Transport and solidification phenomena in molten microdroplet pileup

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This article presents a predominantly numerical investigation of the transient transport phenomena occurring during the pileup (deposition one upon another) of molten, picoliter-size liquid metal droplets relevant to a host of novel micromanufacturing processes. The investigated phenomena last fractions of a millisecond in severely deforming domains of typical size of a small fraction of a millimeter. The prevailing physical mechanisms of the pileup process (occurring simultaneously) are identified and quantified numerically. These are the fluid mechanics of the bulk liquid, capillary effects at the liquid–solid interface, heat transfer, solidification, and thermal contact resistance effects at all interfaces. In terms of values of the Reynolds, Weber, and Stefan number the following ranges are covered: \( Re = 281 - 453 \), \( We = 2.39 - 5.99 \), and \( Ste = 0.187 - 0.895 \). This corresponds to molten solder droplets impinging at velocities ranging between 1.12 and 1.74 m/s having an average diameter of \( \approx 78 \) \( \mu \)m. The initial substrate temperature ranges between 25 and 150°C. The initial droplet temperature is 210°C. The numerical model presented is based on a Lagrangian formulation of the Navier–Stokes equations accounting for surface tension, thermal contact resistance, solidification, and a Navier slip condition at the dynamic contact line. Results of simulations are presented showing the effect of thermal contact resistance and slip at the dynamic contact line on the transients and the outcome of a pileup. Comparisons of the simulated pileup with experimental visualizations are shown, demonstrating good agreement in cases where inertia dominates over capillary effects. For decreasing Stefan number (i.e., higher substrate temperatures) an increasing importance of wettability is observed. For these cases the limitations of the employed popular boundary condition at the dynamic contact line is demonstrated and the need for experimental data (currently nonexistent in the literature) that would yield an improved condition at the contact line accounting for the temperature dependence of wetting phenomena is underpinned. © 2002 American Institute of Physics. [DOI: 10.1063/1.1492019]

I. INTRODUCTION

Research in droplet/surface interactions has received much attention in the past, dating back to the experimental studies of Worthington.1,2 Droplets of various sizes and of different fluids appear in many areas of modern industrial applications. A number of different emerging technologies are based on the controlled deposition of droplets ranging from biotechnologies to microelectronics. A central example of such technologies dealing with the deposition of molten microdroplets is chip packaging in microelectronics. Scale reduction of such packages has predominantly been achieved by replacing wire connections from electrical connector pads by arrays of solder balls/bumps featuring a smaller pitch size and allowing for higher connector densities. A very efficient way of producing such solder bump arrays is to use solder jetting to deposit individual droplets directly onto electrical connector pads.3,4

Another process termed with a variety of names such as digital microfabrication,5 microcasting,6 ballistic particle manufacturing,7 rapid prototyping,7 and solid freeform8–10 exemplifies a different technology based on the deposition of individual molten droplets. The objective of this technique is to deposit a multitude of individual molten droplets at precisely defined locations to build up layers of droplets that solidify upon impact into coherent three-dimensional structures. This technique does not rely on a conventional cast and features high flexibility, the ability to produce near-net shape products, as well as the ability of a rapid conversion of computer aided design object data into a real object. Furthermore, unlike other rapid prototyping processes, processes based on molten droplet deposition produce metal parts. Preliminary experimental studies have shown that metallic objects created by digital microfabrication exhibit higher tensile strength9 and lower porosity than objects created by conventional casting methods.11 Common to the above mentioned processes is the importance of the understanding of transient behavior of droplet interactions with substrates on which they are deposited. Advancement and implementations of such processes are only possible through a rigorous enhancement of the knowledge base associated to these processes. A multiplicity of physical phenomena have to be considered. The fluid mechanics cover a severely deforming free surface interacting with the ambient environment. Furthermore, the fluid flow interacts with a solid of arbitrary shape.
leading to strong deformations, possible breakup (bounce off), recoalescence, and oscillations. It is also necessary to confront the very intricate field of wetting in the contact line region of the droplet. Thermodynamics poses several challenges when considering multiphase and multimode heat transfer (i.e., evaporation, solidification). Chemistry can also come into play through various reactions from combustion to surface oxidation. Hence, droplet physics offers a multitude of serious challenges in basic research and is, at the same time, directly related to significant technology development. An in-depth understanding of the relevant scientific issues can lead to technological advancements with marked beneficial societal implications.

The aim of this work, based on the described industrial processes above, is to shed more light on the complex interplay of the different physical phenomena occurring during the successive (one over the other) deposition (pileup) of molten droplets. More specifically, the work is focused on the process in which a molten droplet impacts on a previously impacted and solidified droplet of the same material. In the light of the processes mentioned above, a two droplet pileup can be considered as the simplest building block in the creation of a complex three-dimensional structures. In addition, there exists many times the need to place more solder material in one position than that contained in a single droplet in chip packaging applications. Since the reliable range of droplet sizes in solder jetting is rather limited, droplet pileup is a viable alternative. A schematic of the investigated process is shown in Fig. 1. As already emphasized in Fig. 1 only the axisymmetric case of the pileup will be considered, which is of course in line with the actual printing approach where the printhead works on a “stop and drop” fashion. Furthermore, the focus is on investigating eutectic melts since the majority of solder materials in electronics manufacturing are eutectic, exemplified by eutectic tin–lead solder (63% Sn–37% Pb).

Unfortunately, there is very little information reported in the literature on droplet impact on nonflat substrates and pileups (Che et al.,12 Orme et al.,8–11 Gao and Sonin,5 Liu et al.,13). Che et al.12 studied numerically the case of droplets impacting on top of each other. The solution method accounted for the conservation of mass, momentum, and energy both for the ambient, gaseous environment, the molten droplets, and the substrate. A fixed grid finite difference scheme was utilized in order to solve the equations numerically. A front-tracking method was employed in order to account for the free surface and the solidification front. Issues like thermal contact resistance, the dynamic contact line, and the release of latent heat in the solidification process that can have a dominant effect on the process were neglected. No experimental results were reported to compare or corroborate their numerical results. No surface structures like ripples were reported as has been shown and verified for single droplets impacting on flat substrates by, for instance, Waldvogel and Poulikakos14 exemplifying the limitations of this study. The work by Orme et al.8–11 is focused on the employment of molten droplets in digital microfabrication. Thus, the work reports more on methods of how to create objects by dropwise buildup and deposition strategies than on the prevailing physical phenomena and neglects the fluid dynamics. However, some analytical/numerical estimations on critical issues like splat cooling, remelting, and thus adhesion properties between subsequent droplets are reported. Gao and Sonin5 reported analytical estimates of cooldown and solidification time scales for columnar wax droplet deposition (i.e., pileup of many droplets, multiple pileups). They also presented endshapes of multiple pileups for candelilla. Liu et al.13 reported numerical studies of droplets impacting on different nonflat substrates. The governing equations for transient, axisymmetric, viscous, incompressible flow, including surface tension effects were solved employing a modified version of Ripple.15 The dynamic contact line, heat transfer, and solidification were not considered. Free surfaces were modeled using the volume-of-fluid method. Simulations were performed using tungsten droplets with diameters in the range of 10–60 μm, at impact velocities on the order of O(100 m/s). The results show only fluid mechanical aspects of the impingement process including the splashing of the droplets. Furthermore, only velocity fields of unverified accuracy are presented. Regarding experiments on the pileup problem, transient visualizations/measurements are nonexistent in the scientific literature.

In contrast to multiple or single droplet impacts on non-flat substrates a large amount of work is reported in the scientific literature on single droplets impacting on flat substrates covering and combining the areas of analytical modeling, experimental visualization, as well as numerical modeling. A very good insight into the impact of single droplets on flat substrates, which is in many respects strongly coupled to the problem of the pileup, is given in the review articles by Haferl et al.,16 and Attinger et al.17

An important very difficult subproblem in droplet deposition problems is that of surface wetting by the droplet. In the field of surface science this topic has received a lot of attention, however, without conclusive results to date. The relevant issue is the dynamic contact line behavior and the controversial determination of dynamic contact angles. Approaches to these connected problems span a wide area from continuum18 down to molecular level.19–22 Another related microscale problem that so far has not been resolved is that of transient thermal contact resistance between an impacting microdroplet and a target object. With reference to the area of dynamic contact line behavior in the presence of heat transfer and solidification, a major research effort is needed...
in the future, because the understanding of molecular phenomena appears to be necessary in order to provide reliable closure conditions for the continuum models.

The focus of this predominantly numerical work is on the fluid mechanics of the impact process, the heat transfer between the impinging and a predeposited already solidified droplet, the subsequent solidification of the molten material, and the related wetting occurring during a pileup of molten solder droplets. A further aim is clearly to create a predictive tool for pileup processes relevant for a host of industrial applications. Thus, the order of magnitude of the two foremost important dimensionless fluid dynamics numbers, Reynolds and Weber, are thereby \( O(100) \) and \( O(1) \), respectively. For molten eutectic solder droplets of picoliter size, this range corresponds to an order of magnitude of the impact velocity and initial diameter of \( O(1 \text{ m/s}) \) and \( O(100 \mu m) \). This is approximately the range covered by the industrial applications described above and experimentally visualized in an accompanying article.\(^23\) These visualizations of the transient impact processes are employed in order to compare and validate the numerical model presented. Furthermore, a systematic study of the employed models for thermal contact resistance and wetting on the pileup process is reported for model cases.

II. The MATHEMATICAL MODEL

A. Outline of the computational domain

The computational domain of the pileup problem is shown in Fig. 2. Since the problem considered is assumed axisymmetric only half of the cross section is taken into account. The entire domain is divided into four regions. For the solution of the fluid mechanics equations, only the domain of the impacting droplet as well as the boundary of the presolidified droplet are considered. The solution of the energy equation covers all four regions in Fig. 2. The heat transfer solution method contains special features necessitated by the two indicated interfaces, which will be discussed below. The multilayer substrate is “manufactured” numerically to duplicate the gold coated wafer substrate employed in the experiments,\(^23\) described in Sec. III, and thus consists of four sublayers (i.e., gold, TiW, silicon oxide, and silicon). The length scales in the computational domain are equal to the experimental cases. The substrate width is truncated at twice the droplet diameter measured from the line of symmetry. It has been shown that this is sufficient for the thermal solution.\(^14,24,25\) Furthermore, based on work in Refs. 25–28 it is assumed that the influence of the inert, gaseous environment on the impact process is negligible (i.e., no heat transfer from the droplet/substrate to the environment, no oxidation of the free surfaces, and no frictional drag form the gaseous environment imposed on the impinging droplet).

B. Fluid mechanics

1. Governing equations for the fluid flow

The fluid dynamics of the liquid region of the pileup is modeled as an unsteady, viscous, incompressible flow with constant thermophysical properties. The mathematical model is formulated to simulate the axisymmetric impact of an initially spherical molten droplet on a presolidified droplet beginning at the instant of contact. The model for the fluid flow is based on the Navier–Stokes equations, applied to the axisymmetric coordinate system shown in Fig. 2, in which \( r \), \( z \), and \( \theta \) are the radial, axial, and azimuthal coordinates. The dimensionless forms of the governing equations in Lagrangian formulation are as follows.\(^29\)

Continuity equation:

\[
\frac{\partial}{\partial t} \rho + \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho \frac{\partial u}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \rho \frac{\partial u}{\partial \theta} \right) + \frac{1}{r} \frac{\partial}{\partial z} \left( r \rho \frac{\partial u}{\partial z} \right) = 0,
\]

Momentum equation in radial direction:

\[
\frac{\partial}{\partial t} \frac{r^2}{\rho} \frac{\partial u}{\partial r} - \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \rho \frac{\partial \bar{u}}{\partial \theta} \right) - \frac{1}{r} \frac{\partial}{\partial z} \left( r \rho \frac{\partial \bar{u}}{\partial z} \right) + \frac{\partial}{\partial \theta} \left( r \rho \frac{\partial \bar{u}}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( r \rho \frac{\partial \bar{u}}{\partial z} \right) = 0,
\]

Momentum equation in axial direction:

\[
\frac{\partial}{\partial t} \frac{r^2}{\rho} \frac{\partial v}{\partial z} - \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \rho \frac{\partial \bar{v}}{\partial \theta} \right) - \frac{1}{r} \frac{\partial}{\partial \bar{u}} \left( r \rho \frac{\partial \bar{v}}{\partial \bar{u}} \right) + \frac{\partial}{\partial \theta} \left( r \rho \frac{\partial \bar{v}}{\partial \theta} \right) + \frac{\partial}{\partial \bar{u}} \left( r \rho \frac{\partial \bar{v}}{\partial \bar{u}} \right) = 0,
\]

where \( \partial_i \) denotes the derivative with respect to the \( i \)th variable. Employing the Lagrangian formulation of the Navier–Stokes equation has the advantage that no convective terms are present in the equations alleviating the need for “up-winding” in the numerical solution. More importantly, it facilitates the handling of the free surface since boundary elements and nodes representing the free surface are always identifiable and do not have to be determined using a front tracking scheme. The artificial compressibility method by Chorin\(^30\) is employed in order to solve the continuity equation. The Mach number \( Ma \) in Eq. (1) can thus be considered as an artificial compressibility parameter. The value of the latter is key for the performance of the method regarding convergence speed, accuracy, and stability of the numerical scheme.\(^24\) It can be related to the speed of the pressure waves in the fluid stemming from the hyperbolic character of the modified equations. Clearly, the larger the value of the artificial compressibility parameter \( Ma \), the more “incompressible” the equations; however, large values of the Mach number make the equations numerically very stiff with the associated problems coupled with numerical stiffness.

To complete the model, the dimensionless stress tensor components \( \sigma_{ij} \) are defined as
\[ \bar{\sigma}_{RR} = - P + \frac{2}{\text{Re}} \partial_R U, \quad \bar{\sigma}_{ZZ} = - P + \frac{2}{\text{Re}} \partial_Z V, \]
\[ \bar{\sigma}_{\theta \theta} = - P + \frac{2}{\text{Re}} \frac{U}{\bar{R}} \quad \bar{\sigma}_{RZ} = \frac{1}{\text{Re}} (\partial_R U + \partial_Z V). \]

The dimensionless initial and boundary conditions are respectively (see Fig. 2),
\[ \tau = 0: \quad U = 0, \quad V = -1, \quad P = \frac{4}{\text{We}}; \]
\[ R = 0: \quad U = 0, \quad \partial_R V = 0, \]
\[ p(s): \quad U = 0, \quad V = 0, \]
where \( p(s) \) describes the surface of the presolidified droplet with respect to the arc length \( s \), Fig. 2. The stress balance at the free surface is formulated as follows:
\[ \bar{n}_R \bar{\sigma}_{RR} + \bar{n}_Z \bar{\sigma}_{ZZ} = - \frac{2 H}{\text{We}} n_R, \]
\[ \bar{n}_R \bar{\sigma}_{RZ} + \bar{n}_Z \bar{\sigma}_{ZZ} = - \frac{2 H}{\text{We}} n_Z, \]
where \( n_R \) and \( n_Z \) are the radial and axial components of the outward unit normal to the surface. The process of making the above equations dimensionless was performed using the following definitions:
\[ R = \frac{r}{d_0}, \quad Z = \frac{z}{d_0}, \quad U = \frac{u}{v_0}, \quad V = \frac{v}{v_0}, \quad \tau = \frac{t}{d_0/v_0}, \]
\[ H = \frac{H}{1/d_0}, \quad P = \frac{P - P_0}{\rho v_0^2}, \quad \bar{\sigma}_{ij} = (\sigma_{ij} + \delta_{ij} P_0) / \rho v_0^2, \]
where \( d_0 \) is the preimpact droplet radius, and \( \bar{\sigma}_{ij} \) corresponds to the dimensionless stresses, \( \delta_{ij} \) is the Kronecker delta, and \( H \) is the mean surface curvature defined as:
\[ 2H = \frac{1}{R_1} + \frac{1}{R_2}, \]
where the two radii of curvature \( R_1, R_2 \) define the curvature of the droplet surface. The latter are being calculated using a nonrational B-spline curve interpolating the free surface nodes of the fluid droplet. The definitions of the dimensionless groups created in the course of the nondimensionalization process are (Reynolds, Weber, Froude, and Mach number)
\[ \text{Re} = \frac{\rho v_0 d_0}{\mu_i}, \quad \text{We} = \frac{\rho d_0 v_0^2}{\gamma}, \quad \text{Fr} = \frac{v_0^2}{d_0 g}, \quad \text{Ma} = \frac{v_0}{c}. \]

2. The boundary condition at the dynamic contact line
The treatment of the contact line needs special attention. Assuming validity of the conventional hydrodynamic no-slip boundary condition over the entire liquid–solid interface, including the contact line, leads to a force singularity at the contact line as has been shown previously by several different authors.\(^{33,34}\) By replacing the no-slip boundary condition by one of the various slip models postulated in the literature,\(^{16,18,35}\) the mathematical difficulties associated with the singularity at the contact line can be circumvented. However, these are only \textit{ad hoc} mathematical models of the physics occurring at the contact line. The method of choice in this investigation for handling the movement of the contact line numerically is to impose the Navier–slip condition at the contact line. This particular boundary condition allows the contact line to attain a velocity which is proportional to the strain rate in the vicinity of the contact line. Thus, the dimensionless Navier–slip boundary condition is formulated as follows:
\[ \bar{\sigma}_{RR} n_R + \bar{\sigma}_{RZ} n_Z = - \frac{1}{\varepsilon} U, \quad \bar{\sigma}_{RZ} n_R + \bar{\sigma}_{ZZ} n_Z = - \frac{1}{\varepsilon} V, \]
where \( \varepsilon \) is the slip parameter, which controls the amount of slippage of the contact line and has to be chosen \textit{ad hoc}, after a trial and error process, such that its value has practically no effect on the transport phenomena in the droplet outside a negligibly small region in the vicinity of the contact line.

C. Energy equation–heat transfer

1. Governing equations for the heat transfer
The governing energy equations can be written in form similar to the governing fluid mechanics equations. Whereas the governing fluid mechanics equations are only solved in the deforming domain of the impinging droplet, the energy equations are solved on the entire domain shown in Fig. 2. It is assumed that a very thin interface of poor thermal conductivity and no specific heat capacity exists between the presolidified droplet and the wafer substrate. This interfacial layer allows for a simple modeling of the heat transfer resistance between the wafer substrate and the presolidified droplet as will be outlined below. In addition to this interfacial layer a second interface between the impinging droplet and the presolidified droplet is assumed as indicated in Fig. 2. This interface is due to modeling of the heat transfer resistance between the impinging droplet and the presolidified droplet.

Using again the Lagrangian approach the equations for all domains have the same form and can be written in dimensionless formulation as follows:
\[ C_i \partial_t \Theta_i - \frac{1}{\text{Pr} \text{Re}} \left[ \frac{1}{R} \partial_R \left( K_i R \partial_R \Theta_i \right) + \partial_Z \left( K_i \partial_Z \Theta_i \right) \right] = 0, \]
\[ i = 1, 2, 3, 4, \]
where \( C_i \) denotes the specific heat, \( K_i \) is the thermal conductivity of the \( i \)th component, and \( \Theta_i \) is the temperature. The different domains in Fig. 2 are numbered as: (1) impacting droplet, (2) presolidified droplet, (3) interface layer, and (4) multilayer wafer substrate. The convective effect on the transfer of energy in the splat is implicit in the Lagrangian formulation. The movement of fluid particles in the deforming droplet represents this convective effect.

The dimensionless initial conditions for the droplet and the substrate domains are
Heat transfer from the exposed droplet and substrate surfaces is neglected such that the following condition applies to all boundaries exposed to the ambient:

$$\frac{\partial \Theta_i}{\partial R} + \frac{\partial \Theta_i}{\partial z} = 0 \quad i = 1, 2, 3, 4. \quad (15)$$

The process of making the energy equations dimensionless was carried out employing the following definition of the dimensionless temperature, heat capacity $C_l$, the thermal conductivity $K_l$, and the Prandtl number Pr:

$$\Theta_i = \frac{T_i - T_{2,0}}{T_{1,0} - T_{2,0}}, \quad C_l = \frac{\rho_l c_{pl,1}}{\rho_i c_{p,1}}, \quad K_l = \frac{k_i}{k_{l,1}}, \quad Pr = \frac{\mu i c_{pl,1}}{k_{l,1}}, \quad i = 1, 2, 3, 4. \quad (16)$$

2. Modeling of thermal contact resistance

The solidified shape of a molten droplet deposited on a substrate is not only influenced by the impact dynamics, but also by the simultaneous heat removal process from the molten material to the substrate.27 A key factor in this process is the resistance to heat transfer arising through imperfect thermal contact at interfaces. The onset, advancement, and completion of, for instance, phase change are dependent on this thermal contact resistance. The cause for the latter is among others surface roughness, surface tension of the melt, impurities on the surface, and gas entrapment alleviating perfect thermal contact.

Two connecting interfaces featuring thermal contact resistance have to be taken into account in the present model as insinuated in Fig. 2. An interface is present between the impinging and the presolidified droplet as well as between the latter and the wafer substrate. Two different approaches are chosen to incorporate thermal contact resistance in the present study. Contact resistance between the presolidified droplet and the wafer substrate is modeled according to Waldvogel and Poulikakos14 by defining a thin layer of arbitrary thickness $d_{il}$ which attaches the two domains. It is assumed that this layer has zero heat capacity and experiences only axial conduction. The dimensionless effective axial thermal conductivity $K_{Z,c}$ between the presolidified droplet and the wafer substrate is related to the contact resistance coefficient $R_{t,c}$ in the following form:

$$K_{Z,c} = \frac{d_{il}}{R_{t,c} k_{l,1}}. \quad (17)$$

With all the subdomains having essentially the same energy equation, the thermal solution of these subdomains can be calculated without having to iterate. These subdomains (heretofore termed together as "substrate domain") can be treated as a single domain having variations in heat capacity and thermal conductivity.

The same approach was not applied on the interface between the impinging and the presolidified droplet (internal boundary, Fig. 2). Defining such a layer on a curved boundary is inappropriate. Furthermore, such a layer would change the boundary curvatures of the presolidified droplet (since it has a finite thickness), resulting in an unacceptable change in geometry of the computational domain. As a clearly better alternative, the interface between the impinging and the presolidified droplet is treated as an internal boundary with the following dimensionless heat transfer condition:

$$K_i \frac{\partial \Theta_i}{\partial R} + K_{Z,c} \frac{\partial \Theta_i}{\partial z} = Bi(\Theta_{i,1} - \Theta_{i,2}), \quad i = 1, 2 \quad (18)$$

where the Biot number models the thermal contact resistance between the impinging and the presolidified droplet and $\alpha$ is the corresponding heat transfer coefficient. The computational domains of the impinging and the presolidified droplet are connected in such a way that the boundary nodes of the respective domains overlap at the interface. Thus, the temperatures $\Theta_{i,1}$, $\Theta_{i,2}$ refer to boundary temperatures of the impinging droplet (1) and the presolidified droplet (2) at the $j$th node of the interface.

3. Modeling of the solidification process

Three assumptions are made with regard to the solidification of the impinging droplet. Since a eutectic solder is under study, a sharp boundary separating fluid and solid regions is incorporated. Furthermore, phase change is assumed to occur at the equilibrium freezing temperature $T_m$ (183 °C). Finally, solid and liquid densities are taken to be identical since the solder under investigation experiences only a contraction of 2.4% upon freezing.14 The exact specific heat method proposed by Bushko and Grosse37 is employed to model solidification. In this approach the dimensionless specific heat is defined as

$$C_l(\Theta_1) = \{C_{l,1}(\Theta_1 < \Theta_m), C_{l,1}(\Theta_1 > \Theta_m)\} + \frac{1}{Ste(1-SHP)} \int_0^\Theta \delta(\Theta_1 - \Theta_m) d\Theta, \quad (20)$$

where $\delta(\cdot)$ is the Dirac delta function, the subscripts $s$ and $l$ refer to solid and liquid, respectively, and the Stefan number Ste and the superheat parameter (SHP) are defined as

$$Ste = \frac{c_{pl,1}(T_m - T_{2,0})}{L}, \quad SHP = \frac{T_{1,0} - T_m}{T_m - T_{2,0}}. \quad (21)$$

where $L$ is the latent heat of fusion. According to Bushko and Grosse37 this approach leads to exact integration of the capacitance terms in the finite element formulation with linear triangle elements, thus, conserving energy very accurately as the droplet solidifies.

D. Solution method

The preceding mathematical models for the pileup are spatially discretized using the finite element method. Triangular linear elements are chosen to discretize the compu-
tional domain. To this end, a commercial mesh generator is used to mesh the computational domain (Hypermesh®, Altair Engineering). The mesh generator employs Delaunay tessellation given the surface distribution of points. Time integration of the flow equations is accomplished using a backward Euler finite difference scheme. The coupling of fluid node locations and of the mass and momentum conservation equations is attained by employing a method proposed by Bach and Hassager. The calculation of a time step $\Delta \tau$ features the following steps:

1. Using the velocities at time step $\tau$ as initial guesses for the velocities at time step $\tau + \Delta \tau$, the new nodal locations are calculated.
2. Based on the nodal coordinates the velocity and pressure fields at time step $\tau + \Delta \tau$ are calculated.
3. The nodal coordinates are re-calculated by employing the derived velocity field.
4. Steps 2 and 3 are iterated until convergence is obtained. The solution is assumed to reach convergence when the maximum difference of velocity and pressure for each node is below 0.05% from one iteration to the next.

Time integration of the energy equation is accomplished using the Crank–Nicholson method. The coupling of the energy equation of the impinging droplet and the substrate domain is attained by Eq. (18). The calculation of a time step $\Delta \tau$ features the following steps:

1. Based on the temperature field in the substrate domain at time step $\tau$ the energy equation of the impinging droplet is solved.
2. Based on the temperature field in the impinging droplet, the energy equation for the substrate domain is solved.
3. Based on the temperature field in the substrate domain, the energy equation for the impinging droplet is solved.
4. Steps 2 and 3 are iterated until convergence is obtained. The solution is assumed to reach convergence when the maximum difference of temperature for each node is below 0.05% from one iteration to the next.

Employing a Lagrangian formulation of the Navier–Stokes equations implies that the mesh nodes in the fluid domain are moving spatially in time. As the solution of the pileup advances in time the mesh is being deformed. Thus, in order to keep a good mesh quality and not to let numerical errors occur due to excessive element distortion, the mesh has to be renewed after a number of time steps. A mesh quality criterion thereby controls the remeshing activity. If the smallest angle in an element decreases below 20°, a remeshing operation is induced. The angle is thereby set arbitrarily. After a remeshing operation the values of the nodal state variables have to be interpolated between the old mesh and the newly generated mesh. This interpolation proceeds in a node-by-node fashion using simple linear interpolation based on the element trial functions on the old mesh.

A detailed study was performed to determine the temporal and spatial discretization necessary to obtain time step and grid insensitive solutions. Mesh independence was achieved for initial meshes containing approximately 700 nodes in the impinging droplet and approximately 2000 nodes in the substrate domain. A dimensionless time step of $10^{-3}$ was found to be sufficient for time step independence. A typical simulation requires 32 CPU hours to complete 25,000 time steps on a Pentium III/600 MHz PC with 256 MByte of internal memory. Energy and mass conservation of the above numerical scheme was found to be well below 1% of the initial energy and mass.

III. EXPERIMENTAL PROCEDURE

Experimental visualization of the transient pileup process is employed in order to compare and validate the numerical model presented above. The techniques employed to visualize the transient pileup process are described thoroughly in Ref. 40. It is based on generating monodispersed picoliter-sized droplets on demand utilizing a modified microdroplet jetting device (Microfab Inc., Dallas, Texas). A schematic of the deployed microdroplet generator device is given in Fig. 3. Therefore, the droplet generation method closely follows the concept of ink-jet printing. The temperature of the molten solder (high-purity 63% Sn–37% Pb solder) in the reservoir section is kept at a constant temperature of 210°C. The melting temperature of the latter is 183°C as can be seen in Table I containing all the relevant thermophysical properties of the materials utilized in this investigation. The characteristic time and length scale of the process is on the order of $O(100 \mu s)$ and $O(100 \mu m)$. The detailed and accurate visualization of the pileup process is achieved using flash video microscopy. This visualization technique requires a high repeatability of the process and is based on reconstructing a single event from multiple, reproducible events by patching together several frames taken at subsequent times (i.e., the impact of a droplet is visualized by recording many droplets once at subsequent times). Using this technique a time resolution on the order of $O(1 \mu s)$ can be achieved.

On account of this, sequences at different impact velocities as well as substrate temperatures were recorded using flash video microscopy. In addition, end shapes of pileups...
were imaged using a scanning electron microscope (Hitachi, S-900) in order to obtain fine surface structures not very visible in the recorded sequences. Basically, two series of experiments were performed covering the influence of the impact velocity on the pileup and, for constant impact velocity and diameter, the influence of the substrate temperature on the outcome of the pileup. For all experiments we attempted to keep the droplet diameter at a size of approximately 80 \( \mu \)m. This is due to the fact that at this diameter the most stable droplet generation was achieved. The following array of experimental cases were visualized in Table II:\textsuperscript{23}

### IV. RESULTS AND DISCUSSION

#### A. Model performance

The complexity of the microdroplet pileup process necessitated modeling assumptions which have to be investigated on their respective performance and validity. These assumptions pertain to the Navier slip model at the dynamic contact line, and the thermal contact resistances at the respective interfaces. To this end, a large number of simulations were performed in order to shed light on the corresponding influential parameters of these models/assumptions on the outcome of a pileup calculation. More precisely, this includes the Biot number for the influence of the thermal contact resistance, and the slip parameter for the Navier–slip model. An identical substrate for all these cases was chosen in order not to make the results dependent on different substrate shapes. To this end, Fig. 4 shows the test substrate (upon which the second droplet in the pileup impacts) employed for this investigation. It corresponds to the solidified shape of a molten droplet impinged on a flat wafer substrate with the following impact parameters: Re=363.9, We=4.17, Fr=3025.2, Ste=0.895 (25 °C). It is assumed for all simulations reported hereafter, that the thermophysical properties (dynamic viscosity, surface tension, thermal conductivity, and specific heat) are not temperature dependent. Changes of the thermophysical properties are only due to phase change (liquid/solid properties). This assumption is reasonable for the relatively narrow temperature range covered by the process.

#### TABLE I. Relevant thermophysical properties\textsuperscript{a}

<table>
<thead>
<tr>
<th>Material</th>
<th>( k ) (W/mK)</th>
<th>( \mu ) (Pas)</th>
<th>( \rho ) (kg/m(^3))</th>
<th>( \gamma ) (N/m)</th>
<th>( T_m ) (°C)</th>
<th>( L ) (J/kg)</th>
<th>( c_p ) (J/kg K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>solder (liquid)</td>
<td>25</td>
<td>0.00262</td>
<td>8218</td>
<td>0.345</td>
<td>183</td>
<td>42 000</td>
<td>238</td>
</tr>
<tr>
<td>solder (solid)</td>
<td>48</td>
<td>...</td>
<td>8240</td>
<td>...</td>
<td>183</td>
<td>42 000</td>
<td>176</td>
</tr>
<tr>
<td>gold</td>
<td>317</td>
<td>...</td>
<td>19 300</td>
<td>...</td>
<td>1064.4</td>
<td>...</td>
<td>129</td>
</tr>
<tr>
<td>silicon</td>
<td>148 (25 °C)</td>
<td>98.9 (125 °C)</td>
<td>94 (180 °C)</td>
<td>...</td>
<td>2330</td>
<td>...</td>
<td>1410</td>
</tr>
<tr>
<td>silicon nitride\textsuperscript{b}</td>
<td>27</td>
<td>...</td>
<td>3180</td>
<td>...</td>
<td>1800</td>
<td>...</td>
<td>712</td>
</tr>
<tr>
<td>T90W\textsuperscript{c}</td>
<td>159</td>
<td>...</td>
<td>14 730</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>178.6</td>
</tr>
<tr>
<td>Thermal Paste</td>
<td>0.84</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

\textsuperscript{a}See Ref. 60.
\textsuperscript{b}Source: www.accuratus.com
\textsuperscript{c}Source: www.puretechnic.com

#### TABLE II. Initial conditions of the experimental cases investigated.\textsuperscript{a} Cases 1–6 represent the impact regime with changing impact velocities and cases 7–12 represent the thermal regime with changing initial substrate temperatures.

<table>
<thead>
<tr>
<th>Case</th>
<th>Impact velocity ( u_0 ) (( \mu )m/s)</th>
<th>Droplet diameter ( d_0 ) (( \mu )m)</th>
<th>Substrate temperature ( T_{2,0} ) (°C)</th>
<th>Reynolds ([-])</th>
<th>Weber ([-])</th>
<th>Stefan ([-])</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.12±0.05</td>
<td>80.10±1.6</td>
<td>25±2</td>
<td>281.39</td>
<td>2.39</td>
<td>0.895</td>
</tr>
<tr>
<td>2</td>
<td>1.26±0.04</td>
<td>76.83±1.5</td>
<td>25±2</td>
<td>303.65</td>
<td>2.91</td>
<td>0.895</td>
</tr>
<tr>
<td>3</td>
<td>1.38±0.03</td>
<td>79.59±1.5</td>
<td>25±2</td>
<td>344.51</td>
<td>3.61</td>
<td>0.895</td>
</tr>
<tr>
<td>4</td>
<td>1.51±0.02</td>
<td>76.83±1.5</td>
<td>25±2</td>
<td>363.89</td>
<td>4.17</td>
<td>0.895</td>
</tr>
<tr>
<td>5</td>
<td>1.63±0.02</td>
<td>77.51±1.5</td>
<td>25±2</td>
<td>396.29</td>
<td>4.91</td>
<td>0.895</td>
</tr>
<tr>
<td>6</td>
<td>1.74±0.02</td>
<td>83.03±1.6</td>
<td>25±2</td>
<td>453.16</td>
<td>5.99</td>
<td>0.895</td>
</tr>
<tr>
<td>7</td>
<td>1.51±0.02</td>
<td>76.83±1.5</td>
<td>25±2</td>
<td>363.89</td>
<td>4.17</td>
<td>0.895</td>
</tr>
<tr>
<td>8</td>
<td>1.50±0.02</td>
<td>77.22±1.4</td>
<td>50±2</td>
<td>363.31</td>
<td>4.14</td>
<td>0.754</td>
</tr>
<tr>
<td>9</td>
<td>1.50±0.02</td>
<td>77.03±1.5</td>
<td>75±2</td>
<td>362.42</td>
<td>4.13</td>
<td>0.612</td>
</tr>
<tr>
<td>10</td>
<td>1.51±0.02</td>
<td>76.17±1.5</td>
<td>100±2</td>
<td>360.77</td>
<td>4.14</td>
<td>0.470</td>
</tr>
<tr>
<td>11</td>
<td>1.52±0.02</td>
<td>75.02±1.5</td>
<td>125±2</td>
<td>357.65</td>
<td>4.13</td>
<td>0.329</td>
</tr>
<tr>
<td>12</td>
<td>1.49±0.02</td>
<td>76.83±1.5</td>
<td>150±2</td>
<td>359.07</td>
<td>4.06</td>
<td>0.187</td>
</tr>
</tbody>
</table>

\textsuperscript{a}See Ref. 23.
cess considered in this investigation as well as for the small changes in the above mentioned thermophysical properties of the employed eutectic solder. However, it must be stressed that the dynamic viscosity changes by orders of magnitude close to the phase change boundary. This transition zone is usually very thin in comparison with the computational domain and is neglected in this investigation.42–44

1. Spreading and the influence of the slip model on the numerical solution

The numerical investigation of the pileup process employs the Navier–slip model at the contact line as described in Sec. II B 2. This purely mathematical model allows the contact line to attain a velocity, which is proportional to the strain rate in the vicinity of the contact line. The proportionality is thereby influenced by the so-called slip parameter \( \epsilon \). The latter serves as a control and possible matching factor and has to be chosen \( \text{ad hoc} \). In order to study the respective influence of the latter on the spreading and thus on the pileup process, a parametric study on the slip parameter was conducted. To this end, the slip parameter was varied in the following range: \( \epsilon = 0.001 \text{–} 100 \). The sample case studied is defined by the following dimensionless impact parameters: \( \text{Re}=376 \), \( \text{We}=4.28 \), \( \text{Fr}=2867 \), and \( \text{Ste}=0.895 \). To this end, Fig. 5 shows the temporal evolution of the spread factor for a host of values of \( \epsilon \). The spread factor is thereby defined as in Ref. 23

\[
\beta(t) = \frac{a_c(t)}{r_0},
\]

where \( a_c \) is the instantaneous travel distance of the contact line and \( r_0 \) is the initial droplet diameter as shown in Fig. 1(b). The instantaneous travel distance of the contact line \( a_c \) is measured in terms of the arc length from the center of impact along the free surface of the presolidified droplet to the contact line. Although the slip parameter spans a range of \( 10^3 \), only negligible differences in the spreading behavior can be observed. The general behavior of the spreading process remains the same, yielding approximately the same final spread factors irrespective of the chosen value of the slip parameter. Hence, the local character of the flow in the very small contact line region, represented by the slip parameter, has practically no effect on the overall pileup process. As follows from inspection of Fig. 5, the numerically calculated spreading process exhibits a sawtooth behavior in the temporal evolution of the spread factor. This behavior is a direct consequence of the employed finite element approach. Changes of the interface area are established through the tendency of the liquid to wet the substrate as well as the fact that the easily deforming, liquid droplet meets the rigid substrate with a certain momentum. In the numerical model the wetting tendency is not modeled but compensated using the Navier–slip condition. Furthermore, and more importantly, the solution of the Navier–Stokes equations accounts for the creation of liquid/solid interface due to the momentum of the impacting and deforming droplet. Since the fluid domain is triangulated into small, linear elements with a finite edge length, every attachment of a free surface node of the impinging droplet to the free surface of the presolidified droplet causes such a step-like increase of the spread factor. Between these step increases a slight receding motion of the contact line can be identified causing the sawtooth shape. This receding motion of the contact line is due to a rolling motion at the contact line. This means that fluid elements at and near to the free surface of the impinging droplet, very close to the contact line, are moving in the direction of the contact line, Fig. 6. At the contact line, the free surface is consumed in favor of the creation of a liquid–solid interface between the impinging and the presolidified droplet as has been observed experimentally by, for example, Dussan et al.34,45–47 The velocity vector at the contact line, Fig. 6, is directed to the interior of the impinging droplet. Also shown is a recirculation zone, an instantaneous vortex in the vicinity of the contact line, further illustrating the rolling motion taking place.

It has to be emphasized that the Navier–slip model only allows for the movement of the contact line but does not account for interfacial wetting phenomena. Uncompensated interfacial forces at the contact line, of the kind for instance modeled by the well-known Young–DuPré equation 48 are not accounted for. Thus, capillary phenomena are not pre-
dictable using the Navier–slip boundary condition only. This is exemplified by the two cases shown in Figs. 7(a) and 7(b). Both cases feature spatial locations of maximum spreading at the instant where recoiling starts. The case shown in Fig. 7(a) corresponds to the following set of Reynolds, Weber, and Froude numbers: Re=2.51, We=0.019, and Fr=0.13. This is equivalent to gently placing a droplet on top of another. As can be seen, the droplet behaves like a rubber ball with an elasticity defined by the surface tension of the fluid. No wetting in the sense of equilibration of the contact angle occurs. Thus, the Navier–slip model only accounts for contact line movement induced and dominated by inertia. This is the case in the impact range of the pileup investigated in this study, Table II, due to the contact line velocities which are approximately on the same order of magnitude as the impact velocity. This situation holds at most stages of the spreading. At later stages, shortly before the contact line is arrested and the final spread factor is obtained, the contact line velocity tends to zero and the influence of inertia at the contact line becomes negligible. During this stage the contact line behavior should be controlled by capillarity. This is equivalent to balancing the Young–DuPré equation, which means that the system is forced to attain equilibrium contact angles. A representative situation of this last stage of the spreading process is shown in Fig. 7(b). As can be seen, the impinging droplet has just changed from the spreading to the recoiling regime. The contact angle thereby features a value of approximately 175°. This corresponds to a nonwetting situation. As is known from the literature, most substances feature a good wettability on substrates of their own kind. It has to be expected that the equilibrium contact angle between liquid and solid solder should gradually adjust to a value much smaller than the above value. However, it has to be emphasized that this period with low inertia effects during which the nonsolidified contact line has stopped moving is short. Numerical simulations, for the sample case mentioned above, predict this time period to be on the order of approximately 15 μs in agreement with experimental visualizations. Thus, it is not expected that in this short time period an equilibration of the interfacial energies, respectively the contact angles, occurs. However, it is expected that the spreading behavior in the last stage could be influenced by capillarity. If the details of the contact line region are of importance capillary phenomena in this region need to be carefully considered and this can only be done with experimental input to numerical simulations, which currently does not exist. In summary, the Navier–slip model is an appropriate approach to circumvent the moving contact line problem. However, it should be kept in mind that it is not capable of accounting for the above described wetting phenomena.

2. Thermal contact resistance

Thermal contact resistance is a key parameter in the deposition of molten droplets as shown by Xiong et al. Unfortunately, accurately predicting or measuring the contact resistance at the time and length scales of the solder jetting technology (less than 100 μm and 100 μm) has proven elusive so far in spite of reasonable efforts in recent years. Experiments by Wang and Matthys on molten copper droplets with weights of about 0.5–0.6 g impinging and solidifying on cold substrates have shown that the thermal contact resistance is strongly time-dependent. Similarly, Xiong et al. obtained contact heat transfer coefficients in the range of 4–350×10^3 W/Km^2 by matching experimental and numerical endshapes of microdroplets impacting on flat substrates.

The two models for the thermal contact resistance introduced in Sec. II C 2, allow for an arbitrary choice of an interface heat transfer coefficient or Biot number respectively. Two regimes are distinguished: (1) melt spreading and liquid cooling and (2) solidification and solid cooling. To this end, a Biot number or a thermal contact resistance is attributed to each of these two regimes. The numerical solution/implementation thereby depends on the respective interface. For the first interface between the impinging and the presolidified droplet this involves monitoring the interfacial node temperatures of the impinging droplet. The conjugate heat transfer from liquid interfacial nodes is treated using the Biot number of the first regime Bi_1. For solidified interfacial nodes the respective Biot number for the second regime is deployed, Bi_2. The second interface between the presolidified droplet and the wafer substrate features no phase change.
and a constant thermal contact resistance is assumed. The value of the latter is estimated as $R_{t,c} = 3 \times 10^{-4} \text{m}^2\text{K/W}$ and was kept constant for all simulations reported herein. To this end, a number of simulations with varying values for the two Biot numbers, $B_i$ and $B_l$, pertaining to the interface between the impinging and the presolidified droplet were performed. The Biot numbers were varied in the following ranges: $B_i = 0.1 - 1$; $B_l = 0.1 - 2$. This corresponds to the following ranges of the thermal contact resistance: $R_{t,c,r} = 3.2 \times 10^{-4} - 3.2 \times 10^{-6}$; $R_{t,c,l} = 3.2 \times 10^{-5} - 1.6 \times 10^{-6}$. The objective was to investigate the influence of the Biot numbers, as predicted by the numerical model, on the spreading process, the solidification process, the transients of the fluid motion, and the final shape of the pileup. To this end, Fig. 8 shows the progression of the spread factor for two representative cases with the following pairs of Biot numbers: (1) $B_i = 2.0$, $B_l = 0.1$; (2) $B_i = 0.1$, $B_l = 0.1$. As illustrated in Fig. 8, the evolution of the spread factor is for both cases almost identical, yielding approximately the same final spread factor, $\beta_{f}$. The temporal evolution of the solid fraction and thermal energy of the impinging droplet feature a similar behavior as shown in Fig. 9. Although the Biot numbers prior to solidification of the second droplet vary by a factor of 20 in the two example cases shown, the temporal evolution of the spread factor, the solid fraction and the thermal energy are practically unaffected. The weak influence of the Biot number prior to solidification $B_i$ can be explained as follows. The time period during which a very good thermal contact between the impinging and the presolidified droplet is established is short compared to the entire process of the pileup as shown by the shaded area in Fig. 9. Correspondingly, the first of the two combined stages, melt spreading and liquid cooling, is short compared to the second stage including the solidification process. The small differences in the temporal evolution of the solid fraction, the thermal energy, and the dimensionless height of the pileup thus have to be attributed to the small difference in the final spread factor shown in Fig. 8. Both cases show approximately the same cooling behavior rendering the aforementioned differences in the temporal evolution of the pileup dependent on the final spread factor. The reason for the similar decrease in thermal energy is that solidification at the interface, for both pairs of Biot numbers, occurs very fast. Thus, the conjugate heat transfer is dominated by the Biot number pertaining to the solidified interface $B_i$. A marked influence of $B_l$ can only occur when the heat transfer during the initial stage has a strong impact on the final spread factor. The spread factor defines the contact area controlling, along with $B_l$, the cooling of the droplet by the underlying substrate. In cases where the spreading process is short compared to the entire pileup process and, more importantly, when solidification is likely to occur very fast upon contact of the melt with the presolidified droplet, leading to a final spread factor controlled primarily by the initial momentum of the impinging droplet, then the Biot number pertaining to the liquid contact has an almost negligible influence on the progression of the pileup. This is illustrated in Fig. 10 showing...

**FIG. 8.** Temporal evolution of the spread factor $\beta(t)$ for different Biot numbers but identical impact parameters: $Re=376$, $We=4.28$, $Fr=2867$, and $Stc=0.895$. $Bi_l=2$, $Bi_s=0.1$.

**FIG. 9.** Temporal evolution of the solid fraction and the thermal energy of the impinging droplet for different Biot numbers but identical impact parameters: $Re=376$, $We=4.28$, $Fr=2867$, and $Stc=0.895$. (a) $Bi_l=0.1$, $Bi_s=2.0$; (b) $Bi_l=0.1$, $Bi_s=0.1$. The shaded area signifies the spreading period.

**FIG. 10.** Pileup endshapes for the following impact parameters: $Re=376$, $We=4.28$, $Fr=2867$, and $Stc=0.895$. (a) $Bi_l=0.1$, $Bi_s=2.0$; (b) $Bi_l=0.1$, $Bi_s=0.1$; (c) $Bi_l=1.0$, $Bi_s=1.0$; and (d) $Bi_l=0.9$, $Bi_s=1.0$. 

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ing the effect of variations of the two Biot numbers on the final shape of a pileup. For identical impact conditions, varying the Biot number for solid–solid contact has a marked influence on the endshape of the pileup.

B. Comparison of numerical and experimental results in the impact regime

The matching of the numerical simulations with the experimental cases 1–6, Table II, is based solely upon varying the Biot number for solid–solid contact $B_{is}$. The Biot number for liquid–solid contact $B_{il}$ has, as shown above, very little influence on the transients and the outcome of the pileup process. The same is true for the slip parameter in the Navier–slip model. For all simulations reported hereafter, the slip parameter was kept constant at $\varepsilon = 0.005$. Optimal values for the Biot numbers were obtained in the same manner as described earlier. A compilation of the relevant dimensionless numbers with the determined optimal values for the Biot numbers for the impact regime is given in Table III.

As follows from Table III the Biot numbers leading to the best conformity between the experimental and numerical cases are practically equal for all cases. The thermal contact resistances are thereby approximately $1.5 \times 10^{-5} \text{ m}^2 \text{K W}^{-1}$ for solid–solid contact and $0.3 \times 10^{-5} \text{ m}^2 \text{k W}^{-1}$ for liquid–solid contact. Furthermore, no dependence between the impact velocity (the Reynolds number) and the corresponding thermal contact resistance (the Biot numbers) exists for the impact regime investigated herein.

In general, a very good agreement between the experimental and the numerical cases is observed for the impact regime using the above values of the Biot numbers. Figure 11 shows, representatively, the oscillatory motion of the center of the impinging droplet $Z_{\text{center}}$ for the experimental and numerical case 1 (see Tables II and III). Considering the complexity of the problem a very good matching in the transient behavior as well as the actual pileup height of the experimental and the numerically simulated pileup process is achieved. The inset in the bottom corner of Fig. 11 clarifies the definition of $Z_{\text{center}}$.

A sequence comparing the experimentally visualized and the numerically simulated pileup process of case 4 is shown in Fig. 12. Again, agreement between the experimental and the numerical results is demonstrated.

A compilation of the final spread factors for the experimentally visualized and numerically simulated cases of the impact regime is given in Fig. 13. The numerical simulations follow the trend observed in the experiments very well.
However, the experimentally measured final spread factors are slightly underpredicted by the numerical solution in all cases. This further points to the effect of the wetting mechanism during the late stages of the spreading process not accounted for by the numerical model as explained earlier. The differences in the final spread factor between the experimental and the numerical results are approximately 10%, which is deemed clearly acceptable for the complex problem at hand.

Before comparing the experimental and numerical times required for complete solidification, the experimental determination of this time needs to be discussed. It is based on visually identifying the instance at which no droplet motion is visible. However, as outlined in Ref. 23 this is not trivial due to the fact that the amount of molten material still oscillating prior to solidification is very small, hence, hardly visible in the visualized sequences. The solidification time is defined in Ref. 23 as the instance where the height $z_{\text{center}}(t)$ of the pileup structure remains constant within $\pm 2\%$ of the initial droplet diameter for a time length more than half a period of its oscillations. This certainly truncates the very last stages of the oscillation and of the solidification process and causes an error in the comparison with the numerically calculated time. A further issue worth mentioning, is the fact that the oscillation frequencies of the molten material converge towards the end of the solidification process to the acquisition rate of the visualization setup, which is 200 kHz. Based on the Nyquist theorem, the sampling rate must be equal to or greater than twice the highest frequency component in the analog signal in order to avoid aliasing of the latter. The analog signal pertains to the temporal evolution of the pileup height $z_{\text{center}}(t)$. The aliasing may again lead to a systematic error in the determination of the experimental solidification times as reported in Ref. 23. The result of applying the two above mentioned criteria for case 6 is shown in Fig. 14. The vertical, dotted lines in Fig. 14 depict, respectively, the experimentally determined solidification time, the acquisition rate limit due to the Nyquist theorem, and the numerically obtained solidification time. The horizontal, dotted lines depict the visibility limit. Oscillations with amplitudes smaller than the bandwidth given by these horizontal lines are not registered in the experiments. The droplet is thus defined as fully solidified. As clearly visible from Fig. 14 both the Nyquist theorem as well as the visibility limit lead to a marked truncation of the oscillation process compared to the numerical result causing a considerable underestimation of the solidification time. A comparison of all experimental and numerical solidification times, including numerical results truncated in the above manner, is shown for the impact regime (cases 1–6) in Fig. 15. Without accounting for the above discussion, the numerically calculated solidification times are considerably larger than the experimentally estimated times. The largest deviation is 23%. Nevertheless, truncating the numerical results in the above described manner to yield a fair comparison, leads to a noticeable improvement in the agreement, with the largest difference in the experimental and the truncated numerical solidification time less than 10%.

Figure 16 shows a comparison of the attained endshapes for all the simulated and visualized pileup cases pertaining to
the impact regime (cases 1–6). A very good agreement between the numerically calculated and the experimentally obtained endshapes is observed.

C. Comparison of numerical and experimental results in the thermal regime

In the thermal regime the Stefan number is the only influential parameter being varied significantly. The Stefan number has, in an implicit manner, a very strong impact on the numerical results and the predictive capabilities of the numerical model, and its significance will be the scope of this entire subsection. If not stated otherwise, the results for the corresponding cases presented herein always pertain to the Biot numbers given in Table IV. The relevant dimensionless numbers for the numerically solved cases of the thermal regime presented herein are also shown in Table IV.

The most important finding is that with decreasing Stefan number the agreement of the experimental and numerical results deteriorates strongly. This deterioration in agreement is shown representatively for the oscillatory motion of the center of the impinging droplet, \( Z_{\text{center}} \) for case 11, Fig. 17. The agreement in case 11 is limited to the first half cycle of the deformation process. Thereafter, a markedly different transient behavior of the numerical and experimental results can be observed. The differences are further underpinned by the numerically simulated and experimentally visualized impact sequences of case 11, Fig. 18.

A compilation of the experimentally measured and the numerically calculated final spread factors is shown in Fig. 19. As is clearly visible, for decreasing Stefan numbers the differences between the experimentally measured and the final spread factors obtained from the numerical simulations increase distinctly. During the first, inertia driven stage of the spreading process a good agreement between the experimentally measured and the numerically calculated spread factors can be observed. During the later stages of the spreading process, where capillary effects become more important, clear differences between the numerical and experimental results are apparent. The disagreement clearly increases with decreasing Stefan number. This means that the importance of capillary effects increases markedly with increasing substrate temperatures (decreasing Stefan numbers). Wetting experiments using metal droplets on metal surfaces (not of the same material) have shown that an increase of the substrate

![FIG. 16. Experimental and numerical endshapes of the impact regime, cases 1–6.](image)

![TABLE IV. The relevant dimensionless numbers and corresponding values of the Biot numbers for the numerical cases of the thermal regime.](table)

<table>
<thead>
<tr>
<th>Numerical case</th>
<th>Reynolds [-]</th>
<th>Weber [-]</th>
<th>Stefan [-]</th>
<th>Biot Bi_s [-]</th>
<th>Biot Bi_l [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>363.89</td>
<td>4.17</td>
<td>0.895</td>
<td>0.2</td>
<td>0.9</td>
</tr>
<tr>
<td>8</td>
<td>363.31</td>
<td>4.14</td>
<td>0.754</td>
<td>0.2</td>
<td>0.9</td>
</tr>
<tr>
<td>9</td>
<td>362.42</td>
<td>4.13</td>
<td>0.612</td>
<td>0.3</td>
<td>1.0</td>
</tr>
<tr>
<td>10</td>
<td>360.77</td>
<td>4.14</td>
<td>0.470</td>
<td>0.5</td>
<td>1.5</td>
</tr>
<tr>
<td>11</td>
<td>357.67</td>
<td>4.13</td>
<td>0.329</td>
<td>2.0</td>
<td>3.0</td>
</tr>
<tr>
<td>12</td>
<td>359.07</td>
<td>4.06</td>
<td>0.187</td>
<td>5.0</td>
<td>10.0</td>
</tr>
</tbody>
</table>

![FIG. 17. Temporal evolution of the dimensionless pileup height for case 11.](image)

![FIG. 18. Experimentally visualized and numerically simulated sequence of case 11.](image)
temperature usually leads to an increased wettability. Furthermore, it has been observed that the spreading time of a droplet depends logarithmically on temperature and that the time decreases with increasing temperature. Similar results were obtained for 60Sn40Pb solder droplets on gold–platinum metal films. The above referenced experiments relied basically on gently placing macroscopic metal droplets on respective metal substrates. Thus, spreading of the droplets was controlled by capillarity. No experiments covering the material system of the present study could be found in the scientific literature. Nonetheless, the above referenced experimental observations point clearly to an explanation for this increasing disagreement of the experimentally obtained and numerically calculated final spread factors for decreasing Stefan numbers.

To investigate the effect of the slip coefficient as a possible cause of the disagreement between the numerical and experimental results in the thermal regime, we performed additional simulations on the effect of the slip coefficient for the lowest Stefan number of the study (Ste=0.187) and by covering a wide range of slip coefficient values, e=0.001–100.0. Our results (specifics are not shown here for brevity) clearly indicated that the slip coefficient effect on the extent of the droplet spreading is indeed minor, much like in the high Stefan number regime. The slip coefficient effect is therefore not responsible for the disagreement between numerics and experiments. It appears that, as already mentioned, this disagreement is indeed due to wetting phenomena not included in the modeling due to lack of experimental data to be inputted into the model.

An additional perhaps more plausible explanation of the disagreement is remelting of the bottom droplet due to the heat input from the top droplet, a phenomenon not included in the numerical model. This is theoretically possible as the substrate temperature (bottom droplet) approaches the melting temperature (the Stefan number approaches zero). However, remelting studies with flat substrates indicate that this effect is unlikely to occur, except perhaps at a thin region at the interface. Hence, it is the wetting mechanism that governs the spreading in Fig. 20 showing the completely solidified cases corresponding to the smallest Stefan numbers of the study (cases 10, 11, and 12).

The influence of the viscosity variation at the phase change boundary (not taken into account in the present macroscopic model) on the fluid mechanics of the spreading process can be safely ruled out. The reason for this is that even for the largest spread ratios obtained numerically, the thinnest fluid layers are still many orders of magnitude thicker than the extremely thin zone affected by the changing viscosity at the phase transition boundary (on the order of nanometers). To give a specific numerical example representative of a low Stefan number spreading, when a droplet with a diameter of 80 μm entirely (until it reaches the flat substrate) spreads on top of a semisphere with the same volume, a semispherical shell with an approximate thickness of 10 μm will be formed which is, as mentioned above, many orders of magnitude thicker than the transition zone of the viscosity. Based on the above, the viscosity variation at the phase transition line cannot explain the differences in the spreading behavior between the experimental and the numerical cases for low Stefan numbers.

V. CONCLUSIONS

In this article, a predominantly numerical study of the transient impact and solidification phenomena occurring during an axisymmetric molten microdroplet pileup was presented. This problem is relevant to a host of micromanufacturing processes. The length, velocity and time scales of the investigated pileup process were \( O(100 \mu m) \), \( O(1 m/s) \), and \( O(100 \mu s) \) respectively. The substrate temperatures were varied in a range of 25–150 °C.

An in depth comparison of experimental and numerical results on the transient fluid dynamics, wetting and solidification of molten microdroplets impinging on presolidified droplets or in more general terms on nonflat substrates is presented.

The numerical model accounting for the incompressible and axisymmetric Navier–Stokes equations, conjugate heat transfer, as well as solidification in a severely deforming domain containing a free surface and three interfaces (including the freezing front) showed desirable stability, accuracy, and mass as well as energy conserving properties.

It was shown that, in addition to the complex shape of the presolidified droplet, the mathematical model features five relevant dimensionless groups. These are the Reynolds number, the Weber number, the two Biot numbers, and the Stefan number. Whereas the Reynolds and Weber number
incorporated in the numerical model and code presented in and phase change. When available, these models could be droplet, in particular in the presence of temperature gradients this model predicts the spreading process well. Presently, no

The thermal contact resistance model employed, showed that the Biot number for solid–solid contact was strongly dominating the conjugate heat transfer over the Biot number for liquid–solid contact. The employed model at the dynamic contact line was the well known Navier–slip model. As long as capillary effects are not dominant (high Stefan number), this model predicts the spreading process well. Presently, no definitive models are available in the literature accounting for the wetting physics of the contact line of an impacting droplet, in particular in the presence of temperature gradients and phase change. When available, these models could be incorporated in the numerical model and code presented in this article.

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