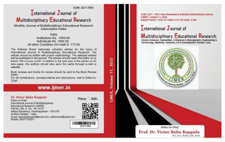

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MULTI-PHASE CORE-SHELL MOLTEN METAL DROP OSCILLATIONS AND ITS NUMERICAL SIMULATION

Saras Salil, Mohd Dawood and Dr. Prabhat Kumar Sinha
Mechanical Department
Sam Higginbottom University of Agriculture Technology and Science
Prayagraj, Uttar Pradesh, India

ABSTRACT

The surface tension of liquid metals was necessary and intriguing framework which affects many mechanical works such as forging, casting, welding and melt spinning processes. Conventional methods for measuring surface tension are crucial to make use for molten metals above 1000 K temperatures. Container less methods are can be used to measure surface tension of molten metals above 1000 K. One such method is Oscillating drop method where a hang droplet is allowed to go through damped oscillations. Using the Rayleigh's theory for the oscillation of force-free infused spherical droplets, surface tension and viscosity of the sample would be able to calculate from oscillation natural frequency and damping respectively.

In the given research work, a numerical model was developed in ANSYS mellifluous for simulation of the oscillations of molten metal droplet multi-phase modelling has done through the volume of fluid approach. The effect of numerical schemes, mesh size, and boundary conditions on the oscillation natural frequency and the surface tension of the liquid were studied. The single-phase model anticipates the surface tension of zirconium within a range of 13% when correlated to the experiment data. The validated single-phase model was elongated to predict the interfacial tension of a core-shell structure drop. The effect of the core and shell orientation was studied at the time of initialization flow. The numerical model that was developed figure out the interfacial tension between copper and cobalt within the range of 6.5% when in comparison to the experimental data. The multiphase model failed to give any conclusive data for interfacial tension enclosed by molten iron and slag.

Keywords: Oscillating Drop Method, Multiphase Model, ANSYS.

INTRODUCTION

The surface tension of liquid metals is an important and scientifically interesting parameter which affects many metallurgical processes such as casting, welding and melt spinning. A first principles calculation of the surface tension would require an atomic scale of the liquid state, including density and free-energy profiles at the surface. Conventionally, surface tension measurement for liquids is done using techniques discussed below which measure the geometry of the drop resting on the substrate or measure the pressure required to force out the droplet from end of a capillary. These experimental data are easy to obtain for liquids at temperatures up to 1000 K. However, for metals with high melting point above 1000 K, measurement becomes difficult due to error in measurement of drop profile, error arising from density determination and physio-chemical effects. Therefore, methods need to be explored where these limitations can be compensated and accurate measurements can be achieved. Containerless processing methods such as oscillating drop method (ODM) is one such method. In this method, a sample is levitated using an electrostatic or electromagnetic levitator and allowed to oscillate by providing external excitation, which is removed and sample is allowed to damp out naturally. The oscillating droplet area at successive small-time intervals is analyzed and mode 2 natural frequency of the oscillations is obtained. The frequency is used to calculate surface tension. The oscillating drop method is extended to multi-phase core-shell molten metal droplets to study interfacial tension between liquids in core and shell. Systems consisting of molten iron and slag is particularly interesting to study due to its application in continuous casting process. During continuous casting process, molten iron and slag layer interact at the inlet nozzle. The flow of iron carries slag into molds and affects the quality of the casting. The knowledge of this interfacial tension between molten iron and slag will help predict the behavior at the interaction of iron and slag and help reduce impurities in the casting. The literature shows that surface tension measurement is critical aspect of processing of materials. The available methods are difficult to use with molten metals at high temperatures in the range of 1500 K and above. ODM was developed and used with levitation to determine the surface tension in a contactless manner thus eliminating difficulties encountered in conventional methods. However, ODM needs extensive setup and controlled environment to successfully process the materials. Computational fluid dynamics (CFD) is a great tool to analyze such problems. A numerical model allows to create conditions close to experiment setup and replicate the physics by solving governing differential equations. This allows to study the effect of various parameters on the outcome of the experiment without the cost of scaled physical setup. CFD studies also help in investigating the problems in extreme environments and validity of assumptions to be used in the scaled model experiments.

The problem of oscillating, levitated drop is controlled by many parameters. For example, the heating of levitated drop is done using a heat source such as laser or magnetic induced electricity. A temperature gradient on the surface of the droplet is developed if the heating is uneven. Secondly, since the sample is levitated and without support from a physical fixture, a small imbalance in force can result in the rotation of the sample. In the problem of oscillating compound drop formed by core-shell structure, CFD provides insights into the processes happening inside the droplet. The opacity of the molten metal does not allow this in the real-life experiment. Simulating the problem using CFD can help design suitable experiments. Insights can be obtained into various limiting factors of the scaled model experiments.

The aim of this study is to simulate the core-shell structured droplet formed by molten iron core and molten slag as shell. The numerical model is validated using the experiment data available in the literature to gain confidence in the numerical schemes used for numerical simulation. Existing research by Egry et al. and Zhao et al. provides data from levitation experiments performed on Copper-Cobalt and Zirconium samples respectively. A numerical model is developed to simulate these experiments using Fluent and validated using the experimental data. The validated model is then used to predict the interfacial tension between molten iron and molten slag. In recent times, experiments were performed using ground based electrostatic levitation at Marshall space flight center (MSFC-ESL) by Zhao et al. The research was aimed at finding the thermophysical properties of zirconium and effect of oxygen on the surface tension of zirconium. The experiment provides data for the surface tension of pure zirconium obtained by containerless processing which is used to validate the numerical model developed in this study.

Masahito Watanabe performed ODM analysis on compound drops where Ag melts formed the core and B2O3 melts formed the shell. The droplets were electromagnetically levitated using a ground-based levitator. However, when the sample was melted, the core-shell structure was lost and a Janus droplet was formed. It was concluded that microgravity conditions are required to maintain the core-shell structure. A numerical simulation was also performed on molten iron and slag system to predict the interfacial tension.

Containerless Method in Materials Science

High temperature analysis and lab processing of metals and alloys is difficult. There are numerous problems in handling of the sample which arise due to contamination of the sample by chemical reaction with the atmosphere or containers, constant heat transfer due to temperature gradient, evaporation of the sample and mass reduction and consistently maintaining the sample at high temperature etc. To avoid these problems, container less methods were developed in which the sample does not get in contact with a container. For container less processing methods, the sample is levitated. This can be achieved by various methods such as electrostatic, electromagnetic, acoustic, aerodynamic or optical forces. The literature reviewed for this study uses electrostatic or electromagnetic levitation. A brief account of these methods is given.

Survey of Levitation Methods

2.2.1 Electrostatic Levitation

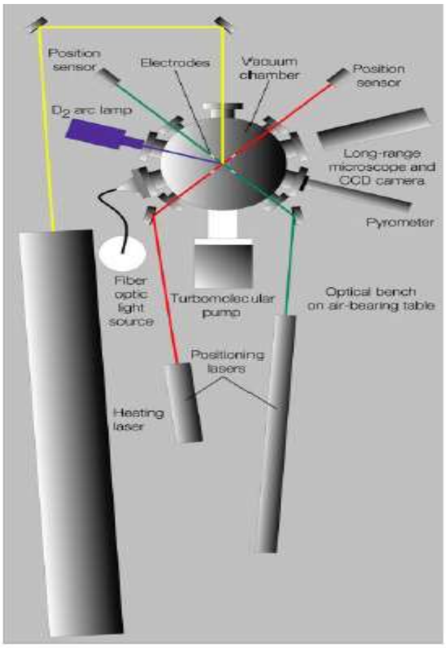
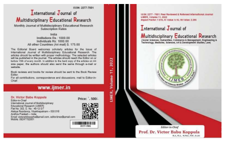


Figure 2.1: Schematic of Electrostatic levitation



Electrostatic levitation (ESL) levitates small samples of approximately 2~3 mm (~40 mg) diameter using a large alternating electric field. The sample is charged by induction. During processing, concept of photoelectric effect is applied using a UV source to refresh the charge on the sample. Therefore, ESL can be used to levitate both conducting and non-conducting samples. Figure 2.1 shows the schematic of the ESL system. The position of the sample is controlled by active feedback control loop, input to which is provided by two dual-axis position detectors. The sample is heated to desired temperature using CO2, Nd:YAG, or diode lasers. Optical pyrometry provides temperature.

measurement required during the containerless processing. Depending on the type of study, the sample is recorded and observed in a video or other suitable means.

Electromagnetic Levitation

The electromagnetic levitation (EML) uses the concept of Lorentz force to levitate and position a sample for containerless processing. An alternating electromagnetic field is used to induce eddy currents in the sample. EML can be used only with conducting samples. Lorentz force is produced due to the interaction of induced eddy currents with the applied magnetic field. By manipulating the applied magnetic field, a non-zero volume integral of Lorentz force is achieved. This net force is used to balance the sample against the gravitational forces. EML setup is used in ground-based experiments or microgravity experiments. The ground based EML, vertical magnetic gradient is applied to balance gravity; where as in microgravity conditions, oscillating electromagnetic field is applied.

Oscillating Drop Method for Surface Tension and Viscosity

The oscillating drop method (ODM) was developed to determine the surface tension and viscosity of the liquid. The basis of ODM is the Rayleigh's theory for surface tension driven oscillations of spherical, force-free and inviscid liquid droplet [4]. The Rayleigh's theory assumes very small amplitude for the oscillation of the droplet. The fluid flow is assumed to be Newtonian, inviscid flow. The ODM needs a form of levitation, electromagnetic or electrostatic to levitate the sample and induce oscillations. The sample oscillations are allowed to damp out naturally. The frequency of oscillation, ω is related to surface tension, σ by the relation given by Lord Rayleigh

$$\omega_\circ = \sqrt{\frac{l(l-1)(l+2)\sigma_\circ}{\rho_\circ R_\circ^{3}}} \qquad (2.1)$$

Where, $\omega0$ is the oscillating frequency for the mode l, for a droplet of surface tension $\sigma0$, density $\rho0$, and radius $R0$.
For a perfectly spherical, non-rotating drop, only mode 2 oscillations are observed and Equation 2.1 reduces to

$$\omega_\circ^{2} = \frac{8\sigma_\circ}{\rho_\circ R_\circ^{3}} \qquad (2.2)$$

The viscosity, η of the droplet fluid is related to damping constant, τ of the oscillations by the relation given by H. Lamb.

$$\tau = \frac{(l-1)(2l+1)\eta}{\rho_\circ R_\circ^{2}} \qquad (2.3)$$

where, η is the molecular viscosity if the flow is laminar, or effective viscosity for turbulent or transitional flow.
The Rayleigh theory assumes inviscid fluid flow and the equilibrium is governed by surface tension forces. When viscosity is included, the equilibrium state are then governed by the linearized equation of motion. The nonlinear analysis by Suryanarayana et al. shows that the surface oscillation depends on parameter $\alpha2$

$$\alpha^{2} = \frac{\omega_\circ \rho_\circ R_\circ^{2}}{\eta} = \sqrt{\frac{l(l-1)(l+2)\sigma_\circ \rho_\circ R_\circ}{\eta}} \qquad (2.4)$$

where $\omega0$ is the Rayleigh frequency defined in Equation 2.1. For mode 2 oscillations, the critical value of $\alpha2$ is 3.69. For the droplet to undergo damped oscillations, the value of $\alpha2$ must be greater than 3.69. For value of $\alpha2$ below 3.69, the droplet will exhibit non-periodic oscillations. Suryanarayana et al. also tabulated the difference in Rayleigh and observed frequency due to viscous effects. For the value of $\alpha2$ greater than 59, the deviation of observed frequency from the Rayleigh frequency is 1%. Therefore, viscous effects can be neglected and Equation 2.1 can be used to compute surface tension from the oscillation frequency of the droplet.

Oscillating Drop Method for Interfacial Tension

Before extending the containerless, the oscillating drop method to determine surface tension and interfacial tension, it is necessary to know the mechanism that forms core and shell structure between two immiscible liquids. The mechanism of core-shell formation is explained in Ref. [3]. Alloys with metastable miscibility gap can be solidified after undercooling to form a core-shell structured droplet. Below the binodal temperatures, the homogenous melt of alloy undergoes demixing forming small droplets of liquid L1 in the matrix of other liquid L2. The small droplets and matrix must follow compositions per the phase diagrams. The liquids L1 and L2 are not pure; L1 and L2 are rich in components 1 and 2 of the alloy respectively. Initial nucleation kinetics gives rise to large number of small droplets which is energetically non-desirable due to large interface area generated. A diffusive mechanism of Ostwald ripening leads to coarsening of the droplets at expense of small droplets thereby changing the structure and leading to two separated phases. Energy stability of the system dictates the liquid with lower surface tension to form the shell which encapsulates the core formed by liquid with higher surface tension.

Another aspect of interfacial tension measurement experiment using container-less and electromagnetic levitation is to maintain the core-shell structure achieved by undercooling and phase separation. However, the large gradient in magnetic field to balance the droplet against the gravitational forces creates stirring effects, destroying the separated core-shell structure. Since core and shell liquids have different densities, the gravitational force acting on them is not equal. This imbalance can also destroy the core-shell structure. The experiment must be conducted by maintaining the separated core-shell structure and this is possible only if gravitational forces acting on the sample are removed by conducting the experiment in microgravity conditions. Various means used to achieve microgravity conditions are parabolic flights, sounding rockets or in recent times performing experiment in international space station (ISS).

Rayleigh's formula in Equation 2.1 gives relation of surface oscillation to surface tension. Since there are two interfaces, one between liquid L1 and L2, other between liquid L2 and the vacuum, the system will oscillate at two fundamental frequencies: $\sigma 0$ attributed to surface tension and $\sigma 12$ attributed to interfacial tension. Work of I. Egry shows ODM can be extended to compound drop formed by immiscible liquids [5]. Based on the theory of Saffren et al. [12], analysis for force-free, concentric spherical drops was worked out. The same nomenclature is used here.

$$\omega^2{}_{\mp} = k \mp \frac{W}{J} \qquad (2.5)$$

where, K and J are dimensionless, while W is frequency squared. W/J is given by:

$$\frac{W}{J} = \frac{\omega_\circ^2 \ \tau^8}{\sigma} \ \frac{1}{(1+\Delta\rho_i)\tau^{10}+\frac{2}{3}\Delta\rho_i} \qquad (2.6)$$

where, $\omega 0$ is the Rayleigh unperturbed frequency (Equation 1) of a simple single phase drop with density $\rho 0$, radius $R0$ and surface tension $\sigma 0$. Figure 2.1 defines additional symbols along with following equations.

$$\tau = \sqrt{\frac{R_\circ}{R_i}} \quad (2.7)$$

is the square root of the ratio between outer and inner radius,

$$\sigma = \sqrt{\frac{\sigma_\circ}{\sigma_{12}}} \ (2.8)$$

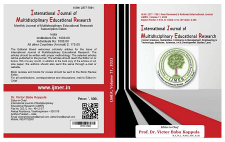
is the square root of the ratio of the surface tension and the interfacial tension, and

$$\Delta\rho_i = \frac{3}{5}\frac{\rho_i - \rho_\circ}{\rho_\circ} (2.9)$$

is the weighted relative density difference between liquids L1 and liquid L2. Expression for K is given by:

$$K\mp = \frac{1}{2}\left(\frac{\sigma m_i}{\tau^3} + \frac{m_\circ \tau^3}{\sigma}\right) \pm \sqrt{\frac{1}{4}\left(\frac{\sigma m_i}{\tau^3} - \frac{m_\circ \tau^3}{\sigma}\right)^2} \ (2.10)$$

The additional symbols $m0$ and mi are given by: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$m_{\circ} = \frac{3}{5}\tau^{5} + \frac{2}{5}\tau^{-5}$ (2.11)
$m_{i} = (1 + \Delta\rho_{i})\tau^{5} - \Delta\rho_{i}\tau^{-5}$ (2.12)

for large σ and small $\Delta\rho i$, approximate equations can be derived for the two frequencies $\omega+$ and $\omega-$:

$$\omega_{+}^{2} = \omega_{\circ}^{2}\left(1 + \frac{1}{\sigma^{2}\tau^{4}}\right) \quad (2.13)$$

$$\omega_{-}^{2} = \omega_{\circ}^{2}\frac{\tau^{6}}{\sigma^{2}}\left(1 - \frac{5}{3}\tau^{-10}\right) \quad (2.14)$$

EXPERIMENTS AND MODEL PARAMETERS

ODM Experiment to Determine Surface Tension of Zirconium

This section describes the experimental setup and results from containerless processing of zirconium by Zhao et al. [6]. A pure zirconium sample was tested using an electrostatic levitator at NASA MSFC. The levitated sample was excited by an electrostatic field alternating near the natural frequency of the sample. A function generator was used to regulate the excitation frequency. After the required excitation was achieved, the function generator was turned off and the sample was allowed to damp freely. The experiment was recorded by high-speed video camera at 1000 frames per second.

The experiment parameters are described here. A vacuum of 10−7 Torr was maintained in the equipment chamber. This avoided contamination of sample by elements like oxygen and sulfur. The zirconium sample of mass 43.288 mg was used. Examination of sample after experiment reported a mass loss of 0.043% due to evaporation. The mass loss is neglected in the simulation model. The density of sample was calculated to be 5891.7 Kg/m3. The sample went through 24 oscillation cycles at temperatures between 1980 0C and 1600 0C. The simulations were recorded using LED back light, high speed camera. The video processing was done using code generated in LabVIEW. Surface tension and viscosity data was calculated using relations in section 2.3.

The Fluent model is developed using the density and sample mass used in the experiments. The data from a single oscillation cycle at 1974 0C is used. The frequency of surface oscillations was found to be 168.88 hz and surface tension and viscosity for this cycle was calculated to be 1.4515 N/m and 0.0081499 kg/m-s respectively.

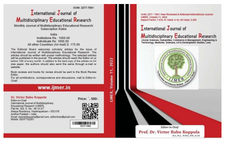
ODM Experiment to Determine Interfacial Tension of Cu-Co Compound Drop

Egry et al. conducted experiments to find interfacial tension between Cu-Co using TEXUS-EML during TEXUS-44 campaign. This section provides details of experiment setup and results from experiments described in Ref. [5]. To accommodate the experiments within 160 seconds of microgravity time available for this experiment, the sample was prepared ex situ allowing for phase separation of Cu-Co and formation of core-shell structure. The composition of the sample was Cu75Co25.

The physical parameters of the sample must be known for analysis of oscillation spectrum. Theoretical calculations for oscillation of phase-separated drop were done by inputting the physical parameters in Equations 2.4 to 2.14. Sample examination provided total mass of sample to be 1.31 g and mass of individual phases, copper and cobalt to be 1.0056 g and 0.3064 g respectively. The composition of the alloy was calculated to be 76.65 wt% copper and 23.35 wt% cobalt. The radius of the droplet was estimated from the total mass, M which is 1.31 g.

$$R_{\mathrm{eff}} = \sqrt[3]{\frac{3M}{4\pi\rho}} \quad (2.15)$$

The density, ρ calculations were done using work of Satio et al. [17], [18] and $Reff$ was calculated as 3.475 mm. This is taken as radius of shell $R0$. By EDX analysis of identical generated samples, composition of shell liquid, L2 and composition of core liquid, L1 was found to be 90 wt% copper, 10 wt% cobalt and 16 wt% copper, 84 wt% cobalt respectively. Using this data, the radius of core, Ri is calculated to be 2.149 mm. The surface tension estimates are calculated using models from Ref. [19]. Inserting the physical values in Equation 2.5, theoretical oscillation frequency obtained for a phase separated drop is 28.75 Hz and 17.26 Hz.



NUMERICAL MODEL AND SOLVER

The governing equations of the fluid flow and the numerical model used to simulate the oscillations of the droplet. The numerical model is divided into three main sections i.e., Pre-processing, Solving, and Post-processing. These terms are associated with any numerical model used to simulate real life fluid flow problem by method of computational fluid dynamics (CFD). The pre-processing includes making suitable assumptions to simplify the numerical model, setting up the geometry, generating mesh, and applying the suitable boundary conditions. The solver includes details of time and spatial discretization, initializing the problem, and selection of discretization schemes. The post-processing includes interpretation of results from the simulation and documenting the results in form of reports, graphs and contours. The chapter provides insights into methods available along with their advantages and limitations. Subsequently, numerical schemes in this study are described.

Fluid Flow and Governing Equations

The fluid flow is governed by non-linear set of partial differential equation based on the principle of conservation of mass and momentum. Collectively they are denoted as Navier-Stokes equations. For an incompressible flow, the Navier-Stokes equations are written as, $\rho\left[\frac{\partial u}{\partial t}+(u.\nabla)u\right]=-\nabla p+\mu\nabla^2 u+\rho g$ (4.1)

and, the continuity equation,

$$\nabla.u = 0 \quad (4.2)$$

where p is the static pressure, μ is the molecular viscosity, u is the fluid velocity, ρ is the density of fluid, and g is the gravitational force acting on the fluid. The Navier-Stokes equations are non-linear coupled equations and almost impossible to solve for complex problems.

Pre-Processing

Assumptions

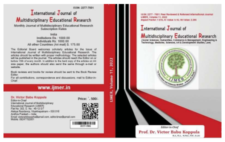
Before CFD study, suitable assumptions were made to simplify the problem. Reasonable assumptions were made such that it simplified the modeling of problem.

The temperature was assumed to be same everywhere in the domain. The temperature gradient on the droplet can cause natural convections and Marangoni convection in addition to the primary fluid flow. In the EML experiments, the primary driving force for the flow is magnetic field. The importance of Marangoni and natural convection was found to be orders of magnitude smaller compared to electromagnetically driven flow [20]. Therefore, its effect on surface tension calculations is neglected. In the ESL experiments, the primary driving force for the fluid flow is Marangoni convection. The natural convection is found to be very small compared to Marangoni flow in ESL experiment of FeCrNi drops [20]. Therefore, from the literature available, it is safe to assume same temperature throughout the domain.

The rotation of the droplet was neglected. The sample is prone to rotation in absence of any fixture support. Even a small imbalance in the forces acting on the levitated sample can cause rotation of the sample. If the sample rotation is below 30 Hz, its effect on surface tension and viscosity can be neglected. However, experiments must be controlled such that the sample does not rotate near its natural frequency which can cause instability due to resonance. If the rotation is considered in the model, a centripetal force term is added to the Navier-Stokes equation. The droplet is assumed to be spherical always. The levitated droplet experiences gravitational and electromagnetic force at the same time. The net resultant force integrated over the volume is equal to zero. However, there resultant residual forces exist on the surface which deform the droplet. This aspherical shape causes Rayleigh frequency to split into three frequencies. The model is developed such that there are no external forces acting on the drop. Therefore, spherical droplet assumption is valid.

Model Material Properties

A numerical model was developed to simulate the oscillation of pure zirconium (Zr) droplet using the ANSYS Fluent package. The values of various material properties used are given in these sections. The diameter of zirconium droplet modeled was 2.412 mm. The materials used in the model were zirconium for the droplet and helium for the region surrounding the droplet. The values of material properties used for zirconium and helium are given in Table 4.1. The mass loss of 0.043% due to evaporation was neglected. Density of the sample used was 5891.73 kg/m3 [6] and working pressure was set to 10−7 Torr. The viscosity of zirconium was calculated using



empirical model in literature and found to be 4.194 mPa-s [8]. The material properties are summarized in Table 4.1.

Table 4.1: Physical model parameters for Zirconium model

Diameter	2.411E-03 m	
Material properties	Helium	Density: 0.1625 kg/m^3
		Viscosity: 1.99E-05 kg/m-s
	Zirconium	Density: 5891.7 kg/m^3
		Viscosity: 0.004194 kg/m-s
Surface tension modeling	1.451 N/m	

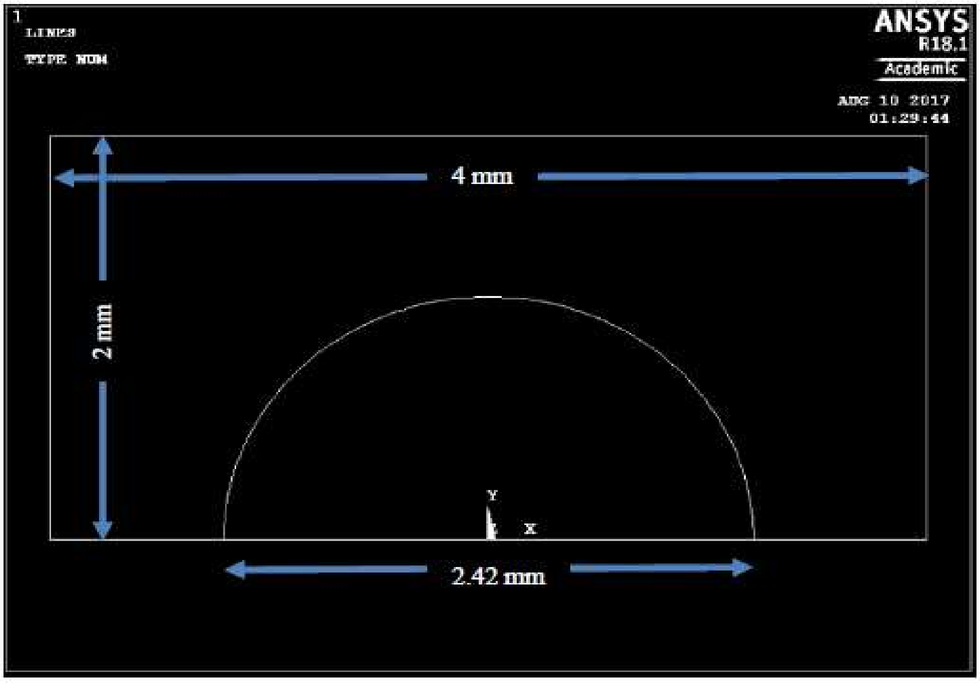
Table 4.1: Physical model parameters for Copper-Cobalt compound drop model

Diameter	Copper (shell)	3.475E-03 m
	Cobalt (core)	2.149E-3 m
Material properties	Copper	Density: 7507.4 kg/kg/m^3
		Viscosity: 0.002012 kg/m-s
	Cobalt	Density: 7618.1 kg/kg/m^3
		Viscosity: 0.004074 kg/m-s
Surface tension modeling	Copper-air	0.21525 N/m
	Copper-cobalt	1.1547 N/m

Computational Domain

The geometries are generated in ANSYS Parametric Design Language (APDL). The computational domain for the zirconium droplet oscillations is shown in Figure 4.1(a). 23

The geometry was set in 2D plane. The problem domain was set in a rectangle of 4 mm width and 2 mm height. The zirconium droplet was generated by a semicircle whose center coincided with the midpoint of lower edge and the horizontal diameter coincided with the bottom edge of rectangular domain. The diameter of the semicircle was 2.4119 mm.



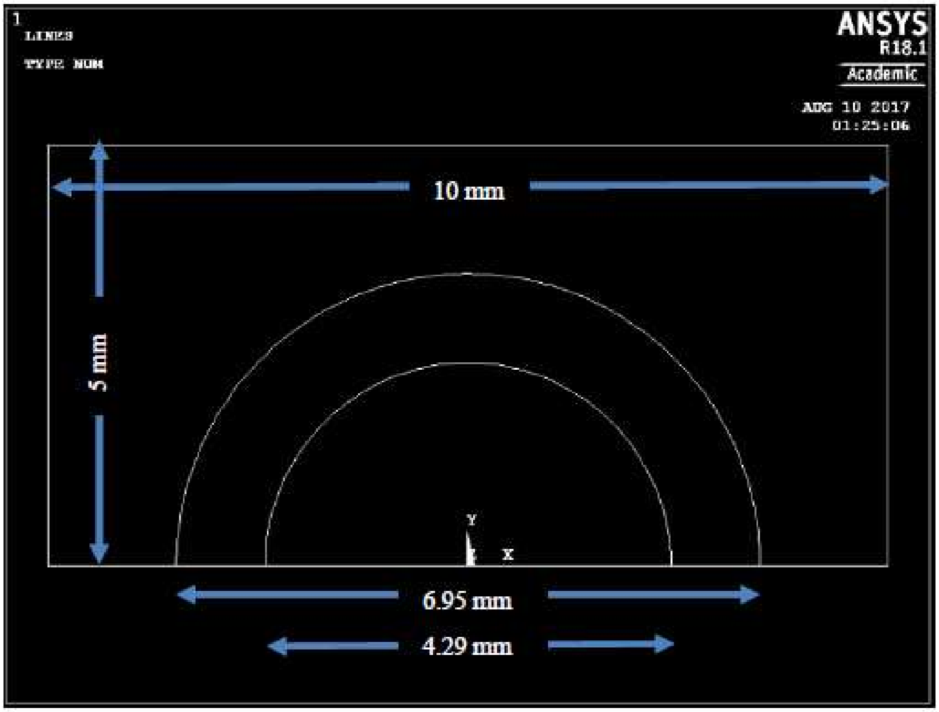


Figure 4.1: Computational domain and (a) Geometry of Zirconium drop (b) Geometry of Copper-Cobalt compound drop

In this study, Volume of fluid modeling was used. VOF modeling allowed to track the interface which was very important in our study. The post processing technique for the same is explained in Section 4.4**.** While implementing the VOF scheme, both explicit and implicit volume fraction formulation were used. Theoretically, explicit formulation is known to exhibit better accuracy compared to implicit formulation. However, explicit formulation is bounded by the value of time step. The time step must satisfy the Courant-Friedrichs-Lewy (CFL) condition. For this study a velocity based CFL condition is used. The CFL condition provides acceptable time steps which are calculated using volume of fluid vf, mesh size Δx, and courant number C. The Courant number was set between 0.1 and 0.25 for various cases. The acceptable time step value is given by,

$$\Delta t = \frac{C\Delta x}{v_f} (4.3)$$

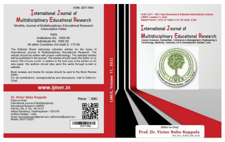
Time Discretization

In an unsteady or transient problem, the plow properties such as pressure and velocity change in the space domain and time domain. The rate at which properties change with respect to time is calculated at small, successive time steps. The flow properties at later time steps are approximated by first order implicit scheme. The information about flow properties at times other than the time steps is not available. Depending on the case setup, time step values between $1x10-5$ s and $1x10-7$ s were used in this study. These time steps were chosen by trial-and-error method to satisfy the CFL condition. A generic information for evolution of property ϕ over time is given by

$$\frac{\partial \emptyset}{\partial t} = F(\emptyset) (4.4)$$

In first order, implicit formulation, the (ϕ) is calculated at future time step,

$$\frac{\emptyset^{n+1} - \emptyset^{n}}{\Delta t} = F(\emptyset^{n+1}) (4.5)$$



The Equation 4.4 is solved iteratively before moving to the next time step. The maximum number of iterations allowed to reach convergence was set to 100. Again, by trial-and-error method it was found that 100 iterations per time step were enough to converge the residuals at each time step.

Post-Processing

A two-step post-processing approach was used. An animation was created which showed the amount of volume fraction of all phases in all cells of the domain at every time step. The generated video was analyzed using code developed in MATLAB. A loop was run where each frame of the animation sequence was analyzed at a time. In experiments, the projected area of the droplet is used to plot the surface oscillations. In the animation sequence, each phase is represented by a color. When analyzing each frame, the number of pixels that lie in each phase is counted. The number of pixels of required phase at all time steps was stored in a variable which is a 1-dimensional array in MATLAB. The number of pixels in a phase and time are plotted on Y and X axis respectively and the oscillation spectrum of the droplet was generated. Further to extract the frequency of oscillation, a fast Fourier transform was performed on the data. In the frequency spectrum, a sharp peak was observed at the frequency of oscillation. The resolution of frequency axis is controlled by the sampling frequency and the number of data points used. For this study, the sampling frequency and number of data points were such that a very coarse frequency spectrum was achieved, which induced an approximation error of 5% to 30% in the predicted oscillation frequency.

In this study, experimental data are used to validate our numerical model. Therefore, the desired value of frequency is known. If the approximate value from the animation sequence was close to data experiment data, a second analysis was performed. The LabVIEW module was used for the area processing of the droplet. A. histo file was generated which contained the histogram of each frame from the animation sequence. This. histo file was then provided as an input to LabVIEW module. The LabVIEW module 34 filtered the data and a fit to the data was generated using 'Levenberg-Marquardt' method. This method provided the frequency of oscillation accurate to 1/100 Hz. The fit also provided the damping constant which was used to calculate the viscosity of the droplet fluid.

RESULTS

Zirconium Single Phase model

Modeling for Surface tension of Zirconium

A general strategy used in this study is explained. For each case, the simulation was run for a small flow time of 0.0173 s, enough to capture approximately 3 oscillation cycles. The post processing of the animation sequence was done using the method explained in Section 4.4 and oscillation spectrum was generated. A fast Fourier transform operation was performed on the oscillation data and an approximate oscillation frequency was obtained. This frequency spectrum was of very coarse resolution approximately 56 Hz. The approximate frequency, oscillation spectrum, and the animation allows to make decisions on the feasibility and physics of the model. If the preliminary results showed that the model followed the physics and the numerical model predicted the oscillation frequency correctly, the simulation was run for a larger flow time such that steady state was reached.

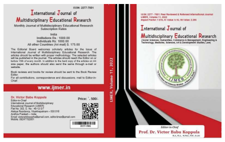
Cases were simulated with an unstructured and a Cartesian mesh (Ref. Section 4.2.4 for grid terminology) with an element size 25 µm. The element size is approximately 2% of the radius of the droplet. In order to initiate the oscillations, the spherical droplet was given a perturbation. This was done by giving an ellipticity to the droplet i.e. squeezing it from one direction. This configuration where the droplet diameter between the poles is greater than the droplet diameter at equator is called as prolate. The Equations 5.1 and 5.2 were used to calculate the dimensions of droplet's major and minor axis after accounting for the ellipticity. The equations are formulated such that the total volume of the droplet remains constant.

Let,

$$\mathrm{e} = \frac{\mathrm{a}-\mathrm{b}}{\mathrm{a}}, \therefore \mathrm{b} = (1-\mathrm{e})\mathrm{a} \quad (5.1)$$

For volume to remain equal,

$$\frac{4\pi}{3}\mathrm{r}^3 = \frac{4\pi}{3}\mathrm{a}^2\mathrm{b}, \therefore \mathrm{a} = \frac{\mathrm{r}}{\sqrt[3]{(1-\mathrm{e})}} \quad (5.2)$$



where, r is the unperturbed radius of the droplet, e is the ellipticity and a, b is the major and minor axes of the ellipse. The ellipticity of 5% was used therefore the mesh resolution was enough to capture the oscillations. Cases with these meshes were modeled using the explicit volume fraction formulation. Comparison between oscillation spectrum for the Cartesian and unstructured mesh shows that the quality of oscillation spectra for Cartesian mesh was superior to unstructured mesh. The spectrum for both the cases shows noise due to localized surface oscillations. Oscillation spectrum for the Cartesian mesh exhibits clean oscillations at peaks and valleys i.e., when droplets change the configuration from prolate to oblate and vice-versa. Therefore, Cartesian mesh was used for further analysis in this study. To reduce the existing localized surface oscillations, the Cartesian mesh was further refined by using element of size 20 μm which is approximately 1.5% of the radius. The effects of initial deformation on the droplet oscillations and the oscillation frequency were studied. Cases were setup by varying the ellipticity value between 1% to 5%. To choose a surface tension modeling method, cases were run using both CSF and CSS methods. The cases were set up with zero ellipticity i.e., without disturbing the equilibrium. The idea was to maintain the droplet equilibrium and allow the simulation to calculate only the parasitic currents. Since the droplet is already in the equilibrium, the surface tension forces should not cause any droplet or surface oscillations and any oscillations generated will be result of numerical formulation of these methods. Cases were run to study any improvement in the droplet oscillation spectrum by further mesh refinement. Two cases were set up with mesh size of 10 μm, 20 μm, 30 μm, 40 μm, and 50 μm. The animation sequence for cases with 30 μm, 40 μm, and 50 μm show that coarser interface was generated after initialization and as the flow progressed, the high frequency surface oscillations were increased. Therefore, results from those cases are not discussed further. 10 μm and 20 μm cases were allowed to run for larger flow time such that the droplet is allowed to damp out naturally and reach the steady state.

The single-phase model developed in this study calculated the surface tension within 8% of the input value. The viscosity calculated from the simulations was found to be 5 times the value provided to the model. Also, the extended multi-phase model calculated the interfacial tension between copper and cobalt within 4% of the experimental data. However, it was found that the problem of high frequency surface oscillations is not handled by the model. The model needs to be improved in that regard and possibilities for the same are discussed in this section.

In the current model, the difference between material properties of phases across the interface is in the order of 104. This difference introduces numerical error in the calculations near the interface regions. In order to reduce the error, the second phase can be replaced by a dummy phase with density closer to, perhaps approximately one-tenth of, the droplet density. The reduction in calculation error may stabilize the surface oscillations.

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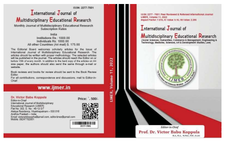

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