Computational modeling of electrically-driven deposition of ionized polydisperse particulate powder mixtures in advanced manufacturing processes

T.I. Zohdi

Department of Mechanical Engineering, 6117 Etcheverry Hall, University of California, Berkeley, CA 94720-1740, USA

A R T I C L E   I N F O

Article history:
Received 27 September 2016
Received in revised form 20 March 2017
Accepted 21 March 2017
Available online 24 March 2017

Keywords:
Powders
Polydisperse
Deposition
Electromagnetics

A B S T R A C T

A key part of emerging advanced additive manufacturing methods is the deposition of specialized particulate mixtures of materials on substrates. For example, in many cases these materials are polydisperse powder mixtures whereby one set of particles is chosen with the objective to electrically, thermally or mechanically functionalize the overall mixture material and another set of finer-scale particles serves as an interstitial filler/binder. Often, achieving controllable, precise, deposition is difficult or impossible using mechanical means alone. It is for this reason that electrically-driven methods are being pursued in industry, whereby the particles are ionized and an electromagnetic field is used to guide them into place. The goal of this work is to develop a model and simulation framework to investigate the behavior of a deposition as a function of an applied electric field. The approach develops a modular discrete-element type method for the simulation of the particle dynamics, which provides researchers with a framework to construct computational tools for this growing industry.

© 2017 Elsevier Inc. All rights reserved.

1. Introduction

A large variety of emerging advanced fabrication methods involve Additive Manufacturing (AM) processes, which are generally characterized as depositing materials onto substrates and bonding them together to create structures, as opposed to classical “subtractive” processes which remove material. The approach was pioneered in 1984 by Hull [40] and was a 2.9 billion dollar industry in 2015, with applications ranging from motor vehicles, consumer products, medical devices, military hardware and the arts. We refer readers to a recent review of the state of the art by Huang et al. [38]. A subclass of AM processes involve “dry” powder deposition. “Dry” powder deposition approaches (where the interstitial space between particles is not saturated with a liquid) do not utilize solvents, since the deposited material will be heated or cured afterwards in order to harden it into place. Interstitial solvents are avoided because they can compromise the resulting hardened material quality, due to gas bubbles, mass-transport induced cracking, etc., during curing. However, the precise deposition of dry powders is difficult. It is for this reason, electrically-driven methods are being pursued, whereby the dry particles are ionized and an electric field is used to guide them into place. A key aspect of these processes are the precise deposition of specialized mixtures of materials. The design of the deposited material properties is enabled by the use of added particles to endow the correct functionality (“functionalization”) to the material (Fig. 1). In many cases, these materials are specially designed mixtures of “dry” powders (particles), whereby one set of particles is chosen with the objective to electrically,
thermally or mechanically functionalize the overall material and another set of finer-scale particles serves as an interstitial filler/binder. The rapid rise in the use of particle-based materials has been made possible by the large-scale production of consistent, high-quality and inexpensive particles, which are manufactured in many ways, such as breakup of liquid streams into solidified droplets or vaporization from a solid or liquid to a gas and recapturing the condensate in the form of particles.

As mentioned, achieving precisely controlled deposition of such dry particulate-based materials is difficult or impossible by solely mechanical means. It is for this reason, electrically-driven methods are being pursued in industry, whereby the particles are ionized and an electric field is used to guide them into place (Fig. 2). The goal of this work is to develop a model and simulation method to investigate the behavior of such depositions as a function of the applied electric field. There is a direct correlation between the ionization strength between particles in the powder and the more fluid-like behavior. Effectively, with no ionization, the system behaves as a loose powder, which is difficult to control. As the ionization is ramped up, the balance between mutual attraction and repulsion leads to surface tension like effects. Thus, the expectation is that at high external fields and high ionization, the deposition will yield coherent aggregate “droplets” of the powder–mixture material.

Dry powders require nonstandard modeling and simulation tools to characterize their behavior as compared to continua. Numerical approaches designed for continuum simulation (such as Finite Element Methods) are not well-suited to describe the dynamics of discrete particles, with overall domains that break apart, coalesce or experience other extremely large configurational changes. One family of methods that is ideally suited to this task are Discrete Element Methods (DEM), which is the approach pursued in this paper. In order for new additive approaches to succeed, such as the one mentioned, one must utilize theory and computation to guide the proper selection of particle mixtures to progress to robust large-scale industrial manufacturing levels. Due to increasingly shorter product development times in the additive manufacturing field, there is a critical need for simulation tools. This has motivated the analysis in this paper, which attempts to develop a DEM computational framework which captures the dynamics of particle-to-particle and particle-to-substrate interaction under the influence of electric fields. As previously mentioned, to enhance control of depositions, the deposited particles are ionized and guided to the surface with an electromagnetic field. Ionization can be achieved through a variety of possible methods, such as by

---

1 For details, see Duran [26], Pöschel and Schwager [66], Ōhata et al. [62–64], Rojek et al. [69,70], Carbonell et al. [9], Labra and Ōhata [45], Mukherjee et al. [55–58] and Zohdi [93–111].
passing the particles through a charged gas or applying an electrified surfactant. We refer the reader to Martin [50,51], Choi et al. [10–13] and Demko et al. [16] for overviews of the various related processes. Generally speaking, this process is physically quite similar to methods used in spray coating technologies (see Martin [50,51] for the state of the art, as well as Sevostianov and Kachanov [73–75], Qian et al. [67], Dwivedi et al. [27], Liu et al. [47,48], Nakamura and Liu [59] and Nakamura et al. [60]). Oftentimes, these types of materials are used to lay down electronic lines or patterns on compliant substrates, such as flexible, “smart”, electronics, such as high-end solar cells. We refer the reader to the literature on printed electronics field found in Gamota [32], Nakanishi et al. [61], Fuller et al. [31], Samarasinghe et al. [71], Ahmad et al. [1], Sirringhaus et al. [76], Wang et al. [82], Huang et al. [39], Choi et al. [10–13] and Demko et al. [16,17] for details.

From a simulation standpoint, the overall particle–mixture system is constructed by coupling submodels for each primary physical process together. An iterative staggering scheme is developed whereby, within every time step, each individual particle is solved, “freezing” the state of the remaining multi-particle system. The state of the particle is then updated and the algorithm moves to the next particle in the system and the process is repeated. The overall process sweeps through the entire system repeatedly until convergence in an appropriate norm. As the system evolves, an error estimate dictates the time-step size that is needed to induce convergence to below an appropriate error level. Essentially, this is an implicit time-stepping scheme, which is combined with an (internal) iterative staggering process. In order to control rates of convergence within a time-step, the algorithm adjusts the time-step size. If the iterative process does not converge within a desired number of iterations, below an error tolerance, the time-step is reduced. The degree of time-step reduction is determined by utilizing an estimate of the spectral radius of the coupled system. Since the construction of model and solution process is modular, one can easily replace physical submodels with other choices, making it easy to numerically experiment with a variety of models.

2. Particle dynamics

Consider a collection of $N_p$ non-intersecting particles which are assumed to be spherical in shape. It is also assumed that the particles are small enough that their rotation with respect to their mass center minimally affects their overall motion (this is discussed further shortly). For an arbitrary $i$th particle in the system, acted upon by

1. $\Psi_i^{\text{con}}$: inter-particle contact forces (see Fig. 3),
2. $\Psi_i^{\text{bond}}$: inter-particle adhesive bonding forces,
3. $\Psi_i^{e+m}$: inter-particle near-field and external electromagnetic forces and
4. $\Psi_i^{\text{drag}}$: particle drag forces from any surrounding gas,

the dynamics are governed by

$$m_i \dot{\mathbf{r}}_i = \Psi_i^{\text{con}} + \Psi_i^{\text{bond}} + \Psi_i^{e+m} + \Psi_i^{\text{drag}} \equiv \Psi_i^{\text{tot}}(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_{N_p}),$$

(2.1)

where $\mathbf{r}_i$ is the position vector of the $i$th particle and $\Psi_i^{\text{tot}}$ is the total of the forces acting on the $i$th particle. The various types of force categories will now be explained further.

2.1. Particle contact forces

A straightforward “overlap” model (Zohdi [103–108]) is used to determine the normal contact forces between particles. Consider the $i$th particle in contact with $N_{ci}$ particles, producing a total contact force of $\Psi_i^{\text{con},n} = \sum_{j=1}^{N_{ci}} \psi_{ij}^{\text{con},n}$, where the
forces are dictated by the separation distance between the particle centers for the particle in contact (Fig. 4), written generally as

$$\Psi_{ij}^{\text{con,n}} = F(||r_i - r_j||, R_i, R_j, \text{material properties}),$$  \hspace{1cm} (2.2)

where $R_i$ and $R_j$ are the radii of the $i$th and $j$th particles in contact. In the literature, there is a large number of contact models for normal force generation. For simplicity, we assume that the contact force is proportional to the distance between the centers of particles $i$ and $j$ (when in contact). Contact is determined by checking if $||r_i - r_j|| \leq R_i + R_j$. We construct an overlap function, $\delta_{ij} = \frac{||r_i - r_j|| - (R_i + R_j)}{R_i + R_j}$, and a corresponding normal contact force$^2$

$$\Psi_{ij}^{\text{con,c}} \propto -K_{p_ij} |\mathcal{E}_{ij}|^{p_p} n_{ij} A_{ij}^c,$$  \hspace{1cm} (2.3)

where $0 < K_{p_ij} < \infty$ is a contact constant, $p_p$ is a material parameter and $\mathcal{E}_{ij}$ is nondimensional strain-like deformation metric

$$\mathcal{E}_{ij} = \frac{||r_i - r_j|| - (R_i + R_j)}{(R_i + R_j)} = \frac{\delta_{ij}}{R_i + R_j},$$  \hspace{1cm} (2.4)

and $n_{ij} = -\frac{r_i - r_j}{||r_i - r_j||} = \frac{r_i - r_j}{||r_i - r_j||}$.

2.1.1. Rotations

Including particle rotations is questionable for extremely small objects (idealized as spherical particles), since in reality they are not perfectly spherical (even if they are manufactured to be as close to spherical as possible) and, importantly, because of rolling resistance. However, for completeness, we illustrate the inclusion of rotational equations of motion (a balance of angular momentum), which augment a balance of linear momentum, $m_i \dot{v}_i = \Psi_{ij}^{\text{tot}},$ $v_i$ being the center of mass velocity. The balance of angular momentum reads

$$\mathbf{H}_{i,\text{cm}} = \frac{d(\mathbf{I}_i \cdot \omega_i)}{dt} = M_{i,\text{cm}}^{\text{tot}},$$  \hspace{1cm} (2.5)

where, for spheres, we have $\mathbf{H}_{i,\text{cm}} = \mathbf{I}_i, \omega_i = \frac{1}{2}m_i R_i^2 \omega_i$. The total moment, $M_{i,\text{cm}}^{\text{tot}}$, is due to interaction forces, contact forces and rolling resistance. In the present analysis, the effects of rotations are generally negligibly small. Regardless, we will formulate the system with rotations, where the important dynamical states of each particle are the $r_i$ is the position and $v_i$ is the velocity of the center of mass and $\omega_i$ is the angular velocity. A critical variable of interest is the velocity on the surface of the “particles” at contact points (with other particles), denoted $v_i^c$ and is computed by $v_i^c = v_i + \omega_i \times r_i - v_i$, where

$^2 A_{ij}^c$ is a contact area parameter (Fig. 5) discussed in Appendix A, along with a review of classical Hertzian and other models.
2.1.3. the relative position vector from the center of mass to the possible point of contact is given by \( r_{i\rightarrow c} \). This is utilized further later in the formulation.\(^3\)

2.1.2. Dissipation during contact

One can incorporate phenomenological descriptions of contact dissipation by tracking the relative velocity of the contacting particles by simply including

\[
\Psi_{ij}^{\text{con},d} = c^d (v_{j,n} - v_{i,n}) A_{ij}^c,
\]

where \( c^d \) is a contact dissipation parameter.

2.1.3. Contact induced friction

At the point of contact, “sticking” induced by friction is captured via the following “regularized” friction algorithm:

1. First one checks the threshold limit for static friction:

\[
K^f ||v_{j,\tau}^c - v_{i,\tau}^c|| A_{ij}^c \Delta t \text{ against } \mu_s ||\Psi_{ij}^{\text{con},n}||,
\]

where \( K^f \) is a tangential contact friction compliance constant, \( ||v_{j,\tau}^c - v_{i,\tau}^c|| \Delta t \) is the relative tangential velocity at (the point of contact), \( \Delta t \) is the time-step used in the numerical discretization,\(^4\) \( \mu_s \) is the static friction coefficient. This step “regularizes” (replaces) a potentially more difficult process of initially assuming no slip, then calculating the no-slip contact forces by solving an entire multibody/multisurface contact problem, \( \Psi_{ij}^{\text{ns}} \), then thereafter checking \( \Psi_{ij}^{\text{ns}} \) against the static friction limit \( \mu_s ||\Psi_{ij}^{\text{con},n}|| \) on each surface.

2. Second, if the threshold limit is not met, namely \( K^f ||v_{j,\tau}^c - v_{i,\tau}^c|| A_{ij}^c \Delta t < \mu_s ||\Psi_{ij}^{\text{con},n}|| \), then one computes

\[
\Psi_{ij}^{\text{con},f} = K^f ||v_{j,\tau}^c - v_{i,\tau}^c|| A_{ij}^c \Delta t \tau_{ij}^c
\]

where (here the subscripts denote the tangential components of velocity)

\[
\tau_{ij}^c = -\frac{v_{j,\tau}^c - v_{i,\tau}^c}{||v_{j,\tau}^c - v_{i,\tau}^c||} = \frac{v_{i,\tau}^c - v_{j,\tau}^c}{||v_{j,\tau}^c - v_{i,\tau}^c||}.
\]

where subtracting away the normal component of the velocity \( \dot{v}_i^c = \dot{v}^c - (\dot{v}^c \cdot \hat{n}) \hat{n} \) yields the contact point’s tangential velocity.

3. Third, if the threshold limit is met (or exceeded), specifically \( K^f ||v_{j,\tau}^c - v_{i,\tau}^c|| A_{ij}^c \Delta t \geq \mu_s ||\Psi_{ij}^{\text{con},n}|| \), then one employs a slip model

\[\text{Consequently, an implicit time discretization reads}
\]

\[
\omega_i(t + \Delta t) = \omega_i(t) + \frac{\Delta t}{I_{i,s}} \left( \phi \dot{M}_i^{\text{rot}}(t + \Delta t) + (1 - \phi) \dot{M}_i^{\text{rot}}(t) \right),
\]

which is discussed further later.

\[\text{Note } ||v_{j,\tau}^c - v_{i,\tau}^c|| \Delta t \text{ has dimensions of length.}\]
\[
\Psi_{ij}^{\text{con.f}} = \mu_d ||\Psi_{ij}^{\text{con.n}}|| \mathbf{r}_{ij},
\]  
\[\text{(2.11)}\]

where \(\mu_d\) is the dynamic coefficient of friction.

2.2. Particle-to-particle bonding relations

As a criterion for particles to bond, we adopt a threshold dictated by exceeding a critical interpenetration distance, computed in the following manner:

1. First, recall that if \(||\mathbf{r}_i - \mathbf{r}_j|| \leq (R_i + R_j)\), then the particles are in contact and \(E_{ij} = \frac{s_{ij}}{\gamma_{ij} + K_{ij}}\).
2. Second, for particles in contact, \(|E_{ij}| \geq E^*\), an adhesive/attractive normal bond (0 \(\leq K_{nb}\) ) is activated between the particles is a bonding constant and \(p_b\) is a material parameter:
\[
\Psi_{ij}^{\text{bond,n}} = K_{nb} |E_{ij}|^{p_b} \mathbf{n}_{ij} \mathbf{A}_{ij}^\alpha.
\]  
\[\text{(2.12)}\]

3. Third, for particles with an activated normal bond, the particles automatically have an activated rotational/tangential bond (similar in form to stick friction) \(^5\):
\[
\Psi_{ij}^{\text{bond,r}} = K_{rb}^{th} ||v_{ij}^{\alpha} - v_{ij}^{\beta}|| \mathbf{A}_{ij} \Delta t \mathbf{r}_{ij}.
\]  
\[\text{(2.13)}\]

2.3. Near-field and electromagnetic forces

We decompose the electromagnetic forces into three parts: (1) Lorentz forces (for charged particles), (2) inter-particle near-field forces and (3) magnetic forces (for magnetic particles). In mathematical form,
\[
\Psi_i^{\text{e+m}} = \Psi_i^{\text{lor.e+m}} + \Psi_i^{\text{mag}} + \sum_{j \neq i}^N \Psi_{ij}^{\text{nf}} = q_i (E_{i}^{\text{ext}} + \mathbf{v}_i \times \mathbf{B}_{i}^{\text{ext}}) + \Psi_i^{\text{nf}} + \Psi_i^{\text{mag}},
\]  
\[\text{(2.14)}\]

where the interaction between particle \(i\) and all other particles \(j = 1, 2, 3...N (j \neq i)\), is \(\sum_j^N \Psi_{ij}^{\text{nf}}\) and the applied Lorentz-induced forces from independent external fields \(E_{i}^{\text{ext}}\) and \(B_{i}^{\text{ext}}\) is \(\Psi_i^{\text{lor.e+m}}\). The terms \(E_{i}^{\text{ext}}\) and \(B_{i}^{\text{ext}}\) are considered to be externally controlled and uncoupled from one another. \(^6\)

Following Zohdi \([95, 97, 98, 102, 104–106]\), a simple form that captures the essential near-field effects is
\[
\Psi_i^{\text{nf}} = \sum_{j \neq i}^{N_p} \left( \alpha_{ij} ||\mathbf{r}_i - \mathbf{r}_j||^{-\alpha_{ij}} - \beta_{ij} ||\mathbf{r}_i - \mathbf{r}_j||^{-\beta_{ij}} \right) \mathbf{n}_{ij},
\]  
\[\text{(2.15)}\]

where the \(\alpha_i\)’s and \(\beta_i\)’s are empirical material parameters. Here, various representations (decompositions) of the coefficients that appear in Equation (2.15) are with \(c_1 = \pm 1\) (a positive/negative multiplier):

1. charge-based: \(\alpha_{ij} = \tilde{\alpha}_{ij} q_i q_j c_i c_j\), where the \(\tilde{\alpha}_{ij}\) are empirical parameters,
2. surface-area based (\(a = \text{surface area}\)): \(\alpha_{ij} = \tilde{\alpha}_{ij} a_i a_j c_i c_j\),
3. volume-based (\(V = \text{volume}\)): \(\alpha_{ij} = \tilde{\alpha}_{ij} V_i V_j c_i c_j\) and
4. mass-based (\(m = \text{mass}\)): \(\alpha_{ij} = \tilde{\alpha}_{ij} m_i m_j c_i c_j\).

There exist a large number of empirical near-field relations that generally fall under the subject matter of the vast field of “Molecular Dynamics” (MD). We refer readers to Frenklach and Carmer \([30]\), Haile \([35]\), Hase \([36]\), Schlick \([72]\) and Rapaport \([68]\), where Leonard-Jones, Mie and Morse potentials (Moelwyn-Hughes \([54]\)) are usually employed, with various extensions such as Tersoff \([78]\) additions and three-body terms (Stillinger \([77]\)). For the remainder of the analysis, we neglect any possible magnetism of the particles themselves. However, we note that such forces can be described by \(\Psi_{i}^{\text{mag}} = V(\gamma B_{i}^{\text{ext}} \cdot B_{i}^{\text{ext}})\) (independently of the Lorentz forces), where \(\gamma\) is a material parameter that is related to the magnetization (magnetic dipole properties, susceptibility, permeability, moment density, etc.) of the particle (see Feynman et al. \([29]\), Cullity and Graham \([15]\), Boyer \([8]\) or Jackson \([41]\)).

\(^5\) Selections for the values of the parameters in these models are given later in the presentation.

\(^6\) For the velocity ranges in the present applications (significantly below the speed of light), self-induced magnetic fields between particles are unimportant (Jackson \([41]\)).
2.4. Drag forces

It is important to note that, ideally, one would like to eliminate a surrounding gaseous environment and run the process in a vacuum. However, this may not always be feasible, thus we include a drag term, potentially due to interstitial and surrounding gas:

$$
\psi_i^{\text{drag}} = \frac{1}{2} \rho_g C_D |v_i^g - v_i|(v_i^g - v_i) A_i,
$$

(2.16)

where \( C_D \) is the drag coefficient, \( A_i \) is the reference area, which for a sphere is \( A_i = \pi R_i^2 \), \( \rho_g \) is the gas density and \( v_i^g \) is the velocity of the surrounding gas medium. We will assume that \( v_i^g \approx 0 \), and that the gas is of extremely low density, relative to the particles. See Appendix B for more details. We refer the reader to Zohdi [98,106] for more detailed calculations on general fluid(liquid)–particle interaction.

3. Solution strategy

With the governing equations established, we integrate Equation (2.1) using a variable-metric (0 ≤ φ ≤ 1) trapezoidal-like rule to obtain the velocity for \( i \)th particle

$$
\mathbf{v}_i(t + \Delta t) = \mathbf{v}_i(t) + \frac{1}{m_i} \int_{t}^{t+\Delta t} \psi_i^{\text{tot}} dt
$$

$$
\approx \mathbf{v}_i(t) + \frac{\Delta t}{m_i} \left( \phi \psi_i^{\text{tot}}(t + \Delta t) + (1 - \phi) \psi_i^{\text{tot}}(t) \right),
$$

(3.1)

and the position for the by applying the integration process again:

$$
\mathbf{r}_i(t + \Delta t) \approx \mathbf{r}_i(t) + \Delta t (\phi \mathbf{v}_i(t + \Delta t) + (1 - \phi) \mathbf{v}_i(t)),
$$

(3.2)

which can be written as

$$
\mathbf{r}_i^{l+1} = \mathbf{r}_i^l + \mathbf{v}_i^l \Delta t + \frac{\phi (\Delta t)^2}{m_i} \left( \phi (\psi_i^{\text{tot},l+1}) + (1 - \phi) (\psi_i^{\text{tot},l}) \right)
$$

(3.3)

which leads to a coupled set equations for \( i = 1, 2, 3...N_p \) particles. This system will be solved iteratively, furthering approaches found in Zohdi [93–111]. Accordingly, we write Equation (3.3) in a more algorithmic form

$$
\mathbf{r}_i^{l+1} = \mathbf{r}_i^l + \mathbf{v}_i^l \Delta t + \frac{\phi (\Delta t)^2}{m_i} \left( \phi (\psi_i^{\text{tot},l+1}) + (1 - \phi) (\psi_i^{\text{tot},l}) \right).
$$

(3.4)

where the superscript \( L \) is a time-step counter. Because the particle mixture configuration system can significantly change during the deposition process, time-step adaptation is crucial. Within a time-step, the solution steps are, based on a global fixed-point iteration (Appendix C provides details on this solution process):

- (1): Set \( i = 1 \) (particle counter) and \( K = 0 \) (iteration counter);
- (2): If \( i > N_p \) then go to (4)
- (3): If \( i \leq N_p \) then (a) compute position \( \mathbf{r}_i^{l+1,K} \) and (b) go to (2) for next particle \( (i = i + 1) \)
- (4): Compute iterative error metrics (see Appendix C):

$$
Z_K \overset{\text{def}}{=} \frac{\sigma K}{TOL} \quad \text{and} \quad \Lambda_K \overset{\text{def}}{=} \left( \frac{\sum_{i=1}^{N_p} ||\mathbf{r}_i^{l+1,K} - \mathbf{r}_i^{l+1,K-1}||}{\sum_{i=1}^{N_p} ||\mathbf{r}_i^{l+1,K}||} \right)^{1/2},
$$

(3.5)

where \( \sigma K \) and \( \Lambda_K \)

- (5): If \( Z_K \leq 1 \) (met tolerance) and \( K < K_d \) (below preset number of iterations): (a) increment time: \( t = t + \Delta t \), (b) construct the next time step: \( \Delta t^{\text{new}} = \Lambda_K (\Delta t)^{\text{old}} \), (c) select the minimum size: \( \Delta t = \text{MIN}((\Delta t)^{\text{lim}}, (\Delta t)^{\text{new}}) \) and (d) update the particle positions and go to (1)
- (6): If \( Z_K > 1 \) (tolerance not met) and \( K < K_d \) (still iterating) then (a) update the iteration counter: \( K = K + 1 \), (b) reset the particle counter: \( i = 1 \) and (c) go to (2)
- (7): If \( Z_K > 1 \) (tolerance not met) and \( K = K_d \) (at the iteration counter limit) then (a) construct a new time step: \( (\Delta t)^{\text{new}} = \Lambda_K (\Delta t)^{\text{old}} \) and (b) restart at time \( t \) and go to (1)

We observe that in step (5), \( \Lambda_K \) may enlarge the time-step if the error is lower than the preset tolerance. Thus, because the scheme can also enlarge the time-steps if the iterative system converges quickly within an existing time step, to ensure the accuracy of the time-stepping scheme, we add an upper bound to control temporal discretization error, i.e., \( \Delta t \leq \Delta t^{\text{lim}} \).
4. Numerical examples of involving polydisperse depositions

In order to characterize the physical process, we select a model problem, where we consider a group of \(N_p\) spherical particles, of two sizes, randomly dispersed, and initially generated within a cylindrical domain of normalized radius \(R = 1\) (diameter \(D = 2R = 2\)) and length \(L = 8\) (Fig. 6). The ratio of smaller particle diameter, \(d_s\), to total domain diameter, \(D\), was \(d_s/D = 0.05\) for and \(d_l/D = 0.2\) for the larger particles. A Random Sequential Addition (RSA) algorithm (Widom [88]) was employed to initially to place particles in the domain. Thereafter, the dynamics of the particles drove them to a more compacted state.\(^7\) Although we have formulated the system with both a simultaneous electromagnetic field, for the examples of interest, consistent with most mainstream industrial processes, we will only include the electric field in the simulations. There are of course applications where an applied magnetic field would be of interest, however, this is beyond the scope of the current paper. The following simulation parameters were chosen\(^8\):

- Magnetic field, \(B_{\text{ext}} = (0, 0, 0)\) Tesla,
- Electric field, \(E_{\text{ext}} = (-100, 0, 0)\) N/C,
- Charge per unit particle surface area, \(q = 100\) C/kg,
- Density of air, \(\rho_g = 1.225\), kg/m\(^3\),
- Particles were distributed randomly in a parallelepiped domain: \((8 \times 2 \times 2)\) m,
- Total number of particles, \(N = 2000\): 1900 small particles and 100 large particles,
- Radius of small particles, \(R_s = 0.05\) m,
- Radius of large particles, \(R_l = 0.2\) m,
- Density of materials, \(\rho_1 = 2000\) kg/m\(^3\) (binder particles), \(\rho_2 = 5000\) kg/m\(^3\) (functionalizing particles),
- Contact damping parameter, \(c_{\text{cd}} = 10^5\),
- Friction contact parameter \(K_f = 10^7\),
- Coefficient of static friction, \(\mu_s = 0.4\),
- Coefficient of dynamic friction, \(\mu_d = 0.3\),
- Normal bond parameter, \(K_{\text{nb}} = 10^6\) N/m\(^2\) and the exponent in the binding law was set to \(p_b = 2\),
- Rotational/tangential bond parameter, \(K_{\text{rb}} = 10^3\),
- Near-field parameters, \(\bar{\alpha}_1 = 0.5\), \(\beta_1 = 1\), \(\bar{\alpha}_2 = 0.01\), \(\beta_2 = 2\), where the \(\bar{\alpha}\) is per unit mass,
- Total simulation event duration, 1.0 seconds,
- Desired number of fixed point iterations, \(K_d = 10\),
- Trapezoidal-like time-stepping parameter, \(\phi = 0.5\),
- Initial time step size, \(\Delta t = 0.0000025\) seconds,
- Time step upper bound, \(\Delta t_{\text{up}} = 0.00025\) seconds and
- Tolerance for the fixed-point iteration, \(10^{-6}\).

\(^7\) For more details on packing of particles, see Torquato [79], Kansaal et al. [44] and Donev et al. [18–22].

\(^8\) This parameter set is not intended to simulate any specific scenario. The units are SI-with properties being the same for the small and large particles, unless explicitly stated otherwise.
Presently, we did not consider thermal effects. However, the algorithm can be modified to account for coupled thermal effects by solving heat transfer equations, in addition to the linear and angular momentum balances. In thermally-active cases, the parameters such as \( K_{pij} \) may be temperature-dependent, and may soften. For example, the normal stiffness constant for the \( i \)th and \( j \)th particles in the contact law can be written as (here \( \Theta^* \) is the temperature, which is fixed in the present analysis and \( \Theta^* \) is a thermal constant):

\[
K_{pi} = \text{MAX} \left( K_{pi0} \left( e^{-\frac{a_i (\Theta^*)}{\Theta_1}} \right), K_{pi}^{\text{lim}} \right),
\]

and for particle \( j \)

\[
K_{pj} = \text{MAX} \left( K_{pj0} \left( e^{-\frac{a_j (\Theta^*)}{\Theta_1}} \right), K_{pj}^{\text{lim}} \right).
\]

and the average taken at the interface, providing the parameter needed in the contact law. \( K_{pij} = \frac{1}{2} (K_{pi} + K_{pj}) \). In the current example, \( K_{pi0} = 10^7 \text{ N/m}^2 \), where \( K_{pj} = \text{MAX}(K_{pi0} \left( e^{-\frac{a_i (\Theta^*)}{\Theta_1}} \right), K_{pj}^{\text{lim}} \) \), where \( \Theta^* = 500 \text{ K}, K_{pj}^{\text{lim}} = 10^6 \text{ N/m}^2 \), exponent in the contact law was set to \( p = 2 \) (the temperature was fixed to be \( \Theta = 300 \text{ K} \) and the thermal sensitivity parameter was set to \( a = 1 \)). There are many possible representations for temperature-dependency. The overall model has a modular structure which allows one to replace models easily. Although a fully coupled thermal model was not considered here, we refer the reader to Zohdi [103–109] for more details in the general area of thermal multiphysics.

Proceeding, for all particles, we applied an initial velocity of \( \mathbf{v}(t = 0) = (-1,0,0) \), projecting them directly towards the substrate, in conjunction with gravity \( (g = -(-9.81,0,0) \text{ m/s}^2) \). The electric field starts below a \( y-z \) parallel flat plane at \( (2,0,0) \). The center of the starting configuration is at \( (3.5,0,0) \) and the substrate is at \( (-2,0,0) \). In the upcoming simulations, the smaller blue particles are “binder” particles and the larger red particles are “functionalizing” particles. We considered four different cases:

- **Example #1: no external electric field and no particle-to-particle ionization interaction effects** – In this case the particle system simply falls due to initial starting velocity. The impact with the substrate is relatively mild, but not controlled (Fig. 7).

- **Example #2: external electric field and no particle-to-particle ionization interaction effects** – In this case the particle system falls due to initial starting velocity and the attraction of the electric field. The impact with the substrate is intense and not controlled. The strong electric field causes more intense impact than before (Fig. 8). To qualitatively consider the added effects of the electric field in the impact velocity, consider an isolated charged mass with position vector denoted by \( \mathbf{r} \), governed by \( \dot{\mathbf{r}} = \mathbf{v} \), \( \mathbf{v} = \mathbf{\dot{v}} \)

\[
m \ddot{\mathbf{v}} = q \mathbf{E}^{\text{ext}},
\]

with \( \mathbf{r}(t = 0) = r_0 \mathbf{e}_1 \), \( \mathbf{v}(t = 0) = v_0 \mathbf{e}_1 \) and \( \mathbf{E}^{\text{ext}} = E_1^{\text{ext}} \mathbf{e}_1 \), consequently, for a single particle:

\[
v_1(t) = v_0 + \frac{q}{m} E_1^{\text{ext}} t = r_0 + v_0 t + \frac{q}{2m} E_1^{\text{ext}} t^2.
\]

This directly shows how the intensity of the impact is correlated to the mass, charge and electric field.

- **Example #3: no external electric field and particle-to-particle ionization interaction effects** – In this case the particle system falls due to initial starting velocity. The mutual ionization forces the stream to collapse upon itself and form three distinct “droplets”. The two upper droplets reconnect. The two connected drops then continue to fall and connect with the lower droplet, and the entire system then progresses to make contact with the substrate. The impact with the substrate is mild (Fig. 9).

- **Example #4: external electric field and particle-to-particle ionization interaction effects** – In this case the particle system falls due to initial starting velocity and the electric field. The mutual ionization forces the stream to collapse upon itself and form three distinct “droplets”. The two upper droplets reconnect while the lower one attatches to the substrate. The two connected drops then continue to fall and connect with the droplet on the substrate. The impact with the substrate is more intense, but controlled by the electric field, which pins the material to the substrate (Figs. 10 and 11). For comparison purposes, we also ran the simulation with only binder particles as well (Fig. 12). The break up is less immediate, primarily because of less Plateau–Rayleigh perturbations (described further momentarily).

Thus, it is observed that there is a direct correlation with the ionization strength between particles in the powder and the more fluid-like behavior, which the electric field can control. With no ionization, the system behaves as a loose powder, which is nearly impossible to control as a deposition. As the ionization is increased, the balance between mutual attraction and repulsion leads to surface tension like effects yielding coherent aggregate “droplets” of the powder–mixture material.
Fig. 7. Example #1: no external electric field and no particle-to-particle ionization interaction effects. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

Remark 1. In the analysis of standard (nonparticulate) fluids, the breakup of a long column of fluid with perturbations (longitudinal waviness) was first investigated experimentally by Plateau in 1873, who found that a vertically falling stream of water will break up into drops if its wavelength is greater than approximately 3.13–3.18 times its diameter. Subsequently, Rayleigh analytically proved that a wavy falling column of non-viscous liquid (with circular cross-section) should break up into drops if its wavelength exceeded its circumference. This type of instability is driven by surface tension, which forces fluids to minimize their surface area. See Papageorgiou [65] and Eggers [28] for more details. In the case of a charged
particulate medium, the degree of near-field strength plays the role of surface tension. An uncharged particulate medium will not exhibit this phenomenon.

**Remark 2.** As we have mentioned previously, because the scheme can also enlarge the time-steps if the iterative system is converges quickly within an existing time step, to ensure the accuracy of the time-stepping scheme, we add an upper bound to control temporal discretization error, i.e., $\Delta t \leq \Delta t^{\text{lim}}$. We started all simulations with extremely small time step sizes, and
allowed the error estimation and time-step adaptation to auto-correct to the proper size. Fig. 13 shows a typical scenario, taken from Example #4, were the sizes were initially small and adapted until they met the limit set by the algorithm (user specified).

Remark 3. The use of so-called interaction lists is advantageous to speed-up calculations and to extend such simulations to very large particle systems. These lists are constructed, for each particle, by taking neighboring particles within a radius of
Fig. 10. Example #4: external electric field and particle-to-particle ionization interaction. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

influence. The list is then updated periodically during the simulations. This significantly reduces the computation time used in contact search and other intra-particle calculations, which are $N^2$ operations. In the simulations that were presented: (a) For each particle, a nearest-neighbor list was constructed at the beginning of the simulation; (b) For a subinterval of time, the interaction for each particle was restricted to these neighbors and (c) the lists were updated after that interval expired and the process repeated. See Zohdi [102–109] for details.
5. Summary and extensions

In related manufacturing processes the particle–mixture may be highly saturated with an interstitial fluid or embedded within a fluid medium. In such cases, the fluid dynamics interaction with the particles can play a major role on the system behavior. This can also involve the evolution of heat. A spatio-temporal discretization can be used to solve coupled fluid particle systems, by employing Finite Element, Finite Difference or Finite Volume methods coupled to Discrete Elements.
Fig. 12. Baseline “Reference” Monodisperse Example: external electric field and particle-to-particle ionization interaction. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

Examples of such particle–fluid systems are colloidal Ferrofluids consisting of ferromagnetic nanoscale particles embedded in an organic solvent.9 Such fluids have a long history (Winslow [89,90]) in industrial applications as electrorheological fluids

9 The particles are usually an iron-based compound, for example hematite or magnetite.
involving hydraulic valves, brakes, actuators and computer screens. Typically, the volume fraction of the particles is under five percent, and they are coated with a surfactant (oleic acid, citric acid, tetramethylammonium hydroxide, soy lecithin, etc.), in order to avoid agglomeration by mitigating near-field interaction effects between particles. We refer the reader to Albrecht et al. [2], Andelman and Rosensweig [3] and Berger et al. [6] for reviews. Extensions to this type of analysis could include parameter studies on the changes in dynamics due to strong magnetic fields. Although magnetic fields are somewhat less used than electric fields for deposition, they have certain unique utility by being able to “bend” sprays and depositions (see Martin [50,51] for the state of the art).

A critical extension of these types of manufacturing processes is to ascertain the effective properties of the particle-functionalized materials after deposition. One of the primary properties of interest is the overall “effective” electrical conductivity, defined via Ohm’s Law:

\[
\langle J \rangle_\Omega = \sigma^* : \langle E \rangle_\Omega. \tag{5.1}
\]

where \(\sigma^*\) is the effective conductivity for the mixture, \(\langle E \rangle_\Omega\) is the volume averaged electric field and \(\langle J \rangle_\Omega\) is the volume averaged current, the averaging operator is defined as \(\langle \cdot \rangle_\Omega = \frac{1}{V_\Omega} \int_\Omega \cdot d\Omega\) over a statistically representative volume element with domain \(\Omega\). Similarly, for other important properties, particularly in the realm of printed electronics, are the effective electrical permittivity (\(\epsilon^*\))

\[
\langle D \rangle_\Omega = \epsilon^* : \langle E \rangle_\Omega, \tag{5.2}
\]

where \(\langle E \rangle_\Omega\) is the volume averaged electric field, \(\langle D \rangle_\Omega\) is the volume averaged electric field flux and the overall magnetic permeability (\(\mu^*\))

\[
\langle B \rangle_\Omega = \mu^* : \langle H \rangle_\Omega, \tag{5.3}
\]

where \(\langle H \rangle_\Omega\) is the volume averaged magnetic field and \(\langle B \rangle_\Omega\) is the volume averaged magnetic field flux. We refer the reader to Torquato [79], Jikov et al. [42], Hashin [37], Mura [53], Markov [49] for theoretical aspects and for more computationally-oriented approaches, Ghosh [33], Ghosh and Dimiduk [34], Zohdi and Wriggers [110], Zohdi [111] and recently, Matous et al. [52] for a review of the state-of-the-art in multiscale methods for nonlinear heterogeneous materials. The determination of effective properties of depositions is currently being pursued by the author.

Finally, in summary, the adoption of discrete and continuum micromechanical models driven by computational methods in additive manufacturing and 3D printing has the potential to bring a level of systematic analysis that can make it a reliable large-scale manufacturing process (see Avila et al. [5], Dornfeld et al. [23], Vijayaraghavan et al. [80], Vijayaraghavan and Dornfeld [81], Dornfeld and Lee [24] and Dornfeld [25]). From the view of practical additive manufacturing simulation tools, there are several hurdles. Additive manufacturing, which is based on deposition, by itself is inadequate for high-precision applications and needs to be combined with classical manufacturing processes, as high-precision surface milling, as outlined in a recent US National Academies Report (Zohdi and Dornfeld [112]). Thus, it is important to realize that guided by simulation, additive and subtractive processes should be combined to model deposition and removal of material, while simultaneously controlling the tolerances, surface finish and, ultimately, product quality. The development of models and simulation tools for additive and subtractive processes is of current actively being investigated by the author.

**Appendix A. Contact area parameter and alternative models**

Following Zohdi [95,97,98,102,104–106], and referring to Fig. 5, one can solve for an approximation of the common contact radius \(a_{ij}\) (and the contact area, \(A_{ij} = \pi a_{ij}^2\)) by solving the following three equations: (a) \(a_{ij}^2 + L_i^2 = R_i^2\), (b) \(a_{ij}^2 + L_j^2 = R_j^2\), and (c) \(R_i + R_j + a_{ij} = D\).
\( R_i^2 \) and (c) \( L_i + L_j = ||r_i - r_j|| \), where \( R_i \) is the radius of particle \( i \), \( R_j \) is the radius of particle \( j \), \( L_i \) is the distance from the center of particle \( i \) and the common contact interpenetration line and \( L_j \) is the distance from the center of particle \( j \) and the common contact interpenetration line, where the extent of interpenetration is \( \delta_{ij} = R_i + R_j - ||r_i - r_j|| \). The above equations yield an expression \( a_{ij} \), which yields an expression for the contact area parameter

\[
A_{ij}^c = \pi a_{ij}^2 = \pi (R_i^2 - L_i^2),
\]

where \( L_i = \frac{1}{2} \left( ||r_i - r_j|| - \frac{R_i^2 - R_j^2}{||r_i - r_j||} \right) \). Alternative models, building on Hertzian-contact which connect the relative proximity of the particles and other metrics to the contact force, \( \Psi_{ij} \), building on, for example, Hertzian contact models, can be implemented with no particular difficulty. For the remainder of the analysis, we shall use the deformation metric in Equation (2.4). For detailed treatments, see Wellman et al. [83–87] and Avci and Wriggers [4]. We note that with the appropriate definition of parameters, one can recover Hertz, Bradley, Johnson–Kendel–Roberts, Derjaguin–Muller–Toporov contact models. For more details, we refer the reader to Johnson [43]. Clearly, a finer resolution of the deformation within a particle, it must be treated as a deformable continuum, requiring, for example the Finite Element Method for the contacting bodies. This requires a large computational effort that is beyond the scope of this paper (see Wriggers [91,92] and Zohdi and Wriggers [110]).

### Appendix B. The effects of drag

For the drag that does not emanate from surrounding particles or that walls, we will employ a general phenomenological model

\[
\Psi_{drag}^i = \frac{1}{2} \rho_g C_D ||\mathbf{v}^g - \mathbf{v}_i|| (||\mathbf{v}^g - \mathbf{v}_i||) A,
\]

where \( C_D \) is the drag coefficient, \( A \) is the reference area, which for a sphere is \( A = \pi R^2 \), \( \rho_g \) is the density of the ambient gas environment and \( \mathbf{v}^g \) is the velocity of the surrounding medium which, in the case of interest, is air. Generally speaking, the drag coefficient, which is an empirical parameter which attempts to represent the action of the gas forces on an object, is not a constant, and would vary with, for example, the Reynolds number. In the zero Reynolds number limit the drag would be that of a Stokesian regime. One possible way to represent the drag coefficient is with a piecewise definition, as a function of the Reynolds number (Chow [14]):

- For \( 0 < Re \leq 1 \), \( C_D = \frac{24}{Re^2} \).
- For \( 1 < Re \leq 400 \), \( C_D = \frac{24}{Re^2 + 0.48} \).
- For \( 400 < Re \leq 3 \times 10^5 \), \( C_D = 0.5 \).
- For \( 3 \times 10^5 < Re \leq 2 \times 10^6 \), \( C_D = 0.000366 Re^{0.4275} \).
- For \( 2 \times 10^6 < Re < \infty \), \( C_D = 0.18 \).

where the local Reynolds number for a particle is \( Re = \frac{d \rho g ||\mathbf{v}^g - \mathbf{v}_i||}{\mu_g} \) and \( \mu_g \) is the gas viscosity. The viscosity coefficient for air is \( \mu_g = 0.000018 \text{ Pa/s} \). Using the hybrid model reduces the drag at the lower Reynolds number regimes, relative to simply picking a constant mid-range value for \( C_D \) (for example \( C_D = 0.5 \)), thus producing less drag than a constant drag coefficient. The piecewise drag law of Chow [14] is a mathematical description for the Reynolds number over a wide range and is a curve-fit of extensive data from Schlichting [72]. As observed in the experimental data, the mathematical function exhibits a discontinuity at \( Re = 3 \times 10^5 \), although in an explosion the time a particle spends at this Reynolds number is almost negligible. In the low velocity (low Reynolds number) limit a Stokesian model is most appropriate, which is what the drag law above attempts to incorporate. The drag forces are significantly smaller with a Stokesian model. Comparing a purely Stokesian drag law, which would be valid for small particles and laminar flow (low Reynolds number)

\[
\Psi_{drag,Stokesian} = c(\mathbf{v}^g - \mathbf{v}_i) = \mu_g 6\pi R_i (\mathbf{v}^g - \mathbf{v}_i),
\]

where \( \mu_g \) is the gas viscosity. We observe the following:

\[
\frac{||\Psi_{drag,Stokesian}||}{||\Psi_{drag}||} = \frac{12 \mu_g}{\rho_g C_D R ||\mathbf{v}^g - \mathbf{v}_i||}.
\]

For typical parameters for air and spherical particles (using \( C_D = 0.5 \), which is a mid-range value from the piecewise drag law introduced earlier), we have

\[
\frac{||\Psi_{drag,Stokesian}||}{||\Psi_{drag}||} = \frac{12 \mu_g}{\rho_g C_D R ||\mathbf{v}^g - \mathbf{v}_i||} \approx 0.0004 \frac{R ||\mathbf{v}^g - \mathbf{v}_i||}{||\mathbf{v}^g - \mathbf{v}_i||}.
\]
which indicates that for extremely small particles and low velocities, the Stokesian model dominates, while for larger particles and large velocities, the phenomenological model dominates.

In order to determine the relative strengths of the gas drag force to the electromagnetic forces acting on the particle, consider the following model for an isolated particle:

\[
m\dot{v} = \Psi^{\text{drag}} + \Psi^{e+m} = C_D \frac{1}{2} \rho_p \frac{V}{m} |v - \bar{v}|^2 \tau + \bar{q} \rho_p \vec{v} (\vec{E}^{\text{ext}} + \vec{v} \times \vec{B}^{\text{ext}}),
\]

where \( \bar{q} \) is the charge per unit mass, \( \rho_p \) is the density of the particle and \( V = \frac{4}{3} \pi R^3 \). The general ratio is

\[
\lambda(v) = \frac{|\Psi^{e+m}|}{|\Psi^{\text{drag}}|} = \frac{|\bar{q} \rho_p V (\vec{E}^{\text{ext}} + \vec{v} \times \vec{B}^{\text{ext}})|}{C_D \rho_p \frac{V}{m} |v - \bar{v}|^2} = \frac{8|\bar{q} \rho_p R}{3C_D \rho_p} \left( \frac{||\vec{E}^{\text{ext}} + \vec{v} \times \vec{B}^{\text{ext}}||}{||v - \bar{v}||^2} \right).
\]

Using the Triangle and Cauchy–Schwarz inequalities:

\[
\lambda(v) \leq \frac{8|\bar{q} \rho_p R}{3C_D \rho_p} \left( \frac{||\vec{E}^{\text{ext}}|| + ||\vec{v}|| ||\vec{B}^{\text{ext}}||}{||v - \bar{v}||^2} \right),
\]

which leads to

\[
||\vec{E}^{\text{ext}}|| + ||\vec{v}|| ||\vec{B}^{\text{ext}}|| \geq \frac{3C_D \rho_p ||v - \bar{v}||^2}{8|\bar{q} \rho_p R}.
\]

If we set \( \lambda = 1 \), we obtain an expression for the electromagnetic forces to have parity with the drag force. In the case when the magnetic field is negligible, we have:

\[
||\vec{E}^{\text{ext}}|| = \frac{3C_D \rho_p ||v - \bar{v}||^2}{8|\bar{q} \rho_p R}.
\]

In the case when the electric field is negligible, we have

\[
||\vec{B}^{\text{ext}}|| \geq \frac{3C_D \rho_p ||v - \bar{v}||^2}{8|\bar{q} \rho_p R||v||}.
\]

Appendix C. Iterative methods

The system represented by Equation (3.4) can be iteratively solved by recasting it in the following form

\[
r_i^{L+1,K} = \frac{r_i^L + \vec{v}_i^L \Delta t + (\phi \Delta t)^2}{m_i} \Psi_i^{\text{tot},L+1,K-1} + \frac{\phi (\Delta t)^2}{m_i} (1 - \phi) \Psi_i^{\text{tot},L},
\]

which is of the form

\[
r_i^{L+1,K} = G(r_i^{L+1,K-1}) + L_i,
\]

where \( K = 1, 2, 3, \ldots \) is the index of iteration within time step \( L + 1 \) and

- \( \Psi_i^{\text{tot},L+1,K-1} \)-defined by \( \Psi_i^{\text{tot},L+1,K-1} = \Psi_i^{\text{tot},L+1,K-1}(r_i^{L+1,K-1}, r_2^{L+1,K-1}, \ldots r_N^{L+1,K-1}) \),
- \( \Psi_i^{\text{tot},L} \)-defined by \( \Psi_i^{\text{tot},L}(r_1^L, r_2^L, \ldots, r_N^L) \),
- \( G(r_i^{L+1,K-1}) = \frac{(\phi \Delta t)^2}{m_i} \Psi_i^{\text{tot},L+1,K-1} \) and
- \( L_i = r_i^L + \vec{v}_i^L \Delta t + \frac{\phi (\Delta t)^2}{m_i} (1 - \phi) \Psi_i^{\text{tot},L} \).

The term \( L_i \) is a remainder term that does not depend on the solution. The convergence of such a scheme is dependent on the behavior of \( G \). Namely, a sufficient condition for convergence is that \( G \) is a contraction mapping for all \( r_i^{L+1,K} \), \( K = 1, 2, 3, \ldots \). In order to investigate this further, we define the iteration error as

\[
\bar{r}_i^{L+1,K} = r_i^{L+1,K} - r_i^{L+1}.
\]

A necessary restriction for convergence is iterative self-consistency, i.e. the “exact” (discretized) solution must be represented by the scheme, \( r_i^{L+1} = G(r_i^{L+1}) + L_i \). Enforcing this restriction, a sufficient condition for convergence is the existence of a contraction mapping

\[
||r_i^{L+1,K} - r_i^{L+1}|| = ||G(r_i^{L+1,K-1}) - G(r_i^{L+1})|| \leq \eta ||r_i^{L+1,K-1} - r_i^{L+1}||
\]

where \( \eta \) is a contraction constant.
where, if $0 \leq \eta^{i+1,K} < 1$ for each iteration $K$, then $\eta^{i+1,K} \rightarrow 0$ for any arbitrary starting value $\eta^{i+1,K} = 0$, as $K \rightarrow \infty$, which is a contraction condition that is sufficient, but not necessary, for convergence. The convergence of Equation (C.1) is scaled by $\eta \propto (\phi \Delta t)^2$. Therefore, we see that the contraction constant of $G$ is (a) directly dependent on the magnitude of the interaction forces $|\nabla W|$), (b) inversely proportional to the masses $m_i$ and (c) directly proportional to $(\Delta t)^2$. Thus, decreasing the time step size improves the convergence. In order to maximize the time-step sizes (to decrease overall computing time) and still meet an error tolerance on the numerical solution's accuracy, we build on an approach originally developed for continuum thermo-chemical multiphield problems [Zohdi [93]], where one assumes: (1) $\eta^{i+1,K} \approx S(\Delta t)^2$, $(S$ is a constant) and (2) the error within an iteration behaves according to $(S(\Delta t)^p)^K \eta^{i+1,K} S = \eta^{i+1,K}$. $K = 1, 2, \ldots$, where $\eta^{i+1,K} \approx r^{i+1,K} = r^0$ is the initial norm of the iterative (relative) error and $S$ is intrinsic to the system. For example, for second-order problems, due to the quadratic dependency on $\Delta t$, $p \approx 2$. The objective is to meet an error tolerance in exactly a preset (the analyst sets this) number of iterations. To this end, one writes $(S(\Delta t)^p)^K \eta^{i+1,K} = TOL$, where $TOL$ is a tolerance and $K_d$ is the number of desired iterations. If the error tolerance is not met in the desired number of iterations, the contraction constant $\eta^{i+1,K}$ is too large. Accordingly, one can solve for a new smaller step size, under the assumption that $S$ is constant,

$$\Delta t_{tol} = \Delta t \left( \frac{TOL}{S(\Delta t)^p} \right)^{1/p} \left( \frac{1}{\eta^{i+1,K}} \right) \approx \Delta t \Lambda K. \quad (C.5)$$

The assumption that $S$ is constant is not critical, since the time steps are to be recursively refined and unrefined throughout the simulation. Clearly, the expression in Equation (C.5) can also be used for time step enlargement, if convergence is met in less than $K_d$ iterations (typically chosen to be between five to ten iterations).

References

K. Y. A. E. K. C.

I. S. V.V. E.A.

T. S.R.

Science

Technol.

Methods

relationships

Sci.

tronics,

Sevostianov,

Eggers,

Nakamura,

Martin,

Martin,

Pöschel,

Ghosh,

Hull,

Oñate,

Mukherjee,

46

Introduction

Handbook

Comput.

Analysis

Molecular

Phys.

Classical

Elementary

Liao,

Gold

Y.

T.I.

Markov,

39

Effects

Engineering

of

the

element

pastoriza-Santos,

pastoriza-Santos,

Pastoriza-Santos,

Kachanov,

Kachanov,

M.J.

3rd

coatings,

the

elastomeric

coating

their

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,

with

the

microstructure,