Simulation and Property Characterization of Nanoparticle Thermal Conductivity for a Microscale Selective Laser Sintering System

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ABSTRACT

Current Additive Manufacturing (AM) technologies are typically limited by the minimum feature sizes of the parts they can produce. This issue is addressed by the microscale selective laser sintering system (μ-SLS), which is capable of building parts with single micrometer resolutions. Despite the resolution of the system, the minimum feature sizes producible using the μ-SLS tool are limited by unwanted heat dissipation through the particle bed during the sintering process. To address this unwanted heat flow, a particle scale thermal model is needed to characterize the thermal conductivity of the nanoparticle bed during sintering and facilitate the prediction of heat affected zones (HAZ). This would allow for the optimization of process parameters and a reduction in error for the final part. This paper presents a method for the determination of the effective thermal conductivity of copper nanoparticle beds in a μ-SLS system using finite element simulations performed in ANSYS. A Phase Field Model (PFM) is used to track the geometric evolution of the particle groups within the particle bed during sintering. CAD models are extracted from the PFM output data at various timesteps, and steady state thermal simulations are performed on each particle group. The full simulation developed in this work is scalable to particle groups with variable sizes and geometric arrangements. The particle thermal model results from this work are used to calculate the thermal conductivity of the copper nanoparticles as a function of the density of the particle group.

INTRODUCTION

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29   Commercially available Additive Manufacturing (AM) systems are typically

30   capable of producing parts with feature sizes on the order of hundreds of micrometers

31   [1,2]. Although these manufacturing processes are versatile, this size limitation prevents

32   their effective use in the microelectronics industry, where parts require manufacturing

33   resolutions on the order of a few micrometers. The leading process used to manufacture

34   two and a half dimensional microelectronics structure consists of a mixture of

35   lithography, etching and chemical deposition steps, which often require intricate setups

36   and suffer from size limitations. The development of a microscale selective laser sintering

37   (μ-SLS) system addresses the need for a more robust, flexible, and easy to use process,

38   while targeting a minimum feature size on the order of a single micrometer [3-6].

39   Selective Laser Sintering (SLS) is an AM process that directly applies laser energy

40   to a deposited powder bed. The laser energy increases the temperature within the bed in

41   the areas of laser application which facilitates the fusion of the powder particles into a

42   solid part. This sintering process is repeated for each subsequently deposited layer until

43   a full part is formed [7]. One of the major advantages of SLS over other commercial AM

44   systems is its ability to build true 3D parts, such as overhanging structures, due to support

45   provided by the unsintered material adjacent to the sintered part. The μ-SLS system

46   functions similarly to traditional SLS processes in that laser irradiation is applied to a

47   particle bed and the process is repeated layer-by-layer. The primary difference between

48   the two processes is the use of nanoparticles as the powdered material in the μ-SLS

49   system, which facilitates the production of parts with single micrometer features. Given

50   the scale of these particles and the required feature sizes, accurate characterization of

51   the nanoparticle beds is needed to optimize the µ-SLS process.

52   Existing simulation work for SLS systems provide insight into the general process

53   of heat transfer in powdered systems [8-10], although they focus on larger particles and

54   typically model a melting and solidification process, as opposed to the solid-state

55   sintering process experienced by the nanoparticles used in the µ-SLS system. These

56   particles are on the order of 10-100 nanometers in diameter. The nanoparticles remain

57   solid as they sinter, with grain boundary and surface diffusion dominating the underlying

58   solid-state sintering mechanisms [11]. The effective thermal conductivity of metal

59   nanoparticles within a powder bed system is found to be dominated by the evolving

60   geometries of the nanoparticles as they diffuse into one another, and the grain boundary

61   thermal contact resistance at the interfaces between adjacent nanoparticles [12]. The

62   work presented in this paper focuses on these two dominating factors, while also

63   considering the conductivities of the individual copper nanoparticles and the surrounding

64   air medium.

65

66   **BACKGROUND**

67   The modeling and simulation of the heat transfer characteristics and material

68   properties of particle systems has been actively researched for years. Two-particle models

69   have been developed to predict heat transfer between adjacent particles in thermal

70   contact using analytical and computational methods. The two-particle model developed

71   by Bachelor and O'Brien [13] was used to determine the effective conductivity of a

72   granular material experiencing steady state heat transfer conditions. The model assumes

73   heterogeneous media which allows it to capture the stochastic nature of particle

74   packings. Sun and Chen [14] developed a particle conduction model that uses Hertzian

75   contact theory to estimate the contact area between two adjacent particles. The model

76   makes a critical assumption that the bodies in contact behave as semi-infinite solids,

77   where changes in temperature due to the contacts between particles are confined to a

78   small region surrounding the contact area. Although it reduces the computational

79   complexity of the model, the semi-infinite solid approximation reduces the accuracy of

80   Sun and Chen's model for larger Fourier numbers. Zhou, Yu and Horio [15] worked to

81   improve the work by Sun and Chen by developing a finite element simulation of a particle-

82   particle collision system to provide a correction factor for large Fourier numbers.

83   Similarly, Shimizu [16] developed a particle conduction model where heat is transferred

84   between adjacent particles through a thermal pipe with a cross sectional area equal to

85   the expected contact area between the two particles. Two particle conduction models

86   like those developed by Bachelor and O'Brien, Sung and Chen and Shimizu can be

87   expanded to particle systems with more than 2 particles given the assumed independence

88   of particle-particle contacts. However, this typically requires extensive computational

89   resources, and fails to account for time-varying, non-spherical geometries and the use of

90   nanoparticles in the models.

91

92   Models capable of estimating the effective material properties and heat transfer

93   characteristics of aerogels consisting of packed nanoparticles have recently been

94   developed. Zhao et al. [17] developed a diffusion-limited colloidal aggregation model to

95   determine particle packing structures in silica aerogels and determine the effective

96   thermal conductivity of the structure using a 3D finite volume numerical model. This work

97   considers the effects of the random particle packing structures, the solid-gas coupling,

98   conduction within solid particles, and the area of contact between adjacent particles to

99   calculate the conductivity values. Guo and Tang [18] built a theoretical model that focused

100   on the relative effects of particle-particle conduction in the solid particle chains, particle-

101   air-particle conduction along the particle chains, and heat conduction from the particle

102   chains to the surrounding pore gas. Although these conductivity models provide insight

103   on relevant heat transfer parameters in nanoparticle systems, they analyze insulative

104   materials and packing structures unlike those found in μ-SLS systems. They also do not

105   consider the geometric evolution of the nanoparticles that occurs during a temperature

106   driven solid state diffusion process.

107

108   Work has also been developed that considers the varying sizes and packing

109   structures of nanoparticles of various materials. These research efforts determine the

110   relationship between the porosity of the nanoparticle structure and its effective thermal

111   conductivity. Wu and Huang [19] prepared SiC nanoparticle beds and measured the

112   thermal conductivity of the system. They also developed a thermal model that accounted

113   for thermal contact resistance between particles and the thermal contact resistance

114   between nanoparticles and the surrounding air. Using this model and corresponding

115   experimental data, Wu and Huang developed a relationship between the particle packing

116 density (represented as porosity) and the effective thermal conductivity of the SiC

117 nanoparticles to quantify SiC capabilities as an insulator. Lin et al. [20] used the transient

118 hot-wire method to measure the thermal conductivity changes of copper and nickel

119 nanoparticle packed beds as a function of the changing porosity during sintering at

120 various temperature. They also analyzed the effects of the tableting pressure in

121 enhancing the conductivity of these structures. Qin et al. [21] also investigated the

122 relationship between porosity and thermal conductivity for groups of metal

123 nanoparticles. Specifically, they investigated the effective thermal conductivity for silver

124 nanoparticles undergoing sintering using multiple analytical methods that factored in the

125 relative conductivities of silver and the surrounding pore media. Although these research

126 efforts developed relationships between particle packing porosity and effective thermal

127 conductivity, they focus on porosity values that are much smaller than those experienced

128 by the nanoparticles in the μ-SLS process. These porosity-thermal conductivity

129 relationships also lack a full representation of the geometric evolution of particle

130 geometries during the early sintering and neck formation stage, where porosities are high

131 and effective thermal conductivities continue to be dominated by particle-particle

132 contact resistances.

133

134   Expanding upon more general conduction models, particle heat transfer

135 simulations for SLS systems have been developed for error minimization and part

136 prediction. Ganeriwala and Zohdi [22] developed a coupled finite difference – discrete

137 element method (DEM) model to predict thermal behavior SLS systems. Particles were

138   assumed to be discrete spheres of various sizes, and the substrate was discretized for a

139   finite difference solution. The model analyzes a subsection of a larger particle bed and

140   provides information from the particle scale model to a larger continuum part scale

141   model. Similarly, Moser et al. developed a DEM model to estimate the effective thermal

142   conductivity of particle packings in an SLS powder bed [23,24]. Moser's model assumes

143   all particles in the system are uniform temperature bodies. The primary heat transfer

144   modes considered by this model are particle-particle conduction, particle-gas-particle

145   conduction and radiation when determining the steady state temperature distribution in

146   the particle system. The output temperature values were used to calculate the effective

147   thermal conductivity of the randomly generated group of particles. Although these SLS

148   models effectively simulating the heat transfer process within powder beads for the initial

149   spherical arrangement of particles, they model a particle melting and solidification

150   process. In the μ-SLS system, sintering is a solid-state diffusion process, where the

151   particles do not undergo phase transformations. Therefore, as the particles diffuse into

152   each other, the resulting geometries of the particle groups change significantly despite

153   the lack of a phase change. This work builds upon previous research efforts and expands

154   their scope to incorporate the complex geometric changes experienced by the

155   nanoparticle powders undergoing laser sintering and estimates the effective thermal

156   conductivity of these particle packings as a function of sintering duration.

157

158   **MODELING METHODS**

159   A Finite Element Model (FEM) was developed in this work to determine the

160   effective thermal conductivity of copper nanoparticles as a function of densification in a

161   μ-SLS system. A steady state heat transfer simulation was performed in ANSYS Mechanical

162   to obtain temperature information within a heated nanoparticle packing. The Finite

163   Element method was selected due to its proven accuracy, setup simplicity and its

164   widespread use in Additive Manufacturing (AM) applications [25,26]. Nanoparticle

165   packings, composed of approximately 20-30 nanoparticles (each on the order of 10-100

166   nanometers in diameter), are analyzed as they transition from discrete spherical particles

167   to a coalesced, sintered unit [27]. Computer aided design (CAD) models are used to

168   represent the packed nanoparticle geometries during the finite element analysis. Surface

169   and grain boundary diffusion is simulated using a Phase Field Model (PFM) to track the

170   morphological evolution of the particles and obtain accurate solid models of the

171   nanoparticles throughout the sintering process [28]. The PFM tracks the evolution of a

172   fixed number of equally spaced points within a defined bounding box. The initial number

173   of particles, the radii of each particle, and the dimensions of the bounding box are variable

174   inputs to the model. For a given particle set, the PFM simulates the entire sintering

175   process and records snapshots of the phase field variables at several timesteps. The

176   captured snapshots provide data files containing a set of phase field variables for each

177   point within the model's bounding box. The data files provide information regarding the

178   particle density, ρ, of the region corresponding to the point in space, as well as a number

179   of η variables equal to the total number of initial particles in the system. Each η variable

180   provides information regarding the contribution each point in the PFM makes to the

181 corresponding particle in the system. Both variables range from 0 to 1 and have cutoffs

182 that determine the geometric arrangement of the nanoparticles. The ρ cutoff determines

183 the threshold between the solid and vapor phases (0 representing vapor, and 1

184 representing solid), and the η variable cutoff determines which points within the

185 bounding box correspond to which particles in the system (0 representing no

186 contribution, and 1 representing full contribution to a specific particle). This dataset is the

187 basis for the subsequent conversion and FEM processes discussed in this paper.

188 **Conversion from PFM Data to CAD Models**

189   Although the PFM data files provide a comprehensive description of the particle

190 geometries throughout the sintering process, the raw data is not a usable geometric input

191 for a finite element solver. FEMs accept solid models and surface bodies in several forms.

192 To use the PFM data in a Finite Element simulation, a conversion process must be

193 established to transform the PFM output data files into usable solid models for FEM input.

194 To accomplish this, a 3-step conversion process is developed. To maintain the accuracy of

195 the PFM output data, this conversion process must provide input CAD files that match the

196 original PFM particle detail. The conversion process must also work for all possible initial

197 particle arrangements, variable particle numbers, and the different geometries that occur

198 as particles diffuse into one another. The steps of the conversion process are as follows.

199 The first step of the conversion process converts raw PFM data into cartesian point

200 clouds. The PFM output data is filtered using the ρ cutoff to determine the solid phase,

201 and the η variable cutoffs are used to divide the domain into individual particles. The

202   resulting collection of filtered points is then run through a surface extraction algorithm to

203   isolate the exterior points and convert the data into a point cloud format. The algorithm

204   evaluates each solid point to see if any of the surrounding points are in the vapor phase

205   and writes a new data file with the filtered surface set. An example of a resulting point

206   cloud is shown in Fig 1a.

207   The second conversion step takes the point cloud coordinate files as input and

208   outputs reconstructed surface meshes for each nanoparticle. These surface meshes are

209   hollow shells composed of small surface triangles and are constructed based on the

210   arrangement of points in the input point cloud. This step in the conversion process is

211   performed in Meshlab, an open-source 3D meshing and reconstruction software, where

212   parameters are adjusted to optimize a tradeoff between surface mesh accuracy and the

213   number and quality of the resulting triangle surface meshes. The major sub steps

214   performed in Meshlab include computation of surface normals, surface reconstruction,

215   surface decimation (a reduction in the number of node points that make up the triangular

216   surface mesh), mesh repair and mesh smoothing. This surface meshing step is often the

217   most critical, as it determines the usability, complexity and surface quality of the

218   downstream solid parts and subsequent finite element meshes. An example of the

219   resulting surface mesh is shown in Fig 1b.

220   The third step of the conversion process takes triangular surface meshes as input,

221   and outputs solid CAD models with defined grain boundaries between adjacent particles.

222   Particle surface meshes are first filled out to form solid CAD models in FreeCAD, an open-

223   source CAD software. The particle models are then checked for holes that result from

224   initial flaws in the surface mesh reconstruction process. Once the particle models are free

225   of defects, a Python script is executed to detect intersections between adjacent

226   nanoparticles in the particle set. This script checks to see if any two particles have

227   overlapping points in their original cartesian point cloud. The information for each pair of

228   overlapping particles is added to an intersection matrix for further processing. The

229   intersection matrix controls a separate Python script with FreeCAD integration that

230   performs particle-particle Boolean cuts. If a small and large particle overlap, the overlap

231   region is removed from the smaller particle to simulate the process of smaller particles

232   diffusing into larger particles. This occurs for each set of intersecting particles recorded in

233   the intersection matrix. These Boolean cuts ensure that the grain boundaries between

234   adjacent nanoparticles in the particle set are correctly modeled, and that the interface

235   between any set of two particles in contact is a perfect fit. This step is critical, as the

236   thermal contact resistance between adjacent nanoparticles in a particle set is one of the

237   most important input parameters in subsequent finite element simulations. This step

238   concludes with two additional operations using FreeCAD Python commands. First, a

239   rectangular block of air with dimensions equal to the PFM bounding box is inserted into

240   the modeling space. This block represents the air surrounding the nanoparticles and is

241   included to allow the FEM to correctly model particle-air-particle conduction. This model

242   neglects natural convection effects due to the extremely small Raleigh numbers, and

243   correspondingly small heat transfer coefficients present in this nanoscale system. In the

244   absence of external forced convection, the surrounding air can be modeled as a solid,

245   thermally conductive medium that remains stationary for the duration of the simulation.

246   To ensure perfect contact between the solid air block and the nanoparticle CAD models,

247   another Python script is used to subtract each particle in the set from the surrounding air

248   block. This forms a hollowed-out block that perfectly envelops the nanoparticle packing.

249   Finally, a second block is inserted beneath the nanoparticles to model the glass substrate.

250   Examples of the completed CAD models are shown in Fig 1.

251   **Conversion from PFM Data to CAD Models**

252   The purpose of converting the PFM output data files to solid CAD models is to

253   prepare the nanoparticle models for FEM. With proper setup, the FEM will solve for a 3D

254   temperature distribution within the nanoparticle models and the surrounding air block,

255   which will be subsequently used to extract the effective thermal conductivity of the

256   particle set. The FEM solver used in this work is configured to solve the heat conduction

257   equation

258     $$\rho c_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla \mathrm{T}) + Q''' \quad (1)$$

259   where k, ρ, c_p, and Q''' represent the thermal conductivity, the density, the specific heat

260   capacity at constant pressure, and the internal volumetric heat generation of a given solid

261   object. The terms on the right-hand side of Eq. 1 describe the second order spatial

262   temperature gradient in the object summed with the heat generation occurring within

263   that object, Q'''. The term on the left-hand side of Eq. 1 represents the transient thermal

264   response of the object given its material properties. To solve this second-order partial

265   differential equation for the spatial temperature distribution within an object, the

266   nanoparticle CAD models must first be transferred to the finite element workspace,

267   where the finite element meshing algorithm will divide each of the bodies into discrete

268   elements and nodes on which an approximate solution can be solved. Then, heat transfer

269   boundary conditions must be specified. Finally, values for the thermal contact

270   conductance between adjacent nanoparticles and at particle-air interfaces are

271   determined and set. Mesh discretization, boundary condition application and thermal

272   contact conductance determination were the key focuses of the finite element setup.

273   *Mesh Generation*

274   Once the nanoparticle models, the enveloping air block and the glass substrate

275   are brought into the finite element workspace, a multistep meshing procedure is

276   implemented. Due to the nature of the conversion process, the resulting particle models

277   have surface profiles that match their corresponding triangular surface meshes. These

278   profiles are composed of many small triangular faces of varying sizes, adding complexity

279   to the geometry of each nanoparticle. The surrounding air block envelops these particles

280   with matching faces at the contact points, and thus has a similar level of geometric

281   complexity. To account for this, heavily refined meshes are generated both within solid

282   volumes and on planar surfaces. The surface and body meshes are generated separately

283   with unique values to ensure an optimal mesh. Within the particle models and the

284   surrounding air block, a fine tetrahedral mesh is applied. This mesh is restricted to a

285   region of influence near the contact surfaces between the particle models and the solid

286   air block. Hollow spheres of influence are generated and imported into the model to

287   accomplish this volumetric meshing procedure. Each of the hollow spheres of influence

288   encompasses the surface of one of the nanoparticles in the particle set. Both the air and

289   particle models are meshed with this method, allowing for a fine mesh near contact

290   regions, and a coarser mesh near the particle centers. This reduces mesh complexity and

291   reduces overall computational time while emphasizing the regions with fine geometric

292   detail. Additionally, a fine surface mesh is applied to all particle- air contact surfaces. This

293   allows the meshing algorithm to handle the large number of small triangular faces present

294   in the model. Finally, the global mesh is instructed to automatically refine the mesh

295   further in tight spaces to improve the success rate of the meshing algorithm and facilitate

296   subsequent Finite Element modeling.

297   *Boundary Condition Application*

298   After the mesh settings are finalized for all surfaces and solid bodies, boundary

299   conditions representing heat transfer processes are applied to the particle model. Prior

300   to importing the solid models into ANSYS, the edges of the nanoparticle group are

301   removed by a vertical cutting plane to allow for the consistent application of boundary

302   conditions to smooth, vertical faces irrespective of initial conditions or timesteps. The

303   locations of the vertical cuts are chosen such that the fraction of the resulting flat surface

304   area that contains particle faces approximates the volume fraction of the nanoparticles

305   with respect to the bounding box. This boundary modification helps maintain a uniform

306   heat flow across different particle geometries. A coordinate direction is chosen, and a

307   constant temperature of 22 °C is applied to the flat particle faces corresponding the

308   minimum value of this coordinate direction. A constant heat flux of $10^4 \frac{MW}{m^2}$ is then applied

309   to the flat particle faces corresponding to the maximum value of the chosen coordinate

310   direction. All remaining boundary faces in the model are insulated to simulate 1-

311   dimensional heat flow through the nanoparticle set. Radiation was neglected in the

312   model, as it has been found to be negligible compared to particle-particle conduction and

313   particle-air-particle conduction. This is largely due to the relatively low average sintering

314   temperature of roughly 700 K [29]. Applied boundary conditions are depicted in Fig 2c

315   and Fig 2d.

316   *Thermal Contact Conductance*

317   The thermal contact conductance values used in the simulation determine the

318   temperature jumps at the interfaces between adjacent bodies in particle-particle contact

319   and in particle- air contact. The thermal contact conductance for particle-air contact is set

320   to $10^5 \frac{MW}{m^2 K}$ to accurately model perfect wetting of the particle surfaces by the surrounding

321   air. The resulting temperature jump at these interfaces should be close to zero to

322   approximate ideal contact between an object and a surrounding fluid. The thermal

323   contact conductance for particle-particle contact is more difficult to estimate. Multiple

324   thermal contacts occur from one end of the nanoparticle packing to the other along the

325   specified coordinate axis. This is especially true for earlier timesteps, where more particle-

326   particle contacts exist along the primary axis of heat flow. A value of $20 \frac{MW}{m^2 K}$ was chosen

327   for the particle-particle thermal contact conductance based on estimations from [29] and

328   [30]. The former used DEM to compare relevant heat flows between spherical metal

329   nanoparticles and selected the value of thermal contact conductance that corresponded

330   to the transition point at which the temperature gradients within individual particles

331   became negligible compared to the temperature drop across particle-particle contacts.

332   The latter measured the thermal contact conductance between deposited metal films (Al,

333   Ag, Sn, Zn, and In) and an underlying copper film using transient thermo-reflectance

334   methods. The values determined from [30] ranged from 8 to 95 $\frac{MW}{m^2 K}$ for the set of tested

335   metals. The value obtained from [29], 20 $\frac{MW}{m^2 K}$ was determined for the same nanoparticle

336   sintering system as covered in this work. Therefore, the value from [29] was used in this

337   paper for consistency.

338   **RESULTS AND DISCUSSION**

339   The densification of 10 one-by-one micrometer beds consisting of anywhere

340   between 21 and 43 particles was simulated using the PFM setup described earlier. The

341   simulation boxes containing the particle sets were 104 by 104 pixels in the x-y plane and

342   ranged from 56 to 86 pixels along the z-axis. PFM output data files representing

343   nanoparticle geometries for each of the 10 sintered beds were saved at 35 timesteps

344   along the sintering timeline. Each output file was converted into a set of particle CAD

345   models with well-defined contact regions. Each of these particle sets was then imported

346   into an ANSYS Mechanical FEM workspace where mesh generation and boundary

347   condition application were performed before ultimately solving each model for the

348   resulting steady-state temperature distribution within the nanoparticle system.

349   Upon completion of the FEM simulation for each timestep, the temperature distributions

350   were analyzed to calculate the effective conductivity of the particle set. To calculate the

351   effective thermal conductivity of a particle set, the steady state, 1-dimensional

352   approximation to Fourier's law of heat conduction was rearranged and solved for the

353   thermal conductivity parameter. This rearranged thermal conduction equation is shown

354   in Eq. 2

$$k_{eff} = \frac{L}{T_2 - T1} \cdot q'' \quad (2)$$

355

356   Where k_{eff} is the effective thermal conductivity, T_2 and T_1 are two independent

357   temperature values located at different locations along the chosen axis, L is the distance

358   between T_2 and T_1 along the same axis, and q'' is the heat flux magnitude at the

359   boundary. The temperature distributions produced by the FEM solver provide

360   temperature data corresponding to every node on every element within the finite

361   element model. To apply Eq. 2 across the full particle set and obtain an effective thermal

362   conductivity value for the given set, the output temperature information must be

363   reduced. The temperature distributions were first filtered to only include the external

364   faces corresponding to the minimum and maximum values of the x coordinates. The

365   temperature datasets were again filtered to only include temperature data from particle

366   nodes. This filtration only included particle faces corresponding to non-insulative

367   boundary conditions and allowed for a more accurate temperature difference calculation

368   across the set of particles. Finally, the filtered temperature data was averaged at both the

369   maximum and minimum x-coordinate locations to obtain a single representative

370   temperature value for each location. These average temperatures correspond to T_2 and

371   T_1 from Eq. 2.

372   Once the output data was simplified to averaged temperature values, the effective

373   thermal conductivity of the particle group could be determined. Eq. 2 provides a first-pass

374   estimate for this value, but it is unable to account for non-uniform surface areas in the

375   primary direction of heat flow, and thus requires modification. The PFM data input is

376   inherently stochastic, meaning the nanoparticle geometries present in each particle set

377   are different. Therefore, the surface areas that are exposed to the constant surface

378   temperature and the constant heat flux boundary conditions can vary considerably

379   between beds and between timesteps for a given bed. Additionally, the cross-sectional

380   area perpendicular to the axis of heat flow changes with location in a unique way for each

381   bed. A correction factor is added to Eq. 2 to account for these sources of variation and

382   provide more comparable values for the effective thermal conductivity of the particle set.

383   The modified formulation is shown in Eq. 3.

384
$$k_{eff} = (\frac{A_S}{V_f}) \cdot \frac{L}{T_2 - T_1} \cdot q'' \qquad (3)$$

385   V_f represents the volume fraction of the PFM bounding box taken up by the nanoparticle

386   models. As represents the surface area corresponding to the constant heat flux boundary

387   condition. T_2 and T_1 correspond to the average temperatures on the filtered external

388   particle faces, q'' represents the applied heat flux of $10^4 \frac{MW}{m^2K}$ and L represents the

389   distance between T_2 and T_1. The fraction shown in Eq. 3 corrects for differences between

390   the surface area where the heat flux and temperature boundary conditions were applied

391   and the average surface area along the axis of heat flow through the particle set. The full

392   calculation was performed on 10 beds for 35 sampled timesteps each. In addition to the

393   thermal conductivity calculation, the density of each nanoparticle packing was calculated

394   using a Python script. This script samples each bed and calculates the ratio of particle

395   volume to the volume of a bounding box. Given the small number of nanoparticles, the

396   bounding box used to determine the total volume was chosen to be a subset of the full

397   PFM bounding box. This reduces the impact of boundary effects on the densification

398   results by focusing on the interior particles that better represent the majority of particles

399   in a much larger bed. The results of the simulations for the 10 randomly generated particle

400   beds were aggregated into a single dataset, shown in Fig. 3.

401   Fig. 3 includes the output data from the thermal simulations and density

402   calculations along with a linear best fit line and prediction bounds. All simulations were

403   run for a fixed number of timesteps, although each bed reached a different density value

404   by the end of the PFM simulation. Many of these density values were substantially larger

405   than the density values achievable during laser sintering (this does not include the

406   annealing process that occurs post-sintering). Therefore, thermal conductivity data points

407   were excluded if their density values exceeded 5000 $\frac{kg}{m^3}$, as this is a reasonable upper

408   bound for achievable densification using the μ-SLS system without post-process

409   annealing. The 95% prediction interval provides a confidence band for future predictions

410   using the linear fit shown in Fig. 3. The large range for prediction results from the

411   considerable uncertainty present for single micron particle beds. In smaller particle

412   systems, boundary effects have considerably more impact on the simulation results. To

413   reduce this uncertainty, a subsection of a larger particle bed can be used at the cost of a

414   significant increase in computational expense. The prediction interval in Fig. 3 will be used

415   to quantify the uncertainty in downstream sintering models that incorporate the results

416   from this work.

417   Due to the random nature of the particle bed generation process, beds have

418   different initial densities. To account for this, the results shown in Fig. 3 are plotted as a

419   function of densification percentage in Fig. 4. Densification percentage refers to the

420   percent increase in density from the density value calculated before the onset of

421   sintering.

422   It is noted that these results describe early necking and initial sintering, but do not

423   include the initial thermal conductivity of the nanoparticle packing before sintering

424   occurs. The values for the initial packing condition were recorded separately. This allows

425   for future simulations to have access to a thermal conductivity value for unsintered

426   copper nanoparticles, in addition to a range of thermal conductivity values for copper

427   nanoparticles as they sinter. The difference in average effective conductivities and

428   densities just before and just after the onset of sintering, along with the associated

429   uncertainties, are 9.42 $\frac{W}{m*K}$ (798.35% change from initial thermal conductivity) and 1.28

430    $\frac{kg}{m^3}$ (.035% change from initial density) respectively. Despite a small average increase in

431   the density during initial sintering, the thermal conductivity increases nearly by a factor

432   of 10. This is the result of the early neck formation that occurs when adjacent

433   nanoparticles surpass the sintering threshold. During this initial necking phase, the

434   contact regions between nanoparticles grow considerably, opening larger paths for heat

435   to conduct between the particles. At the onset of this process, the formation of these

436   heat pathways results in a very small change in the overall geometry, and therefore the

437   density, of the packing.

438   The thermal conductivity results presented in this paper were compared with

439   literature results for silver nanoparticles during densification. Fig. 5 shows three separate

440   estimations for the effective thermal conductivity of silver nanoparticles [21] in addition

441   to the data obtained in this work.

442   The results developed in this work differ considerably from those derived from

443   literature. This is to be expected, since the literature results focus more on densification

444   values close to the bulk material value and lack a comprehensive overview of the early

445   necking process that occurs in the μ-SLS system. The laser sintering that occurs during the

446   μ-SLS process is heavily impacted by the initial necking phase, so it is critical that the rapid

447   increase in thermal conductivity during this phase is accurately captured. A summary of

448   the complete FEA process is shown in Fig. 6.

449   **FUTURE WORK**

450   The results developed in this work describe the relationship between copper

451   nanoparticle densification and the corresponding increase in the thermal conductivity of

452   a nanoparticle group. This relationship has been integrated into a preliminary part-scale,

453   transient finite element model capable of predicting the thermal evolution of a powder

454   bed during sintering. Each copper element of the part-scale model has its thermal

455   conductivity updated at a series of timesteps based on the temperature history of the

456   element. Moving forward, this part-scale model will be validated against experimental

457   results using a combination of IR camera images, profilometer profiles, and

458   experimentally measured electrical properties. Additionally, this multi-scale modeling

459   effort will be used to input laser parameters and predict the resulting sintered part

460   shapes. This predictive capability will allow for model-based control of laser parameters

461   to achieve optimized part shapes with minimized heat affected zones. Additionally, larger,

462   2 micrometer by 2 micrometer beds can be used to reduce the uncertainty of the resulting

463   linear fit line by reducing the impact of boundary effects on the results. By expanding the

464   size of the bed, the thermal model can take a subsection from the larger bed and run the

465   heat transfer analysis on an interior subsection, avoiding the particles along the edges.

466

467   **CONCLUSION**

468   In this paper, a Finite Element Model (FEM) and a fully scalable data conversion

469   smalls sets of copper nanoparticles. This will facilitate the prediction of heat affected

470   zones (HAZ) during microscale selective laser sintering (μ-SLS). Phase Field Modeling

471   (PFM) data was converted into solid particle CAD models, and an automated steady-state

472   heat transfer simulation was used to calculate temperature profiles and effective thermal

473   conductivities of nanoparticle packings. A steady state, 1D thermal conduction

474   approximation was used alongside an area/volume fraction correction factor to extract

475   thermal conductivity information out of resulting temperature data. The results show a

476   rapid increase in the thermal conductivity during early particle necking, and a reduced

477   rate of change for later timesteps. The results also show that there is significant

478   uncertainty in the thermal conductivity of the beds due to the variances in nanoparticle

479   configurations throughout the bed but that there is a clear trend that as be particle bed

480   starts to sinter together and densify, the thermal conductivity of the bed increases.

481   Overall, the thermal conductivity of the bed almost doubles from initial sintering to the

482   maximum densification that is typically achieved in the μ-SLS process.

483

484   **FUNDING**

488 **NOMENCLATURE**

489

ρ	variables should appear in first column with the description in second column, m
η	all variables should appear in italics
c_p	two-letter abbreviations should appear in italics
Q'''	three-letter abbreviations should not appear in italics
K	Reynolds number and similar abbreviations do not use italics
ρ	use the "Tab" key to add more rows to this table
k_{eff}	Effective thermal conductivity of the nanoparticle packing
L	Distance between locations corresponding to T_2 and T_1
T_2	Average temperature on the filtered external particle face
T_1	Average temperatures on the opposite side particle faces
q''	Heat flux magnitude at the boundary
A_s	Surface area corresponding to the constant heat flux boundary condition
V_f	Volume fraction of the PFM bounding box taken up by the nanoparticle models

490

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573   **Figure Captions List**

574

575

Fig. 1   The Full Conversion Process. a). Nanoparticles represented by point clouds; b). Nanoparticles represented by a triangular surface mesh derived from point clouds; c). Nanoparticles represented by solid models derived from triangular surface meshes; d). Nanoparticle solid models resting on a glass substrate; e). Hollowed out air block; f). Full assembly of particles, air, and glass

Fig. 2   3-Dimensional Finalized Solid Models (Note the 180-degree rotation about the z-axis between images c and d). a). Model with enveloping air block; b). Model without air block; c). Constant surface heat flux boundary condition shown in red; d). Constant surface temperature boundary condition shown in blue

Fig. 3   Thermal conductivity vs density with linear fit line and 95% prediction bounds

Fig. 4   Thermal conductivity vs densification percentage with linear fit line and 95% prediction bounds

Fig. 5   Thermal Conductivity vs Density Comparison: This work vs Qin Et. Al [21]

Fig. 6   Summary of Finite Element Setup and Sample Results

576

577   **Table Caption List**
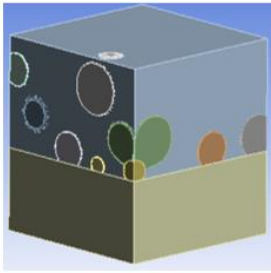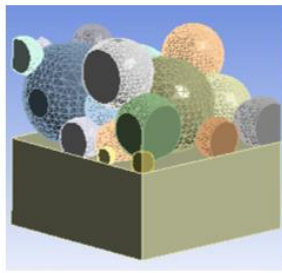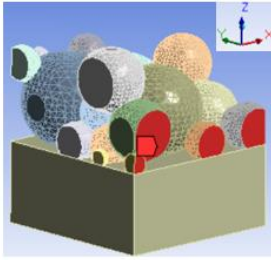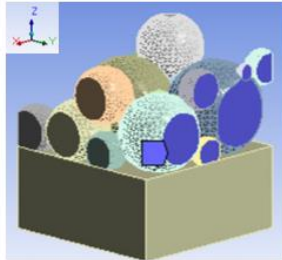578

    Table 1   Capitalize the first word in the caption

579

Fig. 1

Fig. 2

a.

b.

c.

d.

Fig. 3

Fig. 4

Fig. 5

Comparison with Thermal Conductivities from Literature

Fig. 6