

# Solidification of a Liquid Metal Droplet Impinging on a Cold Surface

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**Abstract:** A method for the solidification of a free surface liquid phase is presented and solved with COMSOL Multiphysics 3.2 using a fixed grid. The case of a 2 mm diameter liquid aluminum droplet, initially at 670 °C, spreading onto a flat stainless steel surface kept at a fixed temperature, from 610 °C to 670 °C was studied in 2D axial symmetry. In this case, the Level set method with phase re-injection for mass conservation is used to prescribe the movement of the free surface of the liquid droplet, whereas a modified version of the method presented by Voller and Prakash is used to account for the solidification of the liquid phase, including convection and conduction heat transfer with mushy region phase change, for multicomponent systems or pure materials. The results show that this model is applicable to systems presenting high density and viscosity ratios (aluminum/air) and also for a materials with high latent heat of melting such as aluminum. However, restrictions on the surface tension force model still exist.

**Keywords:** Free Surface Flows, Multiphysics, Fluid Flow, Heat Transfer, Mass Transfer, Level Set Method, Solidification

## 1. Introduction

The study of free surface or multiphase flows are of great importance in the metallurgical and chemical industry. In the production of metals, there are many instances in which multiphase flows and interfacial phenomena are present. Examples of this can be found in processes such as mineral concentration in flotation cells, separation between organic and aqueous phases during solvent extraction in hydrometallurgical plants, generation of immiscible phases during melting and converting of minerals, settling of metallic inclusions in slag, injection of gases to react with the molten phases, etc. In some cases, in addition to deal with multiphase flows, one or

more of the fluids undergoes a phase change due to melting, solidification or chemical reactions, specially in pyrometallurgy of ferrous, non-ferrous or light metals. Some examples correspond to casting, matte granulation, formation of a protective solid slag layer at the walls of the furnace and also during the manufacturing processes employing techniques like thermal spray coating.

The objective of this article is to study the case involving both free surface flow and solidification of a metallic droplet onto a substrate. Although this is a specific problem, the model proposed can be applied to other problems involving multiphase flow with phase change such as the examples mentioned before.

In the literature, similar problems can be found. Pasandideh-Fard et. al. present experimental and computational studies for the solidification of tin droplets [1] and heat transfer from water drops on a hot surface [2] in 2D axial symmetry using the volume of fluid method (VOF) together with an enthalpy formulation for the heat transfer of pure materials. A 3D for the problem of droplet spreading without considering solidification using the VOF method has been described by Bussman et. al. [3]. In the those studies, the contact angle between the liquid and the substrate was imposed as a boundary condition in the model based on experimental observations. Similarly, thermal contact resistance between the droplet and the substrate is modeled through a heat transfer coefficient whose value is based on experimental measurements of the substrate temperature during the impact.

The approach proposed in this article, is a combination of the Level set method described by Sussman et. al. [4] to track the interface between the liquid and gas phases and the

solidification model by Voller and Praksh [5], modified so that the heat transfer equation is based on the temperature instead of the original enthalpy formulation and the latent heat of melting is included as a heat capacity term.

## 2. Governing Equations

### 2.1 Fluid Flow

The flow of the droplet and the gas surrounding the fluid were modeled using the Navier-Stokes equations for incompressible laminar flow.

$$\rho \frac{\partial \mathbf{u}}{\partial t} - \nabla \cdot \mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) + \rho (\mathbf{u} \cdot \nabla) \mathbf{u} + \nabla p = \mathbf{F} \quad (1)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (2)$$

As it will be described later, the interface between the liquid and gas as well as the interface between the liquid and solid, are treated using an Eulerian approach in a fixed grid. Therefore, the position of these interfaces are not explicitly calculated and there is no need to generate a mesh at the interfaces. Finally, only one set of Navier-Stokes equations is required. In these equations, the source term  $\mathbf{F}$ , include the volumetric buoyancy force, due to the density difference between the gas and the liquid, surface tension effect at the liquid-gas interface and finally, additional terms used to model the solidification of the fluid. All these source terms will be detailed in the following sections.

### 2.2 Level Set Method

In the model proposed in this study, the liquid-gas interface is tracked by the Level Set method as described by Sussman et. al. [4]. A brief description of this method is given below:

In this case, a scalar function  $\phi$  is defined such that it takes strictly negative values for the liquid phase, zero at the interface and strictly positive values for the gas phase. The function  $\phi$  is advected by the fluid flow according to the following equation:

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = 0 \quad (3)$$

The change of properties for the different fluids can be calculated as a function of the level set function  $\phi$  for the density, viscosity and thermal conductivity, respectively, as follows:

$$\rho = \rho_g + H(\phi)(\rho_l - \rho_g) \quad (4)$$

$$\mu = \mu_g + H(\phi)(\mu_l - \mu_g) \quad (5)$$

$$k = k_g + H(\phi)(k_l - k_g) \quad (6)$$

where  $H(\phi)$  is a smooth heavy side function defined as:

$$H(\phi) = (1 + \tanh(-\phi/n))/2 \quad (7)$$

in the previous equation,  $H(\phi)$  takes the value of 1 for the liquid phase and 0 for the gas phase with a smooth transition at the interface. The width of that transition is controlled by the parameter  $n$ .

Surface tension effects are included as a body force (or source term) to the Navier-Stokes equations acting only at the interface:

$$\mathbf{F}_{ST} = \gamma \kappa \delta(\phi) \mathbf{n} \quad (8)$$

where  $\mathbf{F}_{ST}$  is a vector field for the body force,  $\gamma$  is the surface tension,  $\kappa$  is the curvature of the interface,  $\delta(\phi)$  is the Dirac delta function and  $\mathbf{n}$  is the unit normal vector to the interface. Equation 8 is approximated by the use of a smooth Dirac delta function which expands in a given width around the interface:

$$\delta(\phi) = \frac{n_2}{\sqrt{\pi}} e^{-n_2^2 \cdot \phi^2} \quad (9)$$

the normal vector and curvature at the gas liquid interface are calculated in terms of the level set function according to:

$$\mathbf{n} = \frac{\nabla \phi}{|\nabla \phi|} \quad (10)$$

$$\kappa = \nabla \cdot \frac{\nabla \phi}{|\nabla \phi|} \quad (11)$$

In the original model of Sussman et. al. [4], it was shown that it is necessary to keep  $\phi$  as a distance function in order to correctly evaluate surface tension force at the interface and more important to ensure mass conservation of the two phases. This is done by reinitializing the level set function  $\phi$  at every time step, so that  $|\nabla \phi| = 1$ . The problem with the implementation of this approach in COMSOL is that it is necessary to stop the solution at every time step or after a given amount of time for which distortions in the level set function are such that reinitialization of  $\phi$  is required, and solve for the steady problem of transforming  $\phi$  into a distance function, then replace this new “initial” condition into COMSOL and continue advancing in time. A different approach named “phase re-injection” has been proposed by K. B. Deshpande et. al [6] in which integration coupling variables are used to compute the mass difference between the original level set function and the actual function at a given time, then this difference is “injected” into the level set equation as a generation or source term for  $\phi$  at the interface, thus ensuring mass conservation. Equation 3 is modified to include the source term:

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = \varphi \delta(\phi) \quad (12)$$

The right hand side of equation 12 represents the generation or source term for the function  $\phi$  applied only at the interface, since it uses the smooth Dirac delta function. The term  $\varphi$  is the difference between the mass of the initial function  $\phi$  and the new level set function  $\phi$ . The advantage of this technique is that it is not necessary to solve the problem of reinitializing  $\phi$  at every time step. In the present model, this is the technique used to avoid mass leakage.

### 2.3 Solidification

The solidification model is derived directly from that proposed by Voller and Prakash [5]. However, in this case, the heat equations is

written in terms of the Temperature rather than enthalpy as in the original method:

$$\rho \cdot c_p \frac{\partial T}{\partial t} + \nabla \cdot (-k \nabla T + \rho \cdot c_p T \mathbf{u}) = 0 \quad (13)$$

Similarly to the level set method, the solidification model does not track the interface between the solid and liquid phases explicitly; it rather uses a fixed grid and modifies the Navier-Stokes equations to represent the flow of a fluid or a solid by introducing additional source terms. Equation 13 is valid for all three phases, solid, liquid and gas, the change in properties for the different phases is captured by equations such as 4 to 6.

First, a solid fraction function ( $F_S$ ) is defined as a function of Temperature. This function takes a value equals to zero when the temperature is below the solidus temperature, and equals to 1 for all temperatures above the liquidus temperature. In the transition zone,  $F_S$  takes values between 0 to 1, in this case  $F_S$  is defined as follows:

$$F_s(T) = \begin{cases} 0, & T \geq (T_m + \varepsilon) \\ (T_m + \varepsilon - T) / 2\varepsilon, & (T_m + \varepsilon) > T \geq (T_m - \varepsilon) \\ 1, & T < (T_m - \varepsilon) \end{cases} \quad (14)$$

where  $2\varepsilon$  is the temperature range between liquidus and solidus.  $T_m$  is the “average” melting temperature, half way between liquidus and solidus. For pure materials,  $\varepsilon = 0$ .

The assumption of the solidification model is that the fluid flow in the transition range, during solidification is similar to the flow in a porous media. A porosity function depending on the temperature is then defined:

$$\lambda = 1 - F_s(T) \quad (15)$$

The porosity function is equal to 1, fully porous media, for temperatures corresponding to the liquid phase ( $T > T_m + \varepsilon$ ) and is equals to 0 for completely solid phase ( $T < T_m - \varepsilon$ ). It will vary linearly between 0 to 1 for temperatures in the range of phase transition. A solidification source

term added to the Navier-Stokes equations is defined as a function of porosity as follows:

$$\mathbf{F}_s = -A\mathbf{u} \quad (16)$$

with  $A$  defined in the following manner:

$$A = \frac{C(1-\lambda)^2}{(\lambda^3 + q)} \quad (17)$$

In Equation 17,  $C$  and  $q$  are constants arbitrarily chosen to achieve the desired effect. This is,  $C$  is big enough and  $q$  is small enough to avoid division by zero. When the temperature of the liquid is above the liquidus,  $\lambda = 1$  and  $A = 0$ , thus no modification to the Navier-Stokes equations is made and the flow corresponds to that of a fully fluid phase. However, when the temperature is below the solidus,  $\lambda = 0$  and  $A$  takes a very high value ( $A=C/q$ ), dominating the transient, diffusive and convective terms of Equation 1, forcing the velocity to be zero, which is the expected solution for a solid in a close cavity. In the intermediate case when  $\lambda$  is between 0 and 1, the Navier-Stokes equations are similar to that of the flow in a porous media.

The complete expressions for the source terms that modify the Navier-Stokes equations are given by the following expressions for each component ( $r$  and  $z$ ):

$$F_r = \gamma\kappa \frac{\partial \phi}{\partial r} \delta(\phi) + F_{s,r} H(\phi) \quad (18)$$

$$F_z = \gamma\kappa \frac{\partial \phi}{\partial z} \delta(\phi) + \rho g_z + F_{s,z} H(\phi) \quad (19)$$

Note that the  $\mathbf{F}_s$  terms in Equations 18 and 19 are multiplied by the heavy side function  $H(\phi)$ , which defines the liquid and gas phases ( $H(\phi)=1$  for liquid). This is done to ensure that the solidification model will act on the liquid phase exclusively and will not affect the gas phase, regardless of the temperature of the gas.

Finally, to account for the latent heat of fusion, a modified heat capacity equation is used:

$$c_{pT} = c_p + c_{pH} \quad (20)$$

In Equation 20,  $c_p$  corresponds to the heat capacity at constant pressure related to the sensible heat in the usual way:

$$h = c_p \cdot T \quad (21)$$

The final term on Equation 20 is an additional term to account for the release of the latent heat of phase change upon solidification. It could be assumed that the latent heat ( $L$ ) is released in a linear fashion as the temperature decreases, thus the enthalpy of phase change as a function of temperature can be written as follows:

$$\Delta H(T) = \begin{cases} L, & T \geq (T_m + \varepsilon) \\ L(1 - F_s), & (T_m + \varepsilon) > T \geq (T_m - \varepsilon) \\ 0, & T < (T_m - \varepsilon) \end{cases} \quad (22)$$

Equation 22 is equivalent to integrate a function similar to the Dirac delta function over temperature around  $T_m$  times the latent heat of melting,  $L$ . In other words, the release of latent heat can be regarded as a sensible heat that is liberated only within the range of transition temperatures (from liquidus to solidus), therefore a heat capacity term can be defined:

$$c_{pH} = L \cdot \delta_{2\varepsilon}(T - T_m) \quad (23)$$

where  $\delta_{2\varepsilon}$  is a smooth Dirac delta function with nonzero values in a range of temperature equal to  $2\varepsilon$  and its integration over temperature is one.

The total heat content (sensible and latent) between temperatures  $T_1$  and  $T_2$  is then calculated integrating the modified heat capacity  $c_{pT}$ :

$$H = \int_{T_1}^{T_2} c_{pT} \cdot dT = \int_{T_1}^{T_2} c_p \cdot dT + L \cdot \int_{T_1}^{T_2} \delta_{2\varepsilon}(T) \cdot dT \quad (24)$$

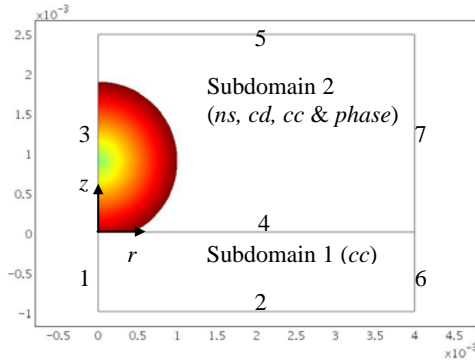
### 3. Numerical Model

The numerical model is implemented in COMSOL version 3.2b, using a 2D axial symmetric geometry (with radial and vertical

coordinates,  $r, z$ ). The case in study corresponds to a 2 mm liquid aluminium droplet spreading onto a stainless steel flat surface, the gas surrounding the droplet is air. Two cases are considered, an isothermal case for constant temperature of the system above the liquidus and a solidification case in which the substrate is at a temperature below the freezing point of the liquid, affecting the spreading of the liquid. In both cases, the initial velocity of the droplet is set to 0, therefore the flow is completely driven by gravitational effect (difference in density). The geometry and dimensions of the system are shown in figure 1. The initial level set function, defining the initial position of the droplet is given by the following expression:

$$\phi(t=0) = \sqrt{r^2 + (z - z_i)^2} - R_i \quad (25)$$

The above expression corresponds to the signed distance function for a circumference of radius  $R_i$  with center at  $(0, z_i)$ .



**Figure 1.** Geometry and initial condition for level set function, surface plot for negative values of  $\phi$  (liquid phase). Scale in m, boundary labels and active models for each subdomain are also shown.

For the implementation of the problem in COMSOL, four models are required, incompressible Navier-stokes ( $ns$ ), convection diffusion ( $cd$ ), convection conduction ( $cc$ ) and general PDE weak form, point ( $phase$ ). The variables for the four models are defined as  $u, v$  and  $p$  for  $ns$ ,  $phi$  for  $cd$ ,  $T$  for  $cc$  and  $lam$  for  $phase$ , using the default elements and order. Physics or subdomain settings are defined by the properties of materials and initial conditions for each model. In the following Tables the

properties, expressions and boundary settings considered for the model are given.

**Table 1. Properties of materials**

Property	Phase			
	gas	liquid	Solid	Substrate
$\rho, \text{kg/m}^3$	1.3	2382		7850
$\mu, \text{Pa s}$	$4.79 \times 10^{-5}$	$1.15 \times 10^{-3}$	--	--
$\gamma, \text{N/m}$	$0.793 \times 10^{-4}$		--	--
$k, \text{W/(m } ^\circ\text{C)}$	0.024	104		44.5
$T_m, ^\circ\text{C}$	--	660		--
$\varepsilon, ^\circ\text{C}$	--	5		--
$L, \text{J/kg}$	--	$3.97 \times 10^5$		--
$c_p, \text{J/(kgK)}$	1005	1090		475

**Table 2. Constants**

Name	Value	Units, Description
$z_i$	$0.9 \cdot R_i$	m, initial position of droplet
$R_i$	$1 \times 10^{-3}$	m, initial radius of droplet
$gz$	-9.8	$\text{m/s}^2$ , acceleration of gravity
$n$	$2.5 \times 10^{-5}$	Parameter for heavy side function
$n2$	$5 \times 10^4$	Parameter for smooth Dirac delta
$T_{wi}$	610	Initial temperature of substrate
$T_i$	670	Initial temperature of liquid
$C$	1600	Parameter for function A
$q$	$1 \times 10^{-3}$	Parameter for function A

**Table 3. Boundary settings**

Boundary	Model		
	$ns$	$cd$	$cc$
1*,3	Axial symmetry		
4	Slip	insulating	Not active
5,6*,7	No slip		insulating
2	Not active		Dirichlet: $T=T_{wi}$

\* only defined for  $cc$  model.

The phase re-injection model uses two subdomain integration coupling variables:  $intphi$  and  $intphi0$ , defined in cylindrical coordinates as  $Hphi * 2 * pi * r$  and  $Hphi0 * 2 * pi * r$ , respectively. In addition, a point integration coupling variable named  $lam$ , defined as the weak form variable  $lam$  is implemented to solve the difference between the volumes  $intphi$  and  $intphi0$ . the weak form-point model is active only in one vertex of the geometry and the weak field is defined as:  $lam_{test} * (intphi - intphi0)$ , ensuring the difference inside the brackets to be zero. To inject this difference back into the level set model ( $cd$ ), in the reaction or source term for  $phi$ , the expression  $lam * dphi$  is added to generate the amount of  $phi$  required to conserve the mass.

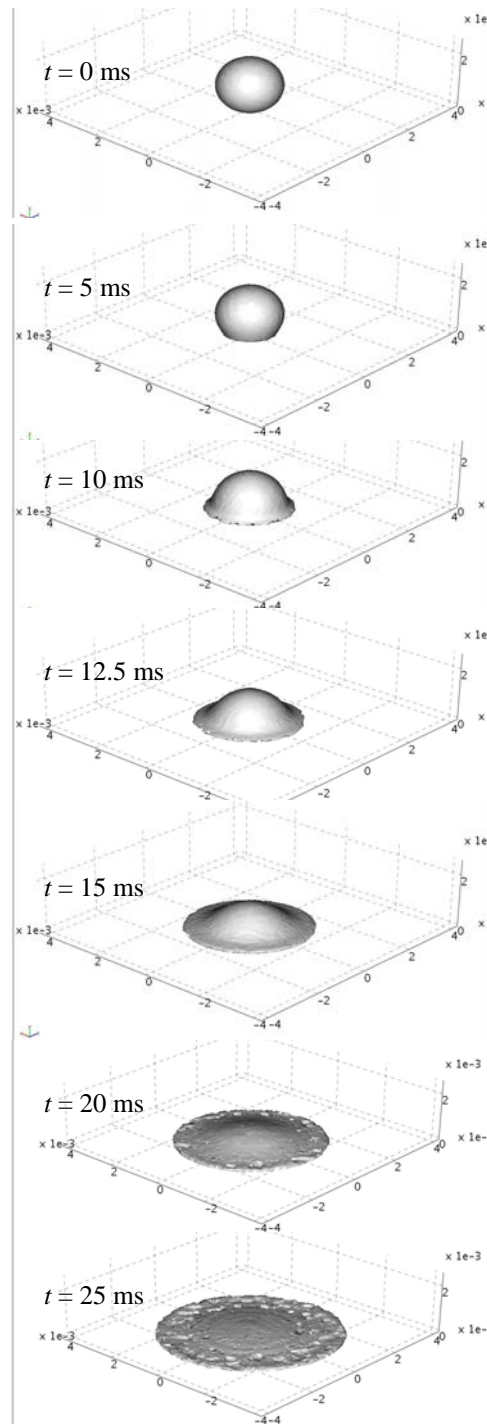
**Table 4. Scalar Expressions**

Name	Expression	Description
phi0	$\sqrt{r^2+(z-z_0)^2}-R_0$	Initial level set function
Hphi	$(1+\tanh(-\phi/n))/2$	Heavy side function
Hphi0	$(1+\tanh(-\phi_0/n))/2$	Initial HS
rho	$\rho_{\text{hog}}+(\rho_{\text{hol}}-\rho_{\text{hog}})*H\phi$	Density
eta	$\eta_{\text{tag}}+(\eta_{\text{tal}}-\eta_{\text{tag}})*H\phi$	Viscosity
k	$k_{\text{g}}+(k_{\text{l}}-k_{\text{g}})*H\phi$	Conductivity
dphi	$n^2/\sqrt{\pi}*\exp(-n^2*2*\phi^2)$	Smooth Delta dirac
kappa	$(\phi_{\text{irr}}*\phi_{\text{iz}}^2-2*\phi_{\text{ir}}*\phi_{\text{iz}}*\phi_{\text{irz}}+\phi_{\text{izz}}*\phi_{\text{ir}}^2)/(\phi_{\text{ir}}^2+\phi_{\text{iz}}^2+\epsilon)^{3/2}$	Curvature
Fr	$-\gamma*\kappa*d\phi*\phi_{\text{ir}}$	Surface tension force
Fz	$-\gamma*\kappa*d\phi*\phi_{\text{iz}}$	
lambd	$(T-T_{\text{m}}+e)/(2*e)*((T<=(T_{\text{m}}+e))*(T>=(T_{\text{m}}-e)))+(T>(T_{\text{m}}+e))$	Porosity Function
A	$-C*(1-\text{lambd})^2/(\text{lambd}^3+q)$	
Sx	$-A*u$	Solid source terms
Sy	$-A*v$	
cpH	$L*\text{fldc}2\text{hs}(T-T_{\text{m}},e)$	Latent heat capacity
cpT	$cp+cpH$	Total heat capacity
Ti0	$T_{\text{wi}}+(T_{\text{i}}-T_{\text{wi}})*(1+\tanh(z/n))/2$	Initial Temperature condition

Finally, an unstructured mesh with maximum element size of  $1 \times 10^{-4}$  (for subdomain 2) is used for the spatial discretization of the domain. The mesh is locally refined at the boundary 4, with maximum element size of  $1 \times 10^{-5}$  on this boundary, generating a mesh consisting of 13707 elements and 84462 DOF for the 4 models.

**4. Results**

The first result, corresponding to the isothermal case is shown in Figure 2, in which the liquid droplet spreads onto the surface without phase change. The problem is solved for a total simulation time of 25 ms with time outputs every 0.5 ms. In this case, only *ns*, *cd* and *phase* models are solved for a total of 56908 DOF



**Figure 2.** Computer generated images of aluminum droplet before and after impact. Isothermal axis-symmetric case. Initial diameter 2 mm.

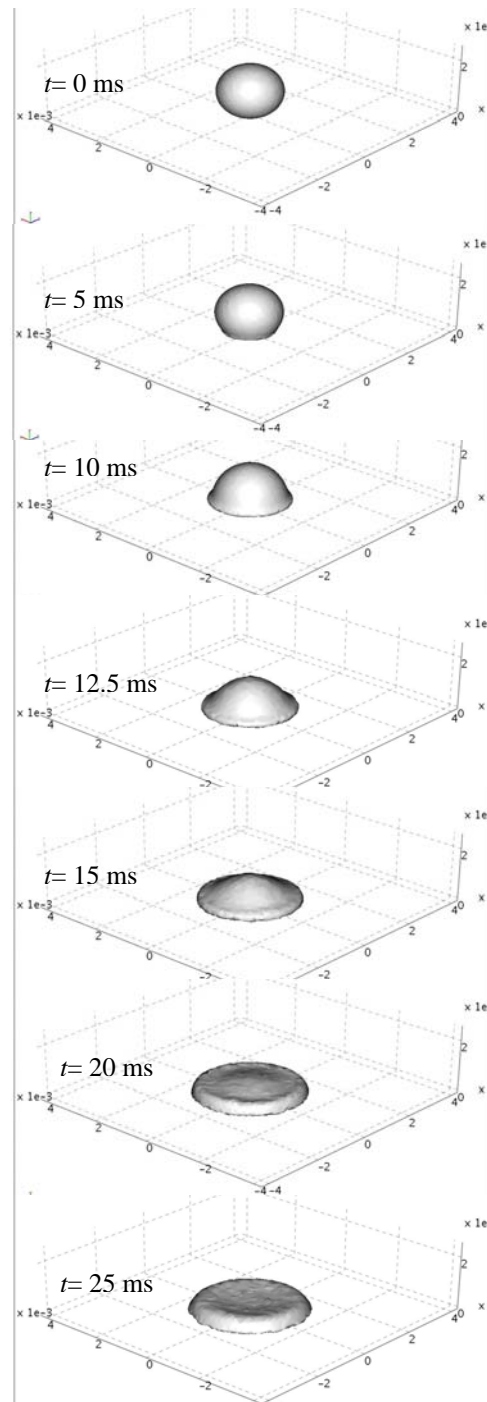
It is observed that the droplet spreads almost completely after the end of the simulation and very little effect of surface tension force is

observed. This is due to the low value used for  $\gamma$ , which is 4 orders of magnitude lower than the real surface tension of aluminum. It was not possible, for the mesh resolution, the accuracy of the elements and the initial condition used in this case, to obtain a reasonable solution for higher values of  $\gamma$ .

The second case adds solidification phenomenon to the previous model. The liquid and substrate initial temperatures are given in Table 2, the initial condition for temperature is given in Table 4. The four models requires to solve for 84462 DOF for the mesh considered. The result is shown in Figure 3 for different times of the solution. The effect of solidification during spreading of the droplet is clear, comparing with Figure 2.

For the problem in study, the main limitation corresponds to the surface tension treatment. To be able to accurately calculate the value of this force, according to Equation 8, it is necessary to evaluate both the curvature and the normal vector at the interface (Eq. 10 and 11). These two expressions depend on the first and second derivatives of  $\phi$ . If  $\phi$  is a distance function, as in the initial condition given in Eq. 25, then  $|\nabla \phi| = 1$  and the normal vector to the interface is simply  $d\phi/dr$  and  $d\phi/dz$  as in Equations 18 and 19. However, as the level set function is advected (with or without phase re-injection) by the fluid in a close cavity, this is not true anymore and oscillating errors are added to the solution. Therefore, evaluation of the unit normal vector is affected. Errors in the curvature calculation are even more pronounced, generating spurious flows to the solution.

Another aspect to consider is the contact angle between the liquid and the substrate. In the present model, this is not considered at all, as a difference with the models of droplet impact using the VOF method [1-3] in which the contact angle is imposed as a boundary condition. This is difficult to do with the level set method, since it would be necessary to modify  $\phi$  at every time step to meet this requirement.



**Figure 3.** Computer generated images of aluminum droplet before and after impact. Substrate at 610°C axial-symmetric case. Initial diameter 2 mm.

## 5. Conclusions

A method for the free surface flow with high density and viscosity ratios and solidification was successfully implemented in COMSOL version 3.2b.

The method, still faces limitations on the surface tension treatment, allowing the use of low surface tension values in the model.

In addition, implementation of boundary conditions at the interface, such as the contact angle between the liquid and the substrate, still requires further development.

## 6. References

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## 7. Nomenclature.

### Vector quantities:

$\mathbf{u}$  : velocity field, m/s

$\mathbf{F}$  : Body (volumetric) force for the Navier-Stokes equations, N/m<sup>3</sup>.

$\mathbf{n}$  : unit normal at liquid-gas interface

### Scalar quantities:

$p$  : pressure field, N/m<sup>2</sup>.

$\phi$  : level set function.

$\rho$  : density, Kg/m<sup>3</sup>

$\mu$  : viscosity, Pa s.

$k$  : thermal conductivity, W/(m K)

$H(\phi)$  : Heavy side function.

$n$  : parameter for heavy side function

$\delta(\phi)$  : smooth Dirac delta function

$n_2$  : parameter for smooth Dirac delta function

$\gamma$  : surface tension, N/m.

$\kappa$  : curvature, 1/m.

$\varphi$  : source term for phase reinjection, 1/m.

$t$  : time, s.

$F$  : Fraction.

$T$  : temperature, K.

$\varepsilon$  : half range of mushy region, K.

$h$  : sensible heat, J/kg.

$c$  : specific heat capacity, J/(kg K).

$L$  : latent heat of melting, J/kg.

$\lambda$  : porosity function.

$R$  : Droplet radius, m.

$g$  : gravity acceleration, m/s<sup>2</sup>.

### Subscripts:

$l$  : liquid phase

$g$  : gas phase

$s$  : solid phase or due to solidification

$m$  : average melting point.

$p$  : at constant pressure.

$T$  : total

$H$  : corresponding to latent heat of melting

$ST$  : surface Tension

$i$  : initial condition.

$w$  : at the wall of the substrate.