The effect of the spacing and angle arrangement of the droplet on the spreading area in simultaneous multiple droplets deposition

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Abstract. Simultaneous multiple droplet deposition involves dispensing of multiple droplets at the same instant on the substrate. A newly defined parameter, arrangement angle is considered as one of the factor that influencing the coalescence of the multiple droplets. However, the effect of such factor on the coalescence is largely neglected in the past. The paper aims to investigate the effect of droplet centre-to-centre distance and arrangement angle on the spreading area of four droplet deposited simultaneously using simulation method. The project starts with single droplet deposition and following by simultaneous multiple droplet deposition. For single droplet deposition, the spreading area of 80% wt aqueous glycerin droplet on the glass substrate were obtained using simulation and experimental methods. A correlation function was introduced to match the simulation result with the experiment result. For simulations of multiple droplet deposition, three droplet centre-to-centre distances as 2.22 mm, 3 mm and 4 mm and three droplet arrangement angles as 90°, 110° and 120° were studied. The larger the droplet spacing, the larger the spreading area for arrangement angle of 90°. The spreading area are largest for spacing of 4 mm, followed by 2.22 mm and 3 mm for arrangement angle of 110° and 120°. The arrangement angle is proven as a factor that affect the spreading area.

1. Introduction

Droplet deposition are classified into vapour and liquid phases. Liquid-phase methods such as inkjet printing, spray coating and drop-casting involve deposition of multiple droplets on a solid substrate. Multiple-droplet drop-casting (MDDC) is simultaneous deposition of highly ordered droplets with the equal size and spacing on the substrate surface. The technique is a simpler and more controllable compared to spray coating in producing continuous thin films.

Controlling the single or multiple-droplet impingement process is important for deposition process. The deposition mechanism of single droplet has been studied in the past researches. When a droplet impacts with a solid surface, the droplet can spread, rebound or splash [1]. A droplet can spread over the surface without splashing and rebounding. The maximum spreading of droplet is affected by several parameters such as droplet initial diameter, surface tension, viscosity, impact velocity and wettability of the substrate surface. These parameters are related to Reynolds and Weber numbers [2]. Lee, Derome, Guyer and Carmeliet [3] found that viscosity and surface tension play a significant effect on maximum spreading ratio of the impinging droplet. Surface roughness and type of surface have minor effect on maximum spreading ratio when the impact velocity is low. When impact velocity increases, spreading velocity increases as well, which results in a larger maximum spread [4].

When two or more droplets overlap, coalescence of droplets occurs which forms a larger droplet or thin film. The neighbouring droplets must be in contact and coalesce during deposition process to prevent discontinuities in dispensing pattern [5]. There are several factors that affect the coalescence of
droplets, which include the parameters affecting deposition mechanism of single droplet and the arrangement of the droplets such as centre-to centre distance and the arrangement angle between the droplets. Lee, Kim, Chandra and Yoon [6] had conducted experiment to study the effect of viscosity on the coalescence of sessile droplets. Graham, Farhangi and Dolatabadi [7] studied the coalescence of falling droplet and sessile droplet on solid surface with different surface wettability. It was found that the maximum spreading length of the coalesced droplet decreased when the surface hydrophobicity increased.

Several researchers have studied the effect of droplet spacing on the coalescence of droplets. Castrejón-Pita, Betton, Kubiak, Wilson and Hutchings [8] studied the effect of droplet spacing on the coalescence of two droplets on a substance. A droplet was deposited adjacent to another droplet resting on the substrate. Dalili A, Chandra, Mostaghimi, Fan and Simmer [9] conducted experiment to investigate the effect of droplet spacing on the coalescence of glycerin-water droplets. The droplets were deposited sequentially on a flat steel plate at a frequency of 1 Hz to form a straight line or square pattern. Even though abovementioned researches studied the effect of droplet spacing on the coalescence of droplets, the droplets are deposited subsequently instead of at the same instant.

Simultaneous multiple droplet deposition involves dispensing of multiple droplets at the same instant to form a pattern. Eslamian and Soltani-Kordshuli [10] studied the surface roughness of poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS) drop-cast films upon drying on a bare glass and a fluorine-doped tin oxide (FTO)-coated glass with different imposed vibration time. The experiment involved equally spaced and equally sized droplets deposited onto the surface at the same time. Thus, there are limited study on the multiple droplet deposition that droplets are deposited at the same instant.

Besides droplet spacing, a newly defined parameter, arrangement angle is considered as an influencing factor to the coalescence. Figure 1 illustrates the arrangement angle θ. The effect of arrangement angle on the coalescence is neglected in the past.

The droplet deposition and coalescence phenomena were realized using numerical simulation method like marker-and-cell [11], Volume of Fluid [12], Lattice Boltzmann Method [13], Smoothed Particle Hydrodynamics [14] and Coupled Eulerian Lagrangian [15]. For Fluid Structure Interaction, Coupled Eulerian Lagrangian (CEL) method is a simple approach to model the interaction between fluid and solid. In Johansson and Ollar [15], a model of the sealing process was generated by applying CEL approach and the applicability of the method was discussed. For droplet deposition, Zhu, Kamnis and Gu [16] conducted a numerical study on impingement of molten and semi-molten ceramic using CEL method.

This research project focuses on how droplets centre-to-centre distance and arrangement angle affect the dispensing area through simulation study. Only the material properties of 80% wt aqueous glycerin droplet and glass substrate are input to the simulation to fix the fluid viscosity and the surface wettability. Four droplets are deposited simultaneously onto the glass substrate. The droplet centre-to-center distance of 2.22 mm, 3 mm and 4 mm and arrangement angle of 90°, 110° and 120° were investigated. Nine sets of combination of the variables were investigated. For each combination, eight spreading area data were obtained for eight different spreading time range from 0-0.1 second. Thus, a total of 72 data were obtained in the study.
2. Methodology
The research methodology is divided into two stages, which are single droplet deposition and simultaneous multiple droplet deposition. The methodology of the research is summarized in figure 2. For single droplet deposition, experiment and simulation works were conducted. The experimental data were obtained to verify the simulation data. After verification, simulations of four aqueous glycerin droplets impacting and coalescing with each other on the glass substrate were conducted.

![Flow chart of methodology.](image)

2.1. Experiment on Single Droplet Deposition
The experiment was conducted by impacting an 80% wt aqueous glycerin droplet on a glass substrate. The density and viscosity of the aqueous glycerin droplet are 1205.45 kg/m³ and viscosity of 0.047 kg/m s respectively [17]. The sound velocity is assumed to be similar to that of pure glycerin, which is 1920 m/s. The apparatus set-up of the experiment is shown in figure 3. A flat rectangular glass plate was placed on a horizontal platform of a retort stand. A microlitre syringe filled with aqueous glycerin solution was fixed on the clamp above the glass substrate. The needle tip of the syringe was positioned 20 mm away from the glass substrate. An aqueous glycerin droplet was formed and separated from the needle tip of the microsyringe once the droplet size was sufficiently large. The high speed camera Photron FASTCAM Mini UX50 was used to capture the droplet deposition. The high speed camera FASTCAM Mini UX50 has an operational pixel of 1280×1024 and exposure time frame per second (fps) can be increased up to 160000fps with reduced pixel. The optical lens is Nikkor Lens, Model: AF Zoom-Nikor 24-85mm f/2.4-4D IF. The experiment was repeated three times to obtain the average results.

The recordings of the deposition process were analyzed using Photron FASTCAM Viewer 4.0 software. The image of droplet before impacting the substrate and the image of droplet deposited on the substrate at the spreading time of 0.1 second are shown in figure 4(a) and figure 4(b) respectively. The scale of the image was calculated by comparing the distance between the needle tip of the microsyringe and the glass substrate surface in the image to the actual distance of 20 mm. The droplet initial diameter was measured based on the falling droplet as shown in figure 4(a). The droplet spreading diameter was measured from the image of droplet on the substrate. Deposited area is calculated using the spreading diameter by assuming the droplet spreads in circular pattern.

2.2. Simulation of Single Droplet Deposition
Simulia Abaqus v6.14 Student Edition software was used to apply finite element method to simulate the droplet impingement behavior with respect to time during impact with glass substrate. CEL method was
used to create a model of single droplet impingement on solid surface. The steps of simulation of single droplet deposition is summarized in figure 5. There are several assumptions are made to simplify the simulation. First, the aqueous glycerin droplet is assumed to remain in spherical shape before it contacts with the glass substrate surface. It is assumed that glycerin droplets have zero initial velocity at the tip of the microsyringe and the effect of air resistance is negligible. The droplet is assumed to spread in circular pattern in deposition.

The 3D models of droplet, glass substrate and Eulerian instance were generated using Abaqus/CAE in Step 1. A hollow sphere with diameter of 2.2 mm was constructed as a droplet reference instance. The diameter of the sphere is equal to the average droplet initial diameter obtained from the experiment. A cuboid of 6 mm × 6 mm × 0.01 mm was created as the glass substrate. A cuboid of 6 mm × 6 mm × 3 mm was generated as an Eulerian instance that defines the fluid region. The Eulerian instance is required to be large enough to cover the glass substrate and the droplet reference instance.

In Step 2, the material properties were assigned to the parts created in Step 1. The substrate was assigned with glass material properties, such as density of 2500 kg/m³, Elasticity modulus of 50 GPa and Poisson’s ratio of 0.22. Assignment of material properties is not required for the droplet reference instance since the part only functions as a container to be filled with aqueous glycerin solution and the instance will be suppressed during the analysis. For 80% wt aqueous glycerin solution, the Eulerian instance was assigned with density of 1205.45

**Figure 3.** Apparatus set up of the experiment.
Figure 4. (a) The falling droplet (b) The droplet on the substrate at the spreading time of 0.1 second.

Figure 5. Flow chart of simulation of single droplet deposition.

kg/m$^3$, viscosity of 0.047 kg/m.s, speed of sound of 1920 m/s and defined with an Equation of State (EOS). In Abaqus, linear $U_s - U_p$ Hugoniot form is used to express the equations of state. The linear Hugoniot slope coefficient, $s$ and material constant, $\Gamma_o$ were both set to zero.
Step 3 involves the assembly of the three parts, such as droplet reference instance, glass substrate and Eulerian instance as shown in figure 6. The droplet reference instance and the glass substrate were positioned within the Eulerian instance to ensure that the fluid material can interact with the glass surface. The droplet reference instance and glass substrate are positioned 0.1 mm apart. The initial velocity of the droplet reference instance was calculated as the impact velocity \( V = \sqrt{2gh} \) where \( g \) is the gravitational acceleration and \( h \) is the falling distance. The impact velocity was calculated to be 0.6264 m/s. The reduce of distance between droplet reference instance and glass substrate from 20 mm as in the experiment with zero initial velocity to 0.1 mm with 0.6264 m/s initial velocity significantly decreased the computational time.

Step 4 involves meshing of the parts. It is suggested by the Simulia general guidelines that the mesh size of the Eulerian part should be at least smaller by a factor of three compared to the smallest feature of interest in Lagrangian mesh [15]. The Eulerian instance was meshed by an approximate global seed size of 0.1 mm using 8-node linear Eulerian brick with reduced integration and hourglass control. The glass substrate was meshed by an approximate global seed size of 0.6 mm and linear in three dimensions. The reduced integration was activated to reduce calculation time [18]. Element deletion was deactivated to prevent element from being removed by the impact of droplet. The droplet reference instance was not required to be meshed since it would be suppressed during the analysis. Finer mesh was not required for glass substrate part since the part was only for interaction with the aqueous glycerine droplet and the mesh was only for continuing the numerical processes such as assigning contact properties, load and boundary conditions. The number of elements in glass substrate and droplet reference instance are 10800 and 100 respectively.

In Step 5, the loads and boundary conditions of the assembly part were defined. The initial fluid location was defined inside the droplet reference instance. To implement fluid-structure interaction, two contact properties were created, which were tangential behaviour and normal behaviour. For tangential behaviour, the contact was given a penalty by applying friction coefficient of glass, which is 0.9. The assembly part was subjected to gravity load of 9.81 m/s\(^2\) in negative y direction as shown in figure 6. The glass substrate was fixed at its original position throughout the simulation. The discrete field created in the Step 4 was assigned with the material properties of aqueous glycerin solution. A node set of Eulerian instance which consisted of 115351 nodes was created. Next, initial velocity of 0.6264 m/s in negative y direction was applied to the Eulerian instance.

In Step 6, a step time of 0.1 second was used to simulate the deposition process. The double precision was used to produce more accurate results. In Step 7, The spreading diameter of the droplet reference instance over time after in contact with the glass substrate were interpreted and analyzed using ImageJ software. The image scale was obtained by comparing the length of glass substrate in the image to the length of 6 mm. The spreading area was calculated from the obtained spreading diameter by assuming the droplet spreads in circular pattern.

2.3. Simulation of multiple Droplet Deposition
Nine sets of numerical simulations were carried out for multiple droplet deposition based on different combinations of droplet spacing and arrangement angle. The droplet spacing values chosen are 2.22 mm, 3 mm, and 4 mm. The arrangement angles selected are 90°, 110° and 120°. For each set of simulation, data were generated for 0.005 second, 0.01 second, 0.02 second, 0.04 second, 0.06 second, 0.08 second, 0.1 second.

The steps of simulation of multiple droplet deposition is similar to those of single droplet deposition. Instead of constructed a hollow sphere, in Step 1, four hollow spheres with the diameter of 2.2 mm were constructed. To make sure that there is enough space for the droplets to spread on the glass substrate, the substrates and Eulerian instances were created with the length and width varies with the droplet spacing. For droplet spacing of 2.22 mm, 3 mm and 4 mm, the length and width of the substrate and Eulerian instance are 12 mm x 12 mm, 15 mm x 15 mm and 20 mm x 20 mm respectively. The height of the substrate and Eulerian instance remaining unchanged.

In meshing of the parts in Step 4, the four aqueous glycerin droplets are merged together to form one part to ease the discrete field assignment process. The original instances are suppressed through this merging step. For meshing of Eulerian instance and glass substrate, the global seed size used also varies for three different sizes of Eulerian instance and glass substrate. The global seed size and number of elements for each sizing are tabulated in Table 1. In Step 6, the process were simulated for 0.005 second, 0.01 second, 0.02 second, 0.04 second, 0.06 second, 0.08 second, 0.1 second.

### 3. Comparison of Experimental and Numerical Results of single droplet deposition

The experimental and numerical spreading areas for single droplet deposition were compared and verified. A correlation function was introduced to compensate the deviation found between both results. A graph of experimental and simulated spreading area against the spreading time was illustrated in figure 7. The time zero refers to the time right before the impact.

The spreading area data obtained from experiment and simulation method can be represented by equation (1) and equation (2) respectively:

\[
\ln y_{\text{simulation}} = \ln 24.511 + 0.0783 \ln x
\]

\[
\ln y_{\text{experiment}} = \ln 9.0739 + 0.0435 \ln x
\]

where \(y_{\text{simulation}}\) is the spreading area obtained from simulation method, \(y_{\text{experiment}}\) is the spreading area obtained from experiment, and \(x\) is the spreading time. The deviation between the data obtained from experiment and simulation method may be caused by the limitation of CEL method and on modelling droplet deposition process. Physical phenomena such as surface behaviors and material behaviors might not be correctly represented using CEL technique. These include adhesion force of droplet to glass surface, surface tension of the droplet impinged and cavitation.

| Droplet Spacing (mm) | Part                  | Global seed size (mm) | Number of elements |
|---------------------|-----------------------|-----------------------|--------------------|
| 2.22                | Eulerian instance     | 0.15                  | 128000             |
|                     | Glass substrate       | 1.20                  | 100                |
| 3.00                | Eulerian instance     | 0.15                  | 200000             |
|                     | Glass substrate       | 1.50                  | 100                |
| 4.00                | Eulerian instance     | 0.20                  | 150000             |
|                     | Glass substrate       | 2.00                  | 100                |
A correlation function was introduced to match the simulation results to the experimental results. The correlation function is given as
\[ \ln y'_{\text{simulation}} = \ln y_{\text{simulation}} - 0.9937 - 0.0348 \ln x \] (3)
where \( y'_{\text{simulation}} \) is the spreading area that close to the spreading area obtained from experiment. To verify equation (3), five spreading time data were used to calculate the spreading area \( y_{\text{simulation}} \) based on equation (1) and equation (3). The calculated results were compared with those obtained from equation (2). Both results are shown in table 2. Based on table 2, the absolute percentage deviation of the spreading area obtained from correlation function and experiment are 0.002% only. Thus, the correlation function is comparable to the experiment and it was used to calculate the spreading area based on the simulated data obtained from multiple droplet deposition.

Table 2. The absolute percentage deviation of the results obtained from the correlation function and experiment.

| Spreading time, \( x \) (second) | \( y'_{\text{simulation}} \) (mm\(^2\)) based on correlation function | \( y_{\text{simulation}} \) (mm\(^2\)) based on experiment | \( y_{\text{experiment}} \) (mm\(^2\)) based on experiment | Absolute percentage deviation of spreading area (%) |
|----------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-----------------------------------------------|
| 0.02                             | 7.65416                                         | 7.65401                                         | 7.65401                                         | 0.002%                                        |
| 0.04                             | 7.88846                                         | 7.88830                                         | 7.88830                                         | 0.002%                                        |
| 0.06                             | 8.02883                                         | 8.02867                                         | 8.02867                                         | 0.002%                                        |
| 0.08                             | 8.12993                                         | 8.12977                                         | 8.12977                                         | 0.002%                                        |
| 0.10                             | 8.20923                                         | 8.20907                                         | 8.20907                                         | 0.002%                                        |

4. Simulated spreading area for multiple droplet deposition
The spreading area versus spreading time for different combinations of droplet spacing and arrangement angle was shown in figure 8. All the lines start from the origin where the spreading area is zero for spreading time of zero. The second spreading area data collected at 0.005 second for all the case. The second spreading area are more than 40 mm\(^2\) for all the case. Therefore, the y-axis is starts from 40 mm\(^2\).
Based on figure 8, the spreading area increased rapidly from 0 to 0.013 second. The slope of the increment decreased and slowly reached the equilibrium as the time went on. This was explained in Dorey [19] that the droplet spreading process could be divided into two stages, which were initial spreading and secondary wet spreading. Initial spreading of droplet depended on the kinetic energy of the impacting droplet which happened in the scale of microseconds. Secondary wet spreading depended on Ohnesorge number, which was influenced by surface tension, inertial and viscous forces. This type of spreading required longer time to reach equilibrium and drying effect might have occurred. The initial spreading occurred before 0.013 second in which rapid spreading of aqueous glycerin droplet occurred. After that, secondary wet spreading took place where the deposited droplet spread slowly on the glass substrate.

An array of droplets has more complicated interaction mechanisms compared to two-droplets interaction due to increasing in number of neighbouring droplets. During coalescence, more interface energy is released by multiple droplets interface, thus increasing the amount of kinetic energy converted. Higher amount of kinetic energy leads to larger deformation of the droplet interface during interaction and splashing of droplets may occur. Due to the existence of additional neighbouring droplets, the time required for contact of droplets to occur is also affected.

It was explained by Zhou, Loney, Fedorov, Degertekin and Rosen [20] that for an array of droplets, the interactions generated more forms of final equilibrium shape compared to two-droplet interaction. The interaction mechanisms were highly sensitive to impact conditions and droplet spacing. For small droplet spacing, the inertial force due to impact on solid surface led to coalescence of fast-moving droplet boundaries. High kinetic energy was gained from the coalescence of the droplet interface. Table 3 shows the droplet deposited pattern at 0.10 second for different combinations of droplet spacing and arrangement angle. It was observed that for image of droplet spacing of 2.22 mm and arrangement angle of 90° in table 3, the droplets had a tendency to spread out in the direction of neck growth due to high kinetic energy generated from coalescence. When droplet spacing was 3 mm, there were larger rooms for spreading which exhausted the inertial force gained from impact during the spreading of droplets. Coalescence of droplets occurred much later compared to droplet spacing of 2.22 mm. Less kinetic energy was gained and thus the droplets spread more steadily and uniform as shown in image of droplet spacing of 3 mm and arrangement angle of 90° in table 3. The droplet spacing of 4 mm generates largest spreading area compared to droplet spacing of 2.22 mm and 3 mm as shown in figure 8. From table 3, however, there are void between the droplets that are not covered up for arrangement angle of 90° and 110°. This is due to when the droplet spacing was further increased to 4 mm, coalescence of droplets could not be done fully due to lack of kinetic energy to energize the coalescence.

5. Effect of Droplet Spacing on Spreading Area

The effect of droplet spacing on the spreading area for three different arrangement angles at spreading time 0.1 second are illustrated in figure 9. The spreading area increased when the droplet spacing increased is only valid for 90° droplet arrangement angle. The finding is supported by the result of two droplets deposition.
Figure 8. Spreading area against spreading time for different combination of droplet spacing and arrangement angle. Obtained by Castrejón-Pita, Betton, Kubiak, Wilson and Hutchings [8] in which the composite length increased as the droplet spacing increased. It was observed that the spreading area for droplet spacing of 4 mm is larger, followed by those for droplet spacing of 2.2 mm and 3 mm for arrangement angle of 110° and 120°.
Table 3. Deposited pattern of deposited droplet at 0.10 second for different combinations of droplet spacing and arrangement angle.

| Arrangement angle, $\theta$ (°) | Droplet spacing, L (mm) |
|---------------------------------|-------------------------|
|                                 | 2.22                    |
| 90                              |                         |
|                                 | 3.00                    |
| 110                             |                         |
|                                 | 4.00                    |
| 120                             |                         |

Figure 9. The spreading area vs droplet spacing for three different arrangement angles.

6. Effect of Arrangement Angle on Spreading Area
The effect of arrangement angle on spreading area is shown in figure 10. When the arrangement angle was increased from 90° to 110°, droplets with spacings of 3 mm and 4 mm displayed a slight reduction on spreading area while only droplets with spacing of 2.22 mm gave increment in spreading area as
compared to that with 90° arrangement angle. For 120° arrangement angle, droplets of all three spacings exhibited reduction in spreading area.

For most of the research, the deposition of an array of droplets was done using arrangement angle of 90°. By changing the arrangement angle of an array of droplets, different deposition patterns can be observed. When the arrangement angle of droplets increases to 110° and 120°, the spacing between the diagonal droplets changed as shown in figure 11. Time for coalescence of droplets differed from each other and thus different kinetic energy was gained for each droplets. This led to more complicated interaction mechanisms compared to ordinary multiple droplet deposition.

![Figure 10. Comparison plot of spreading area vs arrangement angle for three different droplet spacings.](image)

![Figure 11. Diagonal distance of droplets for arrangement angle of (a) 90°, (b) 110° and (c) 120°.](image)

7. Conclusion
The mechanisms of simultaneous multiple droplet deposition were studied using finite element analysis. The research was divided into two stages. For first stage, simulation on single droplet deposition was conducted and was verified using experiment. A correlation function was developed to match the simulation result with the experimental result. For second stage, nine sets of numerical simulations of simultaneous multiple droplet deposition were conducted by using an array consisting of four aqueous glycerin droplets with different combinations of droplet spacing and arrangement angles.

Based on the results obtained, it was found that the spreading area increased as the droplet spacing increased is only valid for arrangement angle of 90°. For arrangement angle of 110° and 120°, the spreading area for droplet spacing of 4 mm is the largest, followed by those for droplet spacing of 2.2 mm and 3 mm. The arrangement angle is proven as a factor that affect the spreading area.

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