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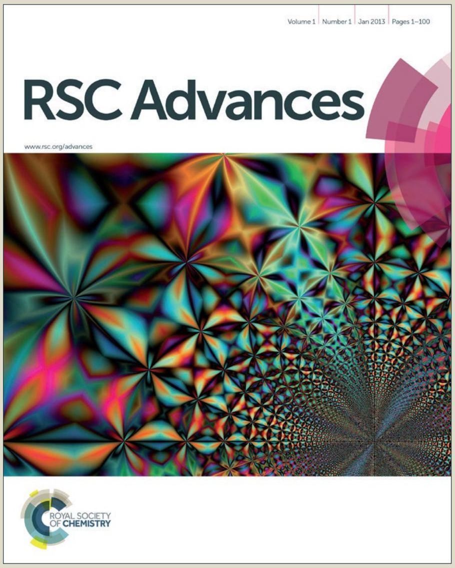

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ARTICLE

Graphite induced periodical self-actuation of liquid metal

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Self-actuation phenomena of liquid metal sphere in NaOH solution including spreading, oscillating and stretching induced by graphite alone were demonstrated for the first time. A liquid metal sphere could spontaneously spread on the surface of the graphite once immersed in the NaOH solution. The surface tension gradient on the sphere induced by the graphite/liquid metal galvanic cell was responsible for such deformation. When a liquid metal sphere was leaned against the side of a piece of graphite, it could oscillate periodically. As the sphere contacted the graphite, it rapidly collapsed while the curvature radius of the sphere at the contact point became smaller. And as the capillary force imposed on the sphere was larger than the friction force, the sphere recovered to its original spherical shape. The surface tension of the liquid metal sphere acted as the restoring force of the oscillatory movement. Further, a phenomenon of resonance could be observed when two spheres were laid respectively on the top and the side of the graphite. The vibration of the top sphere was induced by the vibration of the side one. Such finding provides a novel supplement for making future liquid metal beating heart system and graphite/liquid metal-based batteries or machines.

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1 Introduction

Investigation on the characteristics of the liquid metal including thermal, electrical and rheological properties has gained momentum in recent years.[1] Among them, the rheological characteristic of the liquid metal under electric field[2-4] has obtained special interest. A sheet of liquid metal film immersed in the electrolyte can transform itself from the flattened shape into a sphere as it contacts a cathode electrode,[2] and a liquid metal sphere will spread into a disc-shaped or petal-shaped object as it contacts an anode electrode.[3,5] When a liquid metal sphere is located between the electrified electrodes, it will move from the cathode to the anode while keeping internal self-rotating.[2,4] The deformation of the liquid metal is generated by the electrocapillary force due to the electrowetting effect[6-8] as applying external electric field. In addition, other experiments such as mercury beating heart (MBH) system have been investigated extensively which are based on the chemical redox reaction. MBH is a classical electrochemical oscillator and is first reported by Lippmann as early as 1873.[9] The experimental setup consists of a glass tube containing mercury covered by aqueous solution of sulfuric acid and an oxidizing agent. As a piece of ion or aluminum is brought near to the mercury drop periphery, a sustained pulsating motion of the drop is induced.[10,11] The electrocapillary effect and the surface tension gradient are generally considered to be responsible for oscillations of the mercury drop.[12] Keizer at al[13,14] associated the oscillations in sulfuric acid solutions of strong oxidants with the formation and removal of a film of Hg_2SO_4 on the mercury drop. Subsequently, the MBH system was studied under different experimental conditions including the presence and the absence of an applied voltage.[15-17] Smolin et al[18] observed standing waves in a linear geometry with a number of nodes depending on the potential of the used metal tip. Castillo-Rojas et al[19] investigated the dynamics of the MBH system in the presence of γ-radiation and demonstrated different topological configurations of the mercury drop. However, few studies are about the liquid metal deformation induced by graphite which is a research focus. As a kind of non-metallic functional material, graphite owns superior physical properties including electrical and thermal conductivities which enable its wide application especially in fabricating nanocomposite.[20-23] And the excellent chemical stability enables it a good candidate as the electrode material in making electrochemical devices including fuel cells, batteries and supercapacitors.[24] In this paper, from an alternative, a fundamental phenomenon regarding spreading, oscillating and stretching behaviors of liquid metal immersed in the NaOH solution electrolyte and laid against a piece of graphite is disclosed with the mechanisms interpreted.

2 Results

2.1 Experimental materials

Galinstan (68.5 % gallium, 21.5 % indium and 10 % tin by weight) with melting point about 11 ^oC is taken as the research object. It offers remarkable properties such as high electrical conductivity, high surface tension, and negligible toxicity compared to mercury.[25] The preparation process for making this kind of metal fluid was as follows: three metals of gallium, indium and tin (with high purity of 99.99%) are weighed according to the ratio of 68.5:21.5:10. These

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pure metals are put in a beaker for 2 hours at 150 ℃ in an electric vacuum drying oven. Then, the mixture is stirred in the beaker which is put in water bath at 50-60 ℃ for 30 min to ensure a well-mixed alloy solution. Graphite with ultra-pure of 99.999% is utilized through all the experiments. And NaOH solution with a concentration of 0.5 mol/L is selected as the electrolyte.

2.2. Spreading phenomenon of liquid metal on the graphite surface

A piece of cuboid graphite with a liquid metal sphere with 0.6 g on the surface is located in a petri dish, and NaOH electrolyte solution with concentration of 0.5 mol/L is gradually added into the dish. Variation of the voltage between liquid metal and graphite with the time is measured and shown in Fig. 1(A). As the liquid metal sphere is from unsubmerged (see snapshot 1 in the inset) to submerged (see snapshots 2 and 3 in the inset) into the solution, it rapidly collapses and spreads on the graphite surface (see Movie S1). And meanwhile the voltage value is changed from positive to negative. Such spreading deformation of the liquid metal and the voltage variation can be interpreted as follows: Before the liquid metal is submerged, it is in direct contact with the graphite surface. The contact potential difference V_{lg} between the liquid metal and the graphite can be expressed as: [26,27]

$$V_{lg} = V_l - V_g = -\frac{1}{e}(\Phi_l - \Phi_g) \qquad (1)$$

where e is the charge of an electron. V_l and Φ_l represent the potential and the work function of the liquid metal, respectively. V_g and Φ_g represent the potential and the work function of the graphite, respectively. As the work function of the graphite is equal to 4.7 eV[28] and that of each component (Ga, In and Sn) of the liquid metal is lower than 4.4 eV,[29] V_{lg} is thus a positive value according to Equation (1). It means that the potential of the liquid metal is larger than that of the graphite. An electric double layer (EDL) is formed at the graphite/liquid metal interface when the electrons come to a dynamic balance. Due to that electrons are prone to transfer from the liquid metal which has the smaller work function, the residual electrons are concentrated on the graphite side. This is verified in the curve that the voltage between the liquid metal and the graphite is a small positive value (about 0.031 mV) before the liquid metal is submerged in the NaOH solution. The value of the voltage (about 0.031 mV) is far less than the theoretical calculation result (larger than 0.3 V) obtained from Equation (1). The reason can be attributed to the rough surfaces of the graphite and the liquid metal.[27]

As the liquid metal is immersed in the NaOH solution, its surface oxide (main component: Ga_2O_3[30]) is removed by the electrolyte and the chemical formula can be expressed as:[31]

$$Ga_2O_3 + NaOH \rightarrow NaGaO_2 + H_2O \qquad (2)$$

Pure gallium will be exposed to the solution and gallates like $[Ga(OH)_4]^-$ are produced which make the liquid metal negatively charged. Then the positive ions in the solution are electrostatically attracted by these anions and an EDL is generated at the liquid metal/electrolyte interface.[1,3] Meanwhile, electrons concentrate at the graphite/electrolyte EDL from the liquid metal through the NaOH solution, and the graphite side of the EDL is positively charged. A galvanic cell system is thus formed with graphite, liquid metal and NaOH solution as anode, cathode and electrolyte, respectively. The chemical reactions taking place at the anode and the cathode of this graphite/liquid metal cell can be respectively expressed as:

cathode:[32] $Ga + 4OH^- - 3e^- \rightarrow H_2GaO_3^- + H_2O$ (3)

anode: $2H^+ + 2e^- \rightarrow H_2\uparrow$ (4)

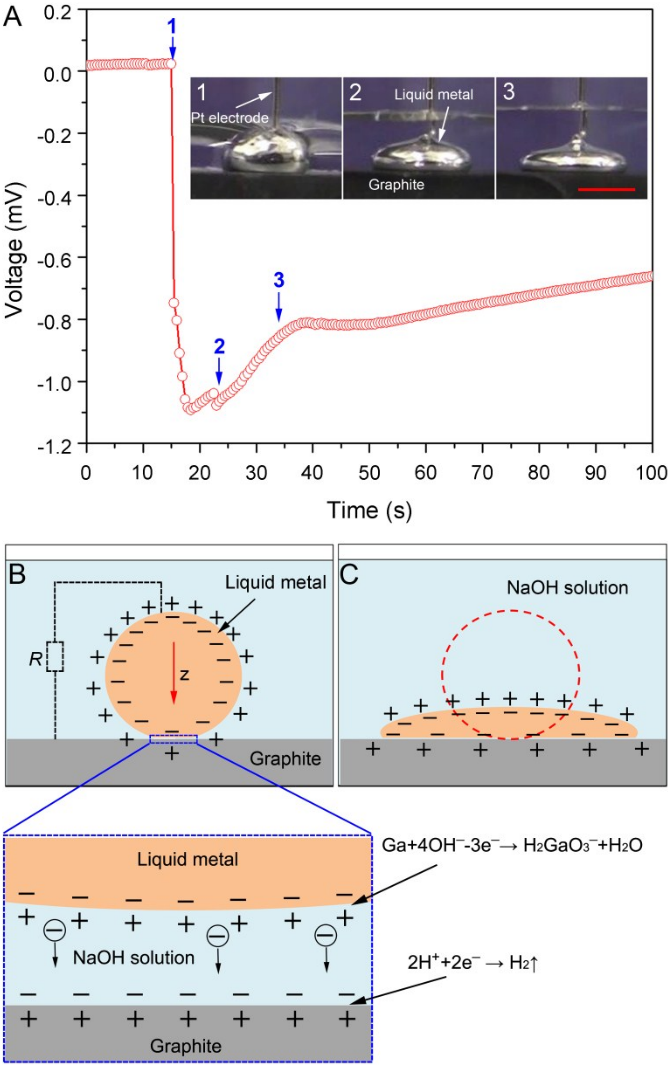

Fig. 1 Spreading deformation of a liquid metal sphere on the graphite substrate immerged in the NaOH solution. (A) Variation of the voltage between liquid metal and graphite before and after their immersion in the NaOH solution. Insets are snapshots of the liquid metal at time points 1, 2 and 3, respectively. Scale bar is 3 mm in length. (B) Schematic of the charge distribution on the surface of the liquid metal in initial state. The enlarged image (marked by the dashed box) is the charge distributions and the chemical reactions on the graphite/electrolyte and the liquid metal/electrolyte EDLs. (C) Schematic of the deformation of the liquid metal in end state. The red dashed-line circle represents the outline of the liquid metal before deformation.

It can be seen that hydrogen gas is generated from the graphite anode. This is verified in our experiment as shown in Fig. 2 that when several liquid metal spheres stay on the graphite for an hour, the surface will be covered with bubbles, compared with the clean surface without liquid metal (see Movie S2). Such graphite/liquid metal cell provides a novel method for producing hydrogen in a convenient way.

The liquid metal can be regarded as an equipotential body for its high electric conductivity.[4] And there will be a uniform charge distribution along the liquid metal/electrolyte EDL without external electric field. However, in the presence of the graphite/liquid metal galvanic cell, there will be a potential gradient with downward

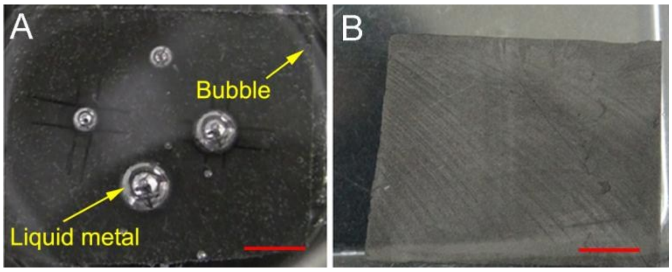

Fig. 2 Comparison between graphite with (A) and without (B) liquid metal droplets on the surface. Scale bars are all 3 mm in length.

direction on the surface of the sphere via the electrolyte whose resistance can be seen as *R*. The surface tension of the sphere is thus altered due to the potential gradient. According to Lippman's equation:[6]

$$\gamma=\gamma_0-\frac{1}{2}cV^2 \qquad (5)$$

where, γ is the surface tension of the liquid metal, c is the capacitance of the EDL per unit area, V is the potential difference across the EDL and γ_0 is the maximum value when V=0. Take derivation to both sides of Equation (5) along z direction as shown in Fig. 1(B), we have:

$$\frac{d\gamma}{dz}=-cV\frac{dV}{dz} \qquad (6)$$

Assuming the direction of V is from the electrolyte to the liquid metal, then V is a positive value while $\frac{dV}{dz}$ is negative in Equation (6). Therefore $\frac{d\gamma}{dz}$ is positive which means that the surface tension on the upper hemisphere of the liquid metal sphere is lower than that on the other half. As a result, the sphere spreads on the graphite and the schematic diagram is presented in Fig. 1(C).

As a liquid metal sphere is laid on the surface of copper in NaOH solution, it will rapidly spread which indicates the formation of copper/liquid metal galvanic cell (as shown in Fig. 3). However, the sphere will brighten up without spreading when the substrate is Teflon which is a kind of insulator. It shows that the deformation of liquid metal cannot be induced by insulators since there is no galvanic effect generated. The deformation characteristic of liquid metal gradually pressed by graphite, copper and Teflon in NaOH solution is also comparatively investigated (see Movie S3 and S4). A liquid metal sphere can be pressed into a thin round cake shape by graphite and copper. And the voltage between them and liquid metal is changed from about 1.2 V to about 0 V, which is presented in Fig. 4. As the liquid metal sphere is pressed by Teflon, it will be squeezed out without flattened and the voltage has no obvious change. The experiments mentioned above indicate the similar electrochemical property of graphite and copper which can both constitute galvanic cells with liquid metal.

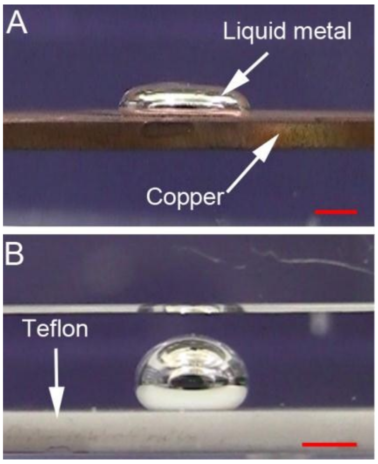

Fig. 3 (A) The lateral spread deformation of liquid metal sphere on the copper in NaOH solution. (B) The liquid metal sphere on the Teflon in NaOH solution. Scale bars are all 3 mm in length.

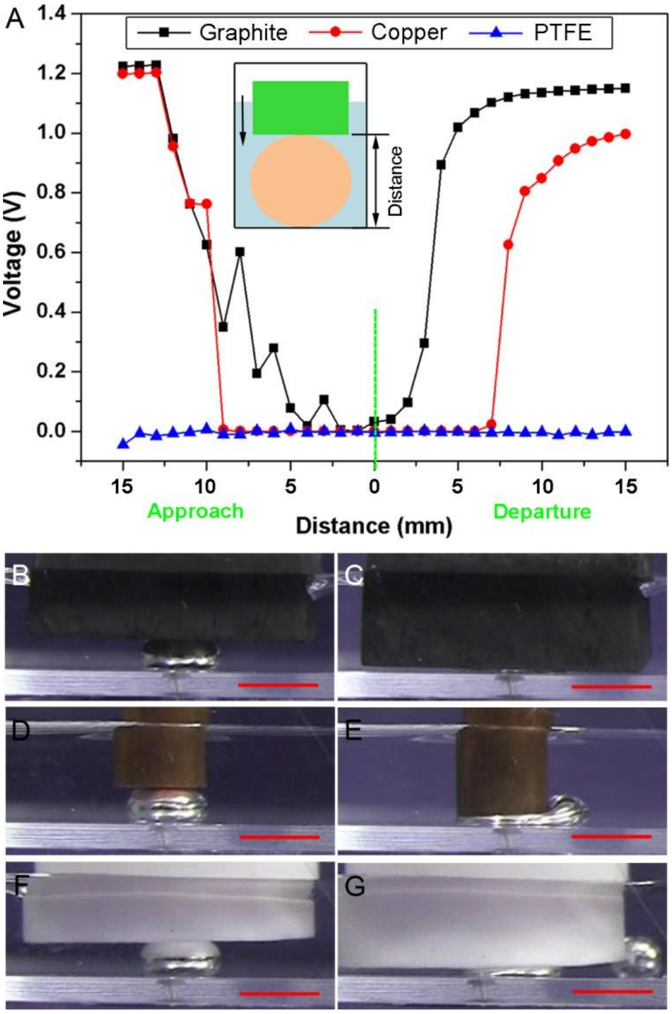

Fig. 4 (A) Voltage-distance relationships between graphite, copper, Teflon and liquid metal when it is pressed. The curves on the left side of the green dotted line represent the approaching process, while the ones on the right side represent the departure process. Inset is the schematic diagram of the experimental setup. (B) Snapshot of liquid metal just contacting the graphite. (C) Snapshot of liquid metal pressed flat by the graphite. (D) Snapshot of liquid metal just contacting the copper. (E) Snapshot of liquid metal pressed flat by the copper. (F) Snapshot of liquid metal just contacting the Teflon. (G) Snapshot of liquid metal pressed flat by the Teflon. Scale bars are all 8 mm in length.

2.3. Self-oscillation phenomenon of liquid metal on the side of the graphite

When a liquid metal sphere is laid on the side of the graphite, periodic oscillation behavior of the sphere can be observed. The substrate is tilted slightly to ensure that the liquid metal sphere could roll toward the graphite spontaneously. The top view snapshots of a liquid metal sphere with 1.2 g (see Movie S5 and S6) are shown in Fig. 5(A), while the side view snapshots of a sphere with 0.9 g (see Movie S7) are shown in Fig. 5(B). Relative variations of the frontal projected area of the 1.2 g sphere and the height of the 0.9 g sphere during oscillation process are shown in Fig. 5(C). The green dashed lines represent the departure of the spheres from the graphite. It can be seen that at the beginning of the oscillation, the liquid metal spheres are in the shape of approximate spheroid with the minimum frontal projected area A_0 and the maximum height H_0. As they are in contact with graphite, the area becomes larger rapidly and the height becomes smaller simultaneously. This is due to the reduced surface tension of the spheres caused by the graphite/liquid metal galvanic cell. Then the area decreases gradually and the height increases simultaneously. When the spheres leave from the graphite, they recover to their original shapes rapidly which is attributed to the surface tension of their own. Therefore the surface tension acts as the restoring force of the oscillatory movement of a liquid metal sphere.

Fig. 6(A) presents the deformation schematic diagrams (images 1, 3 and 5) and the corresponding snapshots (images 2, 4 and 6) of liquid metal spheres in contact with graphite in NaOH solution viewed from the top. The horizontal force diagram of the sphere is shown in the image 1. As the sphere contacts the graphite, it is subjected to the capillary force F_s directed rightward and the friction force F_f directed toward the graphite. The magnitude of F_s which is caused by the surface tension gradient of the liquid metal can be expressed as:[33]

$$F_s \sim \frac{\varepsilon V E_{gl}}{\lambda_D} \tag{7}$$

where E_{gl} is the electric field strength induced by the graphite/liquid metal galvanic cell, V is the potential difference across the liquid metal/electrolyte EDL, ε and λ_D are the electric permittivity and the Debye screening length of the electrolyte, respectively. The friction force F_f includes the viscous force between the sphere and the NaOH solution as well as the frictional force between the sphere and the substrate. $F_s > F_f$ is a necessary condition for pushing the sphere off the graphite and oscillating (see image 1 and 3). Let r represents the curvature radius on the contact point between graphite and liquid metal which is denoted by P. As the surface tension on the left hemisphere is larger than that on the right side, the liquid metal will flow rightward inside the sphere.[4] As a result, r becomes smaller due to the pressure comes from the sphere's surface surrounding the point P. Relative variation of the curvature radius at the contact point P of the spheres with 0.7 g and 1.2 g are presented in Fig. 6(B). The

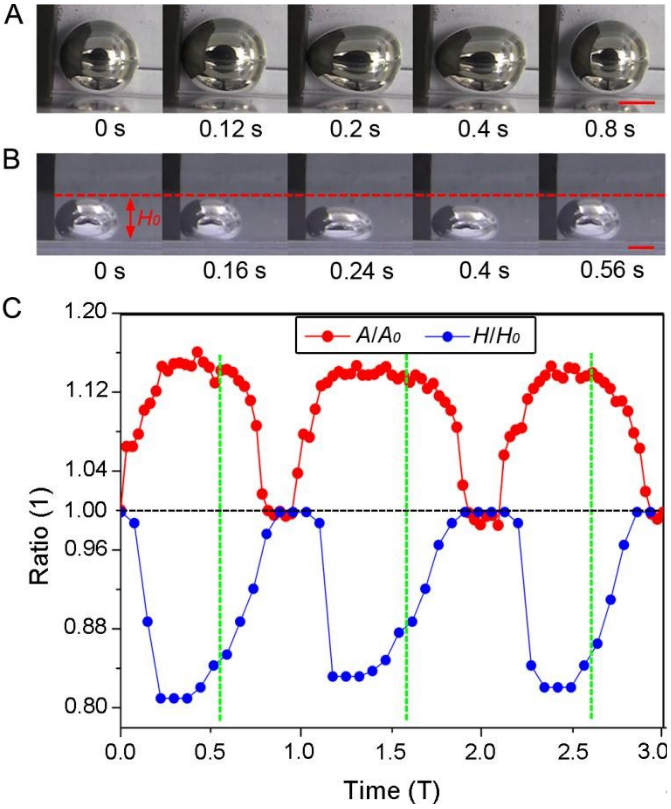

Fig. 5 Oscillation movement of the liquid metal on the graphite substrate immerged in the NaOH solution. (A) Top view snapshots of liquid metal sphere 1 with 1.2 g. (B) Side view snapshots of liquid metal sphere 2 with 0.9 g. (C) Relative variations of the frontal projected area of the sphere 1 and the height of the sphere 2 during the oscillation process. Symbol *A* denotes the frontal projected area of the liquid metal viewed from the top and A_0 denotes the area when *t*=0. Symbol *H* denotes the height of the sphere viewed from the side and H_0 denotes the height when *t*=0. The horizontal coordinate unit T represents the oscillation period of the liquid metal sphere. Scale bars are all 3 mm in length.

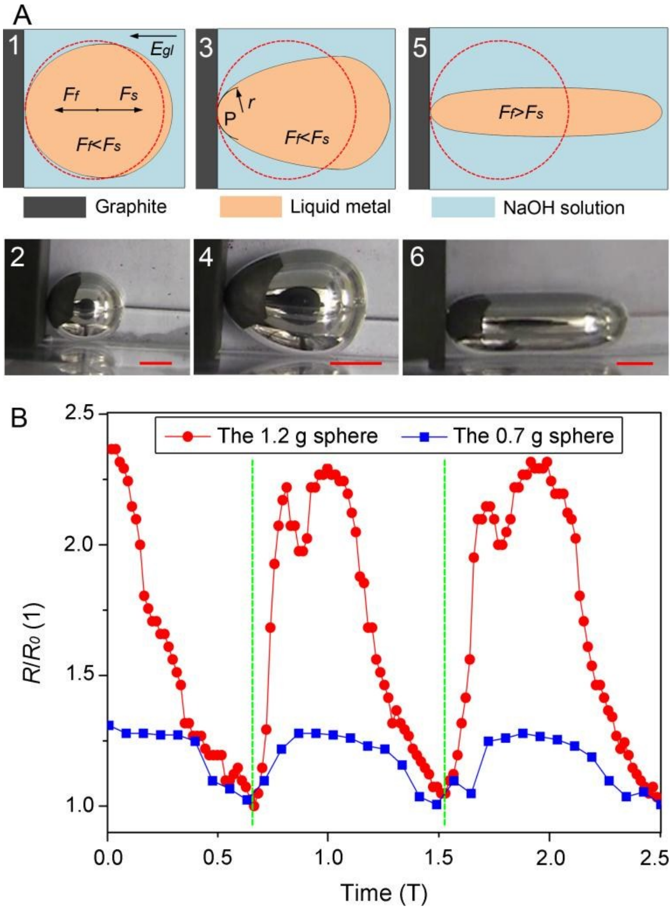

Fig. 6 (A) Deformation schematic diagrams and snapshots of liquid metal in contact with graphite in NaOH solution viewed from the top. The red dashed-line circles represent the outline of the liquid metal before deformation. (B) Relative variation of the curvature radius at the contact point P between the liquid metal spheres and the graphite. Symbol *R* denotes the curvature radius of the contact point viewed from the top and R_0 denotes the minimum value. The horizontal coordinate unit T represents the oscillation period of the liquid metal spheres. Scale bars are all 3 mm in length.

green dashed lines represent the departure of the spheres from the graphite. It can be seen that whatever the mass of the sphere, it detaches away from the graphite when the curvature radius reaches the minimum value. The ratio of the maximum value to the minimum value is 1.30 to the sphere of 0.7 g (images 1 and 2), compared to that of 2.36 to the sphere of 1.2 g (images 3 and 4). This indicates that the sphere with larger mass has larger deformation degree, and it shows a shape of melon seed at the maximum deformation. When F_s<F_f, the capillary force imposed is too small to push the sphere away from the graphite. And the sphere (1.2 g) will be elongated to a wormlike cylinder (see Movie S8) without oscillation as shown in images 5 and 6.

The variation of the oscillation period-time relationship of the liquid metal sphere is investigated and the result is shown in Fig. 7(A). It can be seen that the oscillation period becomes increasingly larger with the time and presents the form of ladder as shown as the blue dashed lines. In the interval of each step there occasionally appear periods with longer time which indicates a slowing down trend of the oscillation movement. The sphere ultimately stops oscillating with slightly but continuously deformation after about ten minutes. However, it oscillates again after the surface layer of the graphite is scraped off. To find the reason, a comparison is performed between the surface of a piece of graphite after oscillation experiment and that of a pure one without experiment. Their scanning electron microscope (SEM) images obtained in the same condition are presented in Fig. 7(B) and 7(C), respectively. It can be seen that the experimented graphite shines brightly and unevenly which indicates the poor conductive of the surface. Energy dispersive X-ray (EDX) is performed to analyze the surface element composition for the graphite after oscillation experiment and the result is shown in Fig. 7(D). There is Na element with 3.18 % by weight on the surface which indicates that some Na$^+$ ions in NaOH solution are adsorbed on the graphite surface. It may be the absorbed ions that destroy the graphite/liquid metal EDL and further weaken the induced electric field strength E_{gl}. The driving force F_s imposed on the sphere is thus reduced to a value lower than the friction force F_f and ultimately results in the termination of oscillating.

2.4. Resonance phenomenon of two liquid metal spheres

As two liquid metal spheres are respectively laid on the top and the side of the graphite, an interesting phenomenon of resonance can be observed (see Movie S9). The vibration of the side sphere denoted as LM2 with 1.0 g can induce the vibration of the top one which is denoted as LM1 with 0.2 g. The schematic diagram and snapshot of the liquid metal double-sphere resonance oscillation experiment are respectively shown in Fig. 8(A) and 8(B). As the side sphere contacts the graphite, its height decreases with the increase of the top one. As the side sphere leaves from the graphite, however, its height increases with the decrease of the top one. This can be seen from the height-time relationship curves of the two spheres presented in Fig. 8(C). The curve crest of LM1 and the curve trough of LM2 correspond to each other which are marked by the green dashed lines. And LM2 is detached away from the graphite at the time corresponds to these lines. Let V_{gl1} and V_{gl2} represent the potential differences across the graphite/LM1 and the graphite/LM2 galvanic cells, respectively. As LM2 is not in contact with the graphite, a loop circuit CL1 is formed which comprises LM1, the graphite and the electrolyte whose equivalent resistance is R_1. The voltage imposed on LM1 is V_{gl1} directed downward, which actuates LM1 to spread. As LM2 contacts the graphite, a loop circuit CL2 is formed which comprises LM1, LM2, the graphite and the electrolyte whose equivalent resistance is R_2. The voltage imposed on LM2 is $V_{gl1}+V_{gl2}$ directed leftward, which actuates LM2 to spread. However, the voltage imposed on LM1 is $V_{gl1}-V_{gl2}$, which is smaller than V_{gl1} and LM1 regains its spherical shape.

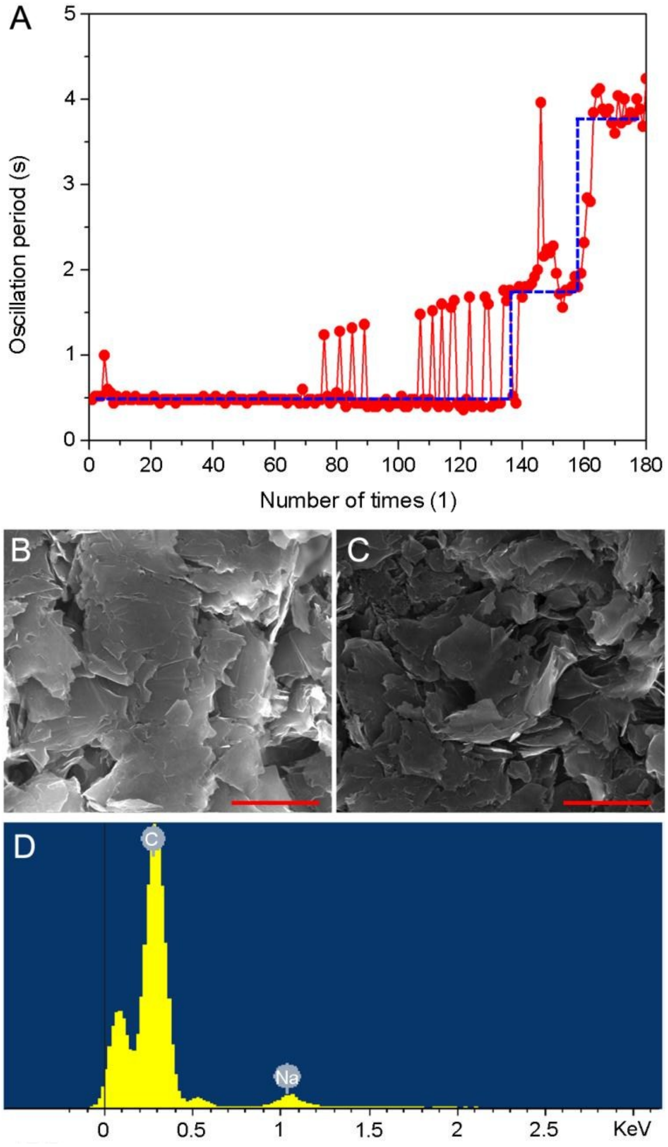

Fig. 7 (A) Variation of the oscillation period of the liquid metal sphere with the number of times. (B) SEM image of the graphite after the liquid metal sphere oscillation experiment. (C) SEM image of the pure graphite. (D) EDX elemental analysis of the graphite after the liquid metal sphere oscillation experiment. Scale bars are all 2 μm in length.

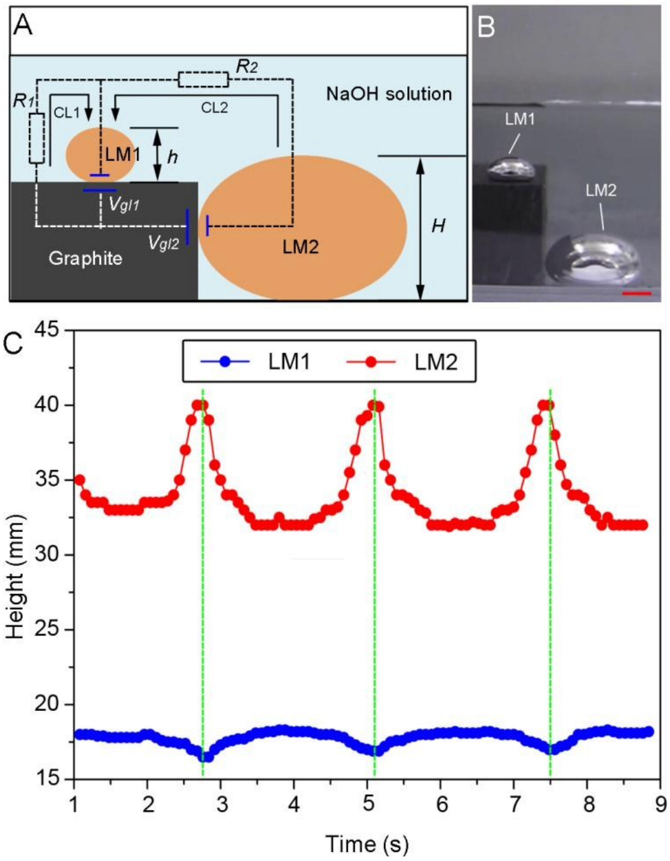

Fig. 8 (A) Schematic diagram and (B) Snapshot of the liquid metal double-sphere resonance oscillation experiment. Scale bar is 3 mm in length. (C) Variation of the heights of the top sphere LM1 and the side sphere LM2 with the time in oscillation state viewed from the side.

As a kind of non-metal conductor, graphite owns more stable chemical properties than common metals such as copper. It cannot fuse or react with the liquid metal to ensure the maintenance of oscillation movement. As to copper, however, a filament will be formed as shown in Fig. 9 when the liquid metal is detached away from it (see Movie S10). The self-actuation of liquid metal demonstrated in this paper is driven by the surface tension gradient which is induced by the graphite/liquid metal galvanic cell. Such mechanism is different from that of traditional mercury beating heat system induced by the oxidation-reduction chemical reaction which has obtained abundant research. Based on the actuation of graphite/liquid metal system, oscillator with complex structure including series-parallel connection can be developed. In addition, graphite/liquid metal-based batteries can also be conceived with a vast application prospect.

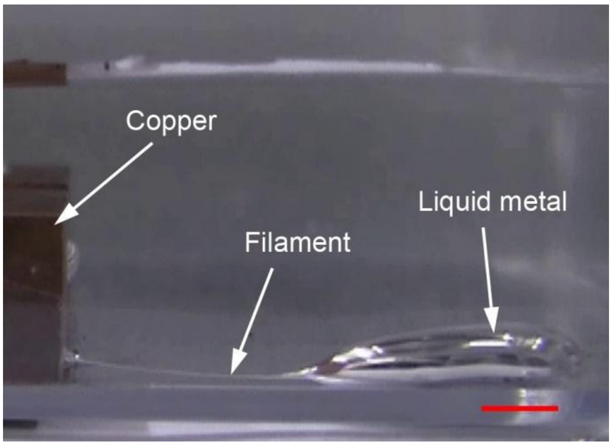

Fig. 9 The adhesion of the liquid metal to the copper. Scale bar is 10 mm in length

3. Conclusions

In summary, the deformation of the liquid metal in the 0.5 mol/L NaOH solution was investigated systematically without external electromagnetic field. When a liquid metal sphere was on the surface of the graphite, it would spread and collapse rapidly. This is due to that a graphite/liquid metal galvanic cell was formed and provided the sphere with a potential directed downward, and the sphere spread because of its reduced surface tension. When a liquid metal sphere was put against the side of the graphite, it would oscillate periodically. Once the sphere was in contact with the graphite, an electric capillary force F_s was induced by the graphite/liquid metal galvanic effect and imposed on the sphere. As F_s was larger than the friction force F_f, the sphere would be detached away from the graphite and the oscillation movement was triggered. And as F_s was lower than F_f, the sphere would be elongated to a wormlike cylinder without oscillating. When two liquid metal spheres were respectively laid on the top and the side of the graphite, a phenomenon of resonance was observed. They oscillated in opposite directions and the vibration of the top sphere was induced by the vibration of the side one. The mechanism of such self-actuation of liquid metal is quite different from that of traditional mercury beating heat system which is ascribed to oxidation-reduction chemical reaction. The oscillating method proposed in the paper provides a novel idea for investigating graphite/liquid metal-based oscillators and batteries, as well as hydrogen production strategy.

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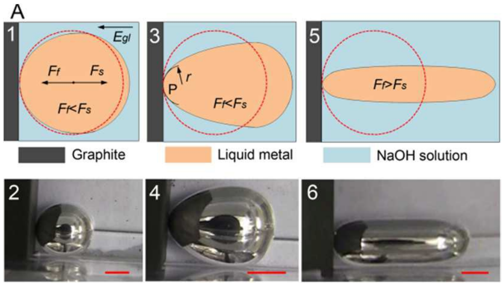

Behaviors of liquid metal spheres in NaOH solution such as spreading, oscillating, stretching and even resonance induced by graphite.