Supplementary Material

Locations of boundaries in state diagrams

<u>Type **II** / **IV** boundary</u>: the boundary between types **IV** and **II** occurs for (n^*, β_{gn}) values at which the first moment of the aggregate size distribution ($\lambda^{(1)}$) equals n^* at a low but nonzero value of m. This is such that $\mathrm{d}m/\mathrm{d}\theta_n$ in Eq. 12 is sufficiently close to zero that the sudden decline in $\mathrm{d}m/\mathrm{d}\theta_{in}$ at long times in type **IV** behavior would not be perceptible by visual inspection of m(t) profiles. With this in mind, the **II** / **IV** boundary is estimated by equating n^* with the value of $\lambda^{(1)}$ in type **II** behavior at $t/t_{50} >> 1$ (i.e., as $m \rightarrow 0$), with $\lambda^{(1)}$ given by

$$\lambda^{(1)}(t) = \frac{1 - m(t)}{\sigma(t)} \tag{S1}$$

$$n^* = \lambda^{(1)}(t/t_{50} >> 1) = \frac{1}{\sigma_{ss}(\beta_{gn}; x, \delta)} \qquad (\textbf{II} / \textbf{IV} \text{ boundary}) \tag{S2}$$

The value of σ_{ss} is a function of only β_{gn} for a given x and δ, and is obtained as described in Sec. 3.2.

<u>Type **IV** / **Ib** boundary</u>: the boundary between types **IV** and **Ib** occurs approximately where the aggregate size distribution first spans the size range from x to n^*. Once σ reaches steady state in type **II** behavior, the a_j distribution grows with essentially a constant shape. Furthermore, the standard deviation (SD_{agg}) of the a_j distribution when $n^* \rightarrow \infty$ empirically follows the relationship $SD_{agg}(t) \approx c_{x,\delta}\lambda^{(1)}(t)$, where $c_{x,\delta}$ is independent of β_{gn} at constant x and δ. The parameter $c_{x,\delta}$ has a weak dependence on x (inset of Fig. 10) and δ (not shown). We approximate the boundary between **IV** and **Ib** by equating the

gap in size between x and n^* (i.e., n^*-x) with $1.5SD_{agg}(t/t_{50}>>1)$. Using Eq. S2 to relate σ_{ss} and the limiting value of $\lambda^{(1)}$ gives

$$n^* \cong x + \frac{1.5c_x}{\sigma_{ss}(\beta_{gn}; x, \delta)} \qquad \text{(\textbf{IV} / \textbf{Ib} boundary)} \qquad \text{(S3)}$$

with σ_{ss} evaluated as noted above.

<u>Type **Ib** / **Ic** & **Ic** / **Id** boundary</u>: type **Ic** behavior is independent of n^* because there is essentially no aggregate growth beyond nucleation. Thus, the boundary between **Ic** and **Ib** or **Id** will be a horizontal line in Fig. 3. We estimate this threshold value of β_{gn} to be that which causes the growth rate to be comparable to nucleation prior to $dm/d\theta_n$ effectively reaching zero (i.e., $xm^x << 1$ in Eq. 12). This motivates the following criterion, which follows by setting the nucleation term in Eq. 12 (xm^x) to ten times that for growth ($\delta\beta_{gn}m^\delta\sigma$) at $t/t_{50} = 1$ ($m = 0.5$) for a given x and δ, and using $\sigma = (1-m)/x$ because type **Ic** behavior holds at the boundary. After some rearrangement the threshold value of β_{gn} is given by

$$\beta_{gn} \cong \frac{0.1x^2(0.5)^{x-\delta+1}}{\delta} \qquad \text{(\textbf{Ic} boundary with \textbf{Ib}, \textbf{Id})} \qquad \text{(S4)}$$

<u>Type **Id** / **II** boundary</u>: the threshold value of β_{gn} below which type **II** behavior is no longer expected to hold is approximated by equating the growth and nucleation terms in Eq. 12 at relatively low extents of monomer loss (m=0.8 in the expression below), assuming σ is reasonably described by σ_{ss} from type **II** behavior.

$$\beta_{gn} \cong \frac{x(0.8)^{x-\delta}}{\delta\sigma_{ss}} \qquad \text{(S5)}$$

We use this expression to define the **II** / **Id** boundary for n^* greater than that given by the **II** / **IV** boundary (Eq. S2). Note that Eq. S5 is an implicit function of β_{gn}, x, and δ because σ_{ss} obtained from Eq. 27 also depends on these three variables.

<u>$x = n^*$ boundary</u>: this boundary exists because $x < n^*$ is physically undefined in the LENP model, independent of β_{gn}. The limiting case of $x = n^*$ necessarily corresponds either to aggregation ceasing at the nucleation stage, or that all aggregates precipitate or grow via only condensation beyond the nucleation stage.