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Microencapsulation of Ascorbic Acid by Spray Drying: Influence of Process Conditions

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***Abstract*—**Ascorbic acid (AA), commonly known as vitamin C, is essential for normal functioning of the body and maintenance of metabolic integrity. Among its various roles are as an antioxidant, a cofactor in collagen formation and other reactions, as well as reducing physical stress and maintenance of the immune system. Recent collaborative research between the Australian Defence Science and Technology Organisation (DSTO) in Scottsdale, Tasmania and RMIT University has sought to overcome the problems arising from the inherent instability of ascorbic acid during processing and storage of foods. The recent work has demonstrated the potential of microencapsulation by spray drying as a means to enhance retention. The purpose of this current study has been focused upon the influence of spray drying conditions on the properties of encapsulated ascorbic acid. The process was carried out according to a central composite design. Independent variables were: inlet temperature (80-120° C) and feed flow rate (7-14 mL/minute). Process yield, ascorbic acid loss, moisture content, water activity and particle size distribution were analysed as responses. The results have demonstrated the potential of microencapsulation by spray drying as a means to enhance retention. Vitamin retention, moisture content, water activity and process yield were influenced positively by inlet air temperature and negatively by feed flow rate.

***Keywords*—**Microencapsulation, spray drying, ascorbic acid.

I. Introduction

RECENT studies have confirmed that water soluble vitamins including ascorbic acid (AA), thiamin, riboflavin, vitamin B6 and folic acid are subject to loss during food processing [1], [2], [3], [4]. Collaborative research between the Defence Science and Technology Organization (DSTO) Scottsdale, Tasmania and RMIT University has sought to overcome the problems arising from inherent instability of AA during processing and storage of foods [5]. The results have demonstrated the potential of microencapsulation by spray drying as a means to enhance retention.

Spray drying is the transformation of a feed liquid from a fluid state (solution, dispersion or paste) into a dried particulate form by spraying the prepared fluid into a hot drying medium. Spray drying techniques have been widely used for drying heat sensitive foods, pharmaceuticals as well as other substances, because of the rapid evaporation of the solvent from the droplets [6].

Spray drying has been used for encapsulation of a variety of food components, including flavouring agents, fats and oils, vitamins and minerals, microorganisms and enzymes as well as sweeteners and colorants. This technique has been increasingly adopted as a technology which is relatively inexpensive and simple to apply to the large scale production of microcapsules.

Microencapsulation technology provides a strategy for enhancing retention of sensitive and expensive food components, including AA through protection from adverse conditions and allowing delivery to the target site at the required time. Microcapsules have the potential to be very widely used in food industry applications and some of these are microencapsulation can protect sensitive core ingredients from the environmental factors, including oxygen, water and light; undesirable interactions with other ingredients; other roles are to control diffusion or to isolate or control the release of an encapsulated ingredient at the right place and the right time; the physical characteristics of the original material can be modified and made easier to handle; evaporation or transfer rate of the core material to the outside environment is decreased or retarded; the core material can be diluted when only very small amounts are required and still achieve a uniform dispersion in the host material; and it can also be employed to separate components within a mixture that would otherwise react with one another [7].

According to Liu, Z., Zhou, J., Zeng, Y., & Ouyang, X. (2004). [8] in order to obtain a good microencapsulation efficiency and even if the wall material is suitable, optimal spray-drying conditions must be used. The main factors in spray-drying that must be optimized are feed temperature, air inlet temperature, and air outlet temperature.

The purpose of this current study has been focused upon the influence of inlet air temperature (80-120° C) and feed flow rate (7-14 mL/minute) on the properties of encapsulated ascorbic acid. Process yield, ascorbic acid retention, moisture content, water activity and particle size distribution were analysed as responses. The effect of inlet air temperature and feed flow rate on encapsulated AA morphology and particle size distribution was also evaluated.

II. Material and Methods

All chemicals used in encapsulating AA as well as for analytical procedures were of analytical grade or of the highest purity available. Details of chemicals and suppliers were sodium tetraborate, di-potassium hydrogen orthophosphate, hydrochloric acid, L-ascorbic acid, rice starch, alginic acid (sodium salt from brown algae) were purchased from Sigma

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Chemical Co,USA and LMP (Pectin type LM – 104 AS, 2200) from CP Kelco, Genu, Denmark. The equipment and instruments were spray drier, type Minor Serial no. 1902 and spray drier rotary atomiser Model: CK 929 (Niro Atomiser, Copenhagen, Denmark), capillary electrophoresis system Model: 270 A-HT Applied Biosystems, Victoria, Australia, environmental scanning electron microscopy FEI Quanta 200 FEI, United States.

A. Preparation of Hydrocolloids for Spray Drying

Typically, 2-6% of binding agents ALG and LMP (1:1) were combined with a known amount of rice starch (RS) (85-96 g) and dissolved in distilled water. The preparation was then continued with each of the solutions of wall materials being combined as required, depending on the blend for the particular trial. In each case the combination was prepared to give a level of total solid material corresponding to approximately 20% (w/v) in the water. Once each solution was homogenous, the core loading material (AA) was added. Finally, the pH of each of the feed solutions for spray drying was adjusted to 4.0. Microcapsules were then prepared by spray drying the solution using the Niro atomiser system under slow agitation at a feed flow rate of 7-14 mL/min, entrance air temperatures 100 - 120 °C and exit air temperatures 70-95 °C, respectively, and an air pressure of 5 kg/cm^2.

B. Preparation of CE Running Buffer

The buffer was prepared by dissolving 2.16 g of sodium deoxycholate in 100 mL of a solution prepared by mixing 50 mL each of potassium dihydrogen orthophosphate (0.02 M) and sodium tetraborate (0.02 M). The pH of the resultant solution was found to be 8.6 ± 0.1 and this was filtered through a 0.45 μm nylon filter prior to use.

C. Ascorbic Acid Analysis

AA was analyzed by capillary electrophoresis using a procedure based upon that of Thompson *et al*. (1995) [9]. An Applied Biosystems instrument (model 270 A-HT) was used with a fused-silica capillary (undeactivated, 75μm internal diameter, Agilent Technologies). The buffer was sodium orthophosphate-sodium tetraborate (0.02 M, pH 8.6) containing sodium deoxycholate. D-Erythorbic acid (D-*iso*-AA) was used as internal standard and the operating conditions were: +15 kV applied voltage, 28°C temperature and 254 nm for detection.

D. Microstructure Morphology Using Environmental Scanning Electron Microscopy

The outer surface structures of microcapsules were observed using the FEI Quanta 200. ESEM at an accelerating voltage of 30 kV; pressure of 0.5 Torr; spot size of 5.0; and working distance of 10 mm. For this, a small quantity of each microcapsule preparation was mounted on a metal stub using double sided adhesive tape and the features viewed under a low vacuum atmosphere and various magnifications.

E. Measurement of particle Size and Distribution by Laser Diffraction

Particle size analysis was measured by laser beam scattering (Mastersizer X, Model MSX025A) using the procedure described by Cornell, Hoveling, Chryss, and Rogers (1994) [10]. For each individual batch of microcapsules, triplicate analyses were performed. Small amounts of microcapsules were used, with the weight being adjusted to provide readings that corresponding to approximately 30% obscuration in order to achieve representative results. The capsules were then dispersed in *iso*-butanol with continuous stirring. This was used for the analysis due to its desirable characteristics as a suspending liquid for microcapsules – these were its ability to effectively act as a dispersant without in any way disrupting capsule structure [11]. The instrument parameters used were: 300 mm range lens (suitable for a range of particle sizes of approximately 1.2-600 μm diameter) and for the analysis the stirred cell option was selected.

F. Measurement of Moisture Content

The moisture analysis of microcapsules was based upon the Official Method of the Association of Official Analytical Chemists (AOAC International) 925.45-44.1.03 [12] using the oven drying procedure. A sample of microcapsules of approximately 2 g test portion was placed in an aluminum moisture dish and transferred into the air oven set and equilibrated at 102 °C. Samples were allowed to dry for 12 hour and then the dried microcapsules were transferred immediately to desiccators, cooled to room temperature and weighed. The microcapsules were then dried for a further hour and this drying process repeated until constant weight was achieved. The results were calculated and the moisture content of the microcapsules expressed as a percentage [12].

G. Measurement of Water Activity

Aw measurements were taken using a Novasina Aw meter (model ms1-aw). Samples of each microcapsule sample (approximately 3 g) were placed into the plastic container provided with the Aw analysis system. Each was placed in the Aw meter for measurement and the results recorded.

III. Results and Discussion

A. Microcapsule Morphology

All capsules prepared by spray drying were evaluated by ESEM particularly in order to assess overall shape, the surface appearance as well as the integrity of the capsules and any evidence of damage or breakage. The typical appearance of capsules is shown in Fig. 1.

The ESEM micrographs demonstrate that most of the microcapsules are basically spherical in shape. The surfaces appear to be continuous and show indentations to varying extents. This is important to provide lower permeability to gases, better protection and core retention. This type of morphology was also reported in the studies of by Wijaya and co-workers, (2011) [4].

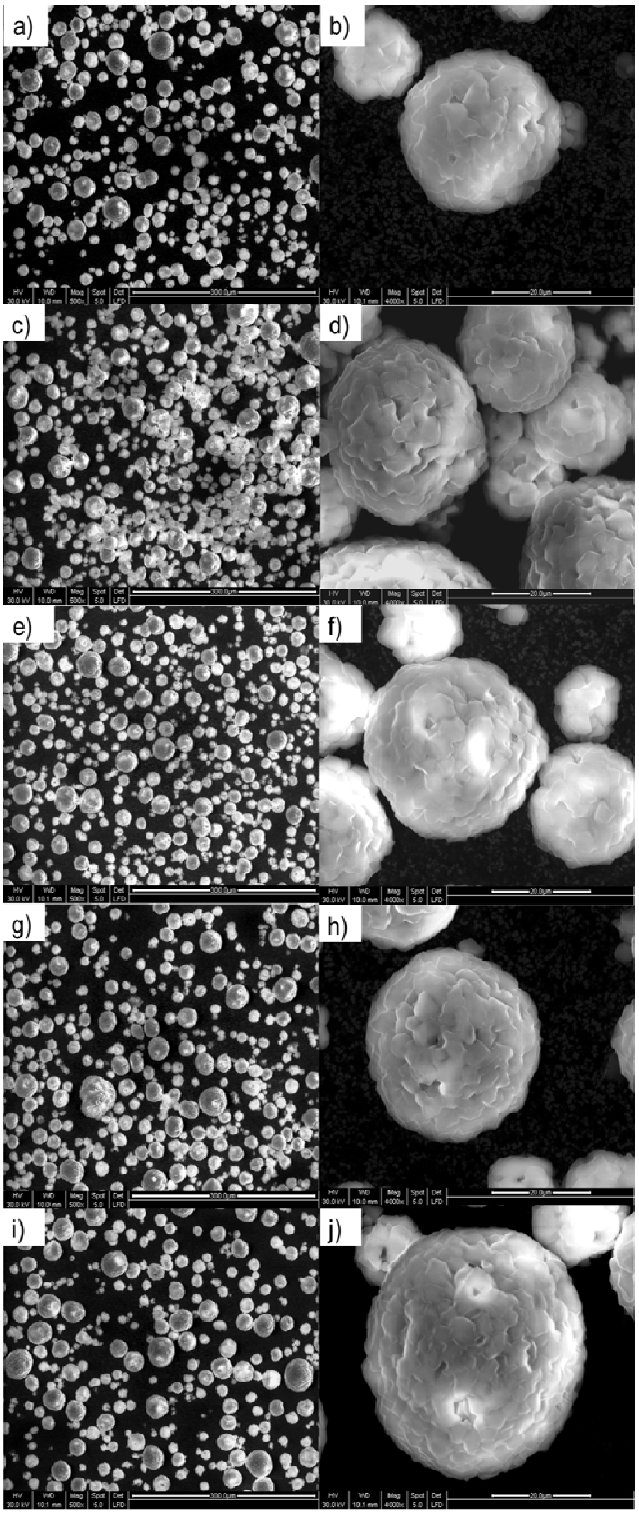

Fig. 1 ESEM images of microencapsulated AA at various inlet air temperature, feed flow rate and magnifications (a) 80°C, 7 mL/min, 500×; (b) 80°C, 7 mL/min, 4000×; (c) 80°C, 14 mL/min, 500×; (d) 80°C, 14 mL/min, 4000×; (e) 100°C, 10.5 mL/min, 500×; (f) 100°C, 7 mL/min, 4000×; (g) 120°C, 7 mL/min, 500×; (h) 120°C, 7 mL/min, 4000×; (i) 120°C, 14 mL/min, 500×; (j) 120°C, 14 mL/min, 4000×

B. AA Analysis Using Capillary Electrophoresis

The electropherogram of ascorbic acid extracted from microcapsules and D- erythorbic acid used as internal standard (Fig. 2) showed retention times of AA and D-erythorbic acid 12-13 minutes and 14-15 minutes respectively.

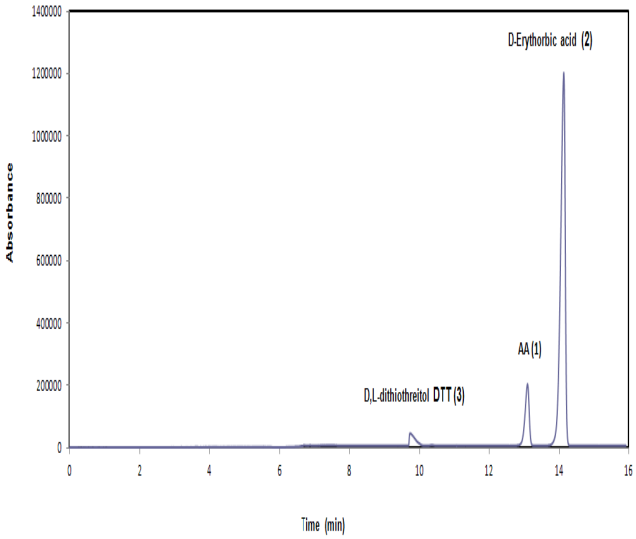

Fig. 2 Electropherogram of ascorbic acid extracted from microcapsules

The method previously validated by Wijaya and co-workers (2011) was adapted [4]. The electropherograms for sample extracts confirm their effectiveness for analysis of AA, giving good separation of the individual components found in the microcapsule samples, as well as the internal standard (Fig. 2). The conditions specifically chosen were: micellar electro kinetic chromatography (MEKC) mode, hydrodynamic injection; running buffer of phosphate-borate and incorporation of sodium deoxycholate at a pH of 8.6 and an applied voltage to the capillary of 15 kV.

C. Response Surface Analysis

There is a significant effect of inlet air temperatures and feed flow rates to the retention of ascorbic acid. Increasing inlet air temperature led to increasing of AA retention (Fig. 3). This is due to lower the moisture content resulting tendency to agglomerate. This reduces the powder exposure to oxygen, protecting the AA against oxidation. This is similar to the results found with spray drying of Amaranthus betacyanin pigments by Cai and Corke (2000) [13].

From Fig. 3 it can be seen the influence of inlet air temperature and feed flow rate on spraying process yield. This response was slightly influenced by these two independent factors. Increasing inlet air temperature and lower feed flow rates led to higher process yield.

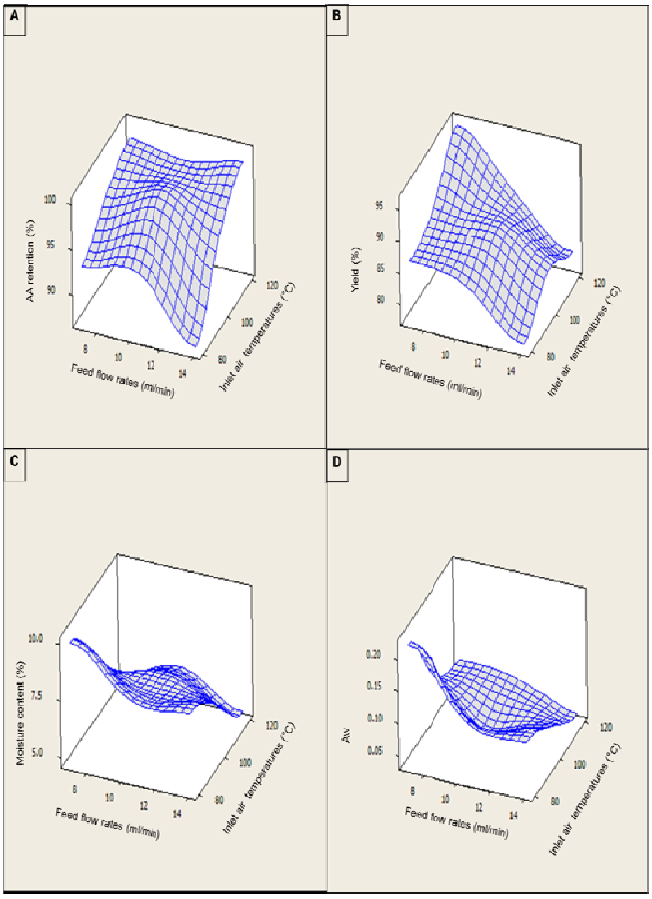

Fig. 3 Response surface for the effect of inlet air temperature and feed flow rate on AA retention, yield, moisture content and aw of microencapsulation of AA respectively by spray drying

Furthermore, moisture content of microcapsules varied from 4-9 % and the aw from 0.06 to 0.2. There was a trend for higher inlet air temperatures to give lower moisture content of the microcapsules. Similar results were also observed for water activity and these observations are attributed to the higher inlet air temperatures.

D. Particle Size Distribution

The resultant microcapsules were then further characterised with a series of analyses including particle size by laser scattering, X-ray diffraction. The results are shown in Fig. 4. The capsules had particle size distributions reflecting uniformity and relatively small sizes, with a range in particle sizes between 5 to 100 µm, predominantly between 30 to 40 µm. The particles size showed a tendency to increasing diameter with lower inlet air temperatures and higher feed flow rates.

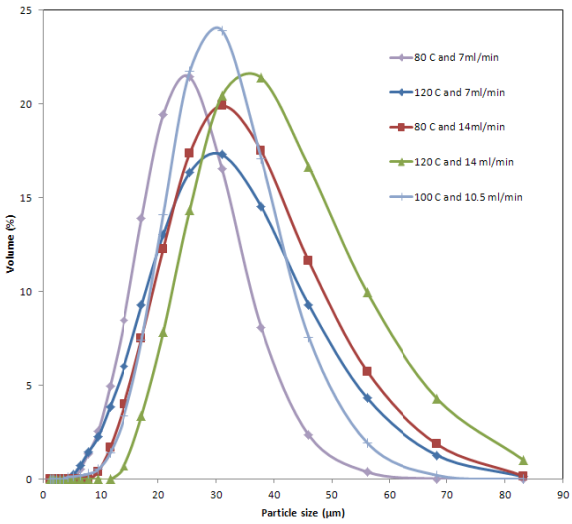

Fig. 4 The effect of inlet air temperatures and feed flow rates for encapsulated AA on particle size distribution

IV. Conclusion

It can be concluded that microcapsules retain good structure and integrity when the inlet air temperature was between 80-120 °C and feed flow rate 7-14 mL/min. The micrographs of the resulting microcapsules showed particles of varying sizes, consistent with the results obtained for analyses of particle size distribution using laser scattering. The results have demonstrated the potential of microencapsulation by spray drying as a means to enhance AA retention. Vitamin retention, moisture content, water activity and process yield were influenced positively by increasing inlet air temperature and negatively as feed flow rate were decreased

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