Directional Motion of Gallium-Based Liquid Metal Induced by Asymmetric Chemical Surrounding

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ABSTRACT

Interfacial, or surface, tension is a significant topic in chemical education. This paper describes the directional motion of gallium-based liquid metal drops resulting from a difference of interfacial tension across the drop. This demonstration can engage students in discovering the underlying chemical principles. A mechanism for the drop's directional motion is proposed to provide insight into this intriguing phenomenon. It appears that unbalanced chemical environments cause different physical or chemical processes to occur on each hemisphere of the drop such as a pH difference, redox reactions, galvanic replacement, or adsorption. As a result, a difference in interfacial tension across the drop is generated, providing the driving force that acts on the drop. This demonstration can be used to introduce the fundamental principles in chemical reactions such as redox activity, electrical double layer formation, and interfacial tension.

GRAPHICAL ABSTRACT



KEYWORDS

First-Year Undergraduate/General, Upper-Divison Undergraduate, Interfacial/Surface Tension, Electrochemistry, Electrical Double Layer, Surface Science

25 INTRODUCTION

Interfacial tension refers to the attractive forces between molecules that exist at the interface between solid-liquid, solid-gas, liquid-liquid or liquid-gas phases. Specifically, the tension that arises at the interface of a liquid and air is known as surface tension. In natural contexts, numerous fascinating phenomena emerge that are related to the surface or interfacial tensions. For instance, water striders effortlessly move across the surface of water. A ring of clear liquid, known as the "tears of wine", can be observed near the top inner rim of a glass of wine from which droplets form and fall back into the wine. Additionally, objects such as paper clips, coins, needles, and razor blades possess the ability to float on the surface of water. Water droplets and bubbles possess a spherical shape. All of these remarkable occurrences are attributed to the presence of surface or interfacial tension.

Surface tension has been a significant topic in chemical education as evidenced by the inclusion of this concept in physical chemistry textbooks¹ and a multitude of papers²⁻⁶ published in this Journal. For instance, some studies have introduced simple apparatuses to demonstrate the effects of surface tension on liquids,⁷ while others have developed various methods to accurately measure surface tension.⁸⁻¹⁰ Additionally, the floatation of glass coverslips on water has been used to explore the selfassembly of molecular monolayers.¹¹ The motion of solids and liquids has been studied as an educational demonstration of surface tension. Researchers have observed the self-motion of sodium benzoate flakes on a water surface¹² and designed a boat that runs on liquids as "fuel".¹³ Another study showed how the self-propulsion of sodium oleate tablets can be driven by interfacial tension imbalance induced by inhomogeneous adsorption of oleate/oleic acid molecules.¹⁴ These findings demonstrate the vast array of techniques and experiments that have been developed to better understand the concepts of surface or interfacial tension.

Liquid metals exist as either an alloy or an elemental metal and remain in liquid form at room temperature. They possess both fluid and metallic properties thus making an interesting topic for study. Traditionally, mercury has been the most recognized liquid metal. The mercury beating heart Page 3 of 15

demonstration has been popularized for electrochemistry education.¹⁵⁻¹⁸ This demonstration shows the oscillations that occur due to the periodic changes in surface tension of mercury droplets caused by redox reactions. Unfortunately, the high toxicity of mercury has limited its use as a classroom demonstration. Gallium-based metal alloys provide an alternative to mercury as they not only have fluidity at room temperature but also have low vapor pressure, and are non-toxic. This means these metals can serve as safer demonstrations.¹⁹⁻²² In a previous demonstration, gallium and its alloys formed a beating heart that occurred in a uniform solution where oscillations were induced by the periodic change of surface tension. Self-propulsion or deformation of the liquid metals resulted from a surface tension gradient²³⁻²⁴ that was derived from ionic imbalance,²⁵ redox reaction,²⁶⁻²⁸ or electric field.²⁹⁻³⁰

In the previous Gallium-based liquid metal beating heart demonstrations, the position of the iron nails or the electrodes in contact with the liquid metal droplets required careful adjustment to trigger a "heartbeat" or oscillation. Here, we present a straightforward and visually striking demonstration to introduce some fundamental principles in chemical reactions, including redox, electrical double layer, and interfacial tension. Adding chemical substances far from one side of the droplets in a uniform aqueous solution can produce directional movement of Ga-based liquid metal. To reduce the necessary amount of chemicals, a small device is employed. This demonstration, simple and visually stimulating, can effectively motivate undergraduate students to explore the underlying principles and provide them with hands-on laboratory experience. Ultimately, we propose a mechanism of droplet movement to interpret these intriguing phenomena.

MATERIALS AND EQUIPMENT

The experimental setup consisted of several reagents and materials, including Galinstan - a Ga-based liquid metal composed of 68.5% gallium, 21.5% indium, and 10% tin, 1 mol/L hydrochloric acid, aqueous hydrogen peroxide at 35% concentration, potassium dichromate, copper sulfate, copper (II) chloride, potassium iodide, and deionized water. The experiment was carried out in a homemade polymethylmethacrylate (PMMA) channel with the dimensions of 4 mm width, 4 mm depth, and 40 mm length, along with a 3-mm radius cylindrical chamber at each end (Figure 1). The PMMA substrate can be purchased from local stores or on line, then fabricated by a numerical controlled machine. Alternativelly, polytetrafluoroethylene (PTFE) is another choice for the substrate. The channel can also be fabricated on a glass slide using polydimethylsiloxane (PDMS) through molding technique, or printed by a 3D printer with polylactic acid (PLA). The reagents were applied to the channel and chambers using a spatula or dropper.



Figure 1. Machine drawing of the channel used in this demonstration (unit: mm).

EXPERIMENTAL PROCEDURE

To perform this experiment, the PMMA channel is first filled with 1 mol/L HCl solution. With the aid of a pipette, a drop of Galinstan with a volume of approximately 20 μ L is added near the center of the channel. Observing and recording what is happening to the drop begins at this point. To create an unsymmetric solution, one chemical chosen from sodium hydroxide, potassium dichromate, copper sulfate, copper (II) chloride, potassium iodide, and aqueous H₂O₂ (35%) is then added to the right chamber. Different unsymmetric solutions are produced by adding respective chemicals to fresh HCl solution with fresh Galinstan drop. An instructor performs the demonstration, and students observe and make note with further discussions. A USB camera is employed to capture the Galinstan drop's directional motion under different conditions. Students are encouraged to carry out this experiment independently or in groups under supervision. Demonstration interaction with questions and discussions, takes place before and during the demonstration to facilitate students' understanding of interfacial tension and redox reaction concepts. The accompanying Supporting Information includes videos of the experiment, learning objectives, and guidlines of demonstration interaction.

HAZARDS AND WASTE DISPOSAL

Unless exposed at a very high concentration, Galinstan is nontoxic.³¹ The disposal of Galinstan does
not pose a significant environmental threat although its potential to adhere to most surfaces can pose a problem. If a surface contains aluminum components, adsorption of the alloys is very problematic and so such surfaces should be avoided. In contrast, chemicals such as sodium hydroxide, hydrochloric acid, hydrogen peroxide, and potassium dichromate are corrosive and can cause significant harm if mishandled. When using potassium dichromate, a highly toxic substance, caution
must be taken. However, less toxic strong oxidants can also be tested as alternatives to trigger the directional motion of the galinstan drop. Copper salts and potassium iodide can also cause skin and eye irritation. Given these hazards, the students must be made aware of the potential risks as well as the precautions that need to be taken before handling these chemicals. They should refer to the material safety data sheet (MSDS)³² for each chemical to fully understand associated hazards. To
prevent accidents, the students are required to follow standard lab safety protocols at all times and proper personal protective equipment, such as goggles, gloves, and lab coats, should be worn during the experiments. Finally, any solutions used in an experiment should be disposed of in appropriate waste containers.

RESULTS AND DISCUSSION

Video images were recorded to demonstrate the directional motion of the Galinstan drop in the following. Upon the addition of NaOH to the right chamber of a channel filled with HCl solution, the Galinstan drop quickly migrated towards the right side (Figure 2). This suggests that the Galinstan drop prefers migrating towards the higher pH region. These observations are consistent with Zavabeti et al.'s findings that the self-propulsion of liquid metal drops occurs due to a pH imbalance resulting in a difference in interfacial tension on opposing hemispheres of the drop.²⁵ Higher surface tension on the HCl side propels the drops towards the NaOH side with lower surface tension. The difference in interfacial tension was confirmed by Handschuh-Wang et al.'s research on Galinstan, which showed a higher interfacial tension in an HCl solution than that in a NaOH solution.²⁹



17 125

37 135

Figure 2. Video images of the directional motion of Galinstan drop in 1 mol/L HCl solution with the addition of NaOH to the right chamber.

When using oxidants, the Galinstan drop was observed to move towards the oxidants. Figure 3 illustrates the displacement of the drop in an HCl solution with the addition of potassium dichromate in the right chamber. Initially, the potassium dichromate dissolves, diffuses, and eventually reaches the liquid metal drop. Following this, the drop begins to elongate and after 203 seconds, it quickly moves to the right side. Similarly, a comparable behavior was displayed by mercury.²⁷ This motion of the mercury drop was ascribed to the asymmetry of the interfacial tension on the two sides of the drop, which resulted from the formation of a concentration gradient of dichromate ions. Gallium, the main component in Galinstan, has a lower standard reduction potential (Ga³⁺/Ga: -0.529 V vs. SHE) than the other two components (In³⁺/In: -0.340 V; Sn²⁺/Sn: -0.138 V). Thus, upon contact with oxidants exhibiting a higher standard reduction potential than gallium, the predominant oxidation reaction involves gallium. The surface of the Galinstan drop can be oxidized by the dichromate ion to form a Ga₂O₃ film. The redox reaction is described as follows:

$$2Ga + 8H^{+} + Cr_2O_7^{2-} \rightarrow Ga_2O_3 + 2Cr^{3+} + 4H_2O$$
(1)

The higher concentration of potassium dichromate on the right side of the Galinstan drop resulted in the formation of a thicker oxide film on the right side than that on the left side. Although the Ga₂O₃ film formed on the Galinstan surface can be dissolved by an acidic solution, when the formation rate of the oxide film exceeds the rate of dissolution, more oxide film remains on the right side than on the left side. The presence of an oxide film on the liquid metal surface can significantly reduce the interfacial tension,^{24, 33} which leads to a higher tension on the left side than the right and ultimately

caused the displacement of the drop towards the right. The presence of hydrogen peroxide solution in the right chamber induces similar redox reactions, resulting in the drop moving to the right side in an acidic electrolyte, as demonstrated in Figure 4. This motion can be again attributed to the difference in interfacial tension caused by the difference in oxide film thickness on the two sides of the drop. The reaction between Ga and H_2O_2 can be represented by the following equation:

$$2Ga + 3H_2O_2 \rightarrow Ga_2O_3 + 3H_2O$$
 (2)



Figure 3. Directional motion of a Galinstan drop in 1 mol/L HCl solution with the addition of $K_2Cr_2O_7$.



Figure 4. Directional motion of a Galinstan drop in 1 mol/L HCl solution with the addition of aqueous H₂O₂ (35%).

The low standard reduction potential of gallium results in galvanic replacement reactions occurring when gallium-based metal alloys encounter many metal salt solutions at the liquid metalsolution interface.³⁴⁻³⁵ In this study, we demonstrate the directional motion of a Galinstan drop in an HCl solution by adding copper sulfate (Figure 5) and copper (II) chloride (Figure 6). As observed, the liquid metal drop rapidly moves towards the right side when the blue (from CuSO₄) or green (from CuCl₂) color reaches the drop. Dark gray or reddish-orange typically appears with time at the interface of the liquid metal and copper salt solutions, with both colors confirmed as presence of copper.^{26, 35} Equation (3) describes the galvanic replacement reaction that occurs with the addition of copper salt. The locomotion of liquid metal is initiated by the surface tension difference associated with the unbalanced interfacial tension, as copper generated in galvanic replacement form a galvanic cell with 23 165 gallium, leading to the reduction of interfacial tension at the right hemisphere.²⁶ Therefore, the unbalanced interfacial tension on the Galinstan drop causes the motion of the drop towards the right side. In the case of CuCl₂, the speed of the liquid metal drop is faster than that in CuSO₄. After 41 seconds, the drop arrives at the right chamber and then quickly migrates towards the left side. In Figure 6, we note that the Galinstan drop always moves towards the areas where the electrolyte is 33 170 green or dark green, reflecting the direction with concentrated $CuCl_2$ solution. When the drop occupies the right chamber, the original $CuCl_2$ solution with high concentration is pushed into the left side of the drop. The Galinstan drop continuously pursues the concentrated $CuCl_2$ solution, as copper is left behind and new galvanic replacement takes place. Thus, the mechanism for this case is likely similar to that of CuSO₄ addition, as copper generated from galvanic replacement on one hemisphere drive the directional motion of the drop by causing the unbalanced interfacial tension on the drop surface.

$$2Ga + 3Cu^{2+} \rightarrow 2Ga^{3+} + 3Cu \tag{3}$$

0 s



72 s

75 s

58 59

60



Figure 6. Snapshots of the motion of a Galinstan drop in 1 mol/L HCl solution with the addition of CuCl₂.

If potassium iodide was added to the right chamber, the Galinstan drop also moves to the right side, as shown in Figure 7. Handschuh-Wang et al. observed that the interfacial tension of Galinstan can be greatly reduced by acidified KI solution, where I⁻ is a surface-active anion.²⁹ When potassium iodide dissolves, diffuses, and gets in touch with the right hemisphere of the Galinstan drop, the iodide anion will be adsorbed on the right hemisphere, increasing the surface charge density of the electrical

double layer and thereby lowering the interfacial tension. This interfacial tension gradient across the Galinstan drop generates a force that propels the drop towards the right side.



Figure 7. Directional motion of a Galinstan drop in 1 mol/L HCl solution with the addition of KI.

When immersed in an acidic electrolyte, the Galinstan drop undergoes a chemical reaction with the acid solution. The dominant and more active metal, gallium, reacts to generate Ga^{3+} ions, positively charging the drop surface. Adsorbed Ga^{3+} cations cause an accumulation of anions in the surrounding electrolyte, leading to an evenly distributed surface charge across the liquid metal-electrolyte interface, forming an electrical double layer (EDL). The interfacial tension (γ) on the liquid metal drop can be expressed by integrated Lippmann equation³⁶:

$$\gamma = \gamma_0 - \frac{1}{2}cV^2 \tag{4}$$

where γ_0 denotes the maximum surface tension when V = 0, *c* is the EDL capacitance per unit area, and *V* is the potential difference across the EDL.

Interfacial tension could be adjusted by directly modifying the EDL by altering the electrolyte surrounding.²⁵ The impact of the surrounding chemical environment on interfacial tension has been demonstrated for mercury³⁷ and liquid gallium³⁸. The maximum surface tension is a function of electrolyte type. When additional chemicals are introduced far from the one side of a Galinstan drop, they dissolve and diffuse to the drop. This results in two distinct hemispheres of the drop with differing chemical environments. The interfacial tension of the hemisphere exposed to the additional chemicals is altered by the factors such as changes in pH, redox reaction, galvanic replacement, and adsorption. This leads to a difference in interfacial tension across the drop.

To unravel the mechanism underlying the drop movement, the force analysis is depicted in Figure 8. The direction of the drop movement towards the right side is defined as positive. The driving force

(dF) imposed on the drop, induced by the unbalanced Young's force, can be given by the following equations:

$$F_{\rm R} = (\gamma_{\rm sa(R)} - \gamma_{\rm sl(R)}) dx = \gamma_{\rm la(R)} \cos \theta_{\rm R} dx$$
(5)

$$F_{\rm L} = -(\gamma_{\rm sa(L)} - \gamma_{\rm s^{\rm l}(L)}) \, \mathrm{d}x = -\gamma_{\rm la(L)} \cos \theta_{\rm L} \, \mathrm{d}x \tag{6}$$

$$dF = F_{\rm R} + F_{\rm L} = \gamma_{\rm l_2(R)} \cos \theta_{\rm R} \, dx - \gamma_{\rm l_2(L)} \cos \theta_{\rm L} \, dx \tag{7}$$

where γ_{sa} , γ_{sl} , and γ_{la} denote the interfacial tensions on the solid/aqueous, solid/liquid metal and liquid metal/aqueous interfaces, respectively. The contact angle between the liquid metal droplet and the PMMA surface is represented by θ , while R and L indicate the right and left sides of the droplet. The thickness of the section of the drop is represented by dx. In a uniform acidic electrolyte, EDL is formed on the interface between the drop and the electrolyte. The absence of the attraction between the drop and the PMMA surface, along with the high interfacial tension y_{la} between the drop and the electrolyte, leads to $\gamma_{Ia(R)} = \gamma_{Ia(L)}$ and $\theta_R = \theta_L > 90^\circ$, as shown in Figure 8A. Thus, dF is equal to 0 and no movement of the drop occurs. If the interfacial tension is reduced on the right hemisphere of the drop due to the addition of chemicals and remains constant on the left hemisphere, i.e. $\gamma_{la(R)} < \gamma_{la(L)}$, the right hemisphere of the drop will spread further towards the right side, causing $\theta_R < \theta_L$. As $\cos \theta_L < 0$ and $\theta_L > 0$ $\theta_{\rm R}$, dF always is positive, and the drop tends to move to the right side. The balance between the driving 37 225 and resistance forces determines the forces acting on the droplet. When the driving force is greater than the resistance force, the drop moves towards the right side (Figure 8B).



Figure 8. Mechanism of directional motion of a Galinstan drop towards the right side without (A) and with (B) the addition of different chemicals.

CONCLUSION

A classroom demonstration featuring directional motions of gallium-based liquid metal drops is presented as an effective tool to introduce fundamental principles in chemical reactions. These principles include redox reactions, the electrical double layer, and interfacial tension. This simple, visually observable experiment requires minimal chemicals and can effectively stimulate students' interest to explore the underlying chemical principles. The proposed mechanism of the drop's directional motions sheds light on these intriguing phenomena. When chemicals are added to only one side of the originally uniform electrolyte, the two hemispheres of the drop encounter different chemical surroundings, resulting in the difference in interfacial tension. The force acting on the drop is then analyzed in terms of the interfacial tension. When the difference becomes significant enough, the driving force will exceed the resistance force and propel the drop to move.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available on the ACS Publications website at DOI:

10.1021/acs.jchemed.XXXXXXX. ACS will fill this in Example brief descriptions with file formats indicated are shown below; customize for your material.

Learning objectives, guidelines of demonstration interaction (PDF, DOCX)

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	video 50. Adding Ki in Her solution (MF4)			
	Video S6: Adding KL in HCl colution (MP4)			
U	Video S5: Adding CuCl. in HCl solution (MP4)			
0	Video S4: Adding CuSO, in HCl solution (MP4)			
	Video S3: Adding H_2O_2 (35%) in HCl solution (MP4)			
	Video S2: Adding potassium dichromate in HCl solution (MP4)			
	Video S1: Adding sodium hydroxide to the right chamber filled with HCl solution (MP4)			

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