Experimental analysis of liquid metal galinstan for electronics actuation

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The experimental interfacial tension measurements of galinstan as a function of applied DC voltages are presented. The measurements are based on a tailored, modified pendent drop setup to make it suitable for this experiment. The findings rationalise Galinstan corresponding to Marangoni flow that actuates under the influence of a change in interfacial tension, resulting from applied voltages. Galinstan is substantiated as an actuation medium to enable ON or OFF states by using a very low applied voltage (~ 0.8V). The illustrated nature of measurements performed in this work has significance for a variety of reconfigurable RF applications, by enabling well-controlled actuations.

Introduction: The liquid metal alloy constituted of gallium, indium, and tin is commercially launched as galinstan by Geratherm Medical AG. Galinstan exists in liquid state at room temperature (a melting point of −19°C) and offers remarkable properties such as high electrical conductivity (3.46×10^6 S/m), extremely low vapour pressure, and good thermal conductivity without posing a safety or toxicity concern [1, 2]. It is, therefore, galinstan emerges as a versatile material for electrochemical research and a potential surrogate material for liquid metal-based RF reconfigurable applications such as resonators, switches, tunable frequency selective surfaces (FSS), power electronics, and a coolant for high temperature devices and applications [3–6].

Despite several attractive traits and properties, the use of galinstan for practical electronic/RF applications is limited by its tendency to get readily oxidised when exposed to air (oxygen in particular) and forms gallium oxide at the surface. The oxidation layer is only a nanometer (nm) thick, but it is non-perforated. This serves as a protective outer layer to inhabit the oxidation of the alloy underneath. The oxidation layer makes galinstan behave like an adhesive gel-like material, as opposed to a perfect liquid [7, 8]. This deviation from a runny liquid behavior is more pronounced at small droplet level, due to higher surface area to volume ratio. The oxidation layer can be stopped from forming by storing galinstan in an inert gas or liquid nitrogen. This sets an inherit limitation to the use of galinstan, for actuation purposes, and subsequently its use as a reconfigurable medium.

The ionisation of oxide layer, in the presence of an electrolyte solution, varies the interfacial tension of galinstan, and this phenomenon can be exploited to switch the material between a gel like substance and a runny liquid. This behaviour adversely influences the retraction mechanism where galinstan is solely used as a reconfigurable medium. The visco-elastic gel like behaviour on a glass surface is shown in Figure 1(a) and (b). The residues can be seen on the glass surface due to adhesion between glass and galinstan. However, when it is used in combination with aqueous sodium hydroxide (NaOH), it immediately turns into a droplet, as shown in Figure 1(c) and (d). The interfacial tension can be also effectively controlled by an external stimulus such as DC voltage.

Galinstan is still an infantile material for its use in reconfigurable RF applications. The interfacial tension measurements of galinstan as a function of applied DC voltages are of significance and facilitates the implementation of galinstan-based reconfigurable systems. Such studies are of valuable importance to provide the bases for its Marangoni fluid nature, by varying the interfacial tension, upon applied voltages [9, 10]. The concept can be effectively employed for a variety of RF reconfigurable applications [11, 12].

Although galinstan's affinity to oxidise limits its usability, the effect can be exploited in micro-switching, by controlling and manipulating the oxidation layer and in turn its flow properties. This can be achieved by immersing galinstan into an electrolyte solution, such as an aqueous solution of NaOH. The behaviour of galinstan, enclosed in an aqueous solution of NaOH (0.5 molar solution), and its electronic actuations for possible RF reconfigurable applications, are investigated.

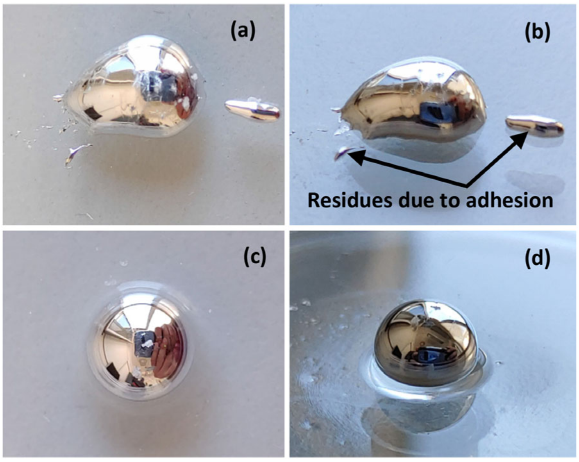

Fig. 1 *Oxidised galinstan turning into a gel like substance, top view (a) and side view (b), galinstan turns into a droplet in the presence of aqueous NaOH, top view (c) and side view (d)*

In the next section, the theoretical background behind the actuation of galinstan's interfacial tension is discussed. The measurement setup and the corresponding interfacial tension of galinstan in NaOH environment as a function of applied voltages are then elaborated. The work is concluded by substantiating galinstan as a Marangoni fluid, capable of actuating under the effect of an external stimulus (discretely applied voltages) and its suitability for reconfigurable RF applications.

Theoretical background: A liquid can be made to flow by creating a change in interfacial tension along the interface between two fluids or the same fluids with different physical properties at the boundaries. The resulting flow caused under the influence of such conditions is termed as 'Marangoni flow'. The 'tears of wine', phenomenon caused by change in alcoholic concentration, is an example of Marangoni effect [13]. The interfacial tension gradient or difference can be achieved in a variety of manners such as temperature difference, addition of surfactants, and concentration difference [14, 15]. The greater the change in interfacial tension, the higher would be the flow rate. Marangoni flow is proportional to change in interfacial tension, as shown in expression 1 [16, 17]. $\Delta\gamma$ is the interfacial tension difference ($\Delta\gamma = \gamma_0 - \gamma_1$) and u is the corresponding flow velocity. γ_0 and γ_1 correspond to initial (at 0V) and final state of interfacial tension (at a certain applied voltage), respectively.

$$u \propto \Delta\gamma. \tag{1}$$

In this work, the change in interfacial tension is carried out by using electric potential, leading to Marangoni flow. This change in interfacial tension phenomenon upon applied voltages is attributed to its electrowetting properties at the surface. Galinstan interfacial tension in NaOH solution is attributed to formation of gallium hydroxide ions. Upon immersion in an electrolyte solution the surface of galinstan is negatively charged and tends to attract the positively charged ions to itself. The presence of oppositely charged particles (ions) in the proximity can be viewed as a capacitive effect. This phenomenon causes an emergence of an electric double layer (EDL) capacitance. Applying a positive voltage across the interface weakens the interfacial tension and galinstan actuates towards the positive applied voltage. It is worth mentioning that the electrolyte solution has a very low conductivity (~ 15 S/m), in comparison to galinstan, and this causes a potential difference along the length of the channel, thus a voltage difference is created. The applied voltage induces an imbalance or change in interfacial tension $\Delta\gamma$, and leads to a pressure difference (Δp) between the two hemispheres of the droplet (one side closer to the positive potential and the other side farther from the positive potential) surrounded by electrolyte. A higher potential difference (ΔV) at the interface results in a higher difference in interfacial tension and subsequently a higher pressure difference is created ($\Delta V^2 \propto \Delta \mathrm{p}$). The pressure difference in turn causes the flow of galinstan.

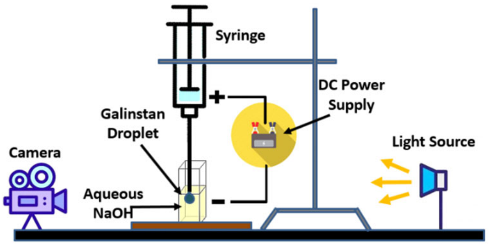

Fig. 2 *Block diagram of the improvised Pendent Drop (PD) method*

Interfacial tension measurements and discussions: Interfacial tension (γ) is the property of a liquid that makes it behave as if the surface is enclosed in an elastic skin [18]. This phenomenon assigns a spherical shape to liquid droplets and subsequently causes a convex or a concave profile at the surface of a liquid contained in a capillary tube.

The voltages across the interface are changed in a progressive manner. It is challenging to determine the electro-wetting properties of galinstan. A probe cannot be directly inserted into the liquid metal, due to higher interfacial tension, while maintaining a stable electric contact. The droplet surface contact angle (a measure of adhesion at the interface between a liquid and a solid surface; an alternative technique to measure interfacial tension) changes, from its neutral state and flattens, as a probe is inserted into the material, due to physical contact. This can lead to erroneous results and affects the accuracy of the interfacial tension [19].

Since the objective of this work is to evaluate the usability of galinstan enclosed in an aqueous NaOH solution, for RF applications, the measurement of interfacial tension between the two immiscible liquids is of special interest. Some of the common techniques used to measure the interfacial tension of fluids are the Nouy-ring method, the Wilhelmy-slide method, and the pendant drop (PD) method [20, 21]. PD method is a robust technique in comparison to Nouy-ring method and the Wilhelmy-slide method, however, it is experimentally complex, and involves mathematical post-processing [22]. In this work, the PD method is chosen, due to its ability to offer better accuracy of the results, and the availability of the setup.

A typical PD measurement setup is comprised a light source and optical arrangements to enable a camera to record the images in high quality. The interfacial tension is computed from the shape of the drop by making it comparable with the upward force, caused by the gravitational acceleration (g) acting on it. The accuracy of the results depends on the selection of images, taken at the time when the droplet is about to detach from the dosing needle under the influence of gravity. These recorded images are post-processed, and the shape of the drop is used to determine the interfacial tension. A schematic of PD method is shown in Figure 2. One of the prominent advantages of PD method is that a very small volume (a few millilitres) of the material under investigation is required to perform the measurements. It is of significance for this experiment due to several sets of iterative measurements performed in a row, which corresponds to different DC voltages.

A state-of-the-art PD measurement setup by ‘DataPhysics Instruments’ is used to perform the experiment. The setup is improvised using a precise computer-controlled digital DC power supply to progressively apply the voltages at the interface between the two immiscible liquids. A small quantity of galinstan (77.63 μl) is filled in a glass syringe. A high precision and high quality SNS 052/026 stainless steel dispensing needle with outer diameter, $d_{\text{outer}} = 0.52$ mm and inner diameter, $d_{\text{inner}} = 0.26$ mm is used to carry out the experiment. The needle material is chosen to ensure that galinstan does not wet the needle surface. Wetting process not only leaves the residues behind but also causes adhesion to the walls of the needle, which can affect the accuracy of the results. The needle is made of stainless steel and causes no adhesion with galinstan. The needle’s tip is submerged in a transparent cuvette. The cuvette is thoroughly washed and dried with nitrogen gas, to ensure there are no contaminates, before it is filled with 0.5 M aqueous NaOH solution. The dispensing system is computer-controlled and provides excellent control over the dosing volume. The steady dosing rate is important to provide accurate results. A slow and steady dosing rate results in an ideal droplet formation, which is used to extract the interfacial tension value. A computer-controlled power supply provides the voltage across the needle and the NaOH aqueous solution interface. Extreme care is taken to minimise external influences such as vibration and external light to ensure the accuracy of the results.

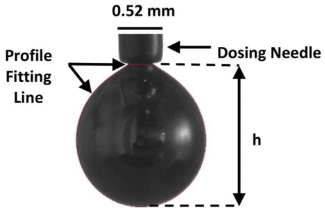

Fig. 3 *Post-processing of the image to extract interfacial tension*

All practical aspects are carefully considered to achieve accurate results, while designing the experiment. The dispensing is performed at an extremely slow and steady rate (~ 0.15 μl/s) in small steps to obtain a drop profile entirely under the influence of gravity. A slow dosing rate emulates quasi-static state and influences the accuracy of the results. Since the interfacial tension measurements are obtained from the post analysis of the images, therefore, a lot of attention is paid to the image quality and resolution. The beam width of the light source is kept larger than the camera’s field of view, and the camera is levelled properly using a liquid leveller. The setup could not be entirely isolated from the vibrations, due to the non-availability of a vibration-isolation platform. The setup was semi-automated by using a computer-controlled dispensing system, computer-controlled power supply, and image capturing software, to minimise the external influences.

The captured images were used for further processing and analysis. In addition, only big enough droplets that were around 60% or more of the projected screen area were considered for post-processing. The captured images were recorded in bitmap file format. The external diameter of the needle was used as a reference scaling factor to evaluate the parameters of the droplet. The self-contained scaling factor estimation is one of the primary strengths of PD method over similar interfacial tension measurement techniques.

The voltage is changed in small discrete steps (100 mV), prior to engaging the dispensing system. The voltage is applied in the range from 0 mV to 800 mV. The interfacial tension measurements of galinstan immersed in aqueous NaOH solution, beyond 800 mV it is not possible to compute because galinstan cannot retain a droplet profile and turns into a runny liquid beyond 800 mV [23].

A Java-based image processing program called ImageJ is used for post-processing of the images [24]. The software tool analyses the shape of an axi-symmetric drop hanging from a capillary tube. The software matches the theoretical profile to the contour of a pendent drop, by applying a profile match. It provides a good estimate of the interfacial tension, volume and/or surface area from the matched profile [25]. The working principal of ImageJ is based on computing the hydrostatic pressure (Δp) caused by g that distorts the shape of a drop from a perfect sphere. A droplet obtained using PD method, during the post-processing using ImageJ, is shown in Figure 3. The accuracy of the post-processed results primarily depends on how accurately the captured droplet’s profile fits to the theoretical contours of an arbitrary droplet. The profile fitting curve is shown as a thin red line in Figure 3. In other words, it provides an inherent quality check mechanism to whether the droplet meets the profile of a freely suspended droplet.

As the droplet profile deviates from a standard shape, the corresponding measurements tend to be erroneous. This is observed at 800 mV and beyond. It can be seen in Figure 4(a) that the droplet deviates from a freely suspended droplet profile. The deviation in profile estimation can be seen at the point, where the droplet tends to detach from the needle. This shows that the balance between the intermolecular forces (interfacial tension) and gravity is no longer at equilibrium. Galinstan behaves as a runny liquid, at 900 mV and beyond as shown in Figure 4(b). Therefore, it is not practical to determine the interfacial tension properties of galinstan accurately and reliably beyond 800 mV.

The interfacial tension values of galinstan in an aqueous NaOH solution, in the voltage range from 0 mV to 800 mV are extracted and plotted in Figure 5.

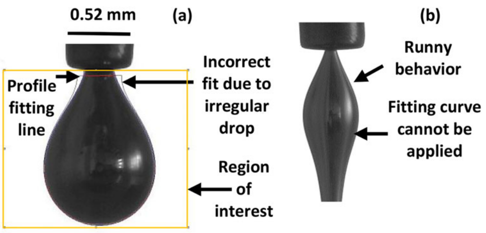

Fig. 4 *Deviation from an ideal droplet shape, and the corresponding erroneous profile curve (a), a runny fluid behaviour of galinstan beyond 800 mV (b)*

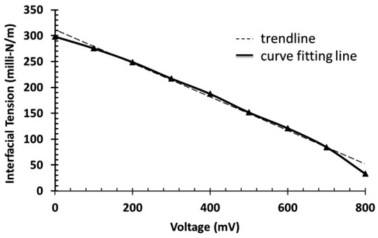

Fig. 5 *Interfacial tension between galinstan and aqueous NaOH solution versus applied voltage*

An error of roughly 10% is estimated based on several factors such as pixels and quality loss during the conversion of images from bitmap to jpg format, limitation of ImageJ to not take input data beyond the third significant digit, and a non-availability of a vibrations-isolation platform. Despite that, a clear decreasing interfacial tension trend is observed, as the voltage is progressively increased (step size of 100 mV). The decreasing interfacial tension trend or in other words interfacial tension difference leads to a flow of galinstan in a channel, according to the expression 1 ($u \propto \Delta\gamma$). The results are in agreement with similar scientific findings that the interfacial tension can be reduced to near-zero due to voltage induced oxidation of galinstan in NaOH solution [26]. Hence, galinstan corresponds to Marangoni fluid phenomenon under the influence of an applied voltage. A trend line for the discretely measured values (100 mV interval each), is shown in Figure 5. The trend line shows that there is a decrease in interfacial tension as the potential difference across the droplet interface is gradually increased.

Conclusion: The theory and physical phenomenon that qualify galinstan as a Marangoni fluid are investigated. Such measurements provide an experimental basis for galinstan and its applications for micro-actuations. The investigation leads to the use of galinstan as a reconfigurable medium for RF applications. In addition, the interfacial tension values are quantified for each voltage. This can serve to estimate flow rate for a given dimension of the reservoir, in which galinstan and NaOH are enclosed. It can be clearly inferred that the approach to introduce a difference of interfacial tension using a corresponding potential difference can be used to perform actuations for RF components such as antennas, filters, and resonators.

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