

# Liquid Metal Droplets

Subjects: Others

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Droplets exist widely in nature and play an extremely important role in a broad variety of industrial processes. Typical droplets, including water and oil droplets, have received extensive attention and research, however their single properties still cannot meet diverse needs. Fortunately, liquid metal droplets emerging possess outstanding properties, including large surface tension, excellent electrical and thermal conductivity, convenient chemical processing, easy transition between liquid and solid phase state, and large-scale deformability and so on. More interestingly, liquid metal droplets with unique features can respond to external factors including electronic field, magnetic field, acoustic field, chemical field, temperature and light, exhibiting extraordinary intelligent response characteristics.

Keywords: liquid metal ; droplets ; stimuli-responsive materials ; smart matter

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## 1. Fabrication of Pure Liquid Metal Droplets

Liquid metal (LM) droplets can be obtained in both micro and nanoscale, due to the fast development of nanoscience and fabrication techniques. In general, pure LM droplets can be fabricated by fluidic jetting, or microfluidic flow focusing. The method chosen for fabricating can be decided quickly by the size of target droplets. Fluidic jetting is a convenient and fast method for obtaining LM droplets. Yu et al. <sup>[1]</sup> proposed a method of injecting LM into water solution with added surfactant by using a syringe at room temperature. The mechanism, which was revealed, is that the surface tension of jetting LM stream overcomes the viscous shear stress, causing the bulk LM to break into micro droplets. The LM droplets fabricated by this method were stable and can be obtained quickly. The key to prevent the LM droplets from coalescing and merging together lies in the introduction of the surfactant sodium dodecyl sulfate (SDS) <sup>[1]</sup>. The jetting flow velocity of LM and the diameter of the syringe needle are two factors which can influence the size of the droplets, and the droplets are mostly at the micrometer scale. According to specific need, such a method was also demonstrated to work well to produce wires or a porous structure. In the following years, more fabrication advancements were also reported <sup>[2]</sup>. Overall, fluidic jetting is straightforward, low cost, and has an important value in LM droplet fabrication <sup>[3][4]</sup>. However, the droplet size is difficult to reduce due to the limitation of the needle size.

Microfluidics are widely used in many application scenarios, such as biological analysis, chemical synthesis, single-cell analysis, and tissue engineering <sup>[5][6]</sup>. However, the fluids mainly used in microfluidics are water and oil <sup>[7]</sup>. LM micro droplets can be fabricated by microfluidic flow focusing as well, because of the satisfactory liquidity of LM at room temperature <sup>[8][9]</sup>. A microfluidic chip was presented, which integrates continuous generation of micro scale LM droplets in glycerol <sup>[10]</sup>. Galinstan micro droplets can be produced continuously in glycerol. The NaOH solution is to remove the oxide layer of LM droplets and prevent further oxidation <sup>[11]</sup>. Moreover, the HCl solution has a similar function and can be used in microfluidics as well <sup>[12]</sup>. The main factors affecting droplet size are the channel dimensions and flow rates of fluids. Hutter et al. <sup>[9]</sup> revealed that different flow rate ratios of EGaIn and the continuous phase had an influence on the droplet diameter. Water containing 20 wt% polyethylene glycol (PEG) and 5 wt% sodium dodecyl sulfate (SDS) was used as the continuous phase and the diameter of the nozzle was 40  $\mu\text{m}$ . With the increase of  $Q_c/Q_d$  value, where  $Q_c$  is the volumetric flow rate of the continuous phase and  $Q_d$  is the volumetric flow rate of LM, the LM droplet volumes decreased quickly and the droplet diameters decreased as well. The diameters of LM droplets fabricated by microfluidic flow focusing can be decreased to  $<30\text{ }\mu\text{m}$  by tuning the size of microchannel. Compared to fluidic jetting, microfluidic flow focusing is a more stable method that can fabricate LM droplets continuously, where the size of droplets is more controllable as well.

Fang et al. <sup>[13]</sup> reported that bulk LM can be separated into discrete droplets by electro-hydrodynamic shooting. When the capillary tube filled with bulk LM was on a direct current (DC) electric field, the bulk LM shot into the solution and formed droplets continuously. The shooting velocity increased as the voltage increased, and the droplet size depended on the diameter of the capillary nozzle. Electro-hydrodynamic shooting provides a convenient way to produce LM droplets quickly, but the diameter of the capillary tube limits the size of droplets.

Metal droplets can be fabricated by ultrasonic cavitation when low-melting-point metals mix with hot silicone [14][15]. Similarly, ultrasonic cavitation is also suitable for fabrication of LM micro/nanodroplets [16][17][18][19]. Organic solvent added to bulk LM forms an immiscible liquid system, and the ultrasonication process induces the dispersion of LM and formation of droplets. Ren et al. [17] reported stable Galinstan droplets with average diameters of 110 nm, which can be suspended in solution for several weeks. The ultrasonication process induced the rapid oxidation of Ga, and the oxide layer covered the droplet surfaces. Moreover, the thiol ligands (R-SH) added into the ethanol solution formed an organic matter layer on the Ga oxide layer by a self-assembly process. The double layer structure protected the LM nanodroplets from coalescence in the neutral solution or atmosphere. The LM droplet diameters depend on the temperature and ultrasonication time [18]. A smaller LM droplet can be fabricated by lower temperature or longer ultrasonication time. However, there is a size limit for LM droplets under different ultrasonication time. Ultrasonic cavitation is a widely used method for fabrication of LM nanodroplets, and is worth further understood in addressing how to break their size limit.

Zhang et al. [20] reported a convenient method for fabricating LM droplets by utilizing an airbrush. The bulk LM was rapidly squeezed through the fluid nozzle by high-pressure air, and then separated into LM droplets. Depending on the diameter of the nozzle, the droplet diameter ranged from 700 nm to 50  $\mu\text{m}$ . The mechanisms of atomized spraying and fluidic jetting are similar; the bulk LM separates when squeezed out of a narrow nozzle and forms micro/nanodroplets due to its high surface tension. Atomized spraying provides a convenient and fast way of LM droplet fabrication in the atmosphere at room temperature. However, because the droplets coalesce rapidly and are difficult to collect, atomized spraying is commonly used to fabricate LM electronics on flexible substrate. Shearing liquids into complex particles (SLICE) is a simple method used to make LM droplets by utilizing emulsion shearing with oxidation. By utilizing mechanical force, bulk LM was broken to small droplets with concomitant surface oxidation and functionalization [21][22]. The LM droplets fabricated by SLICE can be 6.4 nm to over 10  $\mu\text{m}$  in diameter, which is related to the magnitude of shear force. SLICE provides a low-cost, green, facile, and versatile method that can obtain LM micro/nanodroplets of tunable sizes, shapes, compositions, and surface morphologies. However, SLICE is worth being further investigated because the distribution of droplet diameter is uneven. Furthermore, physical vapor deposition based on evaporation and deposition is also an effective preparation method of LM nano droplets, and the droplet size of LM droplets is small and uniform. As Yu et al. [23] report a way to synthesize surfactant-free LM nanodroplets with controlled particle size on a variety of substrates through a facile physical vapor deposition method.

## **2. Liquid Metal Droplets with Core-Shell Structure**

LM droplets with core-shell structure are a nanoscale ordered assembly structure obtained by LM core coated by another nanomaterial through chemical bonds or other forces [24][25]. By utilizing surface modification, LM droplets with core-shell structure can be obtained on the basis of the pure LM droplets. The properties of core-shell structure depend on the core and shell materials, concentrate on the advantages, and make up for disadvantages as well. Moreover, because of forming rapidly, the native oxides of Ga play an important role between LM core and shell material. Generally, Ga and its alloy are covered by a thin oxide layer as Ga can be oxidized easily and rapidly when exposed to the air or neutral solution [26]. Contrarily to the strong surface tension of LM, the oxides of Ga have a much smaller surface tension and greatly change the wettability between LM and substrate. For LM droplets with core-shell structure, the oxide layer can also be considered like a shell, which can connect other materials stably. The LM droplet with core-shell structure fabricated by SLICE is composed of LM core, oxide layer and surfactant shell [22]. The oxide layer highlighted by false-colorization is fractured by an external force.

Zavabeti et al. [27] created a variety of low-dimensional metal oxides by utilizing LM droplets with core-shell structure as a reaction environment. 1 wt% of hafnium (Hf), aluminum (Al), or gadolinium (Gd) was alloyed into LM droplets, and their oxides were easier to form than oxides of Ga. By touching the LM droplets with a solid substrate, the metal oxides adhered to the substrate and were separated from droplets because of the different phase between the LM droplets and solid oxides. Moreover, LM droplets with core-shell structure also can be used as nanomedicines by modifying their surfaces with functional layers. ZrO<sub>2</sub> coated LM nanodroplets were fabricated for photothermal therapy [28]. The ZrO<sub>2</sub> shell can effectively prevent the droplets from coalescence and size variation, and PEG was used to improve the biocompatibility of the droplets. Thus, droplets warm up rapidly under near-infrared (NIR) laser radiation. LM droplets with core-shell structure are confirmed to be a promising vehicle for nanotheranostics, and have attracted widespread attention. In addition, the oxide layer of LM droplets has been measured from 0.5 nm to 5 nm in thickness [29]. As the LM droplet diameter decreases, the mass fraction of LM core decreases as well. It is possible that Ga is completely oxidized and that LM droplets lose original properties and functions. Farrel et al. reported a method to successfully control the growth of a Ga oxide layer on LM nanodroplets by adding thiolated molecules [30]. It was indicated that thiolated

molecules can moderate the growth of Ga oxide via competition with oxygen for surface sites. The results are of great significance for fabricating multifunctional nanodroplets.

LM droplets with core-shell structure also can be fabricated by electroplating. Zhang et al. reported a magnetic soft motor with core-shell structure by electroplating a Ni cap <sup>[31]</sup>. Al foil was added to the droplets as the on-board fuel. The LM droplet can continuously move with a velocity of  $3\text{ cm}\cdot\text{s}^{-1}$  for hours without an external energy source. As a self-propelled droplet motor, it can be controlled under an applied magnetic field or electric field. However, the accurate control of this droplet movement needs to be further known and has important significance in drug delivery. Tang et al. synthesized a LM droplet with a LM core and a Cu shell by electrochemical reduction <sup>[32]</sup>. The preoxidized Cu nanoparticles were first coated on a LM droplet to obtain a LM marble. After alkaline solution was added to the system, the color and shape of the LM marble changed, which is a signature phenomenon of the electrochemical welding process. It was demonstrated that an effective method to cross-link various types of particles by the redox reaction. Thus, it was indicated that LM droplets with core-shell structure can concentrate the properties of core and shell materials. Multifunctional LM droplets can be obtained by adjusting the properties of either core or shell material. In the future, it is necessary to further understand the core-shell interface to obtain more stable droplets. Moreover, it is also expected to achieve more functions by manufacturing droplets with more complex structures, such as controllable motors, droplet micro reaction environments, nanorobots, and so on.

### **3. Liquid Metal Marbles**

LM marbles are droplets that micro- or nanoparticles coat on the LM core. Generally, LM marbles belong to LM droplets with core-shell structure and are a rather prominent category, so it was introduced them in a separate section. The oxide layer of LM formed easily in ambient air has a much lower surface tension than LM, and can adhere strongly to glass, cloth, and silicone <sup>[33]</sup>. In addition, LM is highly corrosive to other metals, which limits its practical application <sup>[34]</sup>. To deal with these problems, it was coated LM droplets with various micro- or nanopowders. Sivan et al. <sup>[35]</sup> introduced LM marbles formed by coating nanoscale powders on the surface of LM droplets. By rolling the LM droplets on a powder bed, LM marbles coated with insulators and semiconductors such as SiO<sub>2</sub> and WO<sub>3</sub> can be obtained. LM droplets formed tips because of the surface oxidation of LM droplets. However, LM marbles coated with WO<sub>3</sub> powder maintained their original spherical shapes, similarly to those treated with HCl solution. In addition, LM marbles coated with Al<sub>2</sub>O<sub>3</sub> powder can suspend on the surface of water.

Chen et al. <sup>[36]</sup> reported a highly elastic and movable LM marble by coating LM core with polytetrafluoroethylene (PTFE) particles. The results of bouncing tests indicated such LM marbles can bounce nine times after falling from the initial height. However, the LM droplet falling from the same initial height stuck to the substrate without any bouncing. Furthermore, the LM marble could scroll on a substrate with an inclined angle of 14°. Chen et al. <sup>[37]</sup> presented a magnetically controllable LM marble, which was coated with ferronickel (FN) and polyethylene (PE) microparticles. The LM core was treated with NaOH solution to remove its oxidized layer and then mixed with FN and PE particles. It was indicated the LM marbles have no corrosive residues on the surfaces of different materials after 12 h. In particular, such LM marbles can be controlled by a magnetic field due to the addition of the FN particle.

Further, Liang et al. <sup>[38]</sup> reported a fluorescent LM marble as a transformable biomimetic chameleon. Fluorescent LM marbles can be fabricated by encasing them with fluorescent nanoparticles. The exhibited color of such LM marble depended on the fluorescent particles. The different colored fluorescent LM marbles could merge together to form a biggish ultimate multicolor LM marble. One entire LM marble could be divided into several small LM marbles as well. The fluorescent particles could also be released from LM marbles under an electric field by adjusting the distribution of the oxide layer. Furthermore, the electric field could lead to a controllable movement of the fluorescent LM marble in a basic electrolyte as well. LM marbles can also be used as nanomedicine <sup>[24][39]</sup>. Hu et al. <sup>[40]</sup> synthesized a LM marble coated with glucose oxidase (GOX) for tumor treatment. GOX can consume intratumoral glucose by oxidation in order to suppress tumor growth. LM has an outstanding photothermal conversion ability and good biocompatibility. By utilizing such LM marbles, Hu et al. brought the two methods together and found that combinational therapy including starvation and photothermal therapy realized outstanding therapeutic effects. Generally, LM marbles can greatly improve the mechanical properties of LM droplets and avoid the corrosivity of LM. In the future, it is necessary to research the method of further kinds of nanoparticles that can be coated on the surface of LM droplets.

### **4. Self-Powered Liquid Metal Droplet Motors**

A self-propulsion phenomenon of LM droplets has been discovered by adding Al. Zhang et al. <sup>[41]</sup> introduced a self-fueled LM motor as a biomimetic mollusk. The LM motor could “eat” Al food and move spontaneously in NaOH solution for more

than 1 h. The bubble recoil force and the surface tension gradient induced by the galvanic reaction propelled the motor forward. The movement direction is opposite to the bubble injection direction, and the velocity can reach up to  $5\text{ cm}\cdot\text{s}^{-1}$ . A large LM motor can be divided into several small ones that can maintain original motion characteristics. In contrast, many small LM motors will coalesce if they collide in the movement [41][42]. Yuan et al. [3] found the random motion of LM motors resembled the Brownian motion. Millimeter scale motors were fabricated by injecting the LM composed of 1 wt% Al and 99 wt% GaIn10 into NaOH solution. The LM motors could roll forward on the substrate propelled by the bubble recoil force.

Under an electric field, these LM droplet motors could move in a controllable direction and speed [43]. The motion trajectory was similar to the distribution of the electric field in solution, and the velocity could reach up to  $43\text{ cm}\cdot\text{s}^{-1}$  under 20 V in a channel filled with aqueous solution. It is an effective method to drive the droplet motor to move at high speed. If the electric field is changed into a magnetic field, the self-powered LM droplet motors will become trapped in the boundary zone of the magnet due to the Lorentz force [44]. Furthermore, the LM droplets with added Al could realize high frequency self-powered oscillations via redox reaction when placed on an iron plate [45]. The oscillation behavior had a frequency of 8 Hz and shows the potential for developing self-powered soft oscillation. Generally, the addition of Al provides a novel strategy for fabricating self-powered LM droplet machines. This method is also expected to promote the development of multifunctional soft droplet machines.

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