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Galinstan liquid metal breakup and droplet formation in a shock-induced cross-flow

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ABSTRACT

Liquid metal breakup processes are important for understanding a variety of physical phenomena including metal powder formation, thermal spray coatings, fragmentation in explosive detonations and metalized propellant combustion. Since the breakup behaviors of liquid metals are not well studied, we experimentally investigate the roles of higher density and fast elastic surface oxide formation on breakup morphology and droplet characteristics. This work compares the column breakup of water with Galinstan, a room-temperature eutectic liquid metal alloy of gallium, indium and tin. A shock tube is used to generate a step change in convective velocity and back-lit imaging is used to classify morphologies for Weber numbers up to 250. Digital in-line holography (DIH) is then used to quantitatively capture droplet size, velocity and three-dimensional position information. Differences in geometry between canonical spherical drops and the liquid columns utilized in this paper are likely responsible for observations of earlier transition Weber numbers and uni-modal droplet volume distributions. Scaling laws indicate that Galinstan and water share similar droplet size-velocity trends and root-normal volume probability distributions. However, measurements indicate that Galinstan breakup occurs earlier in non-dimensional time and produces more non-spherical droplets due to fast oxide formation.

1. Introduction

Understanding the mechanisms that drive liquid metal breakup is essential for a variety of applications including metal powder production (Markus et al., 2002; Luo et al., 2012; Mates et al., 2012), thermal spray deposition (Hussary and Heberlein, 2001; Chen et al., 2012; Newbery and Grant, 2013), conductive microfluidics (Dickey et al., 2008; Liu et al., 2010), metalized solid rocket propellant combustion (Guildenbecher et al., 2014, Chen et al., 2016, 2017b), explosive fragmentation (Rader and Benson, 1988; Zhong et al., 2014), and liquid metal cooling systems (Kondo et al., 1995). In a shock-induced cross-flow, as shown in Fig. 1, the aerodynamic forces imparted by a high speed gas distort the liquid surface while surface tension forces resist deformations. When the aerodynamic forces exceed surface tension, the liquid breaks apart and forms droplets.

For typical fluids like water, this breakup process has been studied extensively both experimentally and in simulation (Hsiang and Faeth, 1992; Joseph et al., 1999; Arienti et al., 2016; Guildenbecher et al., 2017). Several recent numerical investigations have considered high fluid-gas density ratio liquids and liquid metal breakup (Aalburg et al., 2003; Ling et al., 2013; Kékesi et al., 2014; Zhong et al., 2014; Lin et al., 2015; Yang et al., 2016; Arienti et al., 2017). Aalburg et al. (2003) suggest that for density ratios greater than 32, the density ratio has a minimal effect on deformation and breakup properties. Other simulations by Kékesi et al. (2014), however, indicate that higher density ratios produce faster deformation rates. Performing simulations with liquid metals, Lin et al. (2015) show that high density ratios up to 10,000 lead to higher deformation rates and faster growth of surface waves. Yang et al. (2016) further suggest that a higher density ratio leads to more intensive fragmentation but has a non-monotonic effect on droplet deformation. Because these simulations do not incorporate some important physics related to liquid metals (such as surface oxide formation) and point to disparate conclusions, experimental investigations are needed to help understand the underlying physics. Except for a single experimental data point with mercury (Hsiang and Faeth, 1992), there are no known fundamen-

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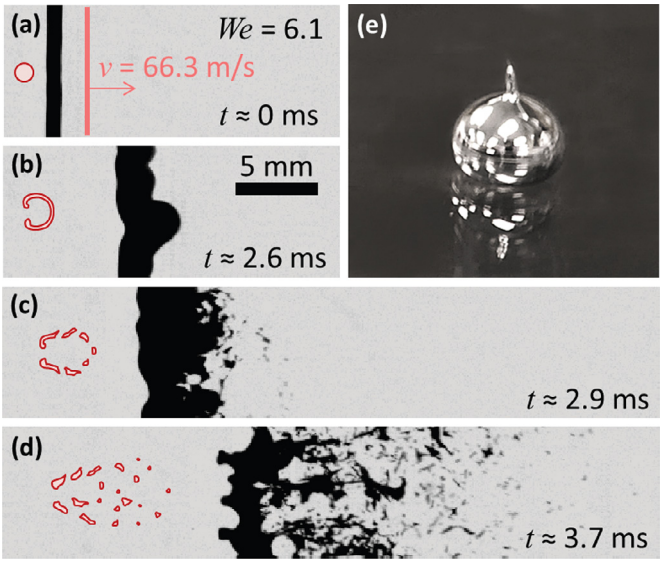

Fig. 1. Liquid Galinstan in a shockwave-induced cross-flow is shown during (a) the initial shock arrival, (b) bag formation, (c) bag breakup and (d) column breakup. The inset of each image shows a top-view schematic of the visually estimated breakup morphology, illustrated in shades of red. (e) A single Galinstan droplet illustrates how oxide skin formation prevents the droplet from becoming fully spherical. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

tal experimental investigations of liquid metals in shock-induced cross-flows.

In addition to high densities, most liquid metals also have high surface tensions, low dynamic viscosities, and the ability to form oxide skins. High temperature molten metals can also exhibit complex physical behaviors including the formation of vapor films (Lin et al., 2015), solidification (Manickam et al., 2017), and combustion (Rader and Benson, 1988). Studying a liquid metal at room temperature minimizes the effect of heat transfer and combustion while focusing on physics driven by high density ratios and surface mechanics.

Galinstan is a non-toxic, eutectic room-temperature liquid metal alloy. This liquid metal has been utilized as a replacement for mercury in thermometers and for research in energy storage, microfluidics and flexible electronics (Dickey, 2014). The material is six times denser than water with a surface tension that is ten times higher. Like aluminum and many other metals, liquid Galinstan forms an oxide skin (Dickey et al., 2008). Fig. 1(e) shows a droplet generated by a pipette that quickly formed an oxide skin, preventing it from becoming spherical. The oxide skin is elastic and has a yield stress that can alter the splash impact (Li et al., 2014) or breakup behaviors (Chen et al., 2017a). Because the influence of the oxide skins on breakup is not well-understood, Galinstan can be used to investigate the effects of surface mechanics and density on breakup behavior.

In this work, the breakup morphology and droplet formation statistics of liquid metal Galinstan is compared with water at a variety of Weber numbers. Back-lit imaging provides a qualitative view of the key differences between the two materials. Digital inline holography (DIH) is then used to provide quantitative measurements of the column deformation as well as droplet velocity and size statistics using automated algorithms. This is the first known paper in archival literature to measure liquid metal breakup in a shock-induced cross-flow. The datasets generated are expected to inform model development and validation while extending scaling relations to liquid metal conditions.

Table 1
Properties for water and Galinstan.

Property	Water	Galinstan	Units
Density ρ_l	1000	6440	kg/m^3
Viscosity μ_l	0.89	2.4	mPa s
Surface tension σ	0.073	0.718	N/m

2. Fluid properties

2.1. Liquid properties

Water is compared with the properties of Galinstan in Table 1. Galinstan (RotoMetals LMP-2) is tertiary eutectic alloy consisting mostly of gallium (68.5%), indium (21.5%) and tin (10%). A eutectic metal melts at a single temperature lower than the melting points of its constituents. Galinstan was chosen for the experiments due to its low toxicity and liquid state at room temperature (melting temperature near −19 °C). The material also essentially has no vapor pressure allowing it to be used in vacuum or in the lab with low inhalation risk. Galinstan has a reported density of $\rho_l = 6440\,\text{kg/m}^3$ (Karcher et al., 2003), which was confirmed in the laboratory and found to be within 3.4% of the reported value.

Surface tension values for Galinstan typically vary from 0.5 to 0.72 N/m depending on measurement conditions. Galinstan quickly forms a thin (5–20 Å) passivating oxide skin (Ga_2O_3 and Ga_2O) with a color that is visually indistinguishable from the fluid in the presence of oxygen above 1 ppm (Regan et al., 1997; Plech et al., 1998; Kim et al., 2013; Dickey, 2014). The skin is elastic and once it yields, the material flows freely, forming fresh oxide. Oxide layer formation rates have been studied on long timescales (Plech et al., 1998), but there is currently no experimental data in the literature for oxide formation rates at the timescales relevant to shock-induced aerodynamic breakup. With less than 1 ppm of oxygen, the measured surface tension is approximately 0.533 N/m (Morley et al., 2008, Liu et al., 2010, 2012). In the presence of hydrochloric acid vapor, the surface oxide is replaced with other alloys ($GaCl_3$ and $InCl_3$) permitting the formation of spherical drops; the surface tension is then measured to be 0.524 N/m (Kim et al., 2013).

In most situations, however, oxygen is present and the effects of the surface oxide must be considered. The surface yield stress of eutectic gallium indium with the same Ga_2O_3 and Ga_2O amorphous skin was measured to be approximately 0.5 N/m (Dickey et al., 2008). The elastic component of the oxide skin provides an additional restoring force to droplets on top of the existing surface tension forces. This causes an upward bias in the measured surface tension. In atmosphere, several authors have measured an effective surface tension of approximately $\sigma = 0.718\,\text{N/m}$ (Kocourek et al., 2006; Karcher et al., 2003). To confirm these measurements in the present study, additional pendant drop experiments were performed and measurements indicate that the effective surface tension is within 2.7% of this reported value (Chen et al., 2017a). These experiments were conducted with a syringe in a setup similar to the final liquid delivery system in the shock tube. In addition, the timescales of the pendant drop formation are similar to the timescales of the liquid column formation for shock-induced cross-flow experiments to increase confidence in surface tension measurements. As will be illustrated later from the experimental results, the formation of the elastic oxide layer is sufficiently fast such that the higher effective surface tension value is believed to be applicable during the entire breakup process studied here.