The Discharge Crucible Method: update on experimental design, measurements, and orifice wetting

Hani Henein

Department of Chemical and Materials Engineering,
University of Alberta,
Edmonton, AB, Canada
hani.henein@ualberta.ca

Abstract

The physicochemical properties, viscosity, density and surface tension, are critical properties of liquid metals and alloys. These properties are needed for thermodynamics, solidification modelling, and materials properties databases. The Discharge Crucible method (DC) developed in 2003 has been used to measure and report these properties for a wide range of liquid metals and alloys, including Sb, Sn, Zn, Al, Al-Cu, Sb-Sn, Sn-Ag, and AZ91D. The results are compared with published data and models that are proposed to predict these property values. This method is based on a mathematical formulation that predicts the velocity of a stream draining from an orifice. The viscous losses are calculated using a discharge coefficient equation and the gas-liquid surface tension is determined using the Young-Laplace overpressure induced in the jet. The model and experiments will be described along with the effect of nozzle shapes on the distribution of forces in the DC method, including the effect of wetting of the orifice. The aim is to define the optimal nozzle design for a good distribution of forces throughout a draining experiment.

Keywords: physiochemical properties, high temperature fluids, surface tension, viscosity

Introduction

With the advent of increased need for process modeling and simulation, the demand is increasing for accurate data for viscosity, gas-liquid surface tension and density of metals and alloys. The values of these properties as a function of temperature, alloy composition, gas atmosphere including the effect of impurities for recycled materials is needed. Numerous methods have been developed to measure one of these properties.[1-14]. These methods are limited in providing the values of one or two of these properties from a single experiment. There is a need to have one method that can measure all three of them using one test. This makes the process of measurement much more efficient as these tests are very time consuming and great care must be taken to ensure the accuracy of the results. Thus, the level of effort to generate all these values is very significant and costly.

Two methods have been developed that can measure all three properties using one test. The first is Electromagnetic Levitation (EML). It is a containerless technique where a droplet of about 6mm is levitated and all three properties may be measured. The advantage of this method is that other fluid properties such as heat capacity may also be obtained. However, the distinct advantage of the EML is that it is containerless and that property values for undercooled liquids may be determined.[15,16] The disadvantage of the EML is that it must be placed in microgravity environment in order to make these measurements.[17,18] This comes at an obvious high cost and time overhead. There is presently an EML unit located on the International Space Station.

An alternative method to obtaining the three properties of a fluid, namely viscosity, gas-liquid surface tension and density is the Discharge Crucible method (DC).[19-26] In this paper, the principle of the DC method will be described. This will be followed by a description of the apparatus, some of the alloy results generated and the effect of experimental design of the apparatus.

Mathematical model

A detailed description of the model developed for the DC method is described elsewhere [19-26]. A brief overview will be presented here. The model is based on using the Bernouli equation to describe the flow of a fluid exiting the bottom of a crucible through an orifice. The forces that must be accounted for are gravity, inertial force of the exiting fluid, viscous forces of the fluid going through the nozzle, and the induced pressure differential due to interfacial phenomena according to the Young-Laplace equation. With the assumption that there is no wetting of the fluid exiting the orifice, and that quai-steady-state is achieved of the descent of the fluid from the crucible, the exit velocity of the fluid may be described by:

$$u_2 = C_d \sqrt{2g\left(h - \frac{\sigma}{\rho g\, r_0}\right)}. \quad (1)$$

Where u_2 is the velocity of the fluid exiting the orifice, C_d accounts for frictional losses to the orifice, g is the gravitational constant, h is the height of the fluid in the crucible, σ is the gas-fluid surface tension, ρ is the fluid density, and r_o is the radius of the orifice. Note that as the crucible empties u_2, C_d and h are functions of time. Thus, as the crucible empties, Equation (1) may be written in the following form:

$$M_{exp}(t) = \pi r_0^2 \rho C_d(t) \sqrt{2g\left(h(t) - \frac{\sigma}{\rho g r_0}\right)}. \tag{2}$$

Where $M_{exp}(t)$ is the mass flowrate of the fluid exiting the crucible. Equation (2) may be written in dimentionless form as shown in Equation (3):

$$F_r + B_o^{-1} = 1. \tag{3}$$

Where Fr and Bo are the Froude and Bond numbers, respectively, and are given by:

$$F_r = \frac{\left(\frac{M_{exp}}{\pi r_o^2 \rho C_d}\right)^2}{2 g h}. \tag{4}$$

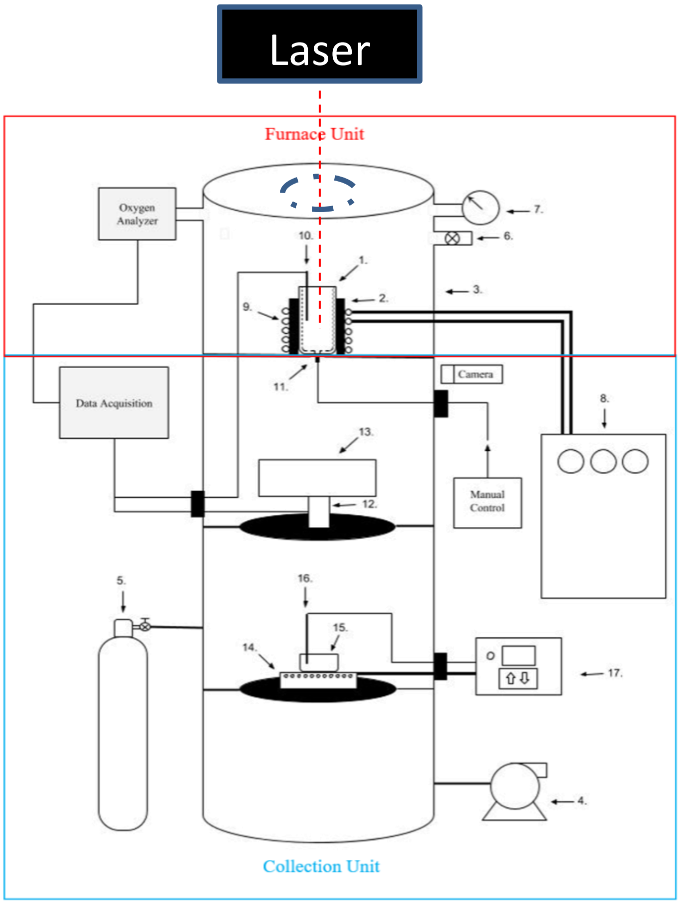
and,

$$B_o = \frac{\rho g r h}{\sigma} \tag{5}$$

The dimensionless form of Equation (2) is useful in designing a particular apparatus for a specific fluid (eg. nozzle size and crucible wall thickness). Note that as the crucible empties the balance between the viscous and surface tension forces changes. Apparatus design assists in maximing the accuracy of the data by ensuring that both forces play an important role during the conduct of the experiment.

Description of DC and sample results

A schematic and a photo of the current apparatus for conduting the DC experiment are shown in Figures 1 and 2, respectively. The apparatus is composed of an induction furnace that heats a susceptor and crucible that contains the melt of interest. A thermocouple is inserted in the melt and a radiation pyrometer are used to monitor the melt temperature. The crucible has an orifice machined in its bottom. To ensure that no flow of liquid occurs during heating and melting, a stopper rod plugs the bottom of the crucible and is removed once the melt reaches the desired temperature. A load cell is placed as close a possible near the bottom of the crucible to capture the stream and provide a mass flowrate as a function of time. More recently a laser has been installed to measure the melt height as a function of time. Previously, the crucile geometry needed to be calibrated and the height change with time was determined by back calculating the height knowing the mass flow measured by the load cell and the initial mass of metal placed in the crucible. An oxygen sensor provides a measurement of the oxygen content in the apparatus before starting to melt the charge in the crucible. During the melting stage, an oxygen getter, placed at the bottom of the apparatus, ensures that the oxygen content remains constant throughout the experiment.



1. Crucible
2. Susceptor
3. Shell
4. Vacuum Pump
5. Gas supply
6. Pressure relief valve
7. Pressure gauge
8. Induction furnace
9. Induction coil
10. Thermocouple-melt
11. Stopper
12. Load Cell.
13. Collection Vessel.
14. Resistance Heater.
15. Ceramic Dish
16. Thermocouple-getter
17. Temperature controller

Figure 1: schematic of DC apparatus.

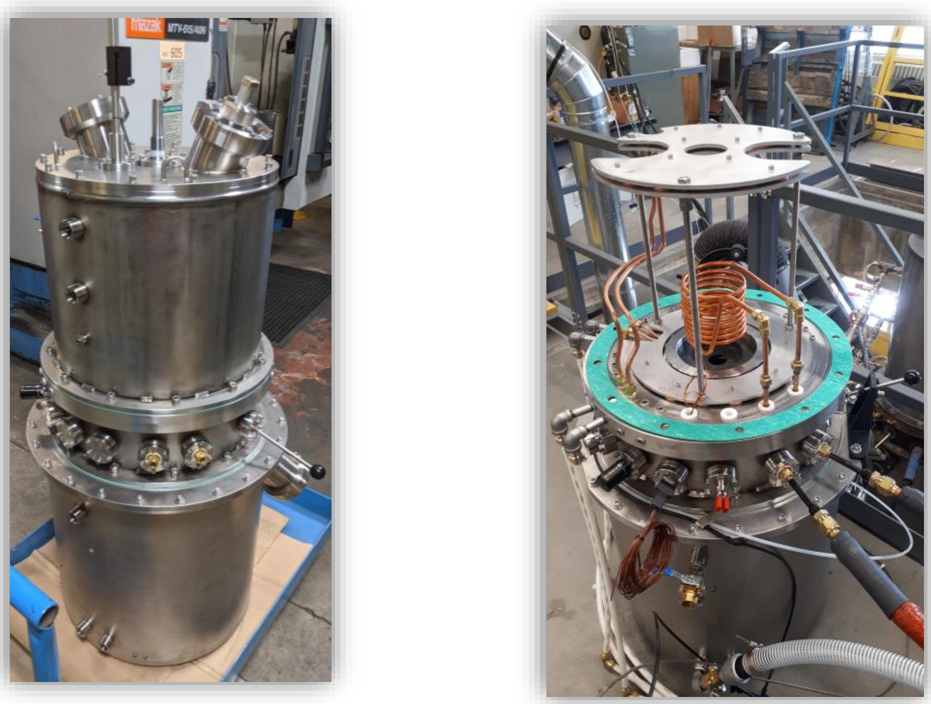


Figure 2: Photo of current apparatus for DC experiment.

Prior to carrying out an experiment with a fluid with unknown properties, a set of calibration runs are carried out with a fluid of known properties. This provides the necessary data in order to obtain a C_d vs *Re* (Reynolds number) plot that characterizes the orifice. Typically, water or other fluid with known properties is used for these calibrations. Once a crucible and its orifice has been calibrated, an experiment can be conducted. The data obtained from a DC experiment is the $M_{exp}(t)$ and corresponding $h(t)$. A non-linear regression is then used to obtain the best fit values of σ, μ and δ at the experimental temperature. For experiments carried out using aluminum, a maximum drop in temperature of the metal of 5K was experienced during the draining of the crucible. This temperature drop does not introduce any significant error in the resulting property values calculated.

The DC method has been used to measure the thermophysical properties of Al, Sn, Sb, and Zn and various alloys, such as AZ91D, Al-Cu, Al-Sn-Ag, Al-Zn, Al-Li-Zn, Sb-Sn, Sb-Sn-Zn, Pb-Sb, Al-Mg, Al-Mg-Zn, Ga-Sn and Ga-Sn-Zn.[19-30] Only a few sample results will be discussed here.

The surface tension of Al was found to be within 1% of averaged values determined by Mills for oxygen saturated elements.[31] With careful control of oygen, Gancarz et al [30] reported a viscosity of Al as a function of temperature that was in excellent agreement with literature values obtained using other techniques. Similar good results were reported for alloy systems such as Sb, Sn, Zn and Sb-Sn [25], Sn-Ag [26], Al-Zn and Al-Li [27], Ga-Sn [28], Pb-Sb [29] and Al-Mg and Al-Mg-Zn [30]. In these results the property values compared very favourably with other measurements reported in the literature using other techniques as well as with theoretical predictions of these properties. There was good agreement with the Butler model for surface tension [32], and reasonable agreement with the Hirai [33] and Kaptay [34] models for viscosity. The ideal soution model was found to be appropriate for alloy density.

Experimental design

Most of the previous experiments reported were carried out using graphite crucibles. More recent work carried out using alumina crucibles for Al and Al-24wt% Cu [23, 35], found that there was significant wetting of the orifice by the liquid metal. This was determined both visually as well as by determining the Capilary number as a function of the height of the metal in the crucible. Even under these circumstances, the surface tension and the viscosity were determined within reasonable accuracy. The model converged on unrealistic values of density. It was found that the model was most sensitive to small changes in density but not those for surface tension or viscosity. Current research is aimed at extending the model for the DC method to include the prediction of dynamic contact angle.[36]

More recent work [36] is focusing on further development of the DC model to incorporate the design of the orifice. Characteristics of the orifice such as bevel design for entrance of the fluid into the orifice and the length of the orifice can play a significant role in the contribution of the *Fr* and *Bo* numbers during the time of drainage of the crucible. This design flexibility can assist in enhancing the capabilities of the apparatus to yield increased accuracy in one of the property values of interest. Current research is also looking at extending the capabilities of the apparatus to carry our measurements on steels.

Summary

A new method for measuring the gas-liquid surface tension, viscosity and density of liquid metals and alloys has been developed and evaluated on a range of alloys and using different gas atmospheres. The method was found to be reliable but sensitive to the wetability of the nozzle and its geometry. Nevertheless, with systems that do not wet the orifice of the crucible and under quasi-steady state for the draining of the crucible, the results are reliable and as accurate as any other method of measurement of these propoerties. Its distinct advantage is that presently air-liquid surface tension, viscosity and density are obtained simultaneously.

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