Numerical Investigation of Grain Coarsening and Coalescence Model

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Abstract. A kinetic nonlinear model of mass transfer, grain coarsening and coalescence with potential applications in sintering processes is studied. The model involves nonlinear differential equations that determine the transport of mass between grains. The rate of mass transfer is controlled by the activation energy (an Arrhenius factor) leading to a nonlinear model of mass transfer and grain coarsening. The resulting dynamical system of coupled nonlinear differential equations with random initial conditions (i.e., initial grain mass configuration) is solved by means of the Runge-Kutta method. An analysis of the fixed points of the two-grain system is carried out, and the solution of the multi-grain system is studied. We incorporate coalescence of smaller grains with larger neighbors using a cellular automaton step in the evolution of the system.

1. Introduction
Many technological materials, including ceramics, are produced by means of nonequilibrium physical processes that generate phase changes. A typical example is sintering, a process during which a powder is transformed into a monolithic material through atomic diffusion mechanisms. The modeling of sintering is a long-standing problem which remains poorly understood to date. Early studies of the kinetics of phase change have focused on interactions between grains that produce a crystal aggregate [1, 2]. Recent approaches use the Discrete Element Method (DEM) to relax assumptions regarding the particle kinematics [4]. In the above, grain coarsening is accounted by considering the overlapping volume of neighboring spherical particles, increasing the radius of the larger particle while decreasing the radius of the smaller by the same amount.

Here we investigate numerically the kinetic, nonlinear model of mass transfer, grain coarsening, and coalescence introduced in [3]. The model comprises a system of nonlinear ordinary differential equations (ODEs) and incorporates local fluctuations in mass transfer rates due to variations in the degree of amorphization of the grains. The initial conditions for the system are the grain masses which are considered to be randomly distributed. The initial value problem is solved by means of the fourth order Runge-Kutta method. The ultimate goal is to use the model in order to predict changes in the grain size distribution as a function of key sintering parameters such as time and temperature, as well as the powder grain size distribution and the non-homogeneity of the transfer rates.
2. Nonlinear kinetic model
A collection of spherical grains such that the surface of each grain (particle) is in contact with at least one other particle is considered. We study a one-dimensional geometry in which \( N \) grains are assumed to be located along a line (the \( x \)-axis), and each grain has two neighbors, one on each side, except for the two boundary grains. The grains are indexed by \( i \) (\( i = 1, 2, \ldots, N \)). Taking into account that the first grain \((i = 1)\) does not have a left neighbor and the last grain \((i = N)\) does not have a right neighbor, we rewrite the original model ([3]) as the following system of ODEs:

\[
\begin{align*}
\dot{m}_1(t) &= e^{-um_2(t)}m_2(t) - e^{-um_1(t)}m_1(t), \\
\dot{m}_i(t) &= e^{-um_{i-1}(t)}m_{i-1}(t) + e^{-um_{i+1}(t)}m_{i+1}(t) - 2e^{-um_i(t)}m_i(t), \quad i = 2, 3, \ldots, N - 1, \\
\dot{m}_N(t) &= e^{-um_{N-1}(t)}m_{N-1}(t) - e^{-um_N(t)}m_N(t).
\end{align*}
\]

In the above, \( m_i(t) \) is the mass of \( i \)-th grain, \( t \) is time, \( N \) is the number of grains and \( u \) represents the normalized grain activation energy. For the mass of \( i \)-th grain we suppose \( m_i(t) = l d \pi r_i^2(t) \rho \), where \( r_i(t) \) is the radius of \( i \)-th grain and \( \rho \) is the mass density of the grains (assumed to be constant); for spherical grains \( d = 3 \) and \( l_d = 4\pi/3 \).

Equations (1) and (3) correspond to the mass transfer process for the boundary grains \((i = 1, N)\). Equations (2) describe the mass transfer process for the grains in the bulk. We assume that the initial grain masses, i.e., the initial conditions for (1)-(3) are random numbers,

\[ m_i(0) = m_{i0}, \quad i = 1, 2, \ldots, N. \] (4)

The initial conditions can be drawn from the normal, lognormal or some empirically determined probability distribution.

If we assume that the grains are arranged along a ring with cyclical boundary conditions so that \( m_0(t) := m_N(t), \quad m_{N+1}(t) := m_1(t) \), then the original model (1)-(3) is written in the following simpler form, whereas the final result of the simulation is not significantly affected by the change,

\[
\dot{m}_i(t) = e^{-um_{i-1}(t)}m_{i-1}(t) + e^{-um_{i+1}(t)}m_{i+1}(t) - 2e^{-um_i(t)}m_i(t), \quad i = 1, 2, \ldots, N. \] (5)

3. Analysis of the two-grain system
In this section we focus on the case \( N = 2 \) and present the results of numerical analysis, which will be helpful for investigating systems with a large number of grains. The initial masses are considered to take values at the nodes of a square lattice \([a, b] \times [a, b]\). According to the numerical experiments, a diffusive regime was observed for \( a \leq 1, \quad b \leq 1 \): the two masses converged to equal values for all \( u \leq 1 \). These results were verified with lattice steps \( h = 0.1, 0.02, 0.005 \). In general, smaller \( u \) values lead to faster convergence. The mass growth regime, i.e., the accumulation of mass on the larger grains and reduction of the smaller, is obtained at large values of \( u \). The critical value of \( u \) depends on the initial masses. For example, if \( u = 0.1 \) the mass growing regime sets at \( m_1(0) \approx 8.5 \) and \( m_2 \approx 12 \) (and vice versa); \( u = 1 \) requires \( m_1(0) \approx 0.85 \) and \( m_2 \approx 1.2 \) and so on. In general, higher values of \( u \) imply that smaller initial mass values lead to the mass growth regime. However, for large \( u \), e.g. approximately \( u_1 m_1(0), \quad u_2 m_2(0) > 10 \) the initial masses change extremely slowly.

For every initial distribution of grain sizes there exists a critical \( u \) (activation energy) that leads to mass growth. For some particular situations the masses change very slowly and a many time steps will be needed for convergence to a final state. By virtue of the numerical results and in light of the equations (1)-(3) (or for the system of equations (5)) it follows that very small or very large values of \( u \) lead to almost stationary processes; for small \( u \) the grains tend to equal mass distribution, whereas for large \( u \) the masses are practically “trapped” at their initial values.
4. Computational algorithm for grain mass transfer
The kinetic model (1)-(3) with the initial conditions (4) is discretized using the fourth order Runge-Kutta method (RK4). An analogous scheme is obtained for the system of equations (5) with the initial conditions (4). For the numerical implementation of the RK4 scheme we use the Matlab ordinary differential equation toolbox, function (*ode45 solver*). The statistical analysis of the resulting grain distributions is based on an ensemble of simulated states.

Below we describe an algorithm for solving the kinetic model including the possibility for grain coalescence.

*Grain mass transfer simulation algorithm*

(i) Set the simulation counter to one.

(ii) Choose an initial configuration of grain masses from a specified probability distribution (e.g., normal, lognormal or empirical distributions).

(iii) Solve (1)-(3), or equivalently (5), with the initial conditions (4) by means of a suitable numerical method (e.g. RK4).

(iv) Check the neighboring mass ratios and implement grain coalescence if needed. Namely, if \( m_i / m_{i+1} < \delta \), where \( 0 < \delta < 1 \) is the coalescence parameter, then the \((i+1)\)-th grain becomes \( m_{i+1} + m_i \) whereas the \(i\)-th grain is removed. If \( m_i / m_{i+1} > \delta \) the \(i\)-th grain accumulates the mass of both grains whereas the \((i+1)\)-th grain is removed.

(v) Reduce the number of the equations in the system (1)-(3), or equivalently (5), by the number of grains removed according to the rules in step (iv).

(vi) Check if the updated number of grains is greater than 1: If it is, go to the next step; otherwise, the simulation terminates.

(vii) Check the number of iterations: If it does not exceed the specified limit go to the next step; otherwise, the simulation terminates.

(viii) Solve the system of equations with the updated number of equations (grains).

(ix) Repeat steps (iv)-(viii) until the simulation stops. Increase the simulation counter by one.

(x) If the value of the simulation counter is less than the specified number of simulations go to step (ii); otherwise stop and calculate statistics.

5. Numerical simulation of multi-grain system
In this section we present numerical simulation results for the equation system (1)-(3), (4) for \( N = 10\,000 \) grains, different values of \( u \) and \( \delta \). We have verified that the numerical solution of the system conserves the total grain mass.

*Example 1.* Figure 1 shows the histogram of the initial masses which are drawn from the normal (Gaussian) distribution with mean \( \mu = 10 \) and standard deviation \( \sigma = 2 \). The maximum number of time steps used is 5,000, and the size of the time step is \( \tau = 0.02 \).

Figure 2 demonstrates the final mass distributions without coalescence. The plots show that small values of \( u \) lead to a diffusive regime which implies convergence of all the masses to the mean of the initial distribution (e.g., see plot 1 with \( u = 0.05 \)). The next higher values of \( u \) lead to growth of some grains at the expense of other grains becoming smaller (Plots 2-6 with \( u = [0.08, 0.1, 0.3, 0.6, 1] \)). Finally, higher values of \( u \) lead to a quasi-static system in which the “final” state does not differ significantly from the initial histogram (Plots 7-9).

Figure 3 demonstrates the “final” mass distributions with coalescence \( (\delta = 0.1) \) after 5,000 steps. The number of remaining grains and the end of the procedure is \( N_{\text{final}} = \{10\,000, 21, 1, 3166, 6036, 9375, 9995, 9998, 9999\} \). The plots of Figure 3 exhibit mass growth for \( u = [0.08, 0.1, 0.3, 0.6, 1] \). For \( u = 0.05 \) the diffusive final state is observed, whereas for \( u = 2, 3, 4 \) the final masses are essentially unchanged with respect to the initial conditions.
Figure 1. Histogram for initial configuration of grain masses for a system comprising $N = 10,000$ grains drawn from the $N(0, 2)$ distribution.

Figure 2. Mass distribution of system comprising $N = 10,000$ grains without coalescence after 5000 steps. The values of the activation energy are $u = [0.05, 0.08, 0.1, 0.3, 0.6, 1, 2, 3, 4]$.

Figure 3. Mass distribution of system comprising $N = 10,000$ grains with coalescence parameter $\delta = 0.01$ after 5000 steps. The values of the activation energy are $u = [0.05, 0.08, 0.1, 0.3, 0.6, 1, 2, 3, 4]$. 
Example 2. Here we investigate the system with different values of the ratio $\delta$. Figure 4 shows the mean value of the grain radius at the final time (one check after 1000 time steps).

![Figure 4](image)

**Figure 4.** The dynamics of changing of the mean value of the radius of grains for $u = 0.05, 0.08, 0.09, 0.1, 0.15, 0.2, 0.3, 0.4, 0.5, 0.8, 1, 3$ with $\delta = \{0.1, 0.05, 0.005\}$ and the initial normal distribution ($\mu = 10, \sigma = 2$) after 5000 time steps.

6. Conclusions
Numerical simulations of a system of coupled, nonlinear ordinary differential equations that describe mass transfer between grains of inhomogeneous sizes have been presented. Three different types of behavior have been identified: a diffusive regime in which all the grains tend to the same mass in equilibrium, a growth-decay regime in which the bigger grains grow whereas the smaller ones lose mass, and a trapping regime in which the initial mass distribution practically does not change. The parameters that control these regimes involve the activation energy, the coalescence ratio and the initial mass distribution of the grains.

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