

Reply to Reviewers

First, we would like to thank the anonymous referees for their comments that will improve the quality of our paper. Our revised version will include several of their suggestions.

Anonymous referee # 1

1. A system with five initial particles is indeed very small (Fig 2). I assume this result is shown for the purpose of demonstrating the method, it is confusing, however, because the system is too small to be of relevance.

Yes, as was remarked by the reviewer, this result (with only 5 particles) is shown for the purpose of demonstrating the method. As the system in this case has only 6 possible configurations, it is much easier to explain the details of the algorithm. In the revised version it will be emphasized that the objective of this section is to demonstrate the method.

2. The Tanaka/Nakazawa equation, which the authors use to calculate the gel mass, is not approximate. It requires, however, a trial-and-error procedure to determine the correct value of i_1 in Eqs 14 and 15. This is because Tanaka/Nakazawa depends on the mean distribution to obtain the gel fraction. If one has access to individual distributions, as the authors have, there is a simpler method: calculate the maximum mass in each individual distribution and obtain their average by weighting each value by the probability of the distribution.

As a matter of fact, we follow a Monte Carlo version of the procedure detailed by the referee, in order to calculate the expected value of the gel mass from Monte Carlo simulations (see equation 16). In this case we have access to the empirical largest droplet distribution at each time. For a fixed time, this distribution is obtained by calculating the mass of the largest droplet in each realization of the Monte algorithm (the sample length is the number of realizations of the stochastic algorithm). The expected mass of the gel is the ensemble mean over all the realizations of the Monte Carlo process (See Eq. 16).

Within the master equation approach, we have access to the joint probability distribution of the random variables $n_1, n_2, n_3, \dots, n_N$, but in order to calculate the average for the largest droplet mass, we would need to calculate the distribution of the maxima. Then, the distribution of the largest droplet (before the sol-gel transition) would be a Gumbel distribution. In the current state of our model, further work is needed in order to calculate the largest droplet distribution.

3. Finite size is not necessary for gelation to occur. The solutions converge when the size of the system becomes large, though these solutions cannot be obtained by traditional KCE and instead require the methods of Lushnikov (2004) or Matsoukas (2015). This means that finite systems, even with as few particles as 40, may be adequately described by these solutions without the need for a detailed (and costly) solution of the master equation.

Yes, we agree with the reviewer that these results may be adequately described without the need for a detailed (and costly) solution of the master equation. However, the analytical techniques developed by Lushnikov (2004), and Matsoukas (2015) work only for a special type of kernels (in that case, when the probability of collision is proportional to the product of the masses). Our method (although it can be computationally expensive) works for any type of kernels.

4. The complete phase space of a population with $N=40$ monomers contains 37338 states. Do the authors solve the Master equation for all of these? Some details need to be offered regarding this calculation.

Yes, we must solve the master equation for the 37338 states. For each state we have to solve the finite difference equation (5), or an equation similar to Eq. (6), but with 40-dimensional state vectors instead of 5-dimensional ones, and of course, a much more complex r.h.s. As suggested by the reviewer, our revised version will include some details of the calculations for this case.