**Supplemental Information**

**Extended log-normal method of moments for solving the population balance equation for Brownian coagulation**

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This section describes the reconstruction of the four size distribution parameters from the first four moments.

For =0, 1, 2 and 3, Eq. (14) becomes

, (S1)

, (S2)

, (S3)

. (S4)

Thus,  is equal to the zeroth moment.

Combining the formula  with Eqs. (S1) to (S4) gives

. (S5)

This indicates that  is the root of . The derivative of  is , so  has two stationary points,  and . For , . For ,

. (S6)

Substituting Eqs. (S1) to (S4) into Eq. (S6) shows that the sign of  is the same as that of . This formula can be easily shown to be no greater than 0, so . Due to the conditions , ,  and ,  has three real roots. Then  should be equal to the second real root  (sorted in descending order), as will be discussed later. Thus,  is expressed as

. (S7)

Substituting Eqs. (S1) and (S7) into Eqs. (S2) and (S3) shows that  and  satisfy the following quadratic equation:

. (S8)

Then  and  can be obtained by solving Eq. (S8):

, (S9)

. (S10)

From Eqs. (S9) and (S10),  should be no greater than  because of the square root, which indicates that  must be the second real root of . Thus, the reconstruction problem has one and only one solution, as shown in Eqs. (S1), (S7), (S9) and (S10).