We study a nonlinear model of two-grain mass exchange. The transition rates follow the Arrhenius equation with an activation energy that depends on the grain mass. We show that the activation parameter can be absorbed in the initial conditions for the grain masses and derive approximate solutions of the resulting coupled nonlinear ODE model in specific neighborhoods of the phase space. We determine that the equilibrium points of the model involve equipartition (diffusive), coarse-graining, and trapped phases. In addition, we demonstrate that coarse-graining is limited by the trapping effect which is unaccounted for by the linearized approximation. Potential applications of the mass exchange model involve coarse-graining during sintering and wealth exchange in econophysics.

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I. INTRODUCTION

Phenomena of grain growth and nucleation are common in many engineering and physical processes [1]. A grain is defined as a contiguous region of material with the same crystallographic orientation, which changes discontinuously at the grain boundaries. Many technological materials, including advanced ceramics, are produced by means of non-equilibrium physical processes that generate phase changes and grain growth. Early studies of the kinetics of crystallization and other phase changes were based on the JMAK (Johnson, Mehl, Avrami, Kolmogorov) equation [2-9]. Solid-state sintering transforms powders into monolithic materials by applying temperature and pressure [4]. Sintering involves diffusion and transport of atoms as well as plastic deformations. During the sintering process, the number of grains is progressively reduced, whereas the average grain radius grain increases, a process known as Ostwald ripening [5-6].

Recent approaches to sintering involve the Direct Multiscale Modeling [7] and the Discrete Element Method (DEM) that relax assumptions regarding the particle kinematics [8-9] and generalized Monte Carlo simulations [10]. In DEM, grain coarsening is incorporated by transferring the overlapping volume of neighboring spherical particles from the smaller to the larger. Existing sintering models are continuum formulations. This is also true of many other diffusion processes such as the Cahn-Hilliard equation [11] that describes phase separation and the phase-field models used to describe solidification [12]. Recently, the self-consistent mean-field kinetic was proposed as an atomic diffusion theory for non-uniform alloys [13]. This theory uses thermally activated transition rates between species and corrects the Cahn-Hilliard model in the case of non-uniformities.

Mass diffusion during sintering is controlled by an activation energy which can be lowered by means of ball milling [14]. A high resolution transmission electron microscope image of an alpha-silicon nitride grain (alloy based on the silicon nitride Si₃N₄ in which some silicon atoms are replaced by Al and corresponding Ni atoms by O) after mechanical activation by ball milling is shown in Fig. 1. The grain includes areas with both oriented and disordered lattice structure which occur both inside and near the boundary of the grain. The existence of such areas can lead to intra-grain as well as inter-grain diffusion.

Motivated by the above observations, we study a simplified model of mass exchange between two discrete grains. The exchange is governed by a kinetic equations which involves transition rates that are based on
the Arrhenius equation $k = \exp(-E_a/k_B T)$, where $k$ is the rate coefficient (reaction constant), $E_a$ is the activation energy, $k_B$ is Boltzmann’s constant, and $T$ the temperature. The kinetic model that we study herein is too simplified to accurately capture properties of actual sintering processes. Nevertheless, it notably exhibits a transition between a diffusive state in which the equilibrium grain masses are equal, and a growth-decay state in which the larger mass grows whereas the smaller one shrinks. These two distinct states may be related, respectively, to normal and abnormal growth regimes observed in sintering \cite{15}.

A. Nonlinear Mass Exchange Model

In \cite{10} we introduced a kinetic model for mass exchange between $N$ grains of different radii. This model involves a system of $N$ coupled non-linear ordinary differential equations (ODEs) with Arrhenius-like transition rate coefficient as follows

$$
\frac{\partial m(x_i, t)}{\partial t} = \sum_{<i,j>} e^{-u m(x_i, t)} m(x_j, t) - \sum_{<i,j>} e^{-u m(x_i, t)} m(x_i, t), \tag{1}
$$

where $m(x_i, t)$, $x$, $t$ and $u$ are the non-dimensional grain mass, position, time, and grain activation energy, respectively, and $<i,j>$ denotes the nearest neighbors $x_j$ of $x_i$. The model’s physical parameters are the grain mass density, $\rho$ (assumed constant), the grain radius, $r(x, t)$, the constant $\lambda$ that determines the scale of the transition rates, the characteristic grain mass and radius, $m_0 = m(x, t = 0)$ and $r_0$, respectively, the dimensionless rate factor $\alpha$, the characteristic activation energy $Q$, the gas constant $R$, and the temperature $T$. The value of $\alpha$ determines the impact of mechanical activation on the activation energy. The dimensionless parameters are $u = \frac{\alpha Q}{R T}$, $t = \lambda t$, $x = \frac{x}{r_0}$, $m(x, t) = m(x, t)/m_0$. The system of eqs. \cite{11} focuses on the exchange of mass between grains through transitions that incorporate non-homogeneous activation energies but neglects plastic deformation effects. In \cite{10}, the ODE system of \cite{11} was numerically solved for a one-dimensional chain using Euler’s first-order explicit method \cite{17} with adaptive step size.

II. TWO-GRAIN MASS EXCHANGE MODEL

We investigate a two-grain system that follows eqs. \cite{11} in order to understand the impact of exponentially varying transition rates on mass exchange. Let us consider $m(x_1, t) = x(t)$ and $m(x_2, t) = y(t)$. Then, the mass transfer between the grains is expressed by means of the following system of two ordinary differential equations (ODEs)

$$
\begin{align*}
\frac{dx(t)}{dt} &= y(t) e^{-u y(t)} - x(t) e^{-u x(t)} \tag{2a} \\
\frac{dy(t)}{dt} &= x(t) e^{-u x(t)} - y(t) e^{-u y(t)}, \tag{2b}
\end{align*}
$$

with initial conditions $x(0) = x_0$ and $y(0) = y_0$. This is a system of first-order, autonomous, nonlinear ODEs. Assuming a positive activation parameter, $u > 0$, by means of the transformations $u x(t) \mapsto m_1(t)$ and $u y(t) \mapsto m_2(t)$, eqs. \cite{12} transform to the following ODE system

$$
\begin{align*}
\frac{dm_1(t)}{dt} &= r_{2 \rightarrow 1}(t) - r_{1 \rightarrow 2}(t) \tag{3a} \\
\frac{dm_2(t)}{dt} &= r_{1 \rightarrow 2}(t) - r_{2 \rightarrow 1}(t), \tag{3b}
\end{align*}
$$

with initial conditions $m_1(0) = u x_0$ and $m_2(0) = u y_0$, and Arrhenius-like transition rates $r_{i \rightarrow j}(t) = m_i(t) e^{-m_i(t)}$ ($i, j = 1, 2, i \neq j$). The transition rates are bounded from above by $1/e$. The differences $r_{2 \rightarrow 1}(t) - r_{1 \rightarrow 2}(t)$ and $r_{1 \rightarrow 2}(t) - r_{2 \rightarrow 1}(t)$ represent the mass transfer rates to $m_1(t)$ and $m_2(t)$, respectively. The phase space of eqs. \cite{12} is determined by the two initial conditions which define the parameter space. The parameter $u$ is irrelevant, because it can be absorbed in the initial conditions: if the solutions $m_1(t)$, $m_2(t)$ of eqs. \cite{12} with initial conditions $m_1(0) = u x_0$ and $m_2(0) = u y_0$ are available, the solution for $x(t)$, $y(t)$ can be obtained for any $u > 0$ from $x(t) = m_1(t)/u$ and $y(t) = m_2(t)/u$.

III. EQUILIBRIUM POINTS

The ODE system \cite{12} conserves mass, as shown by adding the left and right sides of the two equations, respectively, leading to the cumulative mass evolution equation $\frac{d(m_1(t) + m_2(t))}{dt} = 0$. The equilibrium points correspond to

$$
\frac{dm_1(t)}{dt} = \frac{dm_2(t)}{dt} = 0.
$$

The above lead to three equilibrium points that depend on the initial conditions. Whereas eqs. \cite{12} are invariant if both masses are multiplied by the same constant, this scaling affects the initial conditions. The solution of the ODE system with conditions $m_1(0)$, $m_2(0)$ can thus lead to a different equilibrium regime than the solution with initial conditions $\lambda m_1(0)$, $\lambda m_2(0)$. In the following, we use the notation $m_{\text{eq}1} = \lim_{t \rightarrow \infty} m_1(t)$, $m_{\text{eq}2} = \lim_{t \rightarrow \infty} m_2(t)$.
A. Equipartition point

If \( m_1(0), m_2(0) \) are not large, mass diffusion primarily transfers mass from the larger to the smaller grain leading to an equilibrium point at which the grain masses are equal, i.e., \( m_{\text{eq},1} = m_{\text{eq},2} = (m_1(0) + m_2(0))/2 \). If \( m_1(0) = m_2(0) \), transitions between the grains are impeded and \( m_{\text{eq},1} = m_1(0), m_{\text{eq},2} = m_2(0) \). Hence, the two-grain system is at equilibrium at \( t = 0 \).

B. Trapping Point

(i) The system becomes trapped in its current state if the transition rates become equal, i.e., \( r_{2\rightarrow1}(t) = r_{1\rightarrow2}(t) \). This occurs at \( t = 0 \) for identical initial conditions, leading to the static equipartition point above. (ii) Rate balance can also occur for different mass values \( m_1(t), m_2(t) \), since the nonlinear equation of constant transition rate \( m e^{-m} = c \), where \( 0 \leq c < 1/e \), admits two solutions \( m_1^* \) and \( m_2^* \). The dependence of the roots is shown in Fig. 2. Then, if at \( t > 0 \) it happens that \( m_1(t) = m_1^* \) and \( m_2(t) = m_2^* \) or vice versa, the solutions becomes trapped in this state. (iii) For all practical purposes, trapping also occurs if \( m_1(0), m_2(0) \gg 1 \), because the transition rates are then almost zero; as a result, the ODE system is frozen at the initial state.

C. Aggregation point

If \( m_1(0) \gg 1, m_2(0) \approx 1 \), we expect that the larger mass will grow whereas the smaller mass will tend to zero, i.e., \( m_{\text{eq},1} = m_1(0) + m_2(0) \), and \( m_{\text{eq},2} = 0 \). This scenario corresponds to full aggregation of the entire mass on the larger grain. A justification of this equilibrium distribution is based on the fact that initially \( m_1(t)e^{-m_1(t)} \ll m_2(t)e^{-m_2(t)} \) so that \( r_{2\rightarrow1}(t) \gg r_{1\rightarrow2}(t) \). The accumulation of mass on the larger grain implies that the transition rates maintain the transfer of mass from the smaller to the larger grain as the time increases. As the system evolves towards the equilibrium point, however, it can get trapped if the rate balance condition \( r_{2\rightarrow1}(t) = r_{1\rightarrow2}(t) \) is realized. The resulting equilibrium point corresponds to partial aggregation.

IV. DYNAMIC REGIMES

The system of eqs. (3) allows two different dynamic regimes, determined by the initial conditions, which lead asymptotically to the equipartition and aggregation equilibrium points defined above, as well as a static (trapped) regime. Below, we identify these regimes by theoretical analysis for certain combinations of initial conditions. We also integrate the ODE system numerically by means of the fourth-order Runge-Kutta scheme [17] based on the MATLAB ODE toolbox function ode45, in order to determine the equilibrium point for the entire parameter space.

A. Diffusive Regime

Let us assume that \( m_1(0) \neq m_2(0) \) and \( m_1(0) + m_2(0) < 1 \). These relations are preserved during the evolution of the ODE system. The latter is maintained due to mass conservation. The mass transfer rate remains finite, since the two roots of the transfer rate balance equation \( r_{1\rightarrow2}(t) = r_{2\rightarrow1}(t) \) satisfy \( m_1^* + m_2^* > 1 \) (as shown in Fig. 2) for all possible transition rate levels \( c \), whereas \( m_1(t) + m_2(t) < 1 \). Assuming \( m_1(0), m_2(0) \ll 1 \), the ODE system of eqs. (3) is approximated by the linearized equations

\[
\frac{d\hat{m}_1(t)}{dt} = \hat{m}_2(t) - \hat{m}_1(t) \tag{4a}
\]

\[
\frac{d\hat{m}_2(t)}{dt} = \hat{m}_1(t) - \hat{m}_2(t). \tag{4b}
\]

Eqs. (4) conserve mass, as it follows by adding their respective sides. By subtracting the left and right hand sides, respectively, of eqs. (4), it follows that the mass difference \( m_d(t) = \hat{m}_1(t) - \hat{m}_2(t) \), satisfies the ODE

\[
\frac{dm_d(t)}{dt} = -2m_d(t),
\]

with initial condition \( m_d(0) = m_1(0) - m_2(0) \). The above is solved by \( m_d(t) = m_d(0)e^{-2t} \). The respective solutions
for \( m_1(t) \) and \( m_2(t) \) are given by the exponential functions

\[
\hat{m}_1(t) = m_1(0) \left( \frac{1 + e^{-2t}}{2} \right) + m_2(0) \left( \frac{1 - e^{-2t}}{2} \right) \quad (5a)
\]
\[
\hat{m}_2(t) = m_1(0) \left( \frac{1 - e^{-2t}}{2} \right) + m_2(0) \left( \frac{1 + e^{-2t}}{2} \right) . \quad (5b)
\]

The linear approximation remains valid as \( t \) increases because mass conservation implies that \( m_1(t) \) and \( m_2(t) \) are bounded from above by \( m_1(0) + m_2(0) < 1 \). The asymptotic limit of the diffusive solution is the equipartition point. Considering that the activation parameter \( u \) is absorbed in the initial conditions and \( u \propto 1/T \), the diffusive solution is favored by higher temperature.

In Fig. 3 we compare the numerical solutions of eqs. (3) with the explicit solutions of the linearized approximation eqs. (4). There is general agreement between the two solutions, both of which converge to the equipartition point. The linear approximation, however, converges faster to equilibrium than the numerical solution. This is due to overestimation of the magnitude of mass transfer rates by the linear approximation.

The solution of eqs. (6) is given by

\[
\hat{m}_1(t) = m_1(0) + m_2(0)(1 - e^{-t}), \quad (7a)
\]
\[
\hat{m}_2(t) = m_2(0)e^{-t}. \quad (7b)
\]

The above solution predicts growth of the larger and decay of the smaller grain. The mass of the growing grain reaches a plateau determined by mass conservation, as shown in Fig. 4. The solution of the ODE system (5), however, does not achieve this plateau if trapping occurs at some finite \( t \). In Fig. 4 we compare the numerical solution of eqs. (3) and the theoretical solutions of the linearized approximation, i.e., eqs. (6). The difference between the two solutions is due (i) to the linearized approximation of the Arrhenius transition rates and (ii) to trapping of the full solution due to mass transfer balance. Trapping is not captured by the linearized eqs. (6).

C. Static Regime

If \( m_1(0), m_2(0) \gg 1 \) the system is essentially frozen in the initial state, because the transition rates \( r_{1 \rightarrow 2}(t) \) and \( r_{2 \rightarrow 1}(t) \) are nearly zero. This also occurs for mass distribution, i.e., if \( m_1(0) = m_2(0) = m_0 \) for all \( m_0 \). In addition, the static regime is reached if the trapping point is realized. An extreme case of trapping is shown in Fig. 5: the initial masses, \( m_1(0) = 1.66 \) and \( m_2(0) = 0.54 \), correspond to mass transfer rates with magnitude \( \approx 10^{-3} \), so that the system is essentially frozen and the grain masses essentially remain unchanged. The linearized grain coarsening approximation, however, is insensitive to this effect and predicts mass transfer from the smaller to the larger grain.

B. Grain-coarsening Regime

If \( m_1(0) \gg 1, m_2(0) \ll 1 \) or \( m_2(0) \gg 1, m_1(0) \ll 1 \), we can approximate the system of eqs. (3) by means of the following asymmetric, mass-conserving, linearized approximation

\[
\frac{d\hat{m}_1(t)}{dt} = \hat{m}_2(t) \quad (6a)
\]
\[
\frac{d\hat{m}_2(t)}{dt} = -\hat{m}_2(t). \quad (6b)
\]
FIG. 5. Numerical solutions $m_{1}(t), m_{2}(t)$ of the system of eqs. (3) (dotted lines) with initial conditions $m_{1}(0) = 1.66$ and $m_{2}(0) = 0.54$ (trapped state) versus the explicit solution (dots), $\tilde{m}_{1}(t), \tilde{m}_{2}(t)$, given by eqs. (7) of the approximate ODE system (6).

V. PHASE DIAGRAM

The phase diagram of Fig. 6 displays the equilibrium point of eqs. (3) for all points of the parameter plane. Since the total mass is conserved, the crucial state variable is the mass difference, which obeys the following equation

$$\frac{dm_{d}(t)}{dt} = 2 \left[ r_{2\to1}(t) - r_{1\to2}(t) \right].$$

(8)

The initial mass difference is (i) amplified in the grain coarsening regime (ii) reduced in the diffusive regime or (iii) maintained in the static regime. The equilibrium mass difference, $m_{d}(\infty) = \lim_{t \to \infty} m_{d}(t)$, is derived from the numerical solution of eqs. (3) after 10 000 time steps, which is sufficient for $|dm_{d}/dt| \approx 0$ for all $m_{1}(0), m_{2}(0)$ examined. As Fig. 6 shows, in the diffusive regime (lower left), the mass difference tends to zero. In the grain coarsening regime (upper right), one grain concentrates most of the mass. The discontinuous change of the equilibrium mass difference about the main diagonal (upper right corner) is triggered by a small change in the initial conditions: the system moves from the equipartition point (static regime) which extends along the main diagonal to large mass differences (negative above and positive below the diagonal) in the adjacent grain coarsening regime. The curvilinear trace formed by “star” pointers which lie mostly inside the grain coarsening regime marks the trapping boundary.

We plot the time evolution of the masses $m_{1}(t)$ and $m_{2}(t)$ in Figs. 7 and 8 respectively. The plots are generated for fixed $m_{2}(0) = 0.5$ whereas $m_{1}(0)$ varies between 0 and 3. As shown in Fig. 7 for $m_{1}(0) < 0.5, m_{1}(t)$ increases with $t$ marking the diffusion of mass from $m_{2}(t)$ to $m_{1}(t)$. The process is much slower for $m_{1}(0) \approx 0.42$ because the system is close to the static point $(0.5, 0.5)$. For $0.5 < m_{1}(0)$ and $m_{1}(0) \lesssim 1.71, m_{1}(t)$ declines with $t$ due to mass diffusion towards the initially smaller grain. In contrast, for $m_{1}(0) > 1.71, m_{1}(t)$ grows with $t$ again, marking the entrance in the grain coarsening regime. The dependence of $m_{2}(t)$ is diffusive for $m_{1}(0) \leq 1.71$: $m_{2}(t)$ declines for $m_{1}(0) < m_{2}(0)$ and grows for $m_{1}(0) > m_{2}(0)$. For $m_{1}(0) > 1.71$ the system is in the grain coarsening regime, and since $m_{1}(0) > m_{2}(0), m_{2}(t)$ decays by transferring mass to $m_{1}(t)$.

FIG. 6. Difference $m_{d}(t)$ of grain masses at equilibrium as a function of the initial conditions in $[0, 3] \times [0, 3]$. The horizontal and vertical axes denote the initial grain masses, $m_{1}(0)$ and $m_{2}(0)$, respectively. The shading (color online) denotes the mass difference $m_{d}(\infty)$ at equilibrium at the nodes of a $100 \times 100$ grid. Equilibrium values are based on numerical solutions of the ODE system (3) after 10 000 time steps.

FIG. 7. Evolution of $m_{1}(t)$ based on numerical solution of eqs. (3) for ten different values of $m_{1}(0)$ (shown in the legend truncated to second decimal) and $m_{2}(0) = 0.5$. 
The trapping effect is not transparent in Figs. 7-8. To better illustrate it, we plot the trajectories \((m_1(t), m_2(t))\) in Fig. 9. Due to the mass conservation constraint, the trajectories obey the linear relation \(m_2(t) = m_1(0) + m_3(0) - m_1(t)\). Hence, \(m_1(t)\) and \(m_2(t)\), obey decoupled first-order ODEs which, however, are nonlinear and can not be solved analytically. For the two lowest values of \(m_1(0)\) the system behaves diffusively with \(m_1(t)\) growing and \(m_2(t)\) shrinking. For the next three values of \(m_1(0)\), \(m_1(t)\) declines and \(m_2(t)\) grows. The next value, \(m_1(0) \approx 1.71\), also leads to diffusive behavior which is arrested at the trapping boundary. The trapping of diffusive solutions was not captured by the assumption in the linearized approximation of the diffusive regime. Finally, values of \(m_1(0) > 2.03\) lead to grain coarsening which stops at the trapping boundary.

VI. STOCHASTIC TRANSITION RATES

Trapping can be undesirable in certain cases, because it impedes the grain coarsening process. One possibility to escape trapping is by adding noise to the ODE system (3), leading to

\[ \frac{dm_1(t)}{dt} = r_{2 \rightarrow 1}(t) - r_{1 \rightarrow 2}(t) - \eta(t), \]
\[ \frac{dm_2(t)}{dt} = r_{1 \rightarrow 2}(t) - r_{2 \rightarrow 1}(t) + \eta(t), \]

where \(\eta(t)\) is Gaussian white noise with zero mean, i.e., \(\langle \eta(t) \rangle = 0\), and correlation function \(\langle \eta(t) \eta(t') \rangle = \sigma^2 \delta(t - t')\), \(\sigma^2\) being the noise variance. Eqs. (9) conserve the total mass at all times due to the opposite signs of the noise terms. If at \(t > 0\) transfer rate balance is reached, it will almost surely be perturbed by stochastic fluctuations. The mass difference satisfies the equation

\[ \frac{dm_4(t)}{dt} = 2[r_{2 \rightarrow 1}(t) - r_{1 \rightarrow 2}(t)] + 2 \eta(t). \]

If \(r_{2 \rightarrow 1}(t), r_{1 \rightarrow 2}(t) \approx 0\), \(m_4(t)\) is essentially driven by a white noise process and therefore behaves as the classical Brownian motion [18]. In the grain coarsening regime, if the linearized approximation holds, the mass of the second grain based on eqs. (6) satisfies

\[ \frac{d\tilde{m}_2(t)}{dt} = -\tilde{m}_2(t) + \eta(t), \]

which is the Ornstein-Uhlenbeck (O-U) process [19]. The mass of the first grain, on the other hand, is given by the integral of the O-U process. Numerical solutions for both stochastic processes are given in [20].

VII. CONCLUSIONS

We investigated a nonlinear ODE system that describes mass exchange between two grains. The exchange is governed by Arrhenius-type transition rates with an activation energy that has linear mass dependence. The activation parameter that controls the transition rates, however, can be absorbed in the initial conditions. We identified the equilibrium points of the coupled nonlinear ODE system, and we derived linearized ODE approximations which are valid in parts of the diffusive ("high temperature") and grain coarsening ("low temperature")
regimes respectively. The linear approximations provide qualitative understanding of the nonlinear ODE system, which is more accurate in the diffusive regime. In the grain coarsening regime, however, the linear approximation misses the trapping effect. Perturbation analysis of the ODE system in the neighborhood of the trapping boundary is possible but not pursued herein. We also construct the equilibrium phase diagram of the model based on numerical solutions of the ODEs.

We proposed that trapping can be mitigated by adding a stochastic term which represents random fluctuations of mass transfer rates. More detailed analysis is needed to elucidate the system’s evolution of the system under stochastic forcing. We do not expect the trapping effect to be as important in multigrain systems as in the two-grain system, since in the former the static state requires rate balance for all interacting grains.

Based on the analysis above, we suggest that the grain coarsening regime is related to abnormal grain growth such as observed in sintering. Multigrain systems as well as grain coalescence, which are important for sintering applications, are being investigated in ongoing research by our group. Finally, our model could be useful as a component of kinetic models of wealth exchange.

In this context, the activation parameter is proportional to the average wealth of the agents (grains), whereas mass conservation corresponds to total wealth conservation. The existence of two distinct regimes, one corresponding to equipartition of wealth and the other to wealth accumulation by one agent is an intriguing feature of the model.

VIII. ACKNOWLEDGMENTS

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