



# A model for two-component aerosol coagulation and phase separation: a method for changing the growth rate of nanoparticles

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## Abstract

In previous studies of particle growth we have synthesized binary metal oxide aerosols and have observed the evolution of internal phase segregation during growth of molten nanodroplets. We have also generated NaCl/metal aerosols in which the metal nanoparticle is enveloped within a salt droplet. In both systems the nanoparticles were grown in the molten state. In this paper we propose a model which incorporates phase segregation in a binary aerosol. The model assumes that complete phase segregation is the thermodynamically favored state, that no thermodynamic activation energy exists, and that the phase segregation process is kinetically controlled. The results indicate that a steady state behavior can be reached in which the characteristic time for aerosol coagulation and the characteristic time for the growth of the minority phase coincide such that the number of distinct segregated entities within each aerosol droplet is constant.

The results suggest what we believe to be an important concept that can be utilized in materials synthesis. This is that the major phase can be used to moderate the growth rate of the minor phase by changing the characteristic encounter frequency and therefore the eventual growth rate of the minority phase. In particular, temperature, which does not play an important role in aerosol coagulation, is seen to be a very sensitive parameter for the growth of the minority phase nanoparticles. We discuss the parameter space necessary for this to occur. © 2001 Elsevier Science Ltd. All rights reserved.

## 1. Introduction

The study of aerosol dynamics is often limited to homogeneous, single-component aerosol particles. However, it is becoming increasingly apparent that multi-component aerosol particles are of both industrial importance and an area in need of significant research activity. We have been involved in a number of multi-component aerosol dynamics studies with heterogeneous aerosol particles. One of our main goals in this research is to study the evolution of the internal state of the aerosol droplets. For example, we have conducted studies on the formation of binary metal oxide systems with application to removal of heavy metals (Biswas & Zachariah, 1997;

Biswas, Yang, & Zachariah, 1998) as well as the formation of materials with novel and interesting properties (Zachariah, Aquino-Class, Shull, & Steel, 1995; Ehrman, Aquino-Class, & Zachariah, 1999a; Ehrman, Friedlander, & Zachariah, 1999b).

Our initial success in growing interesting microstructures (Zachariah et al., 1995) indicated that further research into the mechanistic aspects of the growth was warranted. In subsequent studies we have employed both in-situ interrogation into the formation process (McMillin, Biswas, & Zachariah, 1996), multi-component aerosol dynamic modeling (Biswas, Wu, Zachariah, & McMillen, 1997) and molecular dynamics computation (Zachariah, Shull, McMillin, & Biswas, 1996). One of the primary conclusions was that at the high temperatures where these materials are typically grown, nanodroplets are in a liquid-like state, and that phase segregation taking place within the

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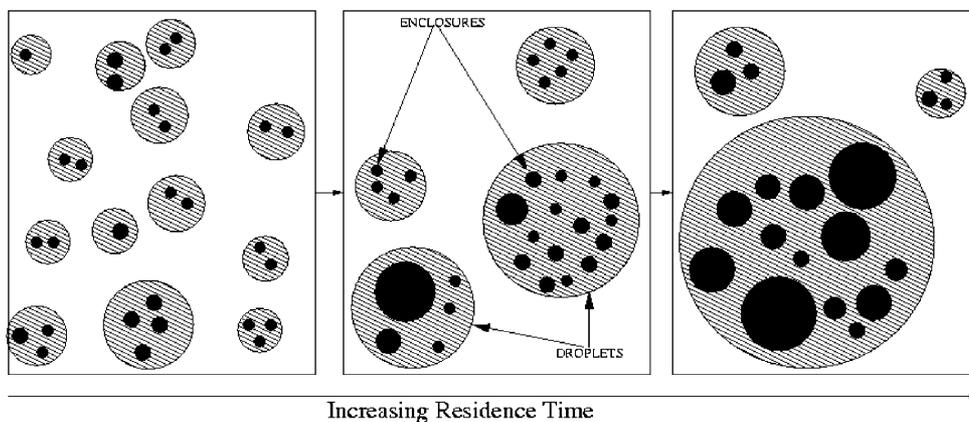


Fig. 1. The schematic illustration of the temporal evolution of a two component aerosol coagulation and phase segregation.

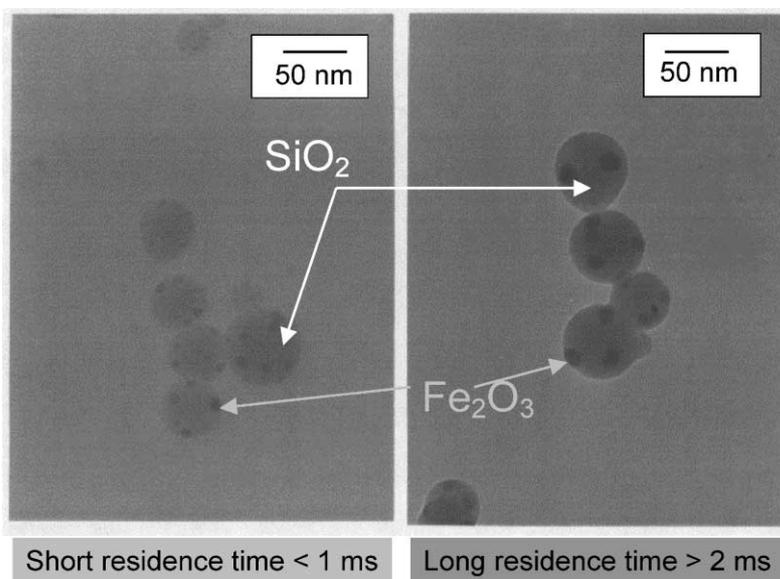


Fig. 2. Evolution of the aerosol ( $\text{SiO}_2$ ) and minor phase ( $\text{Fe}_2\text{O}_3$ ) during the growth of  $\text{SiO}_2/\text{Fe}_2\text{O}_3$  nanocomposites.

nanodroplet was probably limited by the transport within the nanodroplet.

Our goals in this paper are: (1) to develop a simple model, which describes both aerosol growth, and the evolution of the internal morphology of multi-component liquid aerosols in which the phases are immiscible. A more thorough model is detailed in Struchtrup, Luskin, and Zachariah (2001). (2) to evaluate a new strategy for controlling the vapor-phase growth of nanoparticles by changing the particle–particle encounter frequency by embedding the nanoparticle in another matrix which itself resides in an aerosol phase.

In the course of this paper we shall use the terms minor phase and enclosure interchangeable to refer to the component within each aerosol droplet, and droplet or aerosol when referring to the major phase. The enclosures are considered as an aerosol inside the droplet, whose coagu-

lation takes place due to Brownian motion. The temporal evolution of the aerosol phase is schematically depicted in Fig. 1. Due to surface tension effects the enclosures are spherically shaped within the aerosol droplets. In this treatment we will not describe individual enclosures inside individual droplets, but rather supplement the usual statistical formulation for the droplets with the statistics of the enclosures. Our model describes the evolution of the total number density of the droplets along with their volumes and the average number of enclosures per droplet. The main assumptions in the derivation of the model are that the droplets and the enclosures are assumed to be monodisperse, and that the number of enclosures per unit volume in each droplet is assumed to be uniform over all droplets. Using our model we also investigate the volume growth of the minor phase due to the presence of the major phase. It is of particular significance that our results show that introducing the major phase can moderate the

growth rate of the minor phase and leads to interesting possibilities with regard to controlling particle sizes.

The parametric model developed in the paper is applied to the two binary systems, SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> and NaCl/Ge, where SiO<sub>2</sub> and NaCl are major phases and Fe<sub>2</sub>O<sub>3</sub> and Ge are minor phases and for which we have experimental results (Ehrman et al., 1999a, b). An example of the SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> system is presented below in Fig. 2. We can see that at short residence times the dark Fe<sub>2</sub>O<sub>3</sub> enclosures are greater in number and smaller in size than those observed at longer residence times. It should be reiterated that when growing, both phases are in a liquid state.

## 2. Theory

We consider aerosol droplets consisting of two immiscible components in a constant volume ratio. Our goal is to determine the distribution of droplet volumes and the internal state of the droplet with time. In this work we construct a simple model describing the evolution of the aerosol-enclosure system.

Individual enclosures suspended in a droplet, collide and stick together through various mechanisms. Based on the size of the particles and their mean free path, we assume that the enclosures collide/grow as a result of their Brownian motion within the aerosol droplets, and that droplets, which are in free molecular flow, collide/grow based on kinetic theory. The coagulation of the droplets and the enclosures are assumed to occur by instantaneous coalescence of spherical particles. This is a good approximation since most of our experimental work was conducted at high temperature (1200–2500 K) where the components are liquids.

For coagulating aerosols the general dynamic equation characterizes the temporal evolution of the droplet (aerosol) number density,  $N(t, V)$ . The evolution of  $N(t, V)$  is known (Friedlander, 2000) and satisfies the following integro-differential equation:

$$\frac{dN(t, V)}{dt} = \frac{1}{2} \int_0^V \sigma(U, V-U) N(t, U) N(t, V-U) dU - \int_0^V \sigma(U, V) N(t, U) N(t, V) dU, \quad (1)$$

where  $\sigma(U, V)$  is the collision coefficient. For the free molecular and the Brownian regimes the collision coefficient is given

$$\sigma^F(U, V) = \left(\frac{3}{4\pi}\right)^{1/6} \left(\frac{6kT}{\rho}\right)^{1/2} \left(\frac{1}{U} + \frac{1}{V}\right)^{1/2} (U^{1/3} + V^{1/3})^2$$

and

$$\sigma^B(U, V) = \frac{2kT}{3\mu} \left(\frac{1}{U^{1/3}} + \frac{1}{V^{1/3}}\right) (U^{1/3} + V^{1/3}),$$

respectively. Here  $k$  denotes Boltzmann's constant,  $T$  is the temperature,  $\rho$  is the density of the droplets, and  $\mu$  is the viscosity of droplets.

In this work we restrict ourselves to the simplest case and approximate the particles as monodisperse ( $U = V$ ), in which case the collision coefficients reduce to (Fuchs, 1989)

$$\sigma^F(V, V) = 4\sqrt{2} \left(\frac{3}{4\pi}\right)^{1/6} \left(\frac{6kT}{\rho}\right)^{1/2} V^{1/6},$$

$$\sigma^B(V, V) = \frac{8kT}{3\mu}.$$

Since over the time range of the interest the change in  $V^{1/6}$  is insignificant, we take the volume in the expression to be constant and equal to the initial volume of the droplet in our further calculations.

We denote  $N_{\text{tot}}(t)$  to be the total number density of the particles, given by

$$N_{\text{tot}}(t) = \int N(t, V) dV.$$

Integrating (1) for monodisperse droplets over  $V$  we find

$$\frac{dN_{\text{tot}}}{dt} = -\frac{1}{2} \sigma^F N_{\text{tot}}^2. \quad (2)$$

Similarly, for the total number of the enclosure  $n_{\text{tot}}$  in a droplet of volume  $V$  we have

$$\frac{d(n_{\text{tot}}/V)}{dt} = -\frac{1}{2} \sigma^B (n_{\text{tot}}/V)^2. \quad (3)$$

We further assume that the number of enclosures per unit volume of droplet, remains approximately the same for each droplet. The validity of this assumption comes from realizing that the mass ratio of all components in a strictly coagulating aerosol system is a constant in time. Another way to view this assumption is that since the characteristic coagulation time of enclosures in each droplet is given by  $V/\sigma^B n_{\text{tot}}$ , our assumption is equivalent to stating that the characteristic coagulation time of the enclosures in each droplet is approximately the same. Thus,  $n_{\text{tot}}/V$  represents the evolution of the mean number of the enclosures per droplet.

For convenience we drop  $tot$  from the variables  $N$  and  $n$  in our calculations. We denote by  $V(t)$  the volume of the droplet at time  $t$ , so that the conservation of total volume of droplets gives us

$$V(t)N(t) = V_0 N_0, \quad (4)$$

where  $V_0$  and  $N_0$  are initial volume of droplets and their initial number density. The combination of Eqs. (2)–(4)

$$\frac{dN(t)}{dt} = -\frac{1}{2} \sigma^F N(t)^2,$$

$$\frac{d(n(t)/V(t))}{dt} = -\frac{1}{2} \sigma^B (n(t)/V(t))^2,$$

$$N(t)V(t) = N_0 V_0$$

constitute the closed system for  $N(t)$ —the number density of the aerosol droplets,  $n(t)$ —the mean number of enclosures per droplet, and  $V(t)$ —the volume of the droplet at time  $t$ . The initial conditions for the system is given by  $N(t=0) = N_0$ ,  $n(t=0) = n_0$ ,  $V(t=0) = V_0$ .

To solve the system we note  $N(t)$  can be determined from the first equation

$$N(t) = \frac{N_0}{1 + \frac{1}{2}\sigma^F N_0 t}.$$

To find the solution of the second equation we write it as

$$\frac{d}{dt} \left( \frac{n(t)N(t)}{V_0 N_0} \right) = -\frac{1}{2}\sigma^B \left( \frac{n(t)N(t)}{V_0 N_0} \right)^2.$$

The latter can be easily solved for  $n(t)N(t)$ :

$$n(t) = \frac{1}{N(t)} \frac{N_0 n_0}{1 + \frac{1}{2}(\sigma^B/V_0 N_0) N_0 n_0 t}.$$

Substituting the expression for  $N(t)$  we have

$$n(t) = \frac{V_0 n_0 + \frac{1}{2}\sigma^F N_0 V_0 n_0 t}{V_0 + \frac{1}{2}\sigma^B n_0 t}. \quad (5)$$

As  $t \rightarrow \infty$  we have

$$n(t) \rightarrow n_\infty = \frac{\sigma^F}{\sigma^B} N_0 V_0.$$

*This implies and leads to the interesting result that there exists a steady state value for the number of the enclosures in a droplet, which is proportional to the initial mass concentration of droplets, and the relative collision cross sections in the free and Brownian regimes.*

By considering the derivative of  $n(t)$

$$\frac{dn(t)}{dt} = \frac{\frac{1}{2}n_0 V_0 (\sigma^F N_0 V_0 - \sigma^B n_0)}{(N_0 V_0 + \frac{1}{2}\sigma^B n_0 t)^2}$$

we see that  $n(t)$  increases to its final constant value if  $\sigma^F N_0 V_0 > \sigma^B n_0$  and decreases in the opposite case. Note that the condition  $\sigma^F N_0 V_0 > \sigma^B n_0$  represent the relation between the characteristic time ( $t^F$ ) for the coagulation of the droplets in the free-molecular regime and the characteristic time ( $t^B$ ) for the coagulation of the enclosures in the Brownian regime. The ratio between these characteristic times is given as

$$\frac{t^B}{t^F} = \frac{\sigma^F N_0 V_0}{\sigma^B n_0}.$$

Thus  $n(t)$  increases towards its final constant value if the characteristic time for the above ratio is greater than unity, and decreases in the opposite case. In the latter case the model must be supplemented with the condition  $n(t) \geq 1$ .

Based on the asymptotic value of  $n(t)$  we can conclude the following: (1) if the total volume of the droplets increases then the average number of the enclosures per droplet,  $n(t)$ , also increases; (2) if the viscosity of the

major phase increases then  $n(t)$  also increases; (3) if the temperature increases then  $n(t)$  decreases due to a drop in the viscosity of the major phase.

We next turn our attention to the relative volume growth of the enclosures due to the presence of the droplets, i.e., what effect does the major phase have on the growth rate of the enclosures? Assuming that the volume fraction of the enclosures is the same initially for all droplets we have

$$n(t)u(t) = cV(t),$$

where  $n(t)$  is the average number of the enclosures per droplets defined by (5),  $u(t)$  is the average volume of the enclosures,  $V(t)$  is the volume of the droplets, and  $c$  is the volume fraction of the enclosures per droplet and remains constant in time. The volume of the droplet can be calculated from the conservation of the mass,  $N(t)V(t) = N_0 V_0$ , for the droplet as

$$V(t) = V_0(1 + \frac{1}{2}\sigma^F N_0 t).$$

Then taking into account the expression for the evolution of  $n(t)$ ,  $u(t)$  can be calculated

$$u(t) = u_0 + \frac{1}{2}c\sigma^B t,$$

where  $u_0 = cV_0/n$  is the initial volume of the enclosures.

In the absence of the major phase the enclosures coagulate in the free molecular regime. Their volume  $w(t)$  in this case can be readily calculated:

$$w(t) = w_0 + \frac{1}{2}\sigma^F \phi t,$$

where  $w_0 = u_0$  is the initial volume of the particles, and  $\phi$  is the volume fraction of the particles. Now we can calculate the relative volume change for the enclosure due to the presence of the droplets

$$\frac{u(t)}{w(t)} = \frac{u_0 + \frac{1}{2}c\sigma^B t}{u_0 + \frac{1}{2}\phi\sigma^F t}.$$

For large times this ratio becomes  $c\sigma^B/(\phi\sigma^F)$ . Note that we obtain this ratio provided  $n(t) \geq 1$ . It is interesting to note that  $c\sigma^B/(\phi\sigma^F) = 1/n_\infty$ .

We note for clarity in presentation that in this model the change of  $V^{1/6}$  in the collision coefficient  $\sigma^F(V, V)$  has been neglected assuming that over the time range of interest the change in  $V^{1/6}$  is insignificant. The effect of  $V^{1/6}$  can be easily incorporated into the model. In this case the equation for the number density of droplets (Eq. (2)) is given by  $dN_{tot}/dt = -0.5\sigma_0^F N_{tot}^{11/6}$ , where  $\sigma_0^F = 4\sqrt{2}(3/4\pi)^{1/6}(6kT/\rho)^{1/2}(N_0 V_0)^{1/6}$ .

### 3. Numerical results

3.1. In our first example we apply the theory developed above, to the binary  $\text{SiO}_2/\text{Fe}_2\text{O}_3$  system. To implement the numerical examples we consider a case when all silica ( $\text{SiO}_2$ ) droplets are initially at a radius  $r = 5$  nm and a

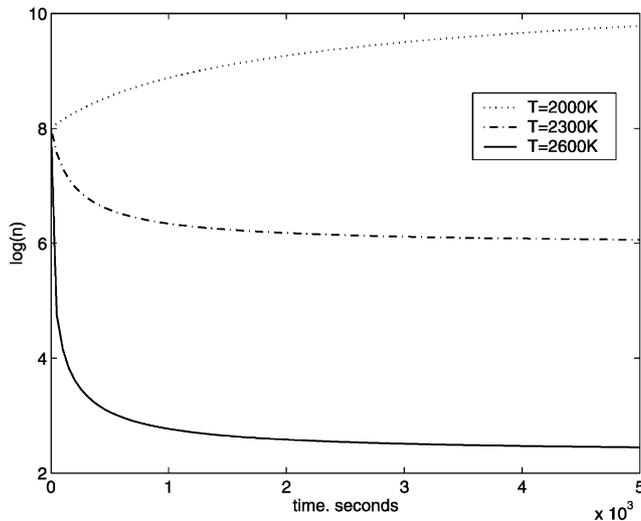


Fig. 3. The average number of enclosures per droplet as a function of time for different temperatures for the SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> system.

number density of 10<sup>12</sup> 1/cm<sup>3</sup>. The droplets constitute the background media (SiO<sub>2</sub>) for the coagulation of the enclosures (Fe<sub>2</sub>O<sub>3</sub>). The viscosity of the silica is highly temperature sensitive and is given by Kingery, Bowen, and Uhlmann (1976)

$$\mu = 10^{-8.7(1-3556K/T)} \text{ kg/m s.}$$

The density of the silica is  $\rho = 2.4 \text{ g/cm}^3$ . For the enclosures, we assume that each droplet initially contains 3000 enclosures. We consider three different temperatures:  $T = 2000, 2300$  and  $2600 \text{ K}$ . For these temperatures we plot the average number of the enclosures per droplet as a function of time in Fig. 3. This plot shows that the number of the enclosures reaches a near constant final value by either increasing or decreasing, depending on the value of  $\sigma^F N_0 V_0 / \sigma^B n_0$ . Denoting the characteristic times for the molecular regime by  $t^F$  and for the Brownian regime by  $t^B$  we have

$$\frac{t^B}{t^F} = \frac{\sigma^F N_0 V_0}{\sigma^B n_0}.$$

We can express this ratio of the characteristic times as a function of the temperature by including the temperature dependence of the collision coefficients and the viscosity, so that

$$\log(t^B/t^F) = \frac{T^*}{T} - \log(T) + C(\rho, N_0, V_0, n_0).$$

Here  $C(\rho)$  is given by

$$C(\rho) = \log(N_0 V_0 / n_0) + \log\left(4\sqrt{2} \left(\frac{3}{4\pi}\right)^{1/6} \left(\frac{6k}{\rho}\right)^{1/2} V^{1/6}\right) - \log(8k/a),$$

where we have used the expression  $\mu = a \exp(T^*/T)$  for the viscosity. The ratio  $\log(t^B/t^F)$  is plotted against  $T^*/T$

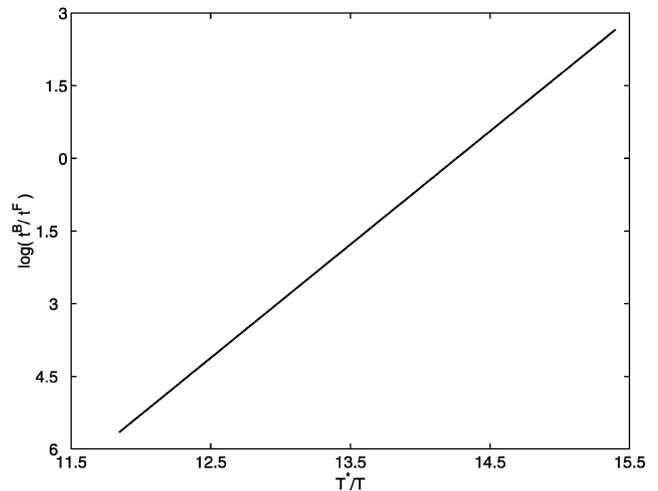


Fig. 4. The ratio of the characteristic times for enclosure and droplet coagulation as a function of temperature for the SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> system.  $T^* = 30800 \text{ K}$ .

in Fig. 4. It is quite clear that for this system the range of ratios of characteristic times spans values both larger and smaller than unity, over what is really only relatively moderate changes in temperature. We discuss the significance of this result in later sections, however, it should be obvious to the reader that such dramatic changes in growth rate could not be achieved in a single component coagulating aerosol system.

Next for this system we calculate the volume growth for the enclosures and compare that to the growth that would be achieved were the major phase absent. As we found in the previous section the ratio of the volume of the enclosures in the presence of the droplets to that in the absence of the droplets is given by  $c\sigma^B / (\phi\sigma^F)$ . A value of unity implies that the major phase has no effect on the growth rate of enclosures. Plotting this quantity as a function of the temperature in Fig. 5 we observe that by introducing the major phase we can moderate the growth of the minor phase by several orders of magnitude. At least qualitatively this is consistent with experiment and suggests an opportunity to change the relative growth rate of nanoparticles by changing the encounter frequency, through the use of a second matrix. Indeed because of the high activation energy of the viscosity of the major phase, moderate changes in temperature lead to significant effects on the average volume of the enclosures.

3.2. In the second example we consider the binary system NaCl/Ge. This example is taken from a generic gas phase synthesis method in which  $\text{Na} + \text{metal halide} \Rightarrow \text{metal} + \text{NaCl}$  (Steffens, Zachariah, DuFaux, & Axelbaum, 1996; Ehrman et al., 1999a). Other researchers and we have used this method to make a variety of metal and metal/carbides/nitrides.

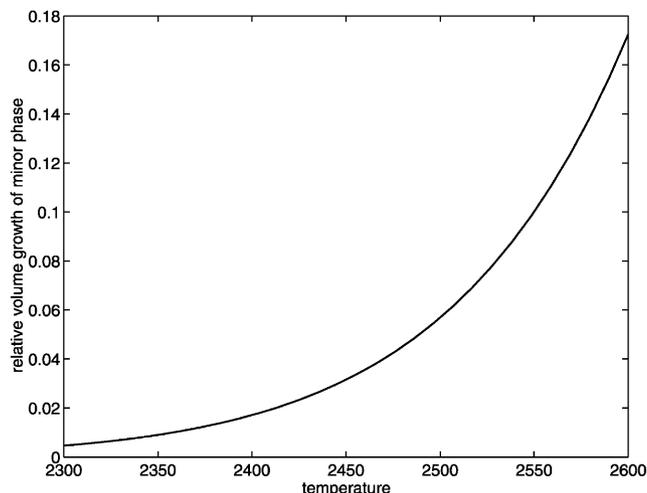


Fig. 5. The relative (to no major phase present) volume growth of the minor phase as a function of temperature for the  $\text{SiO}_2/\text{Fe}_2\text{O}_3$  system.

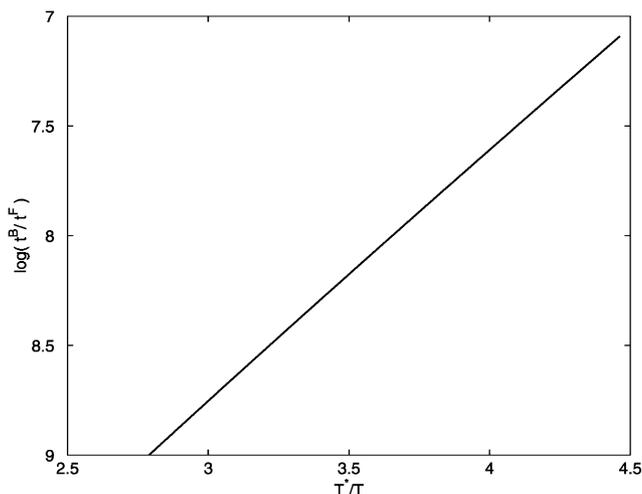


Fig. 6. The ratio of the characteristic times of enclosure and droplet coagulation for the Ge/NaCl system.  $T^* = 4462$  K.

In this example we assume that the volume fraction of the salt (droplet particles) is  $10^{-5}$ , and the relative volume fraction of the enclosures is 25% (based on the stoichiometry of the reaction of sodium and germanium tetrachloride). The viscosity of the salt as a function of the temperature is given by

$$\mu = 2.3e - 5 \exp\left(\frac{4462K}{T}\right) \frac{\text{kg}}{\text{m s}}$$

The temperatures considered here are those consistent with the experiment,  $T = 1100$  and  $1300$  K. Assuming initial radius of the salt particles to be  $2$  nm we find the initial number density for the droplets,  $N = 10^{15}$   $1/\text{cm}^3$ . Similar to the first example we plot  $\log(t^B/t^F)$  as a function  $T^*/T$ , where  $T^* = 4462$  K in Fig. 6. Under these conditions  $\sigma^F N_0 V_0 / \sigma^B n_0$  is small, and thus the coagulation of enclosures is much faster than that of the droplets. We

should expect that the aerosols would evolve to a system in which there will only be one enclosure per droplet. Indeed this is what has been observed experimentally under conditions where the enclosures are melted, so that when they collide they coalesce (Ehrman et al., 1999a). Since there is only one enclosure per droplet the relative growth of the minor phase due to the presence of the major phase is insignificant. The problem in this case is that NaCl, which is a useful material because it is easily removed after synthesis from the minor phase by solvent extraction, has much too low viscosity to control the growth of the minor phase. This implies that in order to have a major phase that can moderate the growth rate of the minor phase we need to use a major phase which is more viscous than NaCl. The data for viscosity of molten salts presented in Jans, Dampier, Lakshminarayanan, Lorentz, and Tomkins (1968) suggests the use of  $\text{ZnCl}_2$ . At  $593$  K for example the viscosity of  $\text{ZnCl}_2$  is  $4525$  cP. Taking the relative concentration of Ge to be  $c = 0.1$ , and the total volume fraction to be  $\varphi = 2e - 5$  we obtain the relative volume growth due to the presence of  $\text{ZnCl}_2$  to be  $0.13$ . This result indicates that were one to employ  $\text{ZnCl}_2$  as the major phase, one should expect to see a significant moderation of the growth rate of the minor phase.

#### 4. Relevance to controlling nanoparticle growth

These modeling results indicate that it is possible to devise systems in which nanoparticle growth rates in an aerosol phase can be slowed down through judicious choice of a matrix that controls the encounter frequency. We saw that for the silica/iron oxide case the growth rate of the minor phase could be significantly impacted. On the other hand the salt/germanium results showed no such moderation. In both cases the model is qualitatively consistent with experimental observation.

The silica/iron oxide system was of interest to us experimentally because it offered the opportunity to make a superparamagnetic nanocomposite (Zachariah et al., 1995). However, more generically the silica/iron oxide system and similar systems has limited value because the minor component cannot be isolated from the major component. On the other hand the use of salt has the potential advantage of ease of removal to release the minor phase. Of course, the results showed that NaCl had minimal or no effect. This implies that in order to have a major phase that can moderate the growth rate of the minor phase we need to use a major phase which is significantly more viscous than NaCl. The data for viscosity of molten salts presented in Jans et al. (1968) suggests that  $\text{ZnCl}_2$  might be a possible candidate with a viscosity of  $4525$  cP at the melting point. Taking the relative concentration of a minor phase to be  $c = 0.1$ , and the total volume fraction to be  $\varphi = 2e - 5$  we obtain a relative volume growth due to the presence of  $\text{ZnCl}_2$  to

be 0.13. Under these circumstance we can conclude that this salt is a good candidate medium for moderating the growth rate of nanoparticles.

## 5. Concluding remarks

In this paper we presented a model which describes both aerosol growth, and the evolution of the internal morphology of multi-component liquid aerosols in which the phases are immiscible. In the derivation of the model we assumed that the number of the enclosures per unit volume in each droplet remains constant over all droplets during growth. Though this assumption imposes a restriction on the evolution of the polydispersity of the system, it should capture the major aspects in the evolution of a heterogeneous aerosol. In particular our model shows that the average number of enclosures per droplet increases when the total volume of the droplets increases, when the viscosity of the surrounding media increases, or when temperature decreases. Using our model we estimated the growth rate of the minor phase due to the presence of the major phase. The parametric model developed here was applied to the two binary cases,  $\text{SiO}_2/\text{Fe}_2\text{O}_3$ ,  $\text{NaCl}/\text{Ge}$  and the results were shown to seem to be consistent with experiments.

Our model suggests that the growth rate of the minor phase in the case  $\text{NaCl}/\text{Ge}$  is insignificant, but that the choice of another more viscous salt such as  $\text{ZnCl}_2$  seems to possess the requisite viscosity.

Finally, we believe that the concept of using a second phase to control the encounter frequency can be used to slow the growth of nanoparticles under conditions where they are liquid like. Temperature plays a much more important role in coagulation in this system than in a single-component aerosol system since it has a very strong influence on the viscosity of the major phase and therefore the internal transport properties within the aerosol droplet. In future work we will generalize our approach by incorporating the size distribution of the enclosures into the model.

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