Built on LDRD Laboratory Directed Research and Development

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LDRD PROJECT TITLE: Peridynamic Theory as a New Paradigm for Multiscale Modeling of Sintering
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ABSTRACT

Sintering is a component fabrication process in which powder is compacted by pressing or some other means and then held at elevated temperature for a period of hours. The powder grains bond with each other, leading to the formation of a solid component with much lower porosity, and therefore higher density and higher strength, than the original powder compact. In this project, we investigated a new way of computationally modeling sintering at the length scale of grains. The model uses a high-fidelity, three-dimensional representation with a few hundred nodes per grain. The numerical model solves the peridynamic equations, in which nonlocal forces allow representation of the attraction, adhesion, and mass diffusion between grains. The deformation of the grains is represented through a viscoelastic material model. The project successfully demonstrated the use of this method to reproduce experimentally observed features of material behavior in sintering, including densification, the evolution of microstructure, and the occurrence of random defects in the sintered solid.

INTRODUCTION

Sintering is a widely used process for the fabrication of ceramic and metal parts. It consists of the heating of powder compacts to about 50% to 60% of the melting temperature. The size of the powder grains is typically 1μm to 50μm. In sintering processes, the powder is compacted mechanically before the temperature is raised. Fluids and other materials are sometimes introduced as aids to the sintering process. Typically, the entire process takes a few hours.

There is inherent variability in the initial distributions of the sizes and shapes of the powder grains. There is also a random character to their mutual contact and placement. Sintered components can therefore suffer from defects that form due to heterogeneous powder packing. These defects are generally voids, although cracks can also occur. They can be of sufficient size to affect the reliability and performance of the finished part, particularly in brittle materials such as ceramics. It is therefore important to be able to understand and predict in detail how these defects form.

The fundamental physical processes, such as the diffusion and transport of mass and voids, can be modeled at the interatomic length scale using molecular dynamics. However, this approach is not feasible at the length scale of grains because of the huge computational effort that would be involved. Conversely, sintering can be modeled at the geometrical length scale of the finished part by smearing out all the grains into a homogenized continuum model. However, this approach does not give sufficient insight into the grain-level processes that lead to defect formation.

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It this study, we performed initial theoretical and software development of a computational model to simulate the sintering process. The method is based on the peridynamic theory of mechanics. In contrast to the traditional theory of continuum mechanics, the peridynamic method allows representation of long-range forces that cause grains to attract and deform. The peridynamic method also allows the incorporation of the mechanical response of individual grains to these attractive forces. The attraction between grains and their resulting merging and deformation lead to densification.

The goals of the project were to provide a theoretical basis and computational demonstration of the application of the peridynamic method to sintering. This consisted first of developing a bond force expression for peridynamic bonds within individual grains and between grains, as described below. It also consisted of determining a suitable material model for the grain deformation as a function of time, temperature, and the internal forces within grains. The project also required development of ancillary capabilities, including

- Models for the initial random packing of grains with a distribution of sizes,
- Evaluation of porosity in a model of a large sample of grains,
- Postprocessing and visualization.

These features will not be described in detail in this report.

DETAILED DESCRIPTION OF EXPERIMENT/METHOD

The distinguishing feature of peridynamics that makes it potentially suitable for application to sintering is that it uses bond forces [1]. Bonds are force interactions between material points separated from each other by finite distances. These bond forces are applied only within some cutoff distance, called the *horizon*, which is a small fraction of the grain size. The peridynamic description is therefore different from the traditional continuum mechanics approach which allows forces only between material particles separated by infinitesimal distances.

Microstructural evolution in sintering is driven by the reduction in surface energies associated with interfaces between grains and with free surfaces. By suitable characterization of the bond forces, the peridynamic continuum model reproduces these surface energies. To explain how surface energies arise from bond forces, consider the schematic diagram in Figure 1. Bond energies contain a contribution from a nonlinear potential that depends only on the deformed lengths of bonds and whether their endpoints belong to the same grain or a different grain. Because systems tend to reduce their potential energy over time, the bonds would tend to approach a length near the minimum of the potential well if there were no other constraints or interactions present.

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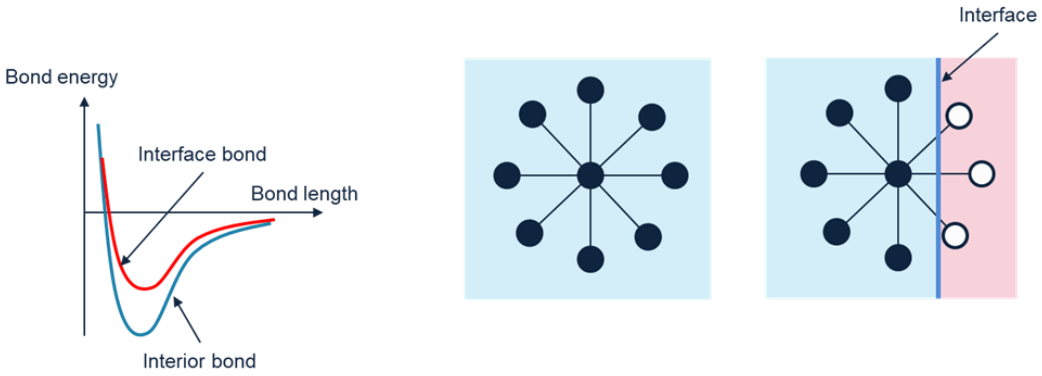

Figure 1. The form of the peridynamic bond potential energy results in free surfaces and interfaces having higher energy than a homogeneous body.

In the interior of a large volume of homogeneous material, most of the bonds connected to some material point have negative potential energy. Near a free surface, some of the bonds are missing and therefore have zero potential energy. Therefore, a configuration of material with more free surface has higher energy than a configuration with less free surface. Thus, a system will tend to reduce its surface area over time.

Similarly, bonds that extend between grains have minima in their bond energies that are slightly higher than bonds that are entirely within a single grain. Therefore, a grain boundary has surface energy, but this surface energy is less than that of a free (pore) surface. The minimum energy state of the system of grains is determined by the need to reduce the total surface energy, comprised of both pore surface energy and grain boundary surface energy. The two types of surface energy compete with each other, resulting in grain shapes that are not perfectly spherical.

The parameters involved in the bond potential model determine the strength of the force interactions that lead to surface and grain boundary energy. An important step in this project was the derivation of explicit expressions for these parameters to match an experimentally measured value of surface energy. Thus, the only inputs for the peridynamic sintering model, aside from the material properties of grains, are the pore surface and grain boundary energies per unit area. The availability of an equation to set the bond parameters avoids the need to perform calibration runs to evaluate them and is therefore a significant result.

For a bond contained entirely within a single grain, assume that the potential energy as a function of the current bond length r has the form

$$w(r) = -w_0 \sigma(r/\delta)$$

where σ is a dimensionless function, w_0 is a constant, and δ is the peridynamic horizon. The calibration in terms of pore surface energy γ_{ps} is carried out by integrating the bond potential

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across a small patch of area on a pore surface. Carrying out the computations leads to an explicit expression for w_0:

$$w_0 = \frac{2\gamma_{ps}}{\pi\delta^4 \int_0^\infty \rho^3 \sigma(\rho) d\rho}.$$

Similarly, bonds that have their two endpoints in different grains are assumed to have their potential energy given by

$$w(r) = -w'_0 \sigma\left(\frac{r}{\delta}\right).$$

The constant w'_0 is then found from

$$w'_0 = w_0 - \frac{2\gamma_{ps} - \gamma_{gb}}{\pi\delta^4 \int_0^\infty \rho^3 \sigma(\rho) d\rho}$$

where γ_{gb} is the grain boundary surface energy. As the grains deform, the configuration of the pore surfaces and grain boundaries changes. These changes are automatically incorporated into the overall model of the system because the bonds generating the surface interactions change according to the deformation. A material particle that starts on a pore surface or in the interior of a grain may end up along a grain boundary due to the evolution of the system. The interaction between this material particle and its new neighbors in another grain is accounted for through the formation of new bonds.

The deformation and rearrangement of the grains to achieve minimum surface energy is resisted by the finite rate of mass diffusion. This rate of diffusion increases exponentially with temperature per the Arrhenius relationship. In the peridynamic model, the diffusive nature of mass transport is represented with a viscoelastic material model. The time dependence of the response of a grain to the surface forces is expressed in terms of stress relaxation, which is essentially the same as creep. The material in the grains has constant bulk modulus, but its shear modulus decreases over time as shown in Figure 2. The temperature dependence of the rate of stress relaxation is assumed to follow an Arrhenius law.

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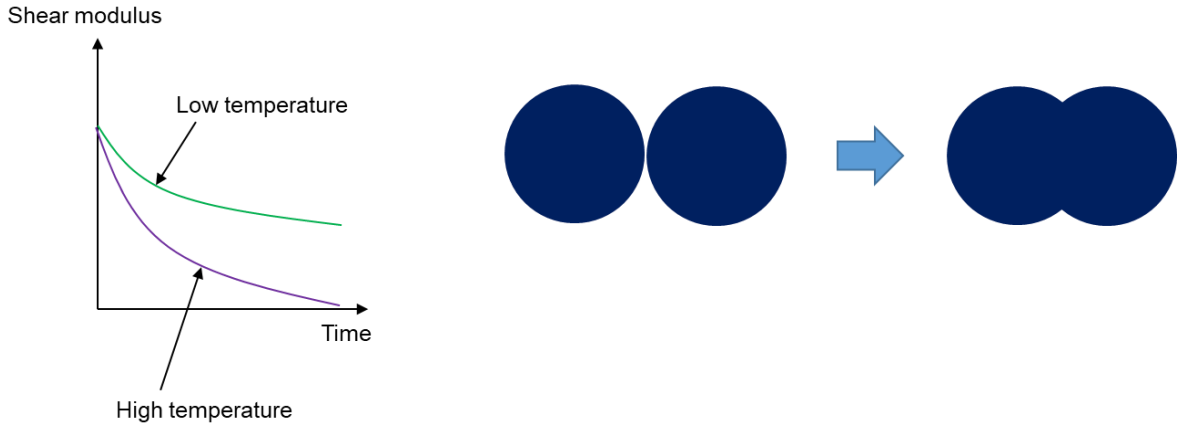

Figure 2 A viscoelastic material model within grains reproduces the time- and temperature-dependent nature of the diffusive transport processes that control sintering.

In the peridynamic sintering model, the grain material is a linear peridynamic solid (LPS) with constant bulk modulus and time-dependent shear modulus as described above. The LPS material model is a standard ordinary state-based material model that allows both the bulk modulus and shear modulus to be prescribed independently of each other [2].

The continuum equations of peridynamics combine the bond force contributions from the surface effects and the deformation of the individual grains. These equations are solved computationally within the meshless peridynamic code Emu. Emu reads an initial grid containing the discretized grains and predicts the motion of the grid over time. Emu uses an explicit solver that is effective in highly nonlinear problems such as this. The solver, when used with a viscoelastic material model, drives the system toward lower potential energy in equilibrium. The use of an explicit solver involves a restriction on the time step, requiring the scaling of the time variable in the relaxation of the shear modulus shown in Figure 2. Since the stable time step is usually less than a microsecond, the simulation time is mapped onto a much longer time scale (hours) that is applied in the material model. This scaling does not affect the results provided that the inertial term in the momentum balance is negligible compared with the other forces arising from the surface terms and the material model. This condition, which is known as the *quasi-static* condition, is easily met in practice by applying damping forces that resist wave-like oscillations.

RESULTS

Our results consist of computer simulations that demonstrate the ability of the peridynamic sintering model to reproduce known behavior in real systems. To show that the method predicts realistic results with simple geometries, we initialized a grid representing two spherical grains of copper, each with a diameter of 50μm. The spheres are initially in contact with each other. The

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pore surface energy and grain boundary energy are 1.65 J/m^2 and 1.30 J/m^2 respectively. The meshless grid used 3624 nodes in each grain. The final configuration predicted by Emu is shown in Figure 3. The exact value for the angle between the grains in the equilibrium solution to this problem is 78 degrees, as shown in the figure. This value is derived from a balance between the pore surface forces and grain boundary surface forces on the corner points where they intersect. The computer simulation does not reproduce this value exactly because of the finiteness of the grid spacing. It is not possible for the grid to accommodate a sharp angle at the interface, so it produces an approximate solution that has a gradual curvature. Farther away from the corner point, the numerical model provides a reasonable approximation to the exact 78 degree angle.

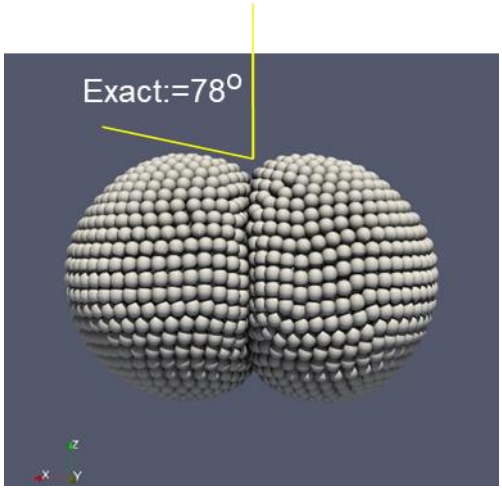

Figure 3. Predicted shape of two spherical copper powder grains after sintering. The exact angle, based on a balance between surface forces, is also shown.

The results of a simulation with four spherical copper grains within initial diameter 50μm in a tetrahedral configuration are shown in Figure 4. The cross-sectional view shows the reduction in pore space due to sintering.

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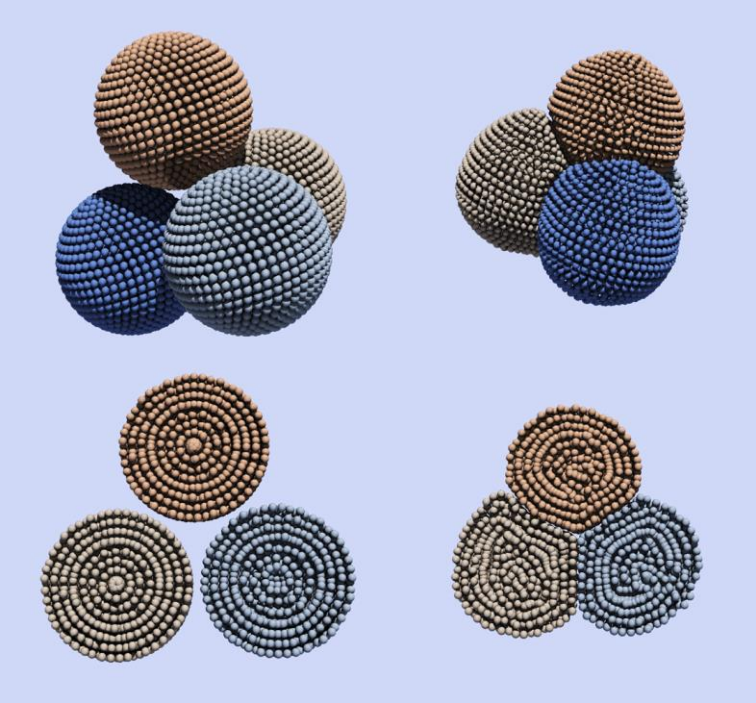

Figure 4. Peridynamic model of the sintering of four spherical grains in a tetrahedral configuration.

A simulation with 1334 copper grains with an initial random distribution of diameters from 10μm to 50μm is shown in Figure 5. The sample is subjected to an initial mechanical compression of about 25%, reducing the initial porosity. Sintering reduces the remaining pore space to about 5%. As shown in the figure, the computed microstructure resembles experimentally observed microstructures in copper. A view from the outside of the sample is shown in Figure 6, illustrating the deformed and distorted shapes of the grains.

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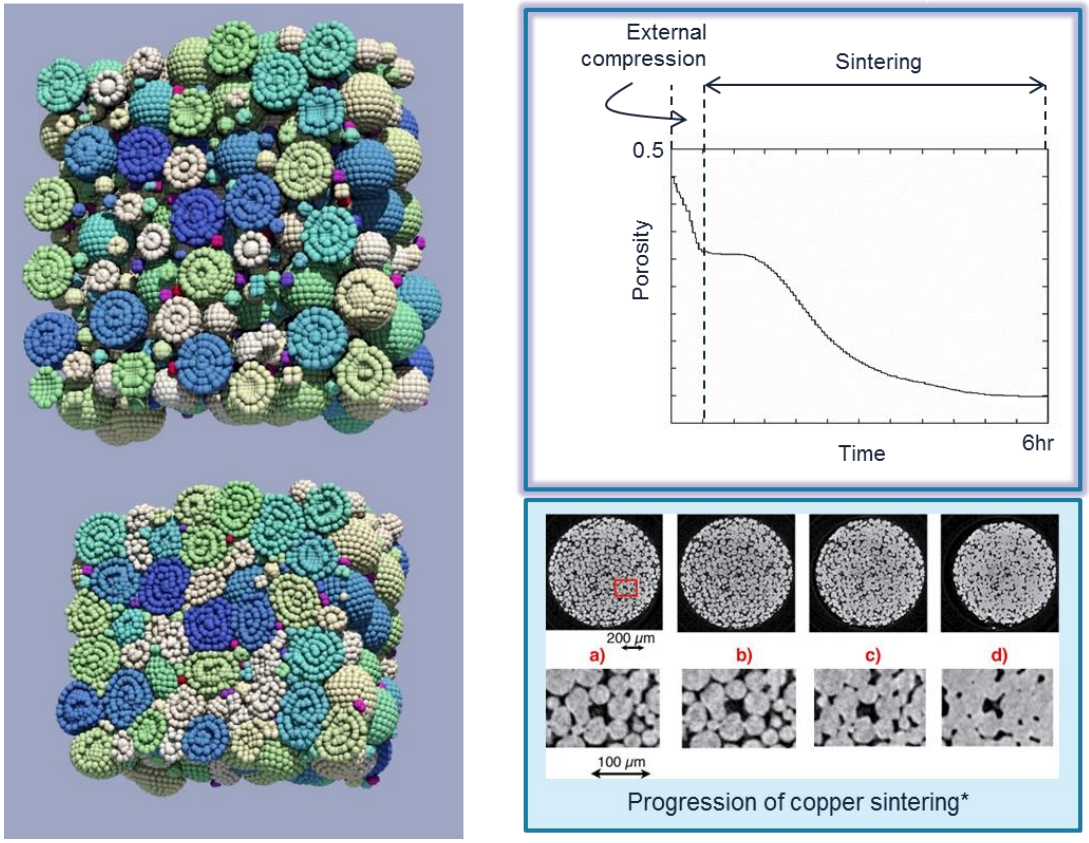

Figure 5. Model of copper sintering with initial mechanical compression.

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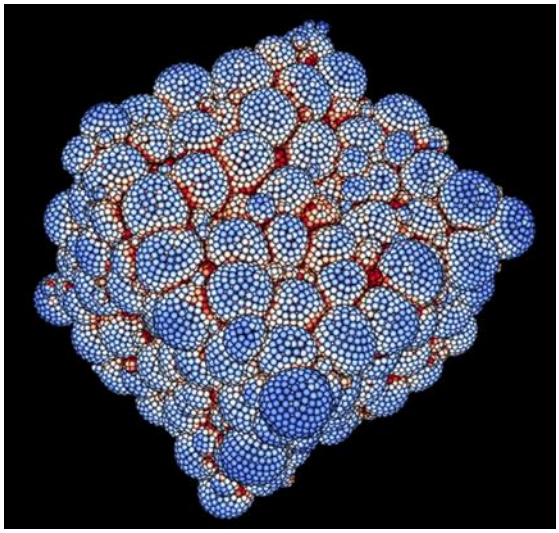

Figure 6. Outside view of a peridynamic model of sintered copper.Colors indicate energy density due to interactions between grans.

DISCUSSION

A key purpose of computational simulation of sintering is to gain insight into the formation of defects, particularly voids. In real sintering processes, usually the desired outcome is to squeeze out all the pore space and create a finished material with as little porosity as possible. However, sometimes the pre-existing voids, rather than disappearing, merge with each other and create larger voids. The details of this sequence of events and the conditions that can mitigate it are of fundamental interest. An example of a peridynamic computational simulation that exhibits this defect formation is shown in Figure 7 on the right. In this simulation, there is no prelimary mechanical compression. However, in the image on the left, initial mechanical compression before sintering helps to avoid the defect in the sintered material.

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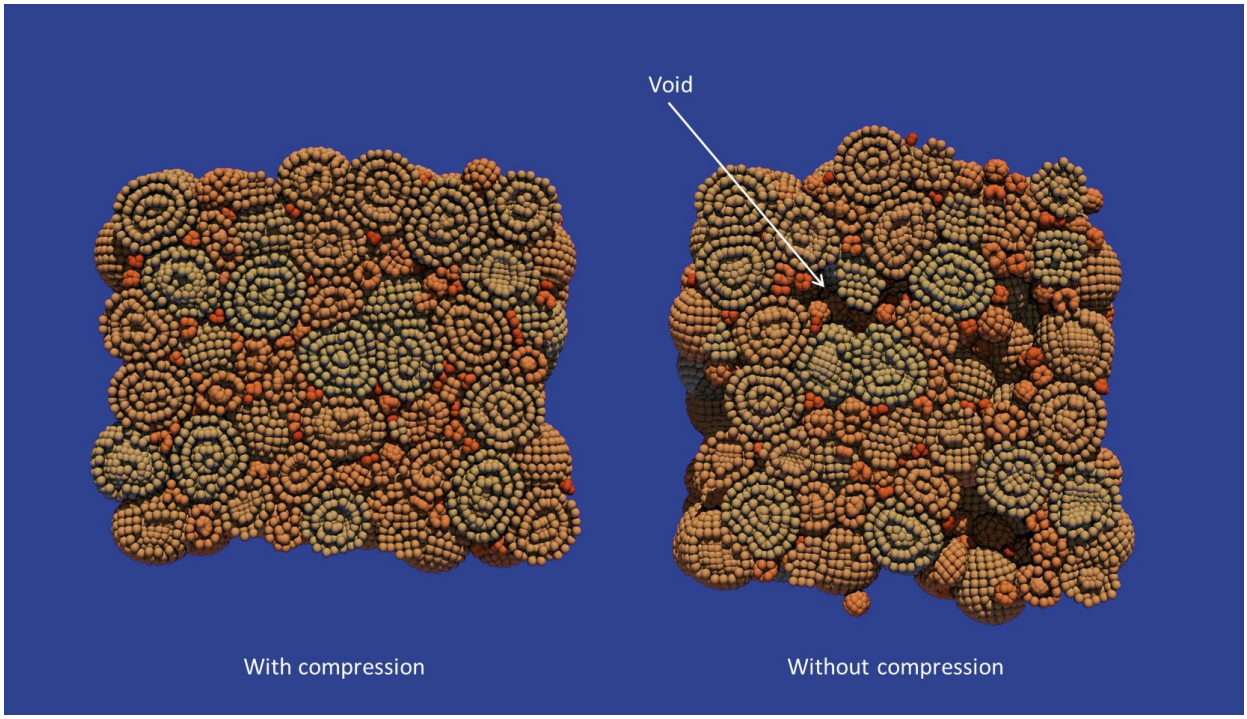

Figure 7. Predicted sintering of copper with and without initial mechanical compression.

The use of a statistically significant number of grains is crucial in understanding the nature and distribution of defects. For example, the simulation shown in Figure 8 uses 4096 grains, compared with 1334 in Figure 7. The larger sample gives greater insight into the distribution of large voids than the smaller sample. The larger model had about 1.7 million nodes and used 24 processors for 12 hours. These modest requirements suggest that much larger, and therefore more credible and useful simulations, could be carried out on larger computational platforms such as exascale.

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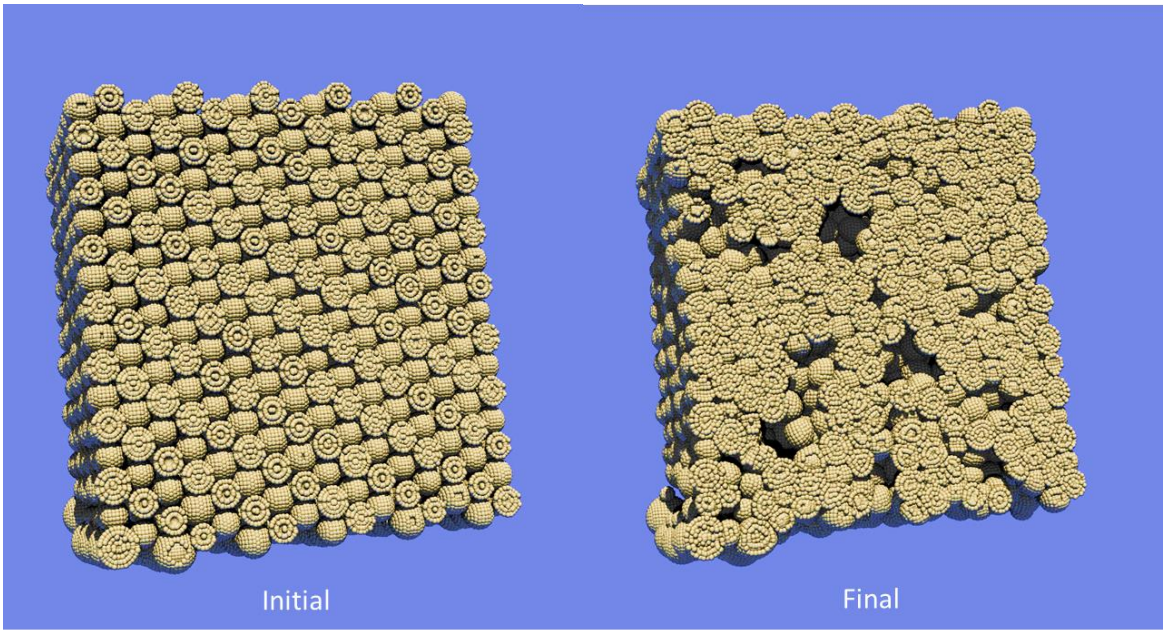

Figure 8. A statistically large sample of grains is required to treat the formation of defects realistically.

ANTICIPATED OUTCOMES AND IMPACTS

This project succeeded in developing a rudimentary theory that expresses surface energy in microstructures in terms of an equivalent mechanical energy through long-range peridynamic forces. It also succeeded in reproducing important qualitative microstructural features that emerge in the sintering of real materials, including certain aspects of defect formation. We anticipate that because of the importance of sintering in the manufacture of weapons components, it should be possible to obtain follow-on funding through a full LDRD project or stockpile programs.

The method developed in this project should also have applications in powder compaction technologies other than sintering. In practice, defects such as density gradients may arise during powder compaction due to frictional effects and other details of processing. The present method, with further development, is expected to offer insight into these processes.

Because of the fundamental importance of surface energy, we anticipate that the present work will find applications in multiscale modeling of material processes beyond sintering. Examples include phase changes and grain growth in metals in conventional metallurgical processes. Another potential way to apply the peridynamic sintering model is through a multiscale approach. In most of the examples discussed in this report, each grain has about 400 nodes. It may be possible to coarsen this detailed model of a grain into a description in which each grain has only six degrees of freedom: a center of mass position vector and three rigid rotation angles. This simplified model, which amounts to a discrete element formulation [3], would involve force interactions with all of

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the neighboring grains that are in contact with it. These force interactions could, in principle, be derived from the more detailed peridynamic model investigated here.

CONCLUSION

In this study, we have developed the essential elements of a peridynamic model for sintering and other powder processes. The main feature is the replacement of a diffusive model for mass transport with a continuum mechanics model in which the diffusive transport is subsumed into a temperature-dependent viscoelastic material model. We showed that the interactions that characterize the bonding of powder grains to each other during sintering can be represented through long-range peridynamic forces. The parameters in the model can be selected to reproduce the surface energy on pore surfaces and grain boundaries. The method appears to provide qualitative agreement with test data on the evolution of microstructures during sintering.

The present model applies only to the early stages of sintering, prior to the coalescence of grains. However, we believe that the model could be adapted to treat the later stages by modifying the force interactions across grain boundaries. This may also require changes to the Lagrangian numerical description currently used in peridynamics because of the large deformations that occur during grain growth.

The current model, by treating surface energy on the same conceptual basis as other forms of energy such as elastic strain energy, offers the possibility of a consistent treatment of mechanical grain deformation and sintering. It also appears possible to extend the present model to include multiple materials, for example fluids that are sometimes used in sintering processes.

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