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Measurement of the Density of Liquid Aluminum-319 Alloy by an X-Ray Attenuation Technique

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Aluminum alloy casting has become an increasingly important field as lighter parts find more widespread use in many areas, especially the transportation industries. As more powerful and inexpensive computers have appeared, simulation of the casting process has become an important part of designing and optimizing processing steps. However, casting simulations cannot hope to accurately describe real processes without the important physical properties of the materials involved in a casting operation.

In this study we have constructed a relatively simple apparatus for measuring the density of aluminum-based alloys in the solid and liquid states, at temperatures up to 900°C. We have measured one of the more important physical properties of a casting alloy, the solidification shrinkage, for a commercial aluminum alloy, Al-319. We find that while the thermal expansion of the alloy in both the solid and liquid states is similar to that of pure aluminum, the density of the liquid alloy is lower than one would estimate by averaging the atomic volumes of the pure liquid components.

Densities were measured by an x-ray attenuation technique originally developed [1] to study density changes in liquid metals that melt at low temperatures, such as Pb. The technique, which has been applied extensively to pure liquid metals and binary alloys [2-9], exploits the change in x-ray attenuation accompanying

changes in density in a sample. When a monoenergetic beam of radiation of intensity I_0 is incident on a material, the transmitted intensity depends on the density ρ , thickness x , and absorption coefficient μ :

$$I = I_0 e^{-\mu \rho x} \quad (1)$$

As the absorption coefficient is independent of temperature, the dependence of the density on temperature can be extracted by measuring I at various temperatures.

The apparatus constructed to perform these experiments is depicted in figure 1. The x-ray source is a 40 mCi ^{241}Am source, which produces a relatively weak 59.57 keV x-ray. The beam is collimated by a 3 mm diameter steel collimator at the source and by a 12 mm diameter lead collimator at the detector; the source to detector distance is approximately 1.2 meters. The low energy and intensity of this source allow it to be used with very little shielding.

The aluminum sample is placed in a boron-nitride coated rectangular graphite (ISO-880U) crucible. The crucible walls were 2.00 ± 0.05 mm thick, and the boron nitride coating layer added an additional 0.15 ± 0.03 mm to the thickness. Boron nitride was chosen as a coating material to prevent adhesion of the aluminum to the graphite during solidification and to prevent reactions between alloy components and the graphite. The crucible is placed in a quartz flash lamp tube furnace, and helium gas is flowed through the tube at 20 liters/hour. The atmosphere inside the tube is contained by two 2 mm thick aluminum plates at the ends of the tube, and the sample thickness is 20 mm (approximately one and a half $1/e$ skin depths for 60 keV x-rays in aluminum). This thickness was chosen to balance reasonable data collection times with good sensitivity to changes in the density of the aluminum. Typical count rates were 250 counts/second, with collection times of 20-40 minutes at each temperature.

Absolute measurements of the density of a sample by the gamma attenuation technique require that the incident intensity be known accurately. However, if the

density of the sample at room temperature is already known, relative changes in the density can be determined without measuring I_0 directly.

The x-rays arriving at the detector have been attenuated by several components in the apparatus: the aluminum endplates, the graphite crucible walls, the helium gas in the quartz tube, and by the aluminum sample itself. Thus the detected intensity I is:

$$I = I_0 e^{-\mu_{Al}\rho_{pl}2x_{pl}} e^{-\mu_C\rho_{cr}2x_{cr}} e^{-\mu_{He}\rho_{He}x_{He}} e^{-\mu_{Al}\rho_{sample}x_{sample}} \quad (2)$$

Here ρ_{cr} and x_{cr} are the density and thickness of the crucible walls, ρ_{pl} and x_{pl} are those of the endplates, ρ_{He} is the density of the gas in the tube furnace, x_{He} is the length of the tube, and ρ_{sample} and x_{sample} are the density and thickness of the aluminum sample. The absorption coefficient μ for the sample is either μ_{Al} (as shown in equation (2), for pure aluminum) or an average μ based on the atomic fractions of each alloy component. Comparing the intensities I_A and I_B measured at two temperatures T_A and T_B (and assuming that the incident intensity I_0 does not depend on the sample temperature):

$$\frac{I_A}{I_B} = \frac{e^{-\mu_C\rho_{cr}(T_A)2x_{cr}(T_A)} e^{-\mu_{Al}\rho_{sample}(T_A)x_{sample}(T_A)}}{e^{-\mu_C\rho_{cr}(T_B)2x_{cr}(T_B)} e^{-\mu_{Al}\rho_{sample}(T_B)x_{sample}(T_B)}} \quad (3)$$

The endplate and helium gas terms present in equation (2) have been omitted here for simplicity, as several factors conspire to make them insignificant: 1) the endplate thickness x_{pl} is quite small (2 mm), 2) the endplates are well outside the furnace and remain at less than 100°C, well below the typical sample temperatures of 300-900°C, and 3) the density ρ_{He} of helium is very low (0.18 mg/cm³ at 1 atmosphere and room temperature). Although ignoring these terms introduces only a 0.15% error in the density, the terms were included in our final calculations.

Taking T_A to be room temperature and T_B the temperature of interest, the density and thickness of the sample and crucible walls are easily measured at T_A .

The density and thickness ρ_{cr} and x_{cr} of the crucible walls are easily calculated at T_B using the linear thermal expansion coefficient α_C of graphite:

$$x_{cr}(T_B) = x_{cr}(T_A) \left[\frac{1 + \alpha_C T_B}{1 + \alpha_C T_A} \right] \quad \rho_{cr}(T_B) = \rho_{cr}(T_A) \left[\frac{1 + \alpha_C T_A}{1 + \alpha_C T_B} \right]^3. \quad (4)$$

Similarly, for temperatures T_B below the melting point of the sample, the density and thickness of the sample can be written in terms of the density and thickness at T_A and the linear thermal expansion coefficient α_{sample} :

$$\rho_{sample}(T_B) x_{sample}(T_B) = \rho_{sample}(T_A) x_{sample}(T_A) \left[\frac{1 + \alpha_{sample} T_A}{1 + \alpha_{sample} T_B} \right]^2. \quad (5)$$

Substituting equations (4) and (5) into equation (3) leaves only one unknown, α_{sample} , allowing the density $\rho_{sample}(T_B)$ to be calculated.

Once the aluminum sample melts, it expands to fill the crucible, the inner dimensions of which are known:

$$x_{sample}(T_B) = x_{cruc}(T_A) \left[\frac{1 + \alpha_C T_B}{1 + \alpha_C T_A} \right]. \quad (6)$$

Here $x_{cruc}(T_A)$ is the inner dimension of the crucible, measured at room temperature. Upon substitution of equations (4) and (6) into equation (3) we are able to solve for $\rho_{sample}(T_B)$, the density of the liquid sample.

As this method requires that the density and thickness be known at room temperature, each specimen was machined to a rectangular block shape, measured, and weighed to determine the room temperature density before melting. The first material studied was pure aluminum (99.99%). Our measured density for pure liquid aluminum is shown in figure 2 with data reported by others [10-12]. We measure a density of liquid aluminum of $\rho_{Al}(T) = 2.368 - 2.64 \times 10^{-4}(T - 660^\circ\text{C})$, which is in quite good agreement with previous measurements. The thermal expansion in the solid is also in good agreement with results of other studies [13].

We next measured the density of an Al-319 alloy with a composition of 6.61% Si, 3.81% Cu, 0.66% Fe, 0.69% Zn, 0.23% Mn and 0.04% Ni (percentages are weight %), and our measurements are shown in figure 2. Although there is no data available for the density of liquid Al-319, measurements [14] of the thermal expansion in the solid of a similar alloy, Al-319.0, are very similar to ours (see figure 2). While the room-temperature density of our Al-319 is only 2.694 g/cm³ (3.4% less than the expected density of 2.79 g/cm³ [14]), this is a typical amount of porosity in a cast ingot.

Although no data exist for the density of liquid Al-319 alloy, an estimate of the density $\bar{\rho}$ can be obtained by averaging the atomic volumes V_i and masses M_i of each of the constituent elements i in the liquid according to their relative fractional abundances x_i [15]:

$$\bar{\rho}(T) = \frac{\bar{M}}{\bar{V}(T)} = \frac{\sum x_i M_i}{\sum x_i V_i(T)}. \quad (7)$$

While measurements of the densities of most liquid metals exist, the melting temperatures of the minor alloy components (such as Si and Cu) are quite high. Thus the $V_i(T)$ used in this calculation are extrapolations of liquid metal density data from much higher temperatures.

The densities measured for pure liquid aluminum and liquid Al-319 are plotted in figure 3 with the expected density calculated using equation (7). Somewhat surprisingly, the liquid appears to be much less dense than expected, indicating that chemical effects have lowered the density of the liquid alloy. We measure a density in the liquid of $\rho_{319}(T) = 2.466 - 3.20 \times 10^{-4}(T - 660^\circ\text{C})$, compared to the calculated density of $\rho_{\text{calc}}(T) = 2.506 - 3.64 \times 10^{-4}(T - 660^\circ\text{C})$. This difference could lead to a sizable underestimation of the solidification shrinkage. Depending on the amount of porosity in the solidified alloy, the estimated shrinkage could be as much as 25% to 55% lower (for 0% and 3.4% porosity, respectively) if the calculated alloy density were used in place of the measured alloy density.

These density measurements have been made with a relatively simple apparatus containing almost no custom parts. Although the precision of the measurements could be improved with modifications to the equipment, even this simple setup is adequate for measuring aluminum alloy densities in the liquid. In addition, the low-intensity radiation source allows the experiment to be conducted with no special radiation shielding.

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Figure 1: Schematic drawing of the experiment. 60 keV x-rays from a 40 mCi Americium-241 source are collimated before and after passing through the sample, which is contained in a graphite crucible in a quartz tube furnace. Two thin aluminum endplates contain the helium atmosphere in the quartz tube.

Figure 2: Densities of pure aluminum (circles) and Al-319 alloy (triangles) in the liquid and solid phases measured in this study. The unbroken lines indicate measurements of the density of pure liquid aluminum by other workers [10-12]. The dashed lines show other measurements of the thermal expansion in the solid for pure aluminum and a similar Al-319.0 alloy. Error bars (not shown) for the data points below 600°C are approximately one half the size of those for the liquid data.

Figure 3: The densities of pure aluminum (circles) and Al-319 alloy (triangles) in the liquid phase compared with measurements of other workers [10-12] (solid lines). The dashed line is an estimate of the density calculated by averaging the liquid atomic volumes of the pure components of the alloy.