

1 **Insights into the impact and solidification of metal droplets in**
2 **ground-based investigation of droplet deposition 3D printing**
3 **under microgravity**

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8 **Abstract**

9 Droplet deposition 3D printing is an additive manufacturing technique offering a great
10 potential for metal parts fabrication in space, of which some preliminary testing is usually
11 performed on ground in the early research. The previous work mimicked the anti-gravity
12 deposition of molten metal droplet through manipulating it into perpendicularly depositing
13 on a vertical substrate. However, the ground-based simulation of droplet deposition 3D
14 printing under microgravity remains an elusive goal, since the spreading and receding
15 processes are still affected by gravity. To address this issue, the prevailing physical
16 mechanisms of gravity effect on droplet impact and solidification are urgent to be defined.
17 Here, we present the studies on the impact dynamics and transient solidification of the
18 molten metal droplet deposited on vertical substrates through numerical modeling and
19 experiments. It is observed that the spreading and retraction of the droplet are asymmetric,
20 besides its solidification shape tilts to gravitational direction. The formation mechanisms
21 of these undesired behaviors are further demonstrated. The results show that the
22 asymmetrical spreading, retraction and solidification shape of the droplet originate from

1 the interaction of gravity and solidification. Moreover, the tilt of the solidified droplet has
2 a correlation with the critical process parameters, i.e. impact velocity, temperatures of
3 droplet and substrate. With a larger impact inertia and a lower solidification rate, the
4 undesired solidification shape can be effectively eliminated. This work provides a
5 foundation for the further investigation of the ground-based physical simulation of outer
6 space droplet deposition 3D printing.

7 **Keywords: 3D printing; metal droplet; solidification; vertical substrate; gravity**
8 **effect.**

10 **1. Introduction**

11 Replacing worn or damaged parts in spacecraft during deep-space exploration or
12 asteroids and Mars visiting is inevitable. However, it is time consuming and costly to
13 transport resupplies as the space is isolated [2–5]. To this end, Additive Manufacturing
14 (AM) is introduced to space manufacturing [6–9] for the advantages of low cost, wide
15 material applicability, high precision, and low-unit on-demand. The droplet-based 3D
16 printing technology [10,11] is believed to be a promising technique for advance in-space
17 manufacturing without using large energy equipment or custom materials [12].

18 Depositing droplets on a vertical substrate perpendicularly is assumed to be a
19 viable method to preliminarily demonstrate the feasibility of droplet deposition 3D
20 printing in microgravity. Because the droplets do not respond to gravity in the direction
21 of deposition, owing to the inertia resulting from impact velocity is separated from the
22 gravity [13,14]. Huang et al. [1] mimicked the anti-gravity deposition of droplets in
23 normal gravity environment by using electric field to manipulate the droplets to

1 perpendicularly deposit on a vertical substrate. However, the droplet is no more subject
2 to electric field force (a balance force of gravity) due to charge transfer at the moment
3 of landing on the substrate. Consequently, the spreading and receding processes will be
4 affected by gravity, resulting in irregular solidification shape of the deposited droplet.
5 That is undesired for the ground-based simulation of droplet deposition 3D printing
6 under microgravity. It is prerequisite to unveil the gravity effects for the proposal of
7 elimination strategies. To this end, the impact dynamics and solidification of droplets
8 impacting onto vertical substrates should be investigated.

9 When a droplet retains on vertical substrates, it is subject to capillary force, viscous
10 force, surface tension, solidification drag, and gravitational force which is a source term
11 that makes the impact dynamics differ from the deposition of droplets on a horizontal
12 solid wall. Considerable works have been reported regarding the effects of gravitational
13 force on the form of sessile droplets. Podgorski et al. [15] identified the dependence of
14 the capillary number (from here on Ca , which is the ratio of viscous force to surface
15 tension, $Ca = \mu v_0 / \sigma$) on tangential Bond number (from here on Bo_c , which is the ratio
16 of tangential gravity to surface tension, $Bo_c = \rho g_c d_0^2 / \sigma$), which governs different
17 states of a droplet staying on or running down along an inclinational substrate. Droplets
18 with small Ca (smaller than the capillary length) and Bo_c numbers show shapes of
19 circles or ovals. By investigating the contact line shapes of sessile water droplets with
20 different scales on inclination surface, Annapragada et al. [16] found that the droplet
21 contact area was almost circular due to the low Bo_c number. Milinazzo et al. [17]
22 numerically studied the shape of a droplet on a vertical wall. Their results show that the

1 wetted area keeps unchanged on condition that Bo number is smaller than a critical
2 value, which is dependent on the static contact angle.

3 In contrast to sessile droplet attached on an inclinational substrate, when a droplet
4 that ejected along free-fall direction impacts on the inclinational substrate, it will be
5 subject to an extra inertia force induced by impact velocity. The droplet spreads, retracts,
6 oscillates, or may even roll and slide on the substrate [18,19]. The dynamic behaviors
7 vary with the inclination angle since the impact velocity of the droplet is not
8 perpendicular to the substrate surface [17,18]. That brings a tangential velocity
9 component which is parallel to the substrate surface. The tangential velocity associated
10 with the inclination angle of the substrate has a significant effect on the droplet impact
11 dynamics [22]. Accordingly, the dynamic behaviors of droplets perpendicularly
12 impacting on vertical substrates would be of difference since the tangential velocity
13 component is 0. Moreover, the difference will be intensified by metal droplet as
14 solidification drag is introduced in the surface flow. Unfortunately, there are limited
15 literatures that reported the impact mechanism of metal droplets on a vertical substrate.

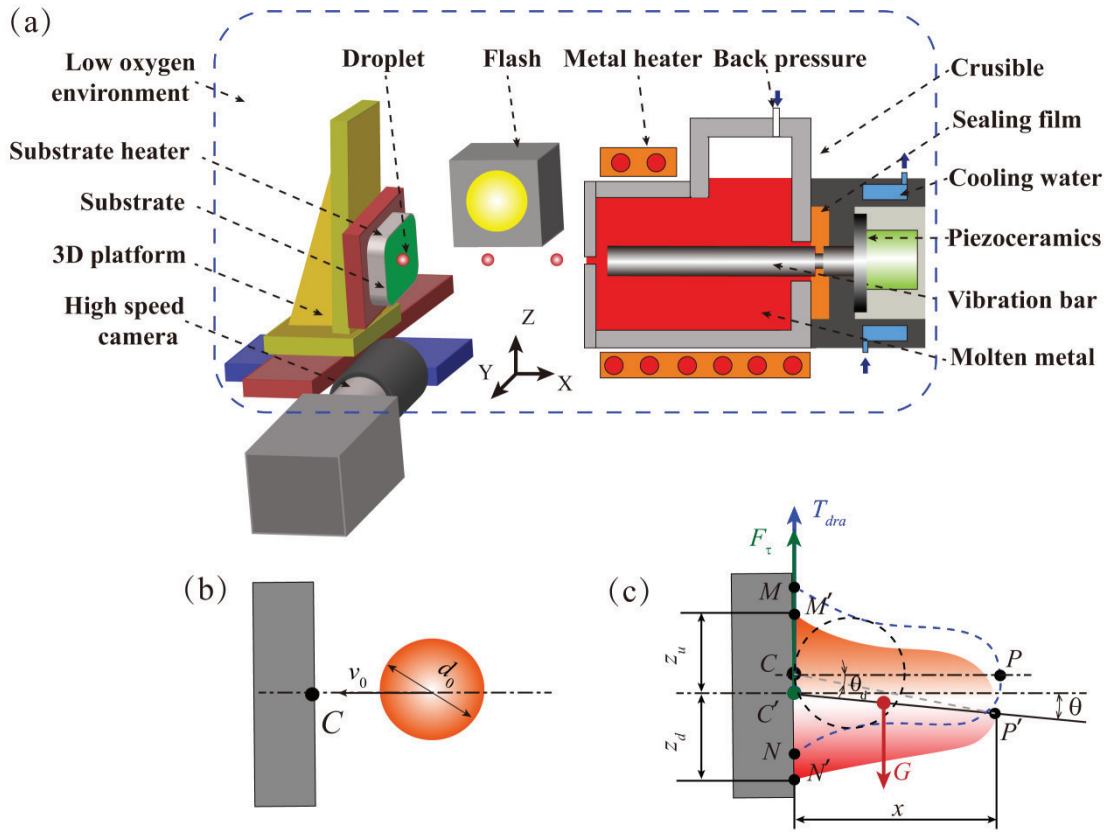
16 The aim of this work is to provide insights into the impact and solidification of
17 metal droplets on vertical substrates. A molten metal droplet horizontal ejection and
18 deposition experimental system is used to eject metal micro-droplets horizontally. As
19 the droplet impacts on vertical substrates, the dynamic behaviors are snapshotted by a
20 high speed camera. The evolutions of spreading factor, vertex height and solidification
21 contour of the dynamic droplet are investigated. Based on these investigations, the
22 gravity effects on impact and solidification are analyzed. Finally, the dependences of the

1 solidification shape of the deposited droplet on the Fr number ($Fr = v_0^2/d_0g$), SHP
2 number ($SHP = (T_d - T_m)/(T_d - T_{sub})$), and Ste number ($Ste = C_l(T_m - T_{sub})/L$) of the
3 droplet are studied. The definition of these dimensionless numbers are from the critical
4 processing parameters[23]: impact velocity, temperatures of droplet and substrate,
5 which are given in Equation (2)-(3).

6 **2. Research methods**

7 *2.1. Experimental approach*

8 The schematic of the deposition system is shown in Fig. 1 (a). It mainly includes a
9 micro droplets horizontal generator, a 3D platform, a low oxygen environment
10 maintenance system (glove box filled with Argon), and a high speed camera with an
11 acquisition rate of 100 kfps (i-SPEED 720, England). The system works as follows: first,
12 raw Sn-63 wt% Pb was melted in a graphite crucible; then, started the generator; under
13 the periodic actuation of the vibration bar, molten metal droplets were ejected on
14 demand. The vibration bar was driven by piezoceramics. The ejection frequency was 1
15 Hz, the orifice diameter was $\sim 200 \mu\text{m}$. The substrate was shifted by the 3D platform,
16 and heated through the substrate heater. The substrate material was copper sheet (50 mm
17 $\times 30 \text{ mm} \times 2 \text{ mm}$), which was mechanically polished before the experiment with the
18 same roughness (see Table 1) to make sure an consistent initial contact condition. The
19 dynamic processes of droplet impacting under three different initial conditions (as listed
20 in Table 1) were snapshotted by the high speed camera.



1
2 **Fig. 1.** Schematics of (a) experimental system, (b) droplet being about to impact, and (c)
3 droplet receding.

4 Fig. 1 (b) and (c) are transient schematics of droplet being about to impact and
5 receding on a vertical substrate, respectively. C is the contact point, while C' is the
6 central point of the contact area of the solidified droplet. M (M') and N (N') denote the
7 upper and lower rim of the deposited droplet, respectively. Assuming microgravity
8 environment, the ideal contour of the receding droplet is $CMPNC$. In contrast, the
9 droplet recedes with a contour of $C'M'P'N'C'$ under gravity. The contour is no more
10 symmetric. Herein, a parameter tilt angle θ is defined to characterize the asymmetry of
11 the solidified droplet induced by gravity effects. θ is the angle between the bisector $C'P'$
12 of the droplet cross-section area and the normal line CP of the vertical substrate surface.
13 The dynamic variation of the tilt angle is characterized as θ_d . Moreover, z and x are
14 introduced to characterize the spread radius and the height of the droplet vertex

1 (denoted by P'), respectively. z_u and z_d are the length of $C'M'$ and $C'N'$, respectively. The
 2 subscripts u and d respectively represent the upward and downward rims of the droplet.
 3 The values of θ , z and x were calculated from the snapshots of the dynamic contours in
 4 droplet impacting via a home-made image processing algorithm programmed with Matlab
 5 R2015a. The main function of the algorithm is to get the coordinate of the key points C ,
 6 C' , M' , N' , P' at first, and then to calculate the geometric relationships.

7 **Table 1**

8 Parameters in experiments and simulation.

| Case | T_d ($^{\circ}\text{C}$) | T_{sub} ($^{\circ}\text{C}$) | Ra (μm) | Substrate Material | v_0 (m/s) | d_0 (μm) | We | Ste/Pr |
|--------|---------------------------------|-------------------------------------|-------------------------|-----------------------|----------------|----------------------------|------|----------|
| Case 1 | 250 | 50 | 0.23 | Cu | 1.70 | 273.7 | 13.5 | 10.8 |
| Case 2 | 250 | 170 | 0.23 | Cu | 1.66 | 279.3 | 13.1 | 4.3 |
| Case 3 | 250 | 240 | 0.23 | Cu | 1.61 | 279.8 | 12.3 | 0.5 |

9 In order to ensure the droplet perpendicularly impact on the substrate, proper
 10 deposition distance is crucial because the velocity increment during droplet flight could
 11 be neglected in such distance. Fig. 2 shows the dependence of the solidification shape of
 12 droplets on the deposition distance with an initial velocity of ~ 1 m/s, an initial droplet
 13 temperature of ~ 250 $^{\circ}\text{C}$, and an initial substrate temperature of ~ 140 $^{\circ}\text{C}$. The tilt angle
 14 of the solidified droplet is constantly equal to $\sim 1^{\circ}$ when the deposition distance is less
 15 than 5 mm, suggesting that the solidification shape is independent of gravity under
 16 certain condition. In our experiment, the distance between the nozzle and the substrate
 17 was ~ 3.5 mm. The velocity in the gravitational direction is at a ratio of $\sim 3.5\%$ to the
 18 ejection velocity (~ 1 m/s). Therefore, the droplet is presumed to perpendicularly impact
 19 on the substrate.

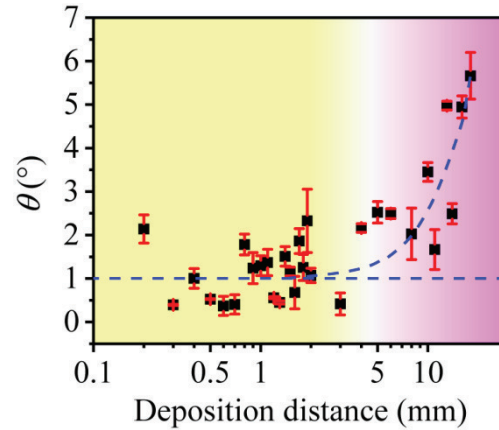


Fig. 2. Dependence of the solidification shape on deposition distance.

2.2. Numerical approach

In order to investigate the flow field during droplet impacting, spreading, oscillating, and solidifying on vertical substrates, a numerical model was established using the volume of fluid (VOF) method [24–26]. The mathematical principle of this simulation model is similar to our previous work [27,28]. The computation domain of the numerical model is shown in Fig. 3. The shape of the droplet was assumed to be a sphere, even though the droplet was slightly oscillating during flight. The initial impact velocity of the droplet was regarded as uniform. The fluid field was assumed as incompressible laminar flow, for the Reynolds number of the metal melt is less than 2300. The incompressible N-S (Navier-Stokes) and continuity equations were used to govern the fluid dynamic processes of the droplet. The enthalpy conservation equation was used to solve the temperature field of the droplet. Since heat transfer to the environment was far less than that to the substrate, the heat dissipation of the droplet was ignored [28].

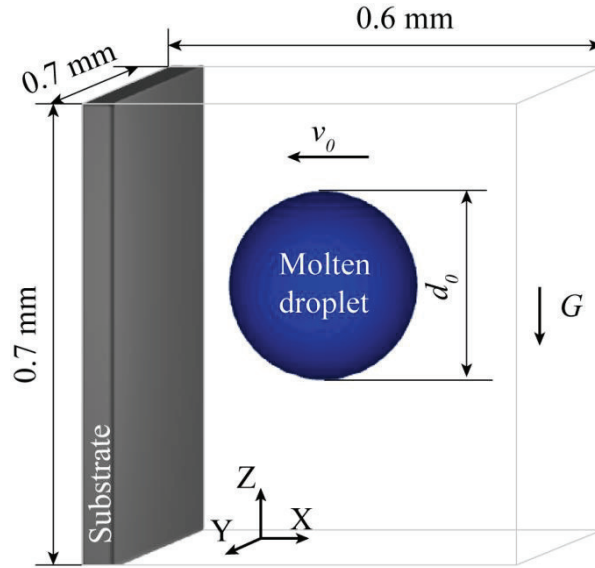


Fig. 3. Computation domain of the numerical model.

Since the computation domain is tiny ($0.7 \text{ mm} \times 0.7 \text{ mm} \times 0.6 \text{ mm}$ in size), the convection between the ambient environment and the droplet could be ignored. The boundary conditions were all set as “continuable” except for the substrate surface. For the thickness of substrate (2 mm in size, and made of copper with a high thermal conductivity of $386 \text{ W/m}\cdot\text{K}$) was about 10 times larger than the droplet diameter, the isothermal “wall” boundary condition was applied to substrate surface. Then, it is noteworthy to note that there is a heat transfer when the molten droplet contacts with the substrate. The heat exchange efficiency is characterized by heat transfer coefficient h_c . h_c is very sensitive to the thermal contact conditions, which are dependent of surface roughness. The coefficient is commonly selected through repeated experimental observations [29–32]. Accordingly, this parameter was set as $6.7 \times 10^5 \text{ W/m}^2\cdot\text{K}$ in the present simulation. For the same reason, the contact angle was set as 150° . The thermo-physical properties of the Sn-63 wt% Pb are listed in Table 2. The subscripts l and s represent the property for liquid phase and solid phase at the temperature of 250

1 °C and 20°C, respectively.

2 A commercial software FLOW-3D 11.2.2.01 was used to implement the numerical
3 model developed above. All initial parameters of droplet, such as the surface tension,
4 contact angle, dimension, velocity, position and temperature can be set in a droplet
5 generating subroutine provided by FLOW-3D. The finite volume method was employed
6 to solve the Navier–Stokes equations for fluid field. Equations were iteratively solved
7 by using a minimum time step of 10^{-9} s. The computational grid consisted of about
8 440,000 rectangular elements, equaling about 30 units per droplet diameter.

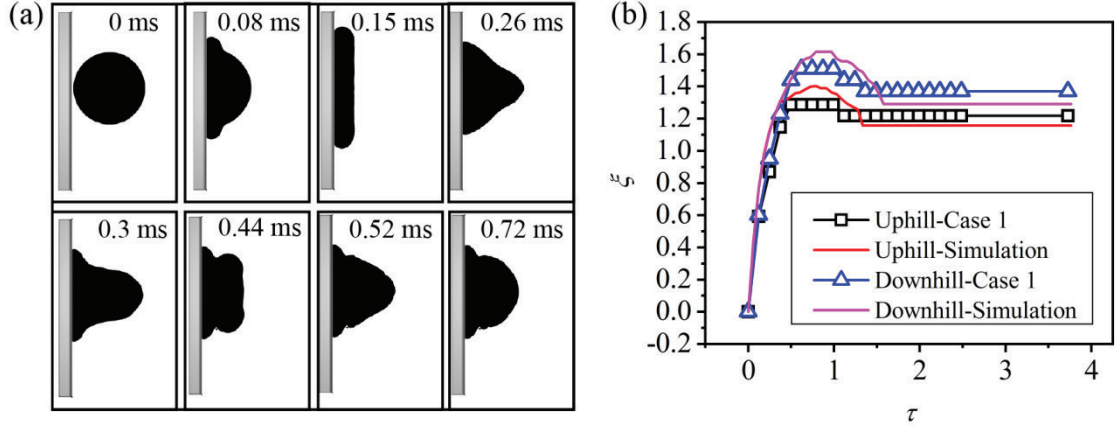
9 **Table 2**
10 Thermo-physical properties of Sn-63 wt% Pb.

| Properties | Value |
|---|----------------------------|
| Density (kg/m^3) | $\rho = 8400$ |
| Dynamic viscosity ($\text{Pa}\cdot\text{s}$) | $\mu = 0.0013$ |
| Surface tension coefficient (N/m) | $\sigma = 0.494$ |
| Latent heat for solidification (J/kg) | $L = 37000$ |
| Heat conductivity coefficient ($\text{W/m}\cdot\text{K}$) | $k_l = 26$ $k_s = 50.9$ |
| Specific heat capacity ($\text{J/kg}\cdot\text{K}$) | $C_l = 203$ $C_s = 167$ |
| Liquidus temperature ($^{\circ}\text{C}$) | $T_{\text{liquid}} = 183$ |
| Solidus temperature ($^{\circ}\text{C}$) | $T_{\text{solid}} = 183$ |
| Contact angle ($^{\circ}$) | $\theta_a = 150$ |

11 2.3. Numerical model validation

12 The simulation results of droplet impacting on vertical substrate are shown in Fig. 4.
13 The initial condition coincides with Case 1 in Table 1. Fig. 4 (a) shows the simulation
14 snapshots of droplet impacting on a vertical substrate. In comparison with the
15 experimental phenomena of Case 1 in Fig. 5, the development of the droplet shapes
16 versus time shows good agreement. Furthermore, the comparison of numerical

1 spreading factors to the experimental results of Case 1 is presented in Fig. 4 (b). They
 2 also agree well with each other, which proved the accuracy and feasibility of the
 3 numerical model.



4
 5 **Fig. 4.** Simulation results of droplet impingement (a) snapshots, (b) comparisons to experimental
 6 spreading factors.

7 **3. Results and discussions**

8 To analyze the droplet impact dynamic and thermal behaviors, dimensionless
 9 parameters including normalized time (τ), spread factor (ξ), dimensionless bump height
 10 (γ), Stefan number (Ste), superheat parameter (SHP) [33], Bond number (Bo), Froude
 11 number (Fr), and Weber number (We) are defined as follows.

$$12 \quad \tau = \frac{tv_0}{d_0}, \quad \xi = \frac{z}{d_0}, \quad \gamma = \frac{x}{d_0} \quad (1)$$

13 where t , v_0 , d_0 , z , x are time, initial velocity of droplet, droplet diameter, spread radius,
 14 height of droplet vertex, respectively.

$$15 \quad Ste = \frac{C_l(T_m - T_{sub})}{L}, \quad SHP = \frac{T_d - T_m}{T_d - T_{sub}} \quad (2)$$

16 where T_m is droplet melting point, T_{sub} , and T_d are initial temperature of substrate and
 17 droplet.

1
$$Fr = \frac{v_0^2}{d_0 g}, \quad We = \frac{\rho d_0 v_0^2}{\sigma}, \quad Bo = \frac{\rho g d_0^2}{\sigma} \quad (3)$$

2 where g is gravity acceleration.

3 *3.1. Impact and solidification of a metal droplet on a vertical substrate*

4 To unveil the gravity effects on impact and solidification, the dynamic processes of
5 metal droplets horizontally impact on vertical substrates with different initial substrate
6 temperatures (as listed in Table 1) were investigated. As shown in Fig. 5, the fluid
7 dynamics processes consist of four stages: impact, spreading, receding and oscillation.
8 When the droplet impacts on a substrate, it spreads outward immediately. After the
9 contact line stretches to the maximum extent, the flow retracts back. The impact
10 processes are analogous with different thermal contact conditions in Case 1- Case 3,
11 whereas the subsequent stages and solidification shapes behave differently. In addition,
12 the contact angles of the upper rims of the droplets are different from the lower ones
13 during the spread and retraction processes. The differences in contact angle vary
14 periodically and appear to be more significant with the increase of the substrate
15 temperature. This phenomenon was not observed in the regime that droplets impact on a
16 horizontal surface [34,35], where the dynamic droplet is axial symmetry, and thus the
17 contact angles of the droplet rim are coincident.

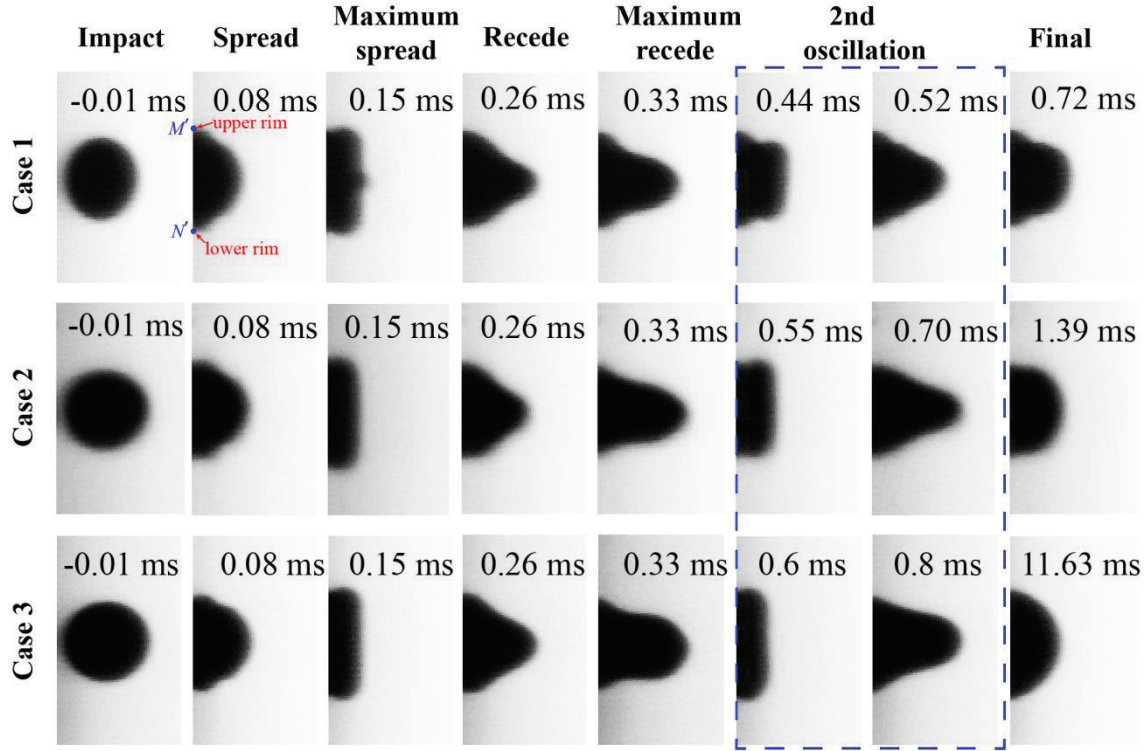


Fig. 5. Snapshots of molten metal droplets impacting on vertical substrates under different thermal contact conditions (Case 1- Case 3).

3.1.1. Mechanisms on the formation of asymmetrical spreading

It is generally known that the spreading factor is a critical parameter characterizing the droplet impact and solidification processes. To further investigate the spreading difference between the upper rim and the lower rim, the evaluation of the spread factor ξ versus dimensionless time τ is investigated. Fig. 6 (a) and (b) show the migration of the upper and lower rims of the droplet, respectively. The maximum spread factors of the upper rim in Case 1- Case 3 are all equal to 1.28, while the lower ones equal to 1.51, 1.52 and 1.58, respectively. The spread factor of the lower rim undergoes an increase with the growth of substrate temperature due to different solidification rate of the contact line of droplet. Compared to surface tension and viscous dissipation, the effect of solidification on droplet spreading can be quantified by a dimensionless parameter ϕ [31]. During the spreading processes, solidification could be neglected if

$$\phi = \frac{Ste}{\sqrt{Pr}} \sqrt{\frac{\eta_s}{\eta_d}} < 1 \quad (4)$$

where $Pr = \mu C_l / k_l$, $\eta = k \rho C$, the subscripts s and d represent substrate and droplet respectively. For Case 1- Case 3, the values of ϕ approximately equal 6, 3.3, and 0.3, respectively. Accordingly, solidification plays an important role in resisting the spreading process in Case 1 and Case 2. The solidified melt around the edges of spreading droplet will form a solid rim that obstructs the surface flow, resulting in that the motion of the contact line ceases during receding. In contrast, in Case 3, the droplet oscillates continually owing to a higher substrate temperature than the melting point of droplet. What is of interest is that for all cases the spread factors of the upper rims are smaller than those of the lower ones, while the retraction of the upper rim is more obvious comparing to the lower ones. We supposed that the differences in the spreading and retraction of the upper and lower rims come from the effects of gravity. To validate this supposition, we investigated the gravity effects on the spreading and retraction velocity and characteristic time for spreading and receding of the impacted droplet.

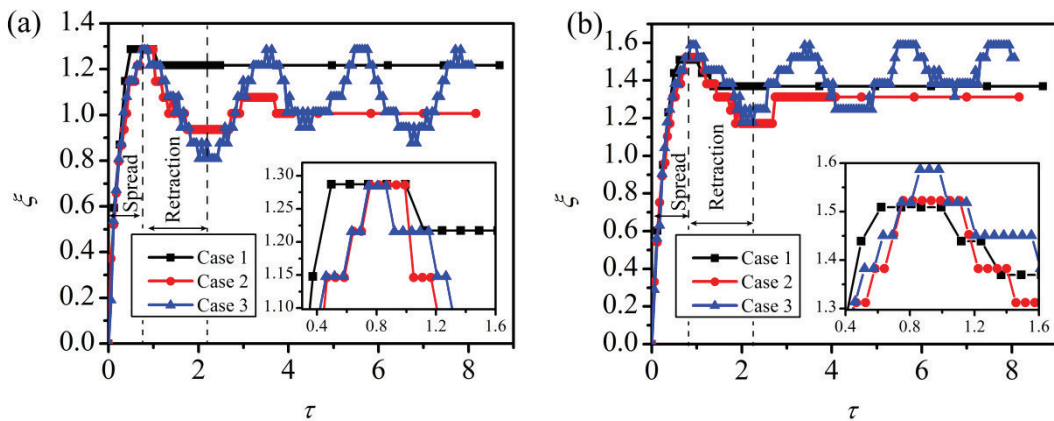
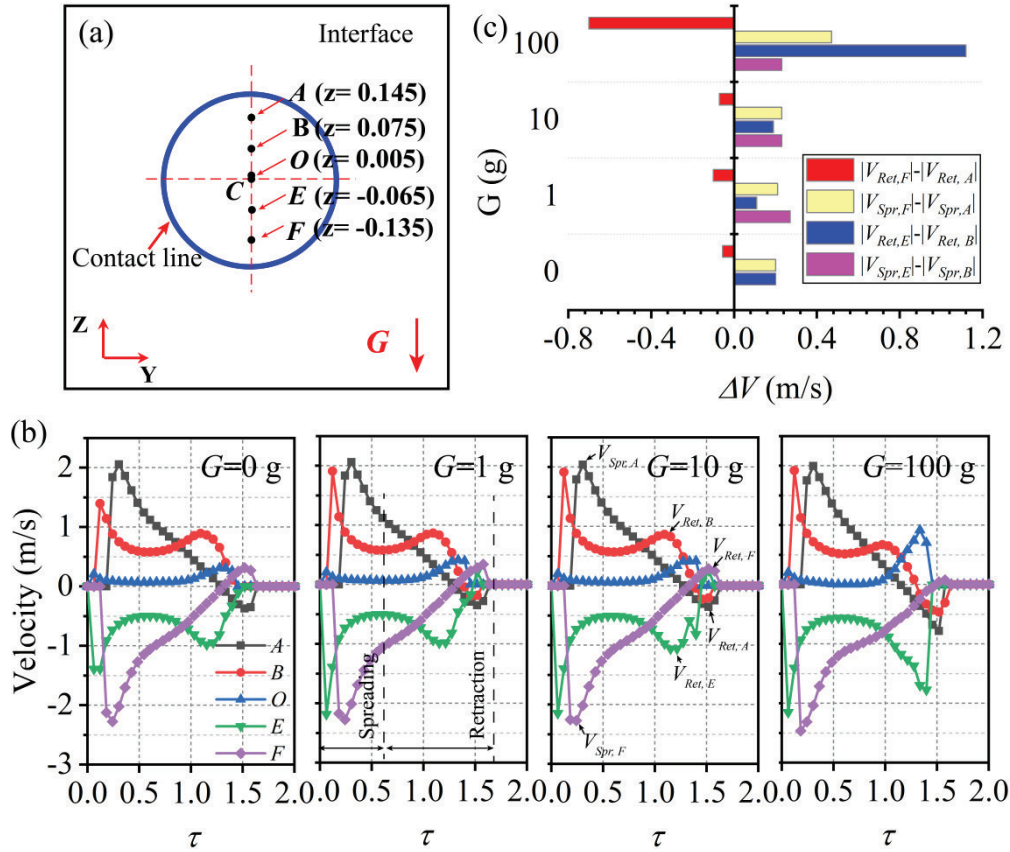


Fig. 6. Variations of the spread factor versus time for the droplet in Case 1-3: (a) uphill spread, (b) downhill spread.

To understand the gravity effects on the spreading and retraction velocity of the

1 impacted droplet, the evolution of the fluid field of the droplet in Case 1 versus
2 spreading time was numerically calculated. We designated five specific points on the
3 interface (labeled as A , B , O , E , F as shown in Fig. 7 (a)) to monitor the velocity field.
4 Points A , B , O are on the route the upper rim of the droplet spreads, and E , F are on the
5 route the lower rim of the droplet spreads. Point O is a symmetrical point of A (B) and E
6 (F). The variations of the velocity of each point versus time under different gravity
7 levels (0 g, 1 g, 10 g, and 100 g) are shown in Fig. 7 (b). $\tau=0$ corresponds to the instant
8 that the droplet impact on the substrate. The negative velocity denotes the fluid flows
9 along the gravity direction, in contrast, the positive velocity represents a reverse flowing
10 direction. Initially, the spreading velocity is small as the velocity curve of point O shows.
11 As the droplet continues to spread forward, the contact line successively pass over point
12 O , E , B , F , A . The velocities at points A and B are respectively smaller than those at
13 points F and E , suggesting that the spreading velocity of the lower rim is faster
14 compared to the upper rim. Based on Newton's law, we can conclude that the upper rim
15 of the droplet experiences a deceleration of gravity, but the lower rim experiences
16 acceleration by gravity. The differences in the velocity peak (marked in Fig. 7 (b)) of
17 each point pairs ($E \rightarrow B$ and $F \rightarrow A$) are shown in Fig. 7 (c). With the increase of the
18 gravity level, the velocity differences tend to be more obvious, especially at droplet
19 retraction stage. It is noteworthy that the velocity of Point B after $\tau=1.3$ turns to be
20 negative, while the velocity of Point E approximates to 0. All these behaviors indicate
21 that the differences in the spreading factor of the upper and lower rims come from the
22 effects of gravity.



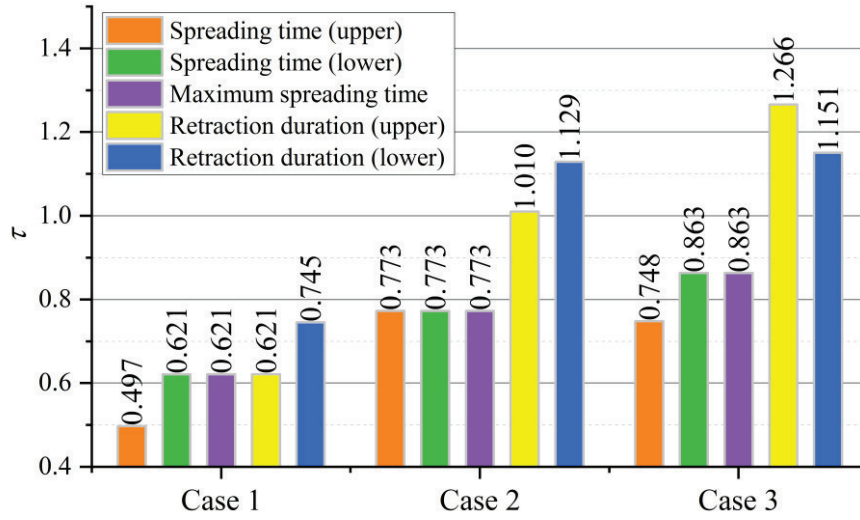
1
2 **Fig. 7.** (a) Coordinate of point A-F; (b) velocity evolution at five characteristic points under different
3 gravity levels; (c) the differences in the velocity peak between E (F) and B (A).

4 Furthermore, Fig. 8 shows the spreading time, maximum spread time, and retraction
5 duration of the first oscillation period of droplets in Case 1-Case 3. These characteristic
6 times show an increment with the increase of substrate temperature, essentially
7 exhibiting a dependence on solidification. Specifically, the spreading time of the upper
8 rim is shorter than that of the lower one as the droplet spreads outward to its limits. The
9 time required for the droplets to reach their maximum spread extent equals the
10 spreading time of the lower rim, which means that the upper rim begins to retract at the
11 moment that the lower rim reaches to its maximum extent. As the droplet retracts back
12 inward, the retraction duration of the upper rim is also smaller than that of the lower rim
13 for droplet in Case 1 and Case 2. As the droplet spreads out, the volume fraction of
14 droplet spreads downhill is more than that spreads uphill because of gravity effects. In

1 other word, the total heat of the downstream fluid is more than that of the upstream.
2 Thus, the upper rim undergoes a fast solidification rate during spreading and retraction
3 processes.

4 But the exception is that the retraction duration of droplet in Case 3 shows an
5 completely opposite regular pattern compared to Case 1 and Case 2, that is the
6 retraction duration of the upper rim is larger than the lower one. The reason is that there
7 is no solidification occurs on the condition that substrate temperature is higher than the
8 droplet melting point, resulting in that the upper rim of the droplet experiences
9 acceleration by gravity during receding as the lower rim does in spreading.
10 Consequently, the retraction extent of the upper rim is certainly larger than that of the
11 lower one. This agrees well with the receding behaviors as shown in Fig. 6. Accordingly,
12 it can be concluded that gravity is a nonnegligible effect on the differences in
13 characteristic time for droplet spreading and receding.

14 Based on the above discussions, it shows that the velocity and characteristic time
15 can indirectly embody the gravity effects on the asymmetrical spreading and retraction
16 of the impacted droplet.



1

2

Fig. 8. Characteristic time for droplet spreading and receding in the first oscillation.

3

3.1.2. Mechanisms on the formation of asymmetrical solidification shape

4

It is found that the solidification shape of the droplet deposited on the vertical substrate is also asymmetric (the droplet tilts to gravity direction). To ascertain the

5

formation of this undesired solidification shape, we investigated the variations of the

6

vertex height D' (as seen in Fig. 1 (c)) and the tilt angle θ of the droplet versus the

7

dimensionless time τ . As shown in Fig. 9 (a)-(b), the vertex D' oscillates with time until

8

the droplet solidified. With higher substrate temperature, the oscillation decays slowly.

9

Under the condition that the temperature of the substrate exceeds the melt point of the

10

droplet (Fig. 9 (c)), the oscillation only decays by dissipation. During the droplet

11

oscillating, θ varies synchronously with the vertex height. The peak values of θ present

12

the instants when the vertex D' descends to the lowest position in every oscillation

13

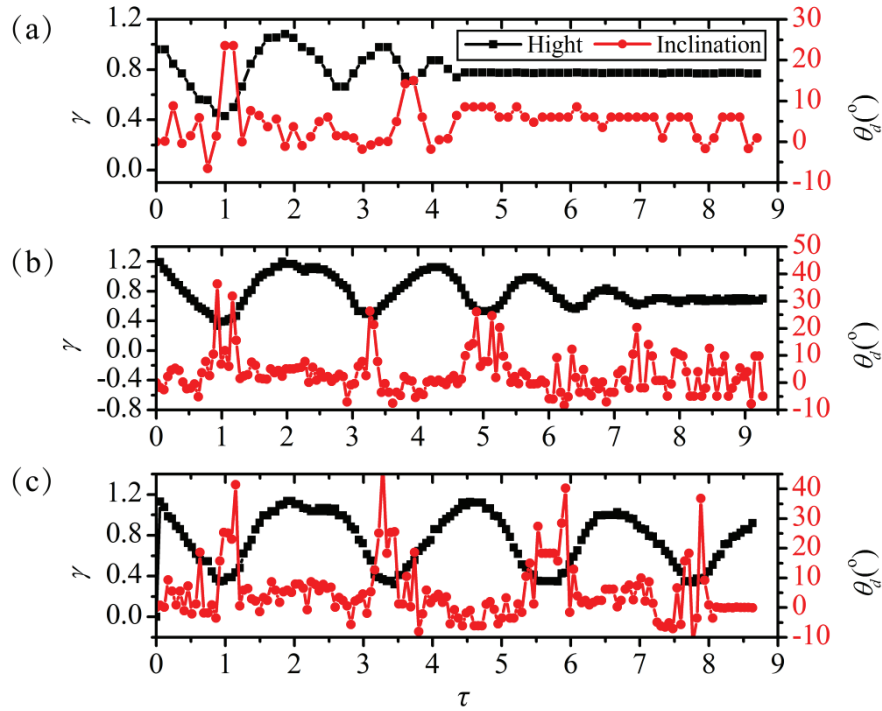
period. These values decrease with the decay of the oscillation. It indicates that the

14

gravity effects behave more significantly when the droplet retracts back. The dynamic

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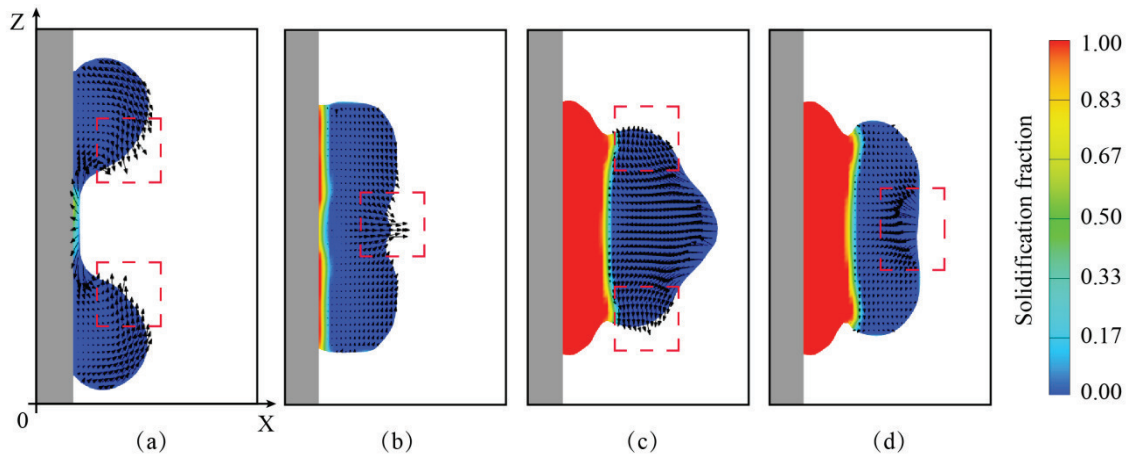
1 droplet inevitably shows an up-down oscillation after impacting on a vertical substrate.



2
3 **Fig. 9.** The apparent dimensionless height and tilt angle of the droplet versus time in (a) Case 1; (b)
4 Case 2; (c) Case 3.

5 To demonstrate the development of the tilt angle θ , the fluid field of the droplet in
6 oscillation was investigated. Four different oscillation moments are shown in Fig. 10,
7 Fig. 10 (a)-(b) are picked out from the first retraction period and Fig. 10 (c)-(d) from the
8 second spreading period. Basically, the velocity field is asymmetric. Affected by gravity,
9 the velocity vectors of the upper part are larger than the lower part when the droplet
10 recedes inward. On the contrary, the vectors of the upper part are smaller than the lower
11 part when the droplet spreads outward. As a result, the droplet oscillates up and down.
12 Synchronously, the droplet solidifies from bottom to top. The up-down oscillation and
13 solidification of the droplet interact with each other. Under the competition of
14 oscillation and solidification, the solidifying phase propagates from bottom to top at the
15 moment the droplet protuberates like a tower (Fig. 11 (b)), which is subject to a

1 deflection induced by bending moment [36]. As a result, the solidified droplet behaves a
 2 noticeable tilt angle of θ . This undesired solidification shape would usually hinder a
 3 good fusion between neighboring droplets, resulting in that some hole-defects emerge.
 4 According to Yi and Qi et al. [37,38], the hole-defect comes from a time-dependent void
 5 region formed among two neighboring droplets and the substrate. It is because that
 6 under the drag effects from gravity, the residual liquid in the molten droplet trends to
 7 flow downwards. That is contrary to the direction of filling in the void region. To this
 8 end, the influence law and elimination strategy for the undesired solidification shape of
 9 droplets on vertical substrates should be studied, which will be discussed in the next
 10 section.



11
 12 **Fig. 10.** Fluid field of the droplet in Case 1 at (a) 0.18 ($\tau=1.1$), (b) 0.2 ms ($\tau=1.2$), (c) 0.37 ms
 13 ($\tau=2.3$), (d) 0.44 ms ($\tau=2.7$).

14 To demonstrate the formation mechanisms of the aforementioned nonuniform fluid
 15 field that brings about the up-down oscillation, the forces (solidification drag, surface
 16 tension, and gravity) exerted on the droplet are analyzed as shown in Fig. 11. The
 17 droplet adhered on the vertical substrate can be regarded as a soft overhanging beam,
 18 which is subject to a bending moment [36] The droplet that protuberates like a tower as

1 shown in Fig. 11 (b) tilts to gravity with a notable deflection during retraction, which
 2 presents as a downward oscillation. On contrast, as the outstretched droplet retracts back
 3 at spreading stage, it shows an upward oscillation (see Fig. 11 (a)). To further
 4 demonstrate this argument, the deflection equation is established. Fig. 11 (c) is an
 5 infinitesimal unit that captured from Fig. 11 (b) at t minute. There is

$$6 \quad \rho A(x,t) \frac{\partial^2 w(x,t)}{\partial t^2} = - \frac{\partial Q(x,t)}{\partial x} \quad (5)$$

7 where ρ is density, $w(x, t)$ is the deflection of a point with an abscissa of x on the tower,
 8 $A(x, t)$ and $Q(x, t)$ are the area and shear force of the cross-section of the infinitesimal
 9 unit, respectively. The rotation equation of the infinitesimal unit is

$$10 \quad Q(x,t) = \frac{\partial M(x,t)}{\partial x} \quad (6)$$

11 where $M(x, t)$ is the bending moment of the infinitesimal unit. Combining Eq. (5) and
 12 Eq. (6), there is

$$13 \quad \rho A(x,t) \frac{\partial^2 w(x,t)}{\partial t^2} = - \frac{\partial^2 M(x,t)}{\partial x^2} \quad (7)$$

14 According to the mechanics of materials, there is

$$15 \quad M(x,t) = EI(x,t) \frac{\partial^2 w(x,t)}{\partial x^2} \quad (8)$$

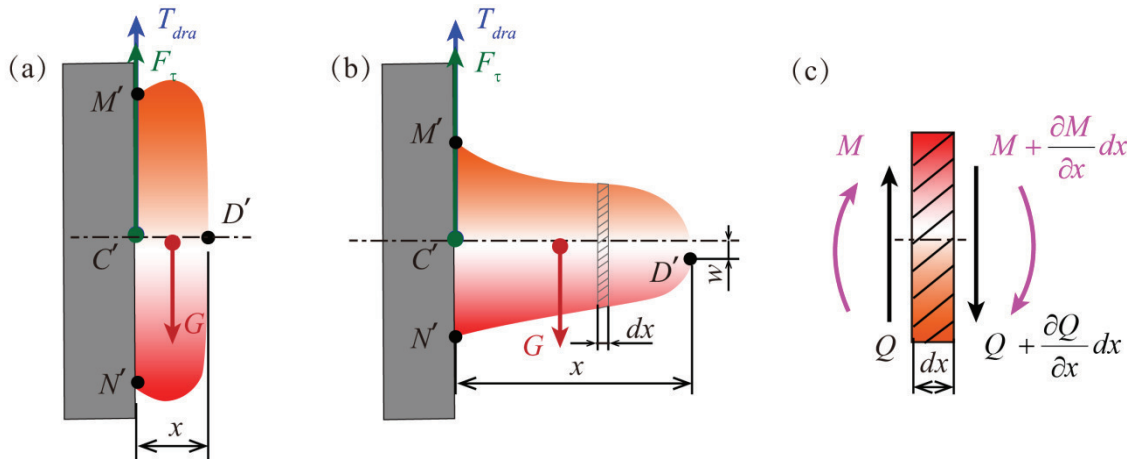
16 where $EI(x, t)$ denotes the bending rigidity of the cross-section. Substituting Eq. (8) into
 17 Eq. (7), we obtained

$$18 \quad \rho A(x,t) \frac{\partial^2 w(x,t)}{\partial t^2} = - \frac{\partial^2}{\partial x^2} \left[EI(x,t) \frac{\partial^2 w(x,t)}{\partial x^2} \right] \quad (9)$$

19 Given a certain instant, the general solution of w can be expressed as

$$20 \quad w_n = K_1 \left[(\cosh \beta_n x - \cos \beta_n x) \right] + \frac{\cos \beta_n x_D + \cosh \beta_n x_D}{\sin \beta_n x_D + \sinh \beta_n x_D} (\sin \beta_n x + \sinh \beta_n x) \quad (10)$$

1 where K_I is determined by the initial conditions of the velocity and displacement of the
 2 beam at $t=0$. x_D is the height of the droplet vertex. $\beta_n = (\rho A \omega_n^2 / EI)^{1/4}$, ω_n are the
 3 natural frequencies of the dynamic droplet. Obviously, the deflection of the overhanging
 4 beam-like dynamic droplet follows vibration function. Accordingly, the droplet behaves
 5 an up-down oscillation.



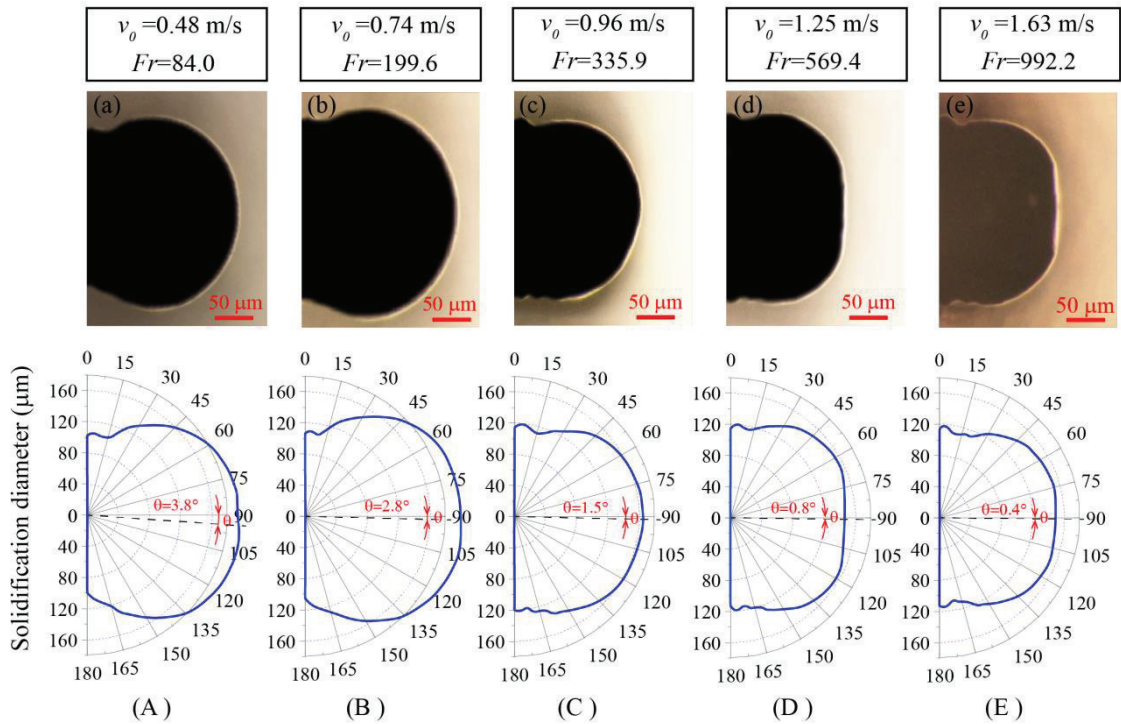
6
 7 **Fig. 11.** Force schematics of droplet at (a) flat pancake shape (spreading) and (b) tower shape
 8 (retraction), (c) an infinitesimal unit captured from (b).

9 3.2 Effects on the undesired shape of the solidified droplets

10 3.2.1. Effects of impact velocity

11 To unravel the influence law on the undesired shape of the solidified droplets, the
 12 dependence of the tilt θ versus processing parameters were studied. As the We numbers
 13 of the droplets in Case 1-3 are approximately equal to 13 and the Ohnesorge numbers
 14 (from here on Oh , which denotes the ratio of viscous forces to surface tension,
 15 $Oh = \mu / \sqrt{\rho \sigma d_0}$) do not exceed 0.01, the droplet spreading is mainly induced by the
 16 dynamic pressure gradient and resisted by inertia [39]. Therefore, the solidification
 17 shapes of droplets with impact velocities varying from 0.48 m/s to 1.63 m/s were
 18 investigated as shown in Fig. 12 (a)-(e). The initial temperatures of the droplet and

1 substrate are $\sim 250\text{ }^\circ\text{C}$ and $\sim 140\text{ }^\circ\text{C}$, respectively. As shown in Fig. 13, the tilt angle θ of
 2 the solidified droplet decreases with the increase of the Fr number. As illustrated in the
 3 previous section, when the rim of the droplet retracts back to its minimal diameter, the
 4 receded droplet is analogous to an overhanging beam, which is subject to a bending
 5 moment because of gravity. Since the spreading diameter of the droplet increases with
 6 the Fr number [40], the height of the top vertex is large under the condition of small Fr
 7 number. Accordingly, the droplet is correspondingly subject to a large bending
 8 deflection. Finally, the droplet solidifies into an obvious tilt shape (characterized as θ).



9
 10 **Fig. 12.** Solidified droplets under various initial velocity: (a) - (e) micrographs, (A) - (E) contours of
 11 (a) - (e), respectively.

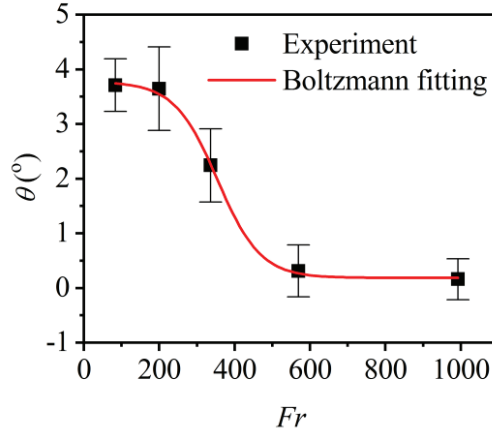


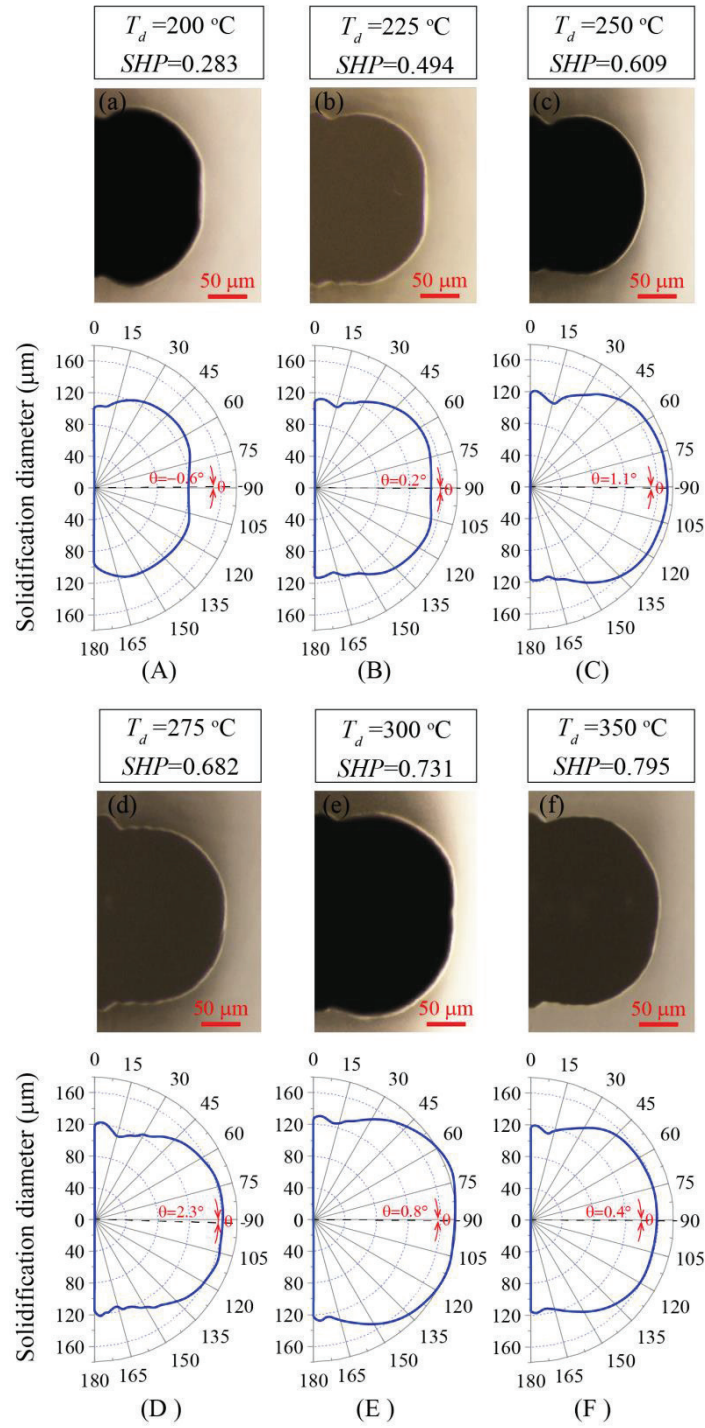
Fig. 13. Dependence of θ on the Fr number of droplets.

When the Fr number of the droplet is larger than 600, θ is less than 0.5° . This small tilt angle could be neglected in the printing process because there might exist an inherent asymmetry [35] of solidified droplets owing to the effects of ejection errors [10] and nonuniform wetting of the substrate surface.

3.2.2. Effects of droplet temperature

As a molten metal droplet deposits on the substrate, the heat of the droplet transfers to the substrate accompanied by fluid dynamics. The initial temperatures of the droplet and substrate have a crucial impact on the final solidification shape of the droplet. As shown in Fig. 14 (a)-(f), by varying the initial temperature of the droplet, different solidification shapes are observed. The dimensionless number superheat parameter (SHP) [33] is used to characterize the heat of the droplet. In Fig. 14, the substrate temperature is 140°C , and the impact velocity is 0.96 m/s . The dependence of θ on SHP is shown in Fig. 15. The value of θ increases with the increase of SHP firstly, but then decreases as the SHP of droplet is larger than 0.65. When SHP is smaller than 0.5 or larger than 0.8, θ is less than 0.5° . In this parameter interval, the deposited droplet could solidify into a good solidification shape. However, as shown in Fig. 14 (a), the solidification angle is

- 1 obviously large with small SHP (<0.5), which is not conducive to printing because of
- 2 the drawback of easily forming hole-defects between two neighbouring droplets and
- 3 substrate. Therefore, only the parameter interval $SHP > 0.8$ is appropriate for printing.



4
5 **Fig. 14.** Solidified droplets under various droplet temperature: (a) - (f) micrographs, (A) - (F)
6 contours of (a) - (f), respectively.

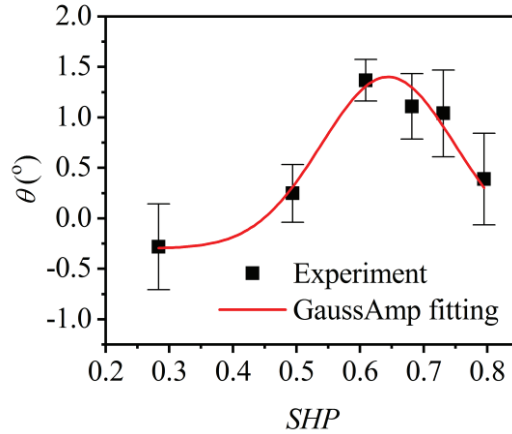


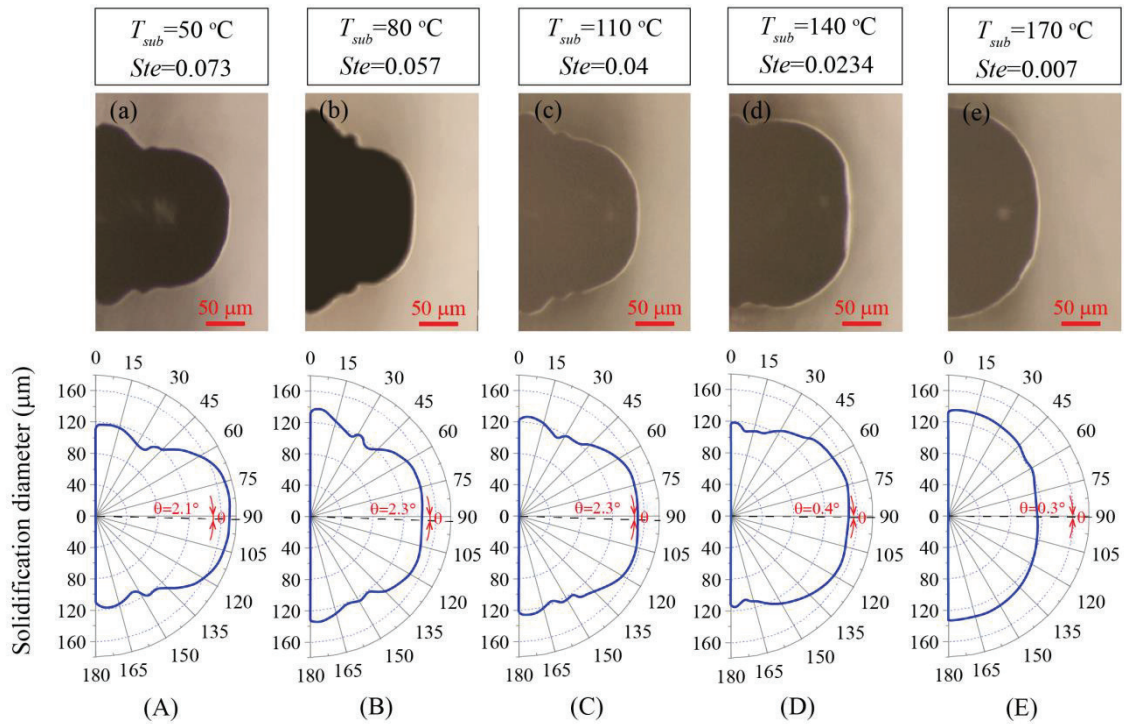
Fig. 15. Dependence of θ on the SHP number of droplets.

In Fig. 15, the variation of θ is attributed to the influence of droplet temperature on solidification time. According to Waldvogel et al. [33], the solidification time increases with droplet temperature. For a lower droplet temperature, the rim of the droplet solidifies before it retracts back due to the rapid solidification of the droplet bottom. Therefore the spread diameter is relatively large, and the height of the top vertex of the droplet is relatively small. Conversely, when the droplet temperature is high, the solidification rate is slow. The kinetic energy of the oscillating droplet reduces to a considerably low level before the rim of the droplet starts to solidify, resulting in low impact of the up-down oscillation of the droplet on solidification shape. For a moderate droplet temperature, the droplet starts to solidify during the retraction process. As a result, the undesired solidification shape shows up.

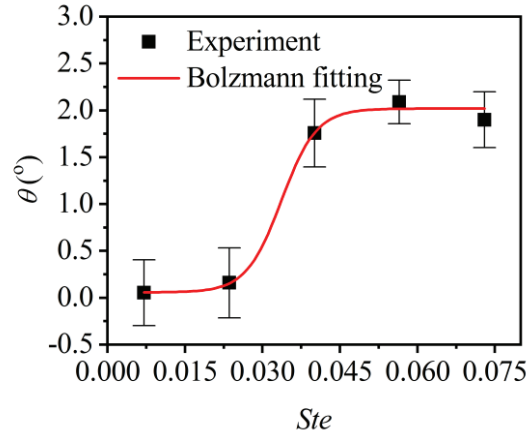
3.2.3. Effects of substrate temperature

In Fig. 16 (a)-(e), we introduced Ste number to characterize the thermal properties that pertains to substrates. The initial temperature of the droplet is 250 °C, and the impact velocity is 1.63 m/s. The solidification shape of the droplet varies with different Ste number. As shown in Fig. 17, θ increases as the Ste number of the substrate

1 increases from 0.3 to 0.4, and keeps constant when Ste is smaller than 0.3 or larger than
 2 0.4. What is different from Fig. 15 is that the droplet solidifies into an obvious tilt shape
 3 when the substrate temperature is low. In this case, we could get a regular solidification
 4 shape as the Ste number is less than 0.057. Similarly, a higher substrate temperature
 5 enables the oscillating droplet have plenty of time to make the kinetic energy reduce to
 6 a considerably low level before the rim of the droplet starts to solidify. That suppresses
 7 the oscillation effects on solidification.



8
 9 Fig. 16. Solidified droplets under various substrate temperature: (a) - (e) micrographs, (A) - (E)
 10 contours of (a) - (e), respectively.



1
2 **Fig. 17.** Dependence of θ on the Ste number of droplet.

3 According to the investigation on the effects of impact velocity, temperatures of
4 droplet and substrate, the undesired solidification shape of the deposited droplet on
5 vertical substrates would be effectively eliminate by moderately improving the inertia
6 and slowing the solidification rate of the droplet on the condition that no splash and
7 over-remelting emerge. In future work, theoretical research should be developed to
8 obtain a mapping between solidification shape and processing parameters. Moreover,
9 the floating zone convection causing the difference of the microstructure in
10 microgravity from that on the earth should also be eliminated by external field (i.e.
11 surface acoustic waves (SAWs) [41], electric field [42], and magnetic fields [43]).

12 **4. Conclusions**

13 In summary, this work unveils the impact and solidification behaviors of molten
14 metal droplets on vertical substrates through experiment and numerical simulation. As
15 the droplet impacts on the substrate, tilt solidification shape arises due to the coupling
16 effects of the asymmetrical spreading, up-down oscillation and solidification. The
17 undesired solidification shape shows a dependence on the processing parameters such as
18 impact velocity, the temperature of droplet and substrate, suggesting that the undesired

1 solidification shape could be suppressed through appropriately selecting the processing
2 parameters. With larger impact inertia or slower solidification rate, the undesired
3 solidification shape would be effectively suppressed. This work provides a foundation
4 to ground-based simulation of droplet deposition 3D printing under microgravity. Future
5 works will be addressed to eliminate the gravity effect on the solidification shape and
6 microstructure of the deposited droplet in ground-based simulation of printing processes
7 through optimizing parameter combination and introducing external field.

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