This requires changing the fluids used, the surfactant concentration, etc. Another way to reduce the second peak in $P(A)$, without having to change the fluid system, is by changing the aspect ratio of the droplet configuration. Arranging the droplets in a slender configuration increases the chance of the propagating front to encounter the boundary more often, which reduces the chances associated with a system-level propagation.

**Propagation depends on the packing**

When droplets of the same size are randomly packed, we observe significant variation in $P(A)$ across different droplet-configurations even when the parameters $\alpha$ and the aspect ratio of the droplet assembly are held constant (see thin lines in Fig. 2i–iii). The thick line (in red) corresponds to the monodisperse hexagonally close packed configuration (hcp) of droplets (same as the results in ref [3]).

When droplets are monodisperse and randomly packed, they exhibit the most variation about the hcp configuration, with few configurations even exhibiting a higher propagation than hcp. However, when droplets are bidisperse, we find both the variation in $P(A)$ between different configurations and their mean propensity for propagation of large avalanches (second peak height) to decrease with increasing bidispersity (see Fig. 2 ii, iii). A configuration’s level of bidispersity can be tuned by changing both the ratio of radii ($sr$) and the proportion of the two types of droplets ($nr$) (see Fig. 2 iv–vi, for average $A$ as functions of $sr$ and $nr$).

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**Fig. 2** Probability of an avalanche $P(A)$ as a function of its size $A$ plotted for: (i) - randomly packed monodisperse emulsion, (ii) - bidisperse emulsions with 30 : 70 composition of droplets with size ratio 1.1 : 1 and (iii) - bidisperse emulsions with 50 : 50 composition of droplets with size ratio 2 : 1. The thick red line in (i–iii) is the $P(A)$ corresponding to the hexagonally close packed configuration of monodisperse droplets (results from ref [3]); the thin black lines correspond to the highest and lowest mean avalanches in a given category; the thin gray lines show the $P(A)$ for the rest. All the simulations here correspond to $\alpha = 1$ and $N = 196$. (iv–vi) Box and whiskers plot showing the spread and the median of Average avalanche sizes for different size ratios and number ratios. The dotted line separates the monodisperse emulsions $sr = 1$ from the bidisperse $sr > 1$.
Emulsions as graphs
When a pair of droplets coalesce, they lead to more such events through their neighborhood. Hence, the number of neighbors available in their immediate vicinity and the angles they make with the recently coalesced pair determine the probabilities associated with propagation. To understand the sustained propagation of an avalanche, one has to not just investigate the nearest neighbors, but also their neighbors, and so on. In other words, a system-level characterization of the droplet configuration is essential to understand why a certain droplet-packing either favors or hinders the propagation of an avalanche.

Tools from graph theory offer a convenient formalism to analyze such system-level aspects of droplet configurations. A concentrated emulsion can be thought of as a graph where the nodes correspond to the droplets and an edge connects every pair of nearby droplets through which coalescence can propagate (see inset of Fig. 3). In this context, the question of interest takes the form: Can one understand the avalanche propagation dynamics from the topology of the underlying graph?

Coalescence avalanches can be interpreted as a cascade on the graph. If \( x_i[t] \) is the probability that a node \( i \) is ‘coalesced’ at time \( t \), then one can write an approximation of the coalescence propagation phenomenon as follows:

\[
x_i[t] = 1 - \prod_{j \in N_i} \left( 1 - w_{ji} x_j[t-1] \right).
\]

The RHS of Eq. 1 quantifies the probability associated with propagation from any of the possible neighbors of \( i \). Here, \( w_{ji} \) is the probability that coalescence will propagate from \( j \) to \( i \); \( N_i \) refers to the neighborhood of \( i \). \( w_{ji} \) is non-zero only when there is an edge connecting \( j \) and \( i \). This equation is similar in spirit to the time-evolution equation of a discrete-state, discrete-time Markov chain [18], where \( x_i[t] \) is the probability of the Markov chain in state \( i \). If \( x = [x_i]_{i=1}^N \) is the vector of all \( x_i \), and \( W = [w_{ij}]_{i,j=1}^N \) is the propagation matrix of the Markov chain, then the propagation equation of the Markov chain can be written as \( x[t+1] = Wx[t] \). However, in the case of coalescence propagation, crucial properties such as the row-stochastic nature of \( W \), and the fact that \( \sum_i x_i = 1 \), do not hold—hence, we have a slightly more involved propagation equation as given above.

A propagating coalescence avalanche could reach \( i \) from \( j \) through any of its neighbors \( k \). Hence, we define \( w_{ji} \) as,

\[
w_{ji} = \begin{cases} 
1 - \prod_{k \in N_j} [1 - p(k, j, i)] & A_{ji} = 1 \\
0 & \text{otherwise}.
\end{cases}
\]

\( p(k, j, i) \) is the probability that a coalescence event between droplets \( k \) and \( j \) results in the coalescence of droplet \( i \). This is identical to the local probability rule, derived from observations of Bremond et al. [1] which is used in the stochastic agent based model (see Fig. 1 A). Since, the probability for propagation from \( j \) to \( i \) depends on the neighbors of \( j \), we expect \( w_{ji} \neq w_{ij} \).

One can immediately see, from the model according to eq 1, that the propagation depends on how the droplets are packed. This is summarized in \( W \), which can be thought of as a weighted adjacency matrix of the underlying graph with elements \( w_{ij} \).

Mean degree explains observed avalanches
A simple way to characterize a droplet configuration is via the degree distribution of the underlying graph [19]. The degree of a node is equal to the number of nearby neighbors of the corresponding droplet, weighted by the probability that coalescence will propagate from that node. When computed for a weighted graph \( W \), this corresponds to the net number of neighbors through which coalescence can effectively propagate. Randomly packing a small number of droplets could lead to variations in the degree distributions between individual configurations that can in turn affect the propagation of avalanches. This is the reason for the variation in the \( P(A) \) curves observed in Fig. 2 i–iii, when droplets are randomly packed.

It is intuitive to expect the configurations with lower avalanche sizes to have smaller mean degree \( d_g \) (averaged over all the droplets in a given configuration). We observe this to be generally true across different configurations, even when they are bidisperse. We observe the
mean degree, when scaled by the system parameter $\alpha$ ($\tilde{d}_g = \alpha \times d_g$), to increase monotonically with the mean avalanche size $A$. We find all the data from our simulations—different configurations for both mono and bidisperse cases, different system sizes, and local propagation propensities $\alpha$—collapse on to a master curve, suggesting a universal relationship between the structure of the graph (defined based on the average degree of a configuration) and the mean avalanche size (averaged over many independent realizations) (see Fig. 3). We build a relationship between $\tilde{d}_g$ and $A$, from this data.

$$\tilde{d}_g = c_1 A + c_2 \left(1 - e^{-c_3 A}\right). \quad (3)$$

We regress a linear model with a correction term (for small $A$), as shown in Eq 3, to account for the phase transition (from autocatalytic to non-autocatalytic propagation) that corresponds with the small avalanche sizes. We find this relationship to hold true for different values of system size $N$, different propensities for propagation $\alpha$ and varying levels of bidispersity defined by the parameters $sr$ and $nr$—yielding a universal relationship.

**Exceptions to the rule** The data-driven model explains the dependence of the mean avalanche size to the packing characteristic $\tilde{d}_g$ accurately for most cases. However, it does not explain how some of the droplet configurations, corresponding to the randomly packed monodisperse emulsions, facilitate avalanches larger than that by the hexagonally packed configuration (hcp) (Fig. 2i and 4i). This is puzzling, since a hcp configuration has the highest possible $d_g$ for a system of a given size. While $d_g$ is small for these configurations with larger $A$, we find them to have different degree distribution. The nodes with 6 neighbors are not as high as in the hcp; however, they have more number of nodes with 5 and 4 neighbors. It is reasonable to expect that this difference in distribution is the cause for the better ‘flow’ that promotes a growing avalanche.

To understand how a droplet configuration as a whole affects the propagation of the avalanche, we look at the conductance of the graph, which is a measure that can be used to quantify how well-connected a graph is [20]. For instance, if a graph has bottlenecks, where there are only a small number of edges across the bottleneck, the conductance will be low. In the context of coalescence avalanches, presence of bottlenecks would decrease the propensity of the system to exhibit large avalanches. Unfortunately, graph conductance as it is formally defined is algorithmically difficult to estimate [21]. Hence, we use algebraic connectivity $C$, i.e., the second smallest eigenvalue of the Laplacian matrix of the graph, which is a proxy for the conductance [20, 22]. Conductance also has interpretations in terms of the diffusion time for diffusion processes on graphs, *i.e* random walks. Since coalescence propagation is a stochastic process, we believe that conductance of a graph (measured as algebraic connectivity) would yield a single measure for the entire graph that can explain the large mean avalanche sizes observed.

**Algebraic connectivity explains the exceptions** In Fig. 4, we plot both the mean degree $d_g$ and the algebraic connectivity $C$, for monodisperse randomly packed configurations, as a function of $A$, the mean avalanche size. We find that algebraic connectivity successfully relates the topology of the graph to the propensity for overall propagation (or flow) of the avalanches: $C$ is found to increase linearly with $A$, similar to $d_g$. Further, algebraic connectivity also explains why some randomly packed configurations facilitate larger avalanches than hcp. In Fig. 4i, hcp configuration (marked as ‘X’) has a higher $d_g$ in comparison to all the randomly packed configurations. However, the configurations that exhibit higher avalanche sizes than a hcp are also found to have greater algebraic connectivity values (see Fig. 4ii).

To understand how the algebraic connectivity, as a measure, works in successfully estimating a slightly lower flow in hcp, we examine its graph connectivity in a little more detail. In the bulk of a hcp configuration, all droplets have the maximum possible number of neighbors, (*i.e.*, 6). Hence, there are no ‘bottlenecks’ in the bulk that could reduce the overall flow in a hcp. However, the droplets near the boundary of the assembly have relatively low degrees (usually 4 or lower), while those for the randomly packed configurations have a wider degree distribution with a considerable number of droplets with a degree of 5.
Now, consider those avalanches that are triggered in the bulk, which propagate towards the boundary. These can become system-spanning only if the boundary is well connected to the bulk. Therefore, these avalanches have a higher chance of propagating further, in randomly packed configurations which have a net higher mean degree near the boundary than a hcp—a potential reason for the observed higher avalanche probabilities than hcp. Algebraic connectivity, being able to characterize the overall flow in the graph, is able to capture this phenomenon well.

Algebraic connectivity $\mathcal{C}$ could have been used as a regressor in the data-driven model (Eq. 3) instead of the scaled mean degree $\bar{d}_g$. However in our investigations, we find that using $\bar{d}_g$ achieves a more accurate fit to observed average avalanche size $A$ than using $\mathcal{C}$; compare the spread associated with the two metrics for a fixed $A$ in Fig. 4 i and ii. While $\mathcal{C}$ explains the exceptions well, it is always affected by the existence of regions on the graph that contribute to smaller ‘flow’—since $\mathcal{C}$ is a global measure. However, $\bar{d}_g$ being an average local measure of the graph topology, is not affected by these regions and serves as a better regressor in Eq. 3. Including both the metrics as regressors does not change the accuracy in explaining the data appreciably.

### 4 Conclusion

In this article, we show how the propensity for coalescence avalanches to propagate in a concentrated emulsion is directly tied to how the droplets are packed. By expressing the droplet configurations as graphs, we are able to employ graph theory formalism to study how the topology of droplet packing can be used to predict the size of coalescence avalanches that these configurations can facilitate. We show the existence of a universal relationship between the scaled mean degree of the graph and the mean avalanche size, which allows us to build a data-driven model that can be used for predicting the avalanche size for a given droplet configuration. Since, the input to the data-driven model is simply how the droplets are packed, this model has the potential to be deployed online, in a real droplet microfluidic setup, to predict the stability of packing as droplets self-organize in a 2D microfluidic channel. We find that there are exceptions to the data-driven model which is based on the mean degree of a graph—which is only an average local property of the graph. These are explained by algebraic connectivity, a proxy for graph-conductance, which takes into account the system-level characteristics of a graph that are linked to a successful propagation of avalanches.

Also, we relate the topology of the droplet configuration via metrics that characterizes its structure to the propagation dynamics. Hence, we expect our approach to be agnostic to the specific choice of the coalescence propagation model used in the study. Though specific details of the hydrodynamics are not considered explicitly, (i) our approach does takes into account the local propagation propensity (estimated from experiments) and extended to systems with different fluid properties and ii) the graph essentially represents the droplet-interaction network where every edge connects every pair of droplets that are within their sphere of hydrodynamic influence. Hence, even the propagation dynamics predicted by a more detailed model—that considers the motion of droplets, thin-film thickness distribution and its effect on triggering a coalescence event, etc.—would still depend on how droplets are packed together and how this configuration gives rise to the propagation of the coalescence avalanches; which is captured by a graphical representation of the droplet ensembles.

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### Data availability statement

No data are associated in the manuscript.

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