

## Supporting information

### Population-balance description of shear-induced clustering, gelation and suspension viscosity in sheared DLVO colloids

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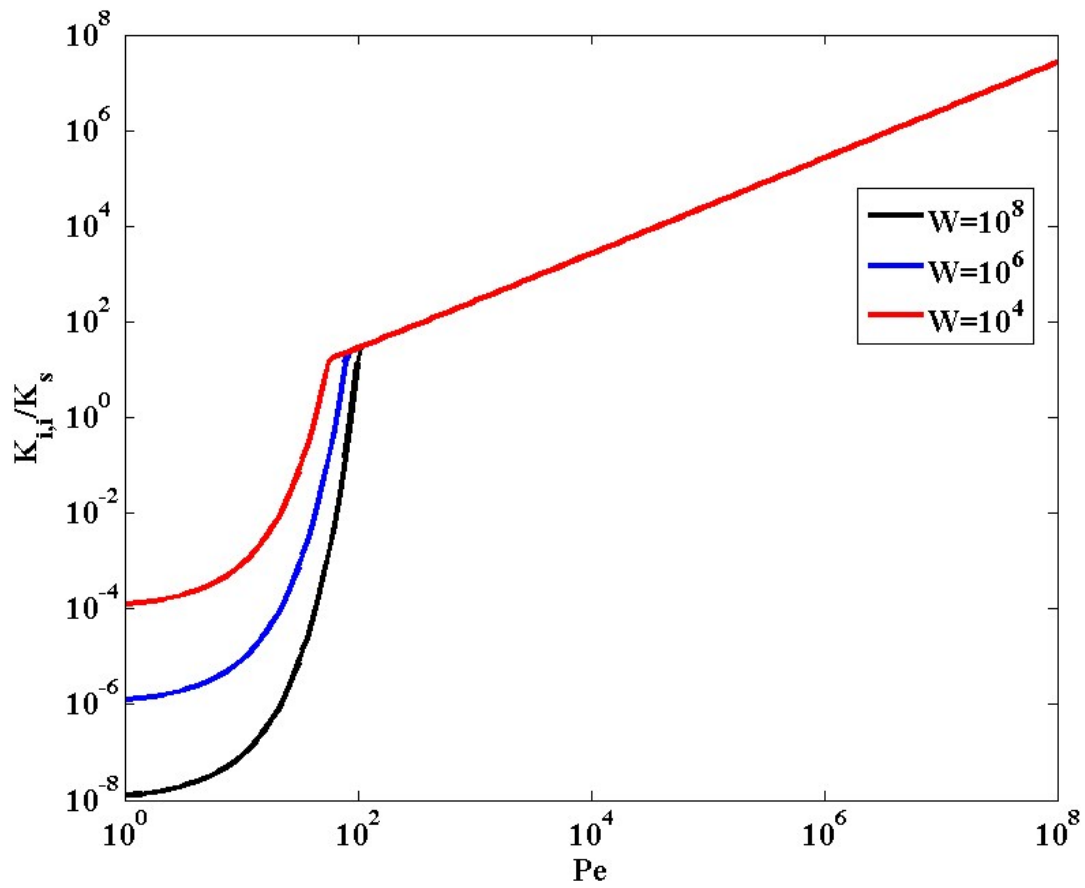
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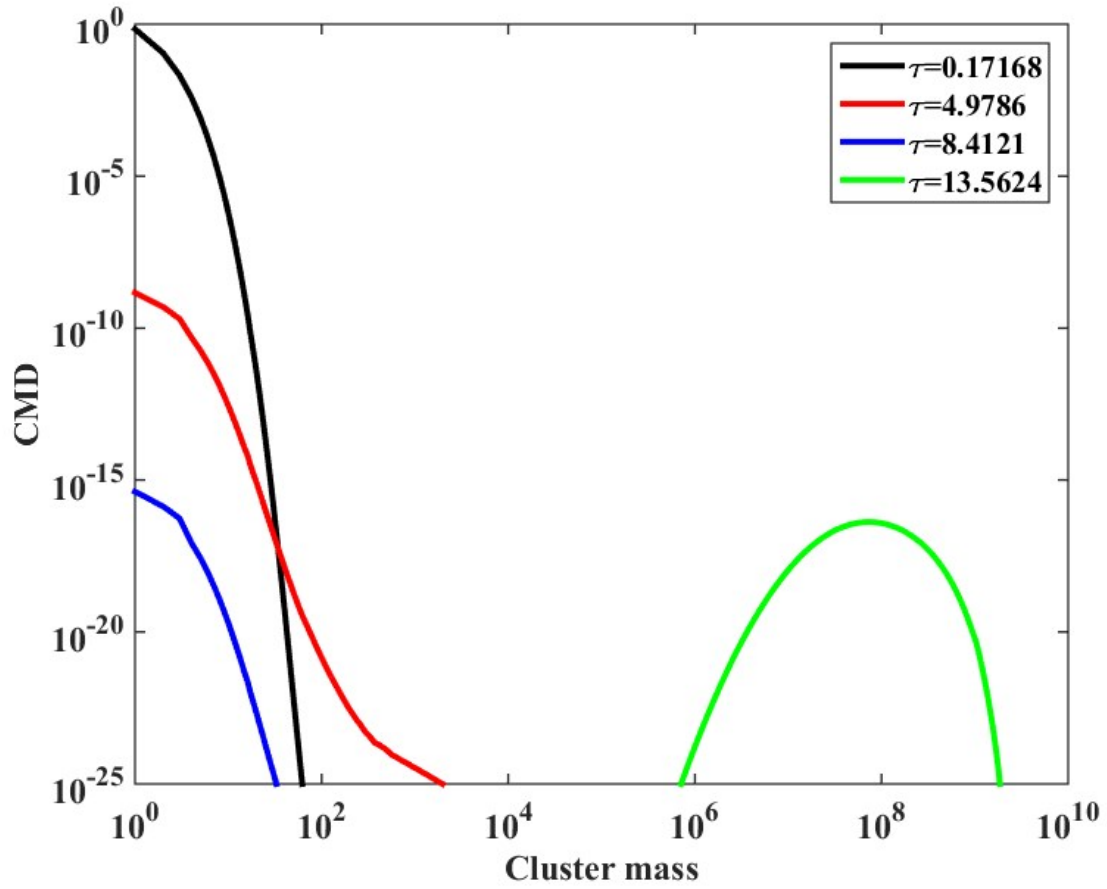
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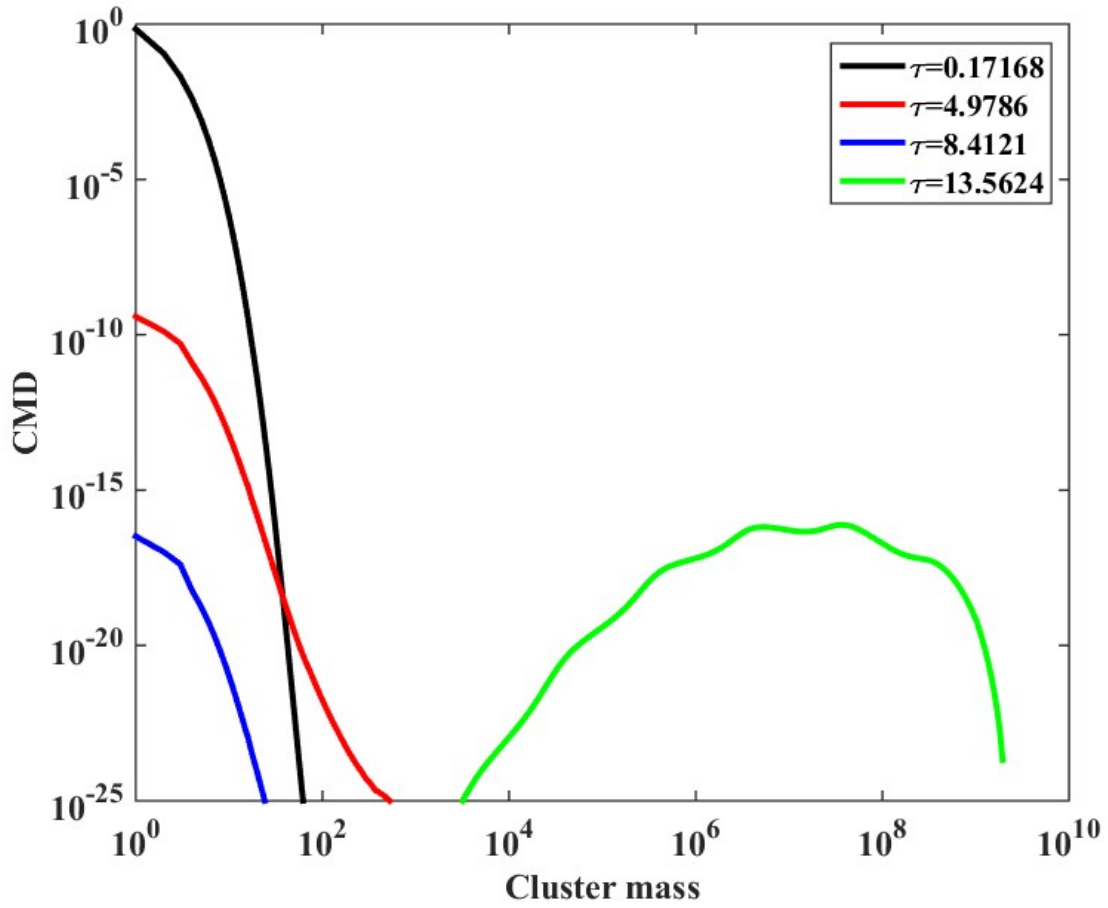
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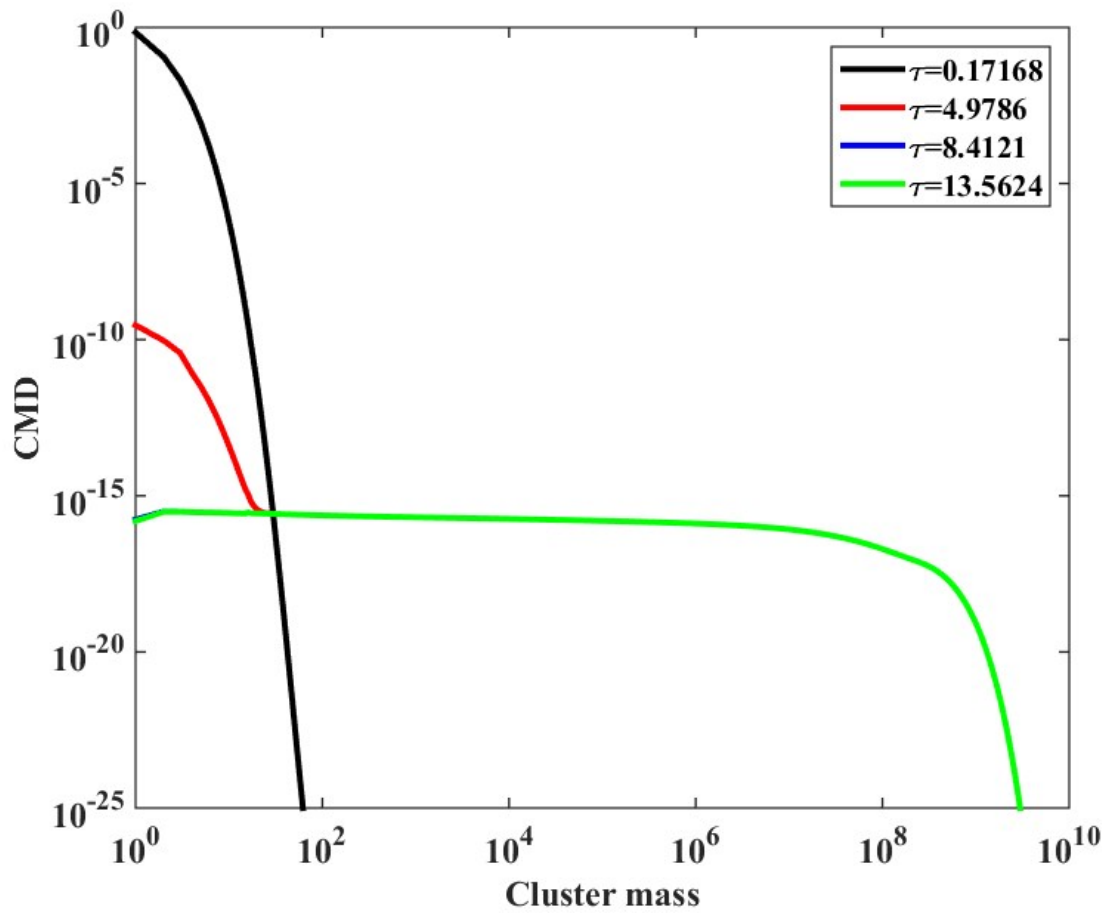
**Figure S1** Normalized rate of aggregation for equal size clusters as a function of  $Pe$  number, according to Equation (2) in the main text, for the three different Fuchs stability ratio values  $W$  indicated in the legend. Calculations have been performed for a particle diameter equal to 120 nm, particle volume fraction equal to 19% and shear rate equal to  $1200 \text{ s}^{-1}$ .  $K_s=8kT/3\eta W$ .



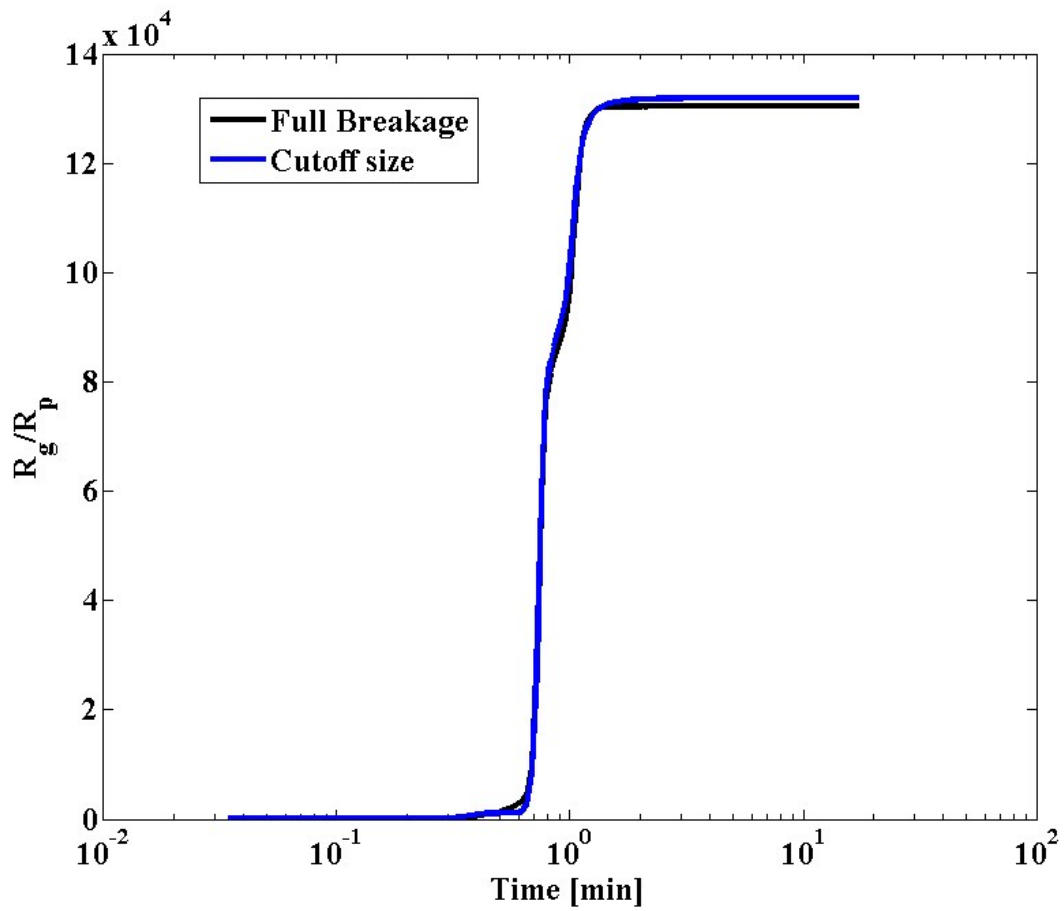
**Figure S2** Cluster mass distribution as a function of the cluster mass (expressed as the number of primary particles), for four dimensionless times indicated in the legend, when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text. The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ . The breakage Kernel is expressed by Equation (6) in the main text, with  $c_f=2.38 \cdot 10^{-10}$ . Symmetric binary breakage.



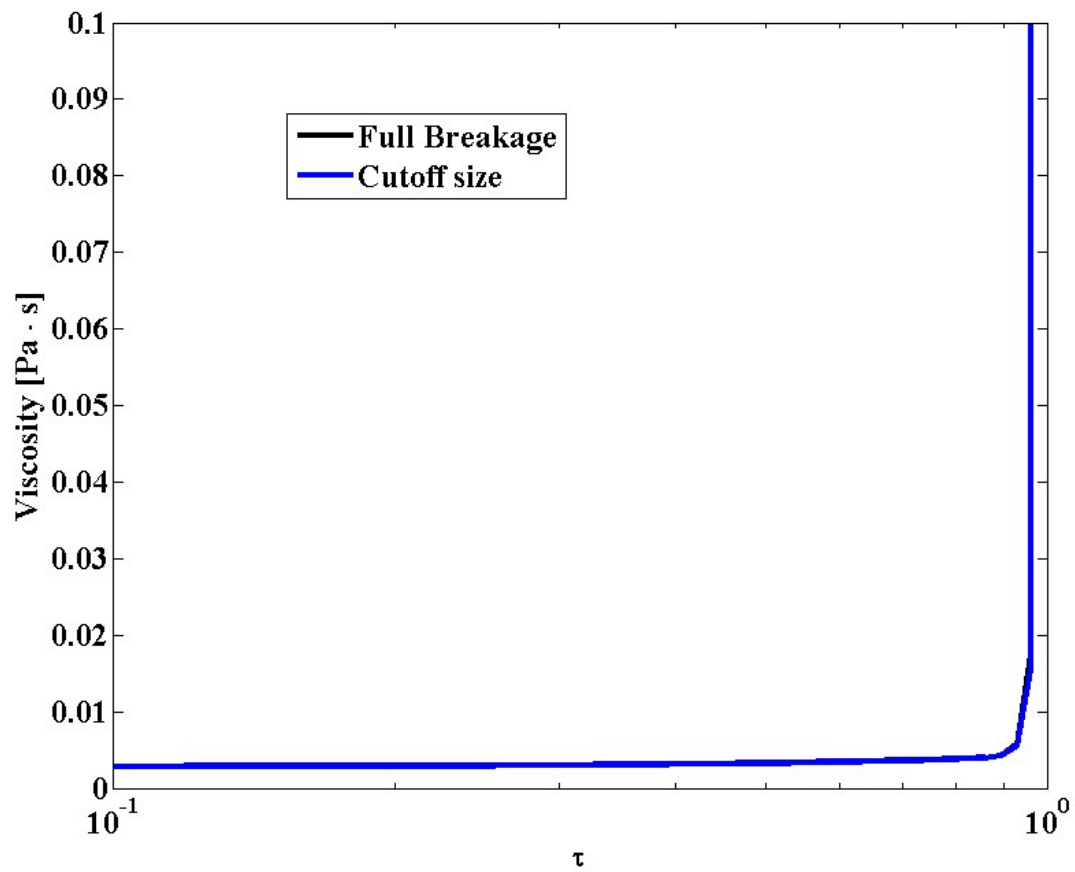
**Figure S3** Cluster mass distribution as a function of the cluster mass (expressed as the number of primary particles), for four dimensionless times indicated in the legend, when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text. The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ . The breakage Kernel is expressed by Equation (6) in the main text, with  $c_l=2.38 \cdot 10^{-10}$ . Asymmetric binary breakage.



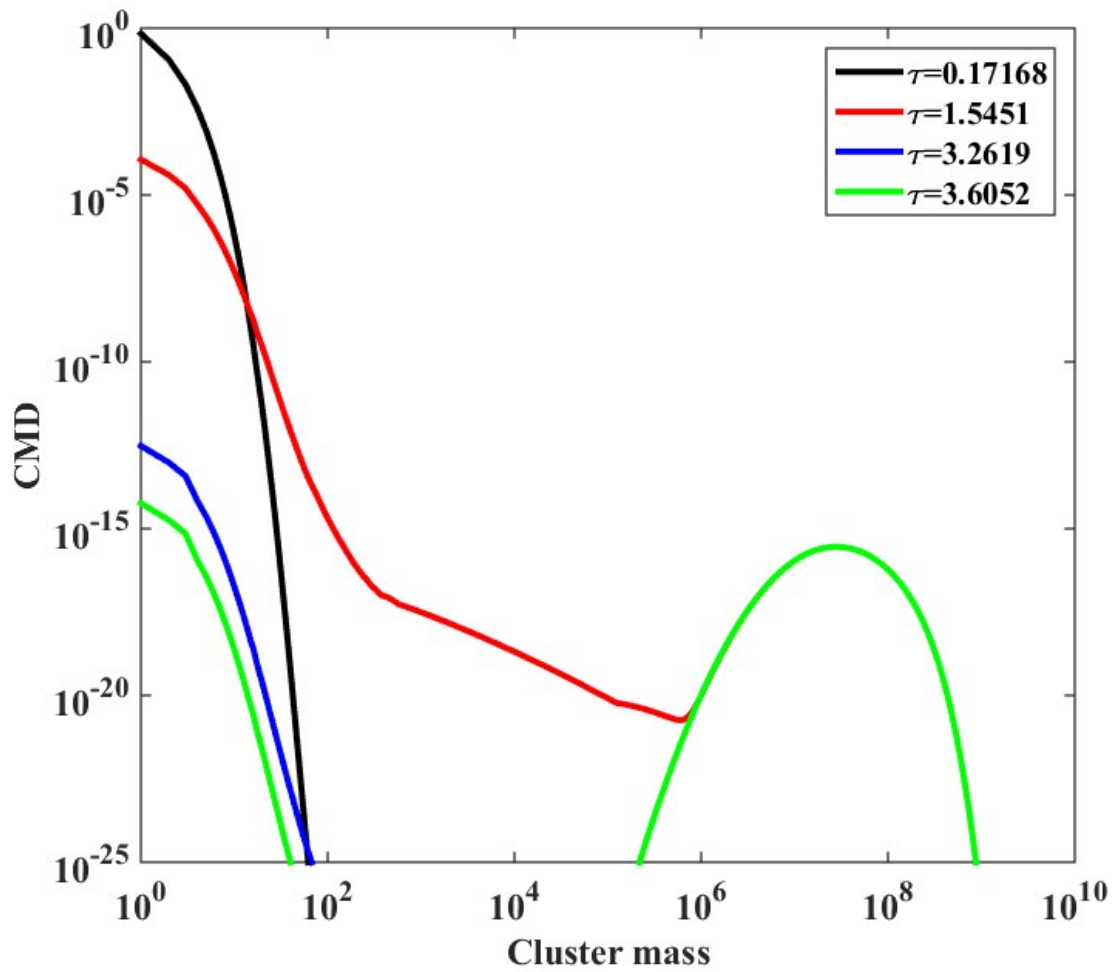
**Figure S4** Cluster mass distribution as a function of the cluster mass (expressed as the number of primary particles), for four dimensionless times indicated in the legend, when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text. The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ . The breakage Kernel is expressed by Equation (6) in the main text, with  $c_1=2.38 \cdot 10^{-10}$ . Broad fragment mass distribution.



**Figure S5a** Dimensionless radius of gyration evolution as a function of time, for two different situation: breakage and aggregation fully active on all sizes, and breakage Kernel expressed by Equation (6) in the main text, with  $c_f=2.38 \cdot 10^{-10}$ , and the presence of a cutoff at a given size ( $R_g^{\text{Max}}=6.2 \cdot 10^4$  nm). The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ , when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text.

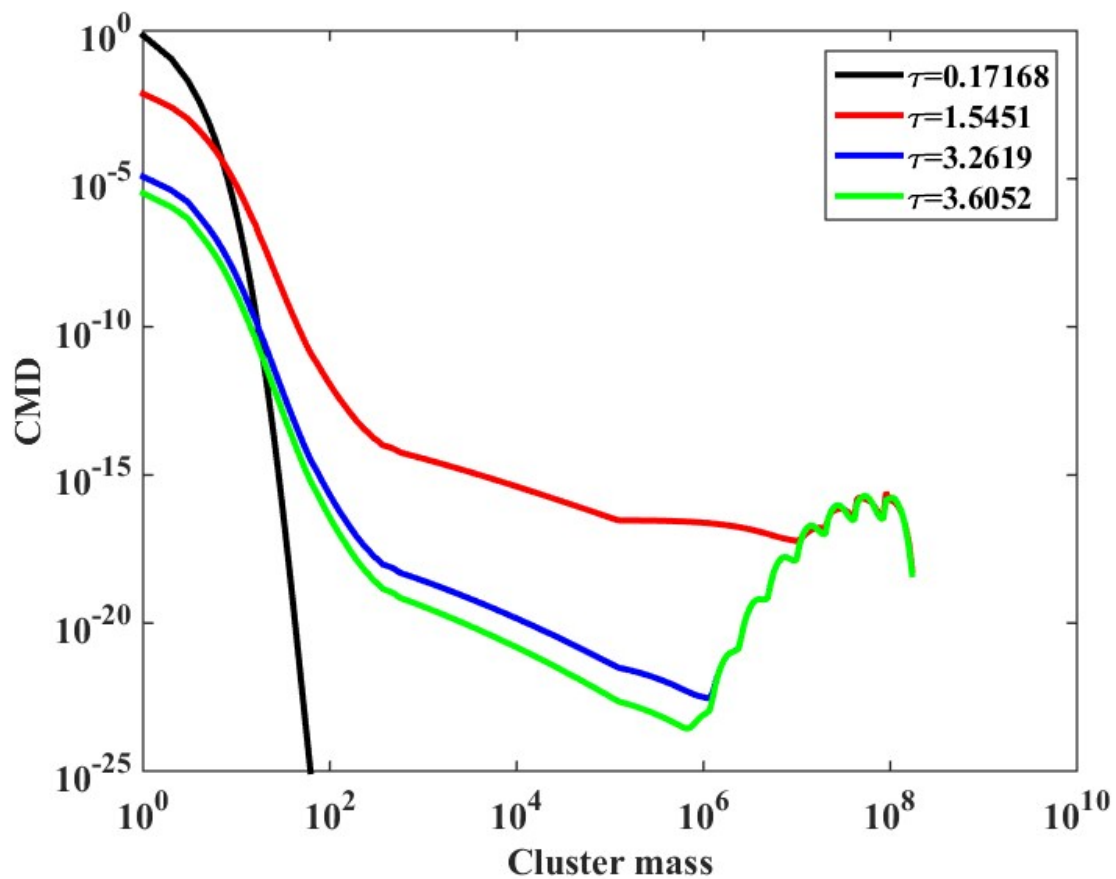


**Figure S5b** Viscosity profiles as a function of time, for two different situation: breakage and aggregation fully active on all sizes, and the presence of a cutoff at a given size ( $R_g^{\text{Max}}=6.2 \cdot 10^4$  nm). The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ , and breakage Kernel expressed by Equation (6) in the main text, with  $c_l=2.38 \cdot 10^{-10}$ , when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text.

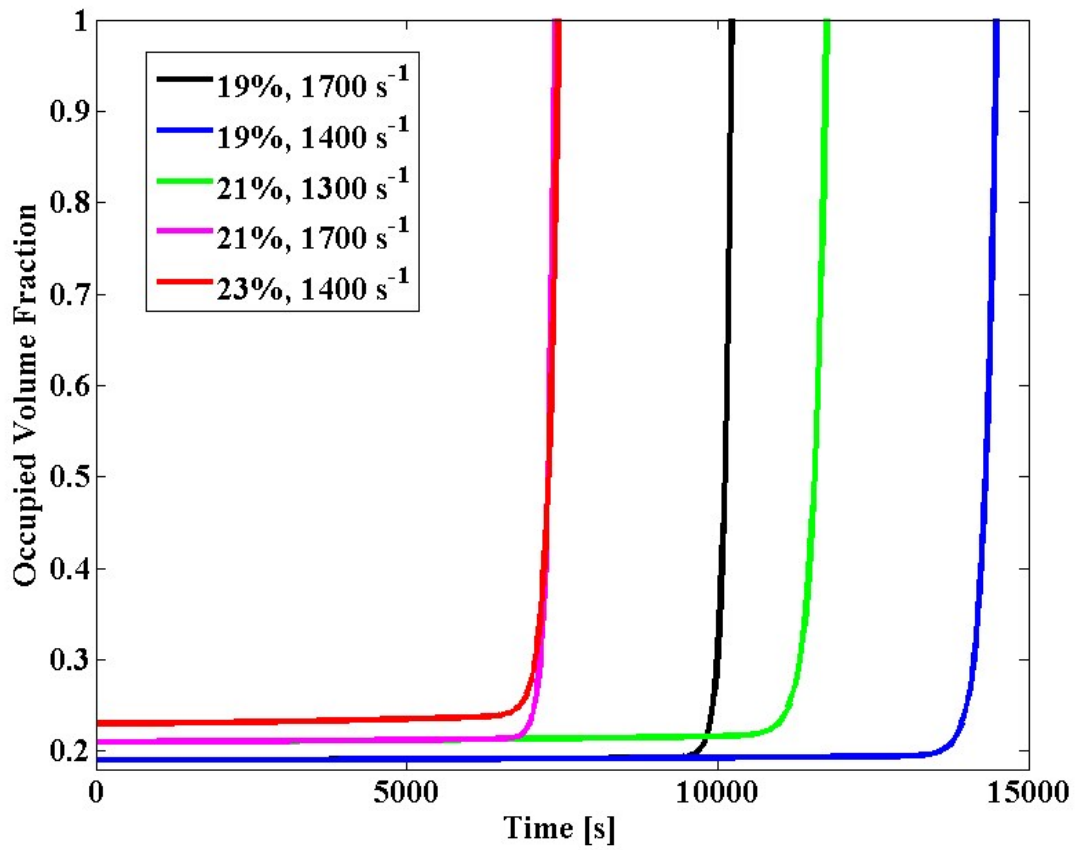


**Figure S5c** Cluster mass distributions as a function of cluster mass, for a few dimensionless time values, as shown in the legend. In this case aggregation is fully active on all sizes, in the case where the aggregation Kernel is given by Equation (2) of the main text. The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ , and breakage Kernel expressed by Equation (6) in the main text, with  $c_I=2.38 \cdot 10^{-10}$ .

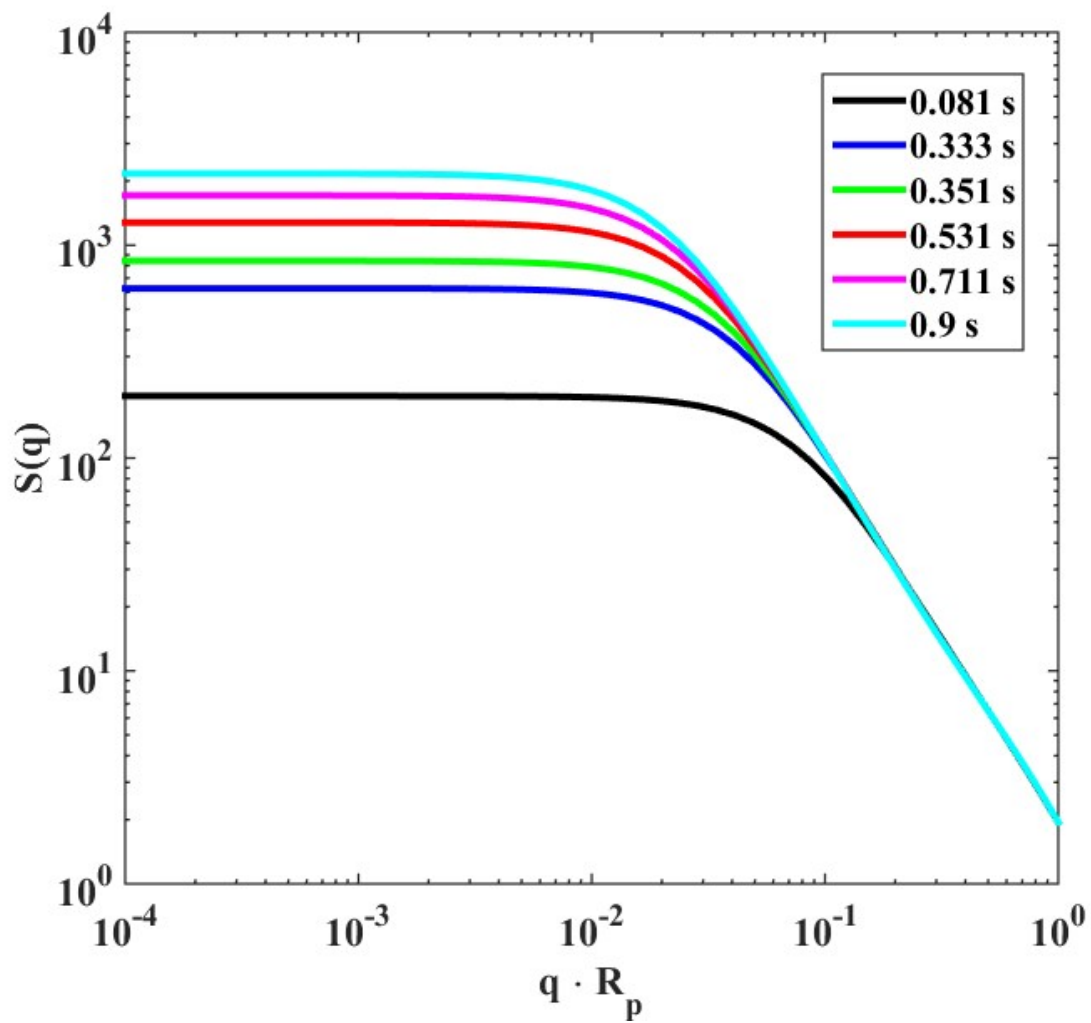




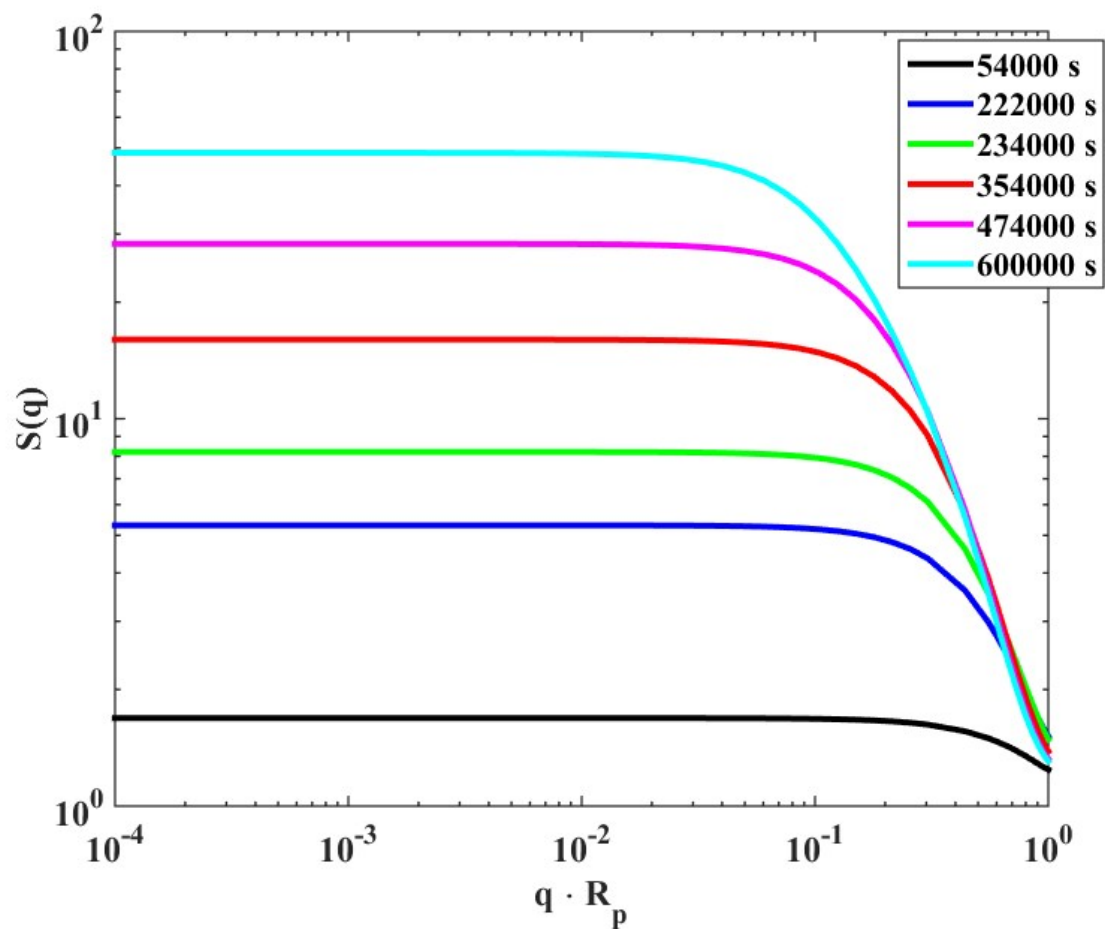
**Figure S5d** Cluster mass distributions as a function of cluster mass, for a few dimensionless time values, as shown in the legend, in the case where the aggregation Kernel is given by Equation (2) of the main text. In this case a cutoff at a given size ( $R_g^{\text{Max}}=6.2 \cdot 10^4$  nm) has been imposed. The calculations have been carried out for particle diameter equal to 120 nm,  $W=10^5$ , particle volume fraction equal to 21% and shear rate equal to  $1700 \text{ s}^{-1}$ , when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text.



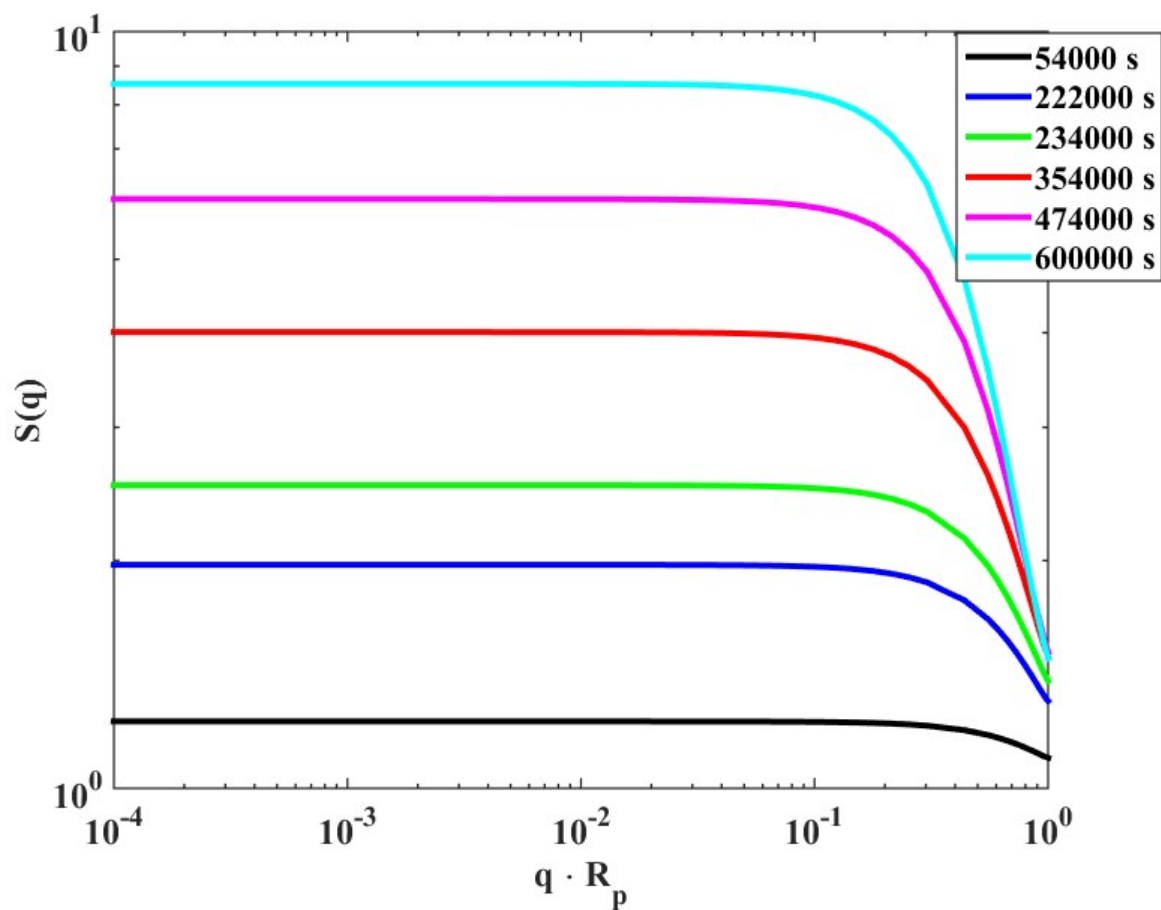
**Figure S6** Occupied volume fraction evolution for the same set of data shown in Figure 4 of the main text. The calculations have been carried out when aggregation is modeled using the aggregation Kernel given by Equation (2) in the main text, with the following stability ratio values:  $W=1.38 \cdot 10^8$  for particle volume fraction equal to 19%,  $W=10^8$  for particle volume fraction equal to 21% and  $W=6.5 \cdot 10^7$  for particle volume fraction equal to 23%.



**Figure S7a** Average Scattering Structure Factor as a function of the dimensionless scattering wave vector, for the time indicated in the legend. The calculations have been carried out for the following conditions: particle volume fraction equal to 21%, fractal dimension equal to 1.8, diffusion-limited aggregation kernel, no breakage.



**Figure S7b** Average Scattering Structure Factor as a function of the dimensionless scattering wave vector, for the time indicated in the legend. The calculations have been carried out for the following conditions: particle volume fraction equal to 21%, fractal dimension equal to 2.7,  $W=10^8$  and simple shear aggregation kernel, with no breakage.



**Figure S7c** Average Scattering Structure Factor as a function of the dimensionless scattering wave vector, for the time indicated in the legend. The calculations have been carried out for the following conditions: particle volume fraction equal to 21%, fractal dimension equal to 2.7,  $W=10^8$  and simple shear aggregation kernel with cluster breakage active, modeled by Equation (6) in the main text.