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The solidification of encapsulated jasmine extract using electrostatic atomizer (Electrospray)

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Abstract

In this work, a non-thermal solidified method of Jasmine extract was developed via the electrostatic atomizer or electrospray technique. The electrospray technique is dependent on two continuous processes which are water evaporation and droplet fission. To verify the water evaporation rates of the atomized droplets, the deposition trajectory distance were varied from 10 to 30 cm. The morphology and size distribution of the deposits were observed and analyzed. From the experimental results, it was noticed that the evaporation rates of the atomized droplets reduced from 2.65 x 10^{-10} - 1.1 x 10^{-10} g/s as the trajectory distances of the atomized droplet increased from 10-30 cm. Meanwhile, the droplet fission is increased from 12.5 to 16 disintegrations as the distance went further. The morphology and particles size of the solidified compound were analyzed by SEM and ImageJ software respectively.

Keywords: electrospray, encapsulation, droplet charge, Rayleigh limit, coulomb fission

INTRODUCTION

Natural fragrance extracts from plants such as Jasmine flower with its scientific name *Jasminum*, is one of the rejuvenating aroma that commonly used in food industry as a flavor and fragrance. The Jasmine extract in liquid form is more difficult to be handled compared to the dried and solid form. The benefits of having the bioactive compound in solid forms are the storage and shipment process will be more cost efficient and it gives stability to the active substance (Oliveira and Bott, 2006). Other than that, the preservation of the Jasmine extract will be enhanced if the bioactive compound is subjected to the solidification (Aoki and Us, 2004).

Various techniques of solidification have been applied in industry such as spray drying, freeze drying, coacervation, and spray chilling (Jafari et al., 2008). However the applications of these conventional techniques are unfavourable due to the several drawbacks. Spray drying for example, is not preferable for heat sensitive bioactive compound because of the heat application (Zuidam and Nedovic, 2010). Meanwhile, freeze drying that is considered as an alternative technique for heat-sensitive compound had major disadvantages which are high energy consumption, long processing time, and the obtained open porous structure do not act as a good barrier between the active compound and the surroundings (Zuidam and Nedovic, 2010). In addition, the process is up to 30 to 50 times more expensive compared to spray drying process (Gharsallaoui et al., 2007). After considering the disadvantages of conventional solidification techniques, this work will study on the feasibility of using electrostatic atomizer (electrospray) for solidification of bioactive compound. Many advantages can be obtained by using electrospray technique which are producing fine monodispersed droplet within nanometer scale, minimum droplet aggregation and high deposition efficiency (Gu, 2013).

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Electrospray applies high capacity of voltage instead of heat to solidify the particles. It is an atomization process that takes place when a strong electric field is exerted on the liquid that cause liquid dispersion into a fine aerosol (Hiraoka, 2013). Taylor cone, which is the characteristic of the charged droplet, was forming when the coulombic repulsion of surface charge is equal to the surface tension of the solution. This condition is known as Rayleigh limit (Cech and Enke, 2002). When the atomized droplet leave the Taylor cone zone, the excess charge dissipate together with the solvent vapor and cause the primary droplet to eject out the fine progeny droplets in nanometer scale. During the process, the ejected droplets are surrounded by the positive charges that were obtained from the electrostatic generator. At this stage, the water removal occurs and leads to the reduction of droplet size due to the evaporation and follows by droplet fission. The evaporation and droplet fission will continuously occur until the droplets solidified and deposited onto the collector that consist of grounded aluminum substrate.

In this work, the evaporation rate and droplet fission are studied in order to obtain the optimum condition that contributes to the solidified deposits. Atomization distance are varies and their relation with water removal mechanism is scientifically observed.

MATERIALS AND METHODS

Sample preparation

Bioactive compound was extracted from Jasmine flower (Jasmine flowers were obtained from Universiti Putra Malaysia's garden) by using the Ultrasonic Assisted Extraction (UAE). 3 g of flower was cut finely and put in a 100 ml of a clean beaker. Then, 20 ml of deionized water (DI) was poured into the beaker and ready for sonication. After ultrasonication, it is necessary to centrifuge the Jasmine extract to get consistent particle size and to separate between the bigger and smaller particles. The centrifuge process was done at 10 000 rpm, 20 ^{o}C, for 30 minutes. Figure 1 shows the step of the extraction.

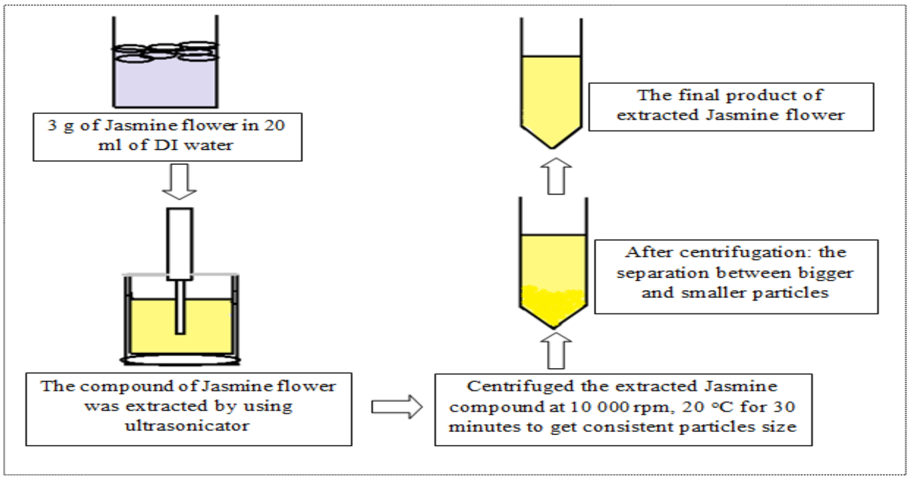

Figure 1. Preparation of Jasmine extract.

Electrospray

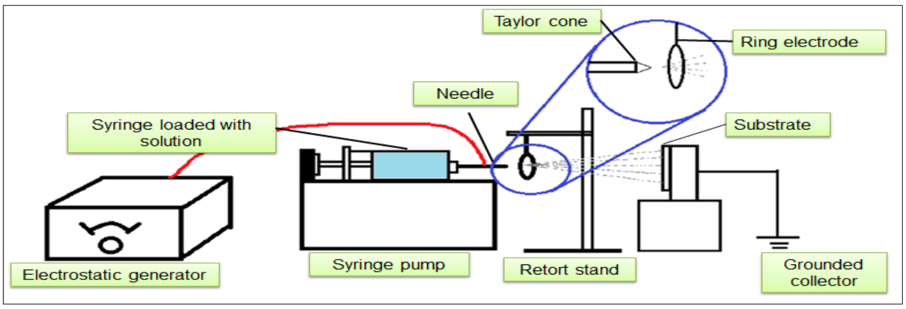

Figure 2. The setup of electrospray.

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Figure 2 shows the setup of electrospray. The solution of Jasmine extract was loaded in a 12 ml plastic syringe that connected to the stainless steel needle which acted like a nozzle. The flow rate of the solution was controlled by an infusion syringe pump (NE-300, New Era Pump System Inc., New York) at the value of 0.20 ml/hr. An electric field with the positive polarity was applied to the nozzle using a high voltage supply (AMI-10K3P, Max-Electronics Co. Ltd., Tokyo). A ring electrode was placed perpendicular to the needle tip to reduce the potential differences between the cone and the grounded collector (Yurteri et al., 2010). The ring electrode used has a 7 mm inner diameter and 20 mm outer diameter.

The electrospray was performed under Taylor cone jet mode at 20ºC and the relative humidity (RH) was between 35 to 36%. The distance between the needle tip to the aluminium substrate was varied from 10, 20, and 30 cm. Aluminium substrate with the dimension of 9.5 cm x 9.5 cm was used as a collector for the deposits and was connected to the ground. The conditions of electrospray for this research as shown in Table 1.

Table 1. The condition of electrospray.

Parameter	Range
Voltage (kV)	4 – 5
Flow rate (ml/hr)	0.2
Needle tip to substrate distance (cm)	30
Electrospray duration (hr)	1

After the electrospray process, the solidified powder was analysed by Scanning Electron Microscopy (SEM), ImageJ software, and the calculation on evaporation rate and droplet fission were also done.

Scanning Electron Microscopy (SEM)

The morphology of the collected particles were observed by using SEM (JSM 6510, JEOL, TOKYO) at the accelerating voltage of 10 kV and the magnification of 20000. In order to avoid the charging effect during the SEM observation, the samples were sputter-coated with gold (JFC 1200, JEOL, TOKYO) under vacuum condition. ImageJ (NIH, Bethesda) was used to determine the particles size and was reported in average value.

Droplet size for evaporation rate calculation

Droplet size is one of the important parameter in order to get the evaporation rate and it is was calculated by using the equation proposed by Hartman *et. al* (Hartman et al., 2000):

$$d_d = [(\rho \varepsilon_o Q^3)/YK]^{1/6} \qquad (1)$$

where d_d is a droplet size diameter in (nm), ρ is the density of solution (kg/m^3), ε_o is an electrical permittivity of the vacuum (8.8x10^{-12} C^2/N/m^2), Q is the flow rate of the solution (ml/hr), Y is the surface tension of the liquid (N/m), and K is the conductivity of the solution (S/m). The value of initial droplet is summarized in Table 3.

Evaporation rate for evaporation

During the electrospray process the incorporation of electrostatic charge contributes to water evaporation and coulomb fission which drive the atomized mixture components to solidify. Equation (2) is used in calculating the evaporation rate.

$$E_R = \Delta V / \text{t} \qquad (2)$$

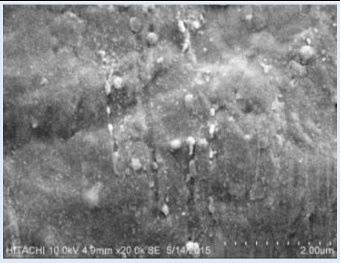
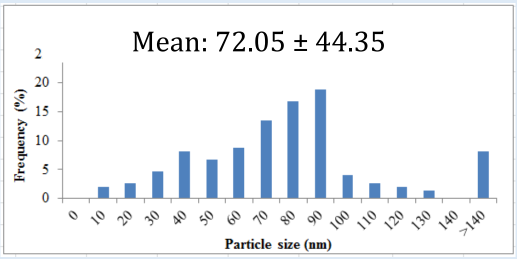
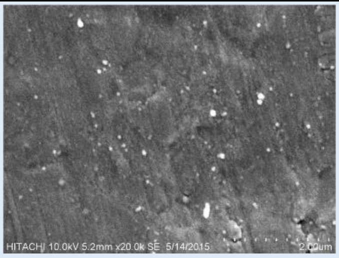
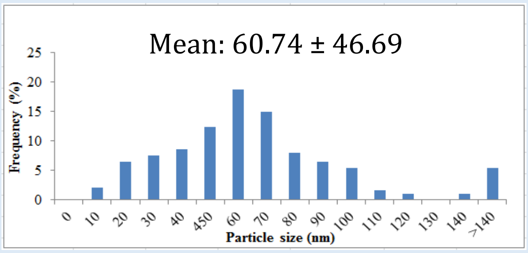
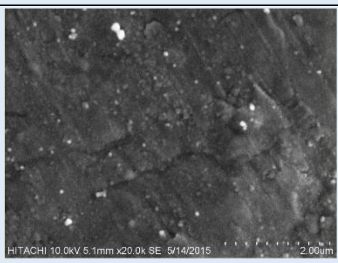
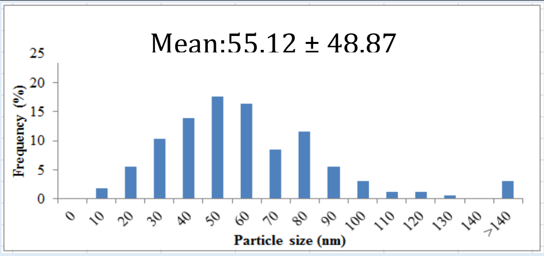
Where ΔV is the volume difference (nm^3), t is the time taken (s), and E_R is the evaporation rate in g/s.

RESULT AND DISCUSSION

SEM analysis and particle size distribution

In order to understand the morphology of particles and particles size distribution after electrospray, all samples were analysed by using SEM and ImageJ respectively. Table 2 shows the result on SEM and particle size distribution.

Table 2. SEM images and particle size distribution.

Distance	SEM Images	Particle size distribution
10		
20		
30		

The SEM images in Table 2 demonstrated the solidification of droplets at varies distances. The solidified powder is getting finer from 10 to 30 cm. 'Wet' deposition and aggregation was observed at the distance of 10 and 20 cm while less aggregation and finer particles can be seen at 30 cm. The wet deposition and aggregation phenomena that were observed at the shorter distance were due to the incomplete water evaporation. Bock et al. (2011) reported that wet deposition usually produces inhomogeneous semi-solid, flat particles that solidify after deposition. When the distance from needle tip to collector is increase at 30 cm, complete solidification occurs, and causes the particles to well distributed, and only little aggregates particles were observed.

Particles size distribution graph at different distances were plotted after calculating the particles size using ImageJ. The graphs were illustrated as shown in Table 2. Based on the particle size distribution graphs, it was observed that the particles size were shifted from the right to the left side (from bigger to smaller particle size) when the distance were increased from 10 to 30 cm. Having said that, any increase in distance will lead to the decrease in average particle size due to the long trajectory distance that allows the droplets to evaporate and experience the droplet fission. According to Hazeri et al. (2012), the continuous occurrence of evaporation and droplet fission are the mechanism that contributed to the smaller average of particles size. It was found that the longest distance produced the smallest particles size and contributed to the narrow size distribution that confirmed the shrinkage of droplets due to the evaporation and droplet fission occurrence before the deposition of particles onto the aluminium substrate (Saallah et al., 2014).

Evaporation rate of the deposits

In order to obtain the evaporation rate, the initial droplet that diffused out from the needle tip need to be known. The initial droplet can be calculated by applying equation (1). By using the calculated initial droplet size obtained from equation (1), the droplet volume can be calculated, while the volume of deposits can be obtained from the average particle diameter as listed above. Table 4 shows the result on the evaporation rate and the parameters needed to find them.

Table 3. Evaporation rate.

Distance (cm)	10	20	30
Volume of droplet x 10^8 (nm^3)	3.8100	3.8100	3.8100
Volume of deposit x 10^5 (nm^3)	1.9590	1.1737	0.8771
Volume differences x 10^8 (nm^3), ΔV	3.8081	3.8088	3.8091
Time taken to reach collector (s), t	0.0014	0.0023	0.0034
Evaporation rate x 10^{-10} (g/s), Er	3.64	1.82	1.21

From the equation (2) and Table 3, it can be understood that the evaporation rate is highly influenced by volume difference and the time taken for the droplet to reach the collector. The volume difference between the initial droplet and the deposit is assumed to be equal to the amount of water lost due to evaporation and droplet fission mechanism.

From the obtained data we noticed that the evaporation rate is getting slower from 10 to 30 cm. Theoretically, the water at the droplet surface which loosely held by the chemical binding is easy to be removed by the evaporation. When the water molecules at the surface are gradually removed, the moisture content at the surface is decreased and the remaining water inside the droplet will be bound stronger and tighter to the solid compound inside the droplet (Earle, 2013). Therefore, when the droplet travel for a long distance, the moisture and water content inside the droplet will be reduced, and leads to the strong binding between solid content and the water molecules. Consequently, the droplets that take more time and far distance to reach the collector will cause the slower evaporation rate.

From the obtained data in Table 3, the evaporation rate was higher, but contributes to the large particles size deposit. Despite on the discussion on the theory of moisture and water evaporation above, it can be explained by concerning on the distance taken for the particles size to reach the droplet. Even though the drying rate was faster at the lower concentration, but due to the shorter distance, it does not allow evaporation and the droplet fission to happen at the fullest. Since this electrospray process involves continuous evaporation and droplet fission, thus the droplet disintegration by droplet fission must be counted as well.

Droplet fission

The occurrence of droplet fission was due to the decrease in volume by water evaporation of a highly charged droplet (Hiraoka, 2013). The rapid evaporation of water increases the surface charge density and increase the repulsion between the surface charges (Scholten et al., 2011). High charged droplet which formed by electrospray tend to undergo the fission to produce progeny droplets. Therefore, when the distance between the needle tip and the collector is increasing, more droplet fission is expected to occur and produced smaller size deposits compared to the shorter distance. To observe the phenomena, the droplet fission value indicates the amount of droplet disintegration occurrence from the needle tip to the aluminum substrate (collector).

The results were presented in Table 4. The increase in distance from 10 to 30 cm contributes to the smaller particles size due to the large number of droplet fissions occurrence. For example, at 10 cm, due to the short distance; only low numbers of droplet fission occurrence are noticed thus resulting in large particles size deposit. In contrast, at 30 cm; the numbers of droplet fission occurrence is high and contributes to the fine particles size deposit.

Table 4. Droplet fission.

Distance (cm)	10	20	30
Droplet fission	12.5	14.8	16

CONCLUSIONS

The following conclusions can be drawn from the study:

– A solidified and well distributed fine particles size was observed at the 30 cm distance between the tip and the collector. Wet and aggregated deposition was observed at the 10 cm due to the short trajectory distance.

– Droplet disintegration by water removal and droplet fission allow maximum fission that result in fine solidified particles.

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