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# Role of chemical reaction and drag force during drop impact gelation process

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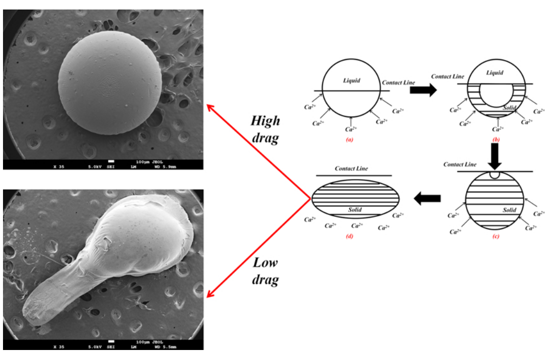
## GRAPHICAL ABSTRACT

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## ABSTRACT

An instantaneous chemical reaction and solidification have been experimentally explored during sodium alginate drop impact on calcium chloride liquid pool through a high-speed imaging system. Here, different concentrations of sodium alginate are used to study the effect on gelation process. As soon as the alginate drop touches the calcium chloride pool, a crosslinking polymerization reaction starts under the liquid pool surface. The consequence of crosslinking is rapid formation of calcium alginate gel which denotes instantaneous freezing of liquid drop. The drop impact solidification immediately results in the formation of a strong gel layer in front of it which is having significant role in spherical shape recovery. This layer formation is favoured by increasing sodium alginate concentration in the drop and thus crater growth is hindered also. The initial reaction time which is determined as soon as the drop touches the surface, is found to be dependent on the drop kinetic energy. However, the drop gelation time reduces with drop concentration and it controls the final shape and morphology of gel. Such reduction in gelation time is inferred from the enhanced rate of crosslinking reaction. The motion of liquid drop through liquid pool while interacting with the surroundings influences the drag around the spherical drop. The shape evolution in the solidifying gel is governed by the dynamics between the fluid drag and chemical reaction. The morphology of the gel surface has also been studied and its roughness variation has been measured from fractal box counting method.

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## 1. Introduction

Modern day applications show lots of involvement of phase transition like solidification, freezing, boiling, evaporation, liquefaction, melting etc. Freezing is a type of solidification when liquid changes into solid as its temperature is lowered below its freezing point. Boiling is extensively studied in the scientific community for its wide range of practical applications [1,2]. Similarly, extensive studies on boiling, evaporation, Leidenfrost effect are available in quite abundance [3–5]. Solidification in the context of supercooled drop is also studied but is limited to cold and solid surfaces [6–9]. However, solidification study on liquid pool is not abundantly found in existing literature. Here, the difference is in the absence of latent heat dissipation, chemical reaction takes place. Thus, it can be termed as solidification as there is no observation of drop temperature depression.

The application of phase change is an obvious phenomenon observed in different applications like encapsulation, drug delivery, cosmetics, food application [10–13]. The requisites of this concept are drop impact chemical reaction and subsequent phase change. Rather, it becomes very interesting to study while a liquid drop upon impacting on liquid pool reacts and solidifies suddenly. Hence, it is very much scientific to acquire the in-depth physics involved during this drop impact chemical reaction and phase change.

The crater which is formed under the liquid pool surface is an interesting aspect which is studied under the hydrodynamics part of this research. Another feature is the solidification phase change which is a direct consequence of the crosslinking chemical reaction upon liquid drop impact on liquid pool. Even the distinctive effect of chemical reaction on crater dynamics is newly studied by comparing with non-reactive drop impact on the same liquid pool. Dynamics of crater has been studied since very early ages by Engel [14,15], van de Sande [16] Ogawa et al. [17], Berberovic et al. [18] and very recently Michon et al. [19]. These studies are limited to the energy transition during crater formation and the theoretical prediction of the maximum crater depth.

The phase change event during wax formation only was studied also in the context of fluid dynamics [20]. On the other hand, the gelation studies are found in the works of various researchers but not related to fluid dynamics. Very recently, Davarcı et al. [21] have analysed drop dynamics study in context of gelation by varying the viscosity and surface tension of liquid pool. They used high-speed camera and measured the penetration depth of solidified gel. They also found that highly viscous calcium chloride solution reduces the sphericity of the gel. The relation of vortex ring formation during the gelation process has been found by Bremond et al. [22]. The effect of viscosity and impingement height on the critical shell thickness of the gel has also been found from their work. Also, the effect of gelation time, sodium alginate and calcium chloride concentration on the thickness, diameter of the gel during oil encapsulation has been done by Abang et al. [23]. The role of hardening time, temperature, concentration of calcium chloride solution on the size, shape and morphology of calcium alginate gel have been also optimized by Smrdel et al. [24]. Even the different shape evolution of gel-like mushroom, teardrop, ring, sphere has been observed by Zhang et al. [25]. Rezende et al. [26] also determined the influence of sodium alginate on the gelation process regarding the application of tissue engineering. Deng et al. [27] worked on the bubble entrapment with the gel just after the chemical reaction between alginate and calcium chloride. The reason for bubble entrapment is the crater surface collapse due to bulk focusing flow around the frozen crater bottom. The drop of Ohnesorge number above 0.29 produces spherical shaped gel bead post impact on the liquid pool [28]. Meiser et al. [29] studied the time -dependent variation of bead velocity, horizontal width, and bead acceleration during clinical grade microencapsulation using high-speed camera. However, Tien-Chu Lin [30] proposed that the final shape of the gel is dependent on the sodium alginate concentration and the droplet penetration only. The simultaneous spreading and gelation phenomena of polymer drops and its dependency on substrate temperature have been studied by de Ruiter et al. [31] Also, the role of calcium chloride concentration on gel formation and swelling has been experimentally and theoretically found recently [32].

The objective of the current work is to study the effect of sodium alginate concentration on the drop impact gelation process and shape formation of the final calcium alginate gel. The hydrodynamic features have been determined, and it is related to the chemical reaction during gelation process. Also, the shape and morphology of the solidified gel are convoluted from the idea of fluid drag during the movement of spherical liquid drop through another liquid. The novelty of this work is the correlation of chemical reaction with the fluid dynamics.

## 2. Experimental methods

The experiments are conducted using a glass beaker (Borosil), drop dispensing system and a high-speed camera (Phantom V 7.3 of Vision Research Inc.) (Fig. 1). The drop dispensing system consists of a Rame-Hart microsyringe (p/n 100-10-20), held by an aluminium stand and driven by a stepper motor. A stainless steel needle of gauge 14 (OD: 2.11 mm, ID: 1.60 mm) is attached with the microsyringe and placed at the top of the liquid filled beaker. The drop of sodium alginate is formed from the needle tip and impacts on the pool of calcium chloride. The diameter of the drop is calculated by counting the number of drops by dispensing a fixed amount of sample volume. Then, from the formula of sphere is used to determine the volume of each single drop. Also, the drop diameter is verified with the measurement from the image of the drop captured in high-speed camera. The Nikon autofocusing zoom lens (24–85 mm) with f-stop number (f/2.8-4D) is plugged with the camera and placed in parallel with the liquid level of the pool to capture the sub-surface phenomenon. A flicker-free LED light source (MultiLED LT High Power 24 LED Lamp Head of GS Vitec) is kept at the backside of

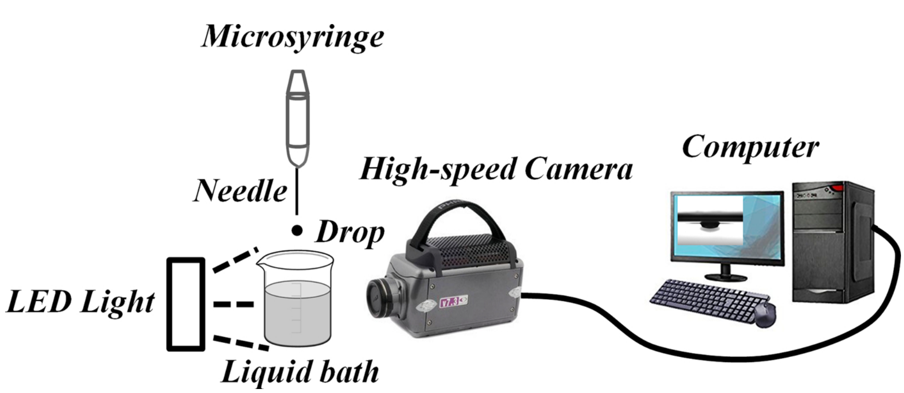


**Fig. 1.** Schematic diagram of experimental setup which consists of microsyringe, liquid bath of calcium chloride, high-speed camera, data acquisition computer and LED light.