# Encapsulation of aqueous herbal extract for drying in a packed bed system

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## INTRODUCTION

Freeze-drying and spray-drying are widely used to produce dried herbal extracts. However, these methods carry certain disadvantages. It is necessary to develop an alternative drying method for herbal extract which would offers qualities such as low capital and operating cost, easy to operate and mild process condition. Packed bed drying method is fulfilling these criteria and it is commonly applied in drying of medicinal plants and food products. However, utilisation of packed bed drying for dehydration of liquid products has never been reported since the application of this drying method is only limited to the naturally occur solid materials. In this study, a new approach to dry a model aqueous herbal extract by using the packed bed drying system was demonstrated. The aqueous herbal extract was first converted into semi-solid material by using encapsulation within ca-alginate hydrogel beads and followed by drying in a packed bed drying system. The effect of process variables on the drying kinetics of encapsulated extract was studied. The antioxidant content of the herbal extract was also monitored in order to evaluate the degree of product degradation during drying.

## MATERIALS AND METHODS

The aqueous extract of *Piper sarmentosum* (Furley, Malaysia) was used as the model herbal fluid. The herbal antioxidants content was selected as the major response parameter. Based on existing method, an improved DPPH method for quantification encapsulated herbal antioxidant was used (Yim et al., 2009). Encapsulation of herbal extract was achieved by using two approaches, i.e. dripping-gelation and absorptive encapsulation method (Chan et al., 2009; Yim et al., 2009). Diffusion of herbal antioxidants during gelation is the main limitation when dripping-gelation method was used. The release mechanism of the herbal antioxidants was determined based on Ritger and Peppas equation that derived from Fick's Law: $M_t/M_\infty = kt^n$ (Equation 1), where $M_t/M_\infty$ is the fractional release of drug at time t; $k$ is a constant which incorporating characteristics of the macromolecular network system and the drug and n is the diffusional exponent, which is indicative of the transport mechanism of polymeric systems. The process variables studied were particle size, alginate M/G ratio, concentration, gelling cation concentration and extract strength. Based on this information, the feasibility of the method was determined and improved process strategies in controlling the antioxidant release were proposed. The encapsulated extract produced by absorption method (Chan et al., 2009) was used in the study of packed bed drying system which was fabricated in-house. The effect of drying variables (i.e. drying temperature, relative humidity of air, air flow rate and bead size) on the drying on both encapsulated extract and blank ca-alginate beads was evaluated. Also, the quality of the final products in terms of the preserved antioxidant content was also determined.





## RESULTS AND DISCUSSION

The effect of the process variables on the release kinetics of herbal antioxidants during gelation was studied. In all cases, a sharp release of the encapsulated antioxidant (about 80%) was observed during the initial gelation period (i.e. in the first 20 minutes) (data not shown). After this time, the antioxidant release was significantly reduced. The amount of antioxidant that could be retained within the beads after prolonged gelation time was about 10-20%. Varying the process variables did not obviously increase the encapsulation efficiency although some marginal improvements could be observed in some cases. The rate of release of antioxidant from the beads could be explained by the concentration gradient and the different stages in the bead formation process. During the initial gelling period, the rapid release of antioxidant was due to the large concentration gradient between the internal bead and the external medium. In addition, the thin gel-layer that was instantly formed at the drop surface at this stage could not create an effective mass transfer barrier to slowdown the release. The gel network could also be loose and may have large molecular cut-off. When the gelling period was prolonged, the rate of release decreased because the concentration gradient has become smaller. At the same time, the resistance to mass transfer has increased due to the formation and consolidation of gel networks throughout the beads. One way to solve this problem is by pre-loading the antioxidant into the gelation bath in order to reduce the concentration gradient between bead and gelation bath. However, the main disadvantage of this method is the hold-up of active compounds in the gelation bath. This method may not be feasible if it involves large volume of gelation bath or expensive active compounds. Another way to increase the encapsulation efficiency is through absorption with blank calcium alginate bead, as reported in our recent work (Chan et al., 2009). This technique is particularly suitable for aqueous extract which has low molecular weight.

| Variables | | n | k, $min^{-n}$ | $R^2$ |
|---|---|---|---|---|
| Alginate concentration, % m/v | 2 | 0.42 | 0.30 | 0.93 |
| | 3 | 0.40 | 0.31 | 0.92 |
| | 4 | 0.32 | 0.40 | 0.91 |
| Bead diameter, mm | 2.2 | 0.42 | 0.30 | 0.93 |
| | 3.5 | 0.41 | 0.21 | 0.97 |
| $CaCl_2$ concentration, % m/v | 1.5 | 0.42 | 0.30 | 0.93 |
| | 7.5 | 0.48 | 0.27 | 0.99 |
| | 15 | 0.17 | 0.40 | 0.95 |
| Alginate type | High-G | 0.42 | 0.30 | 0.93 |
| | High-M | 0.40 | 0.36 | 0.96 |
| Extract loading | 1.00x | 0.42 | 0.30 | 0.93 |
| | 0.75x | 0.39 | 0.37 | 0.93 |
| | 0.50x | 0.64 | 0.28 | 0.95 |
| | Mean | 0.40 | 0.32 | |

**Table 1: Calculated parameters in Ritger and Peppas equation**

Ritger and Peppas equation (1987) was used to determine the release mechanism of the herbal antioxidant during gelation (Table 1). Under the process conditions, the average diffusional exponent, n is about 0.40. Theoretically, the solute release from a spherical device is described by Fickian diffusion if the diffusional exponent n has the limiting value of 0.43. This reinforces the earlier explanation that the antioxidant release during gelation was caused by the concentration gradient. Meanwhile, the characteristic constant, k of the polymeric system composed of ca-alginate-*P. sarmentosum* extract in $CaCl_2$ is in the range from 2.1–4.0 x $10^{-1}$ $min^{-n}$. The modeling of the release of herbal antioxidants from the encapsulated matrix during gelation is important to facilitate and optimise the design of effective encapsulation system by using dripping-gelation method. The average diffusional exponent, n was calculated for 60% of the fractional released herbal antioxidant by setting $M_t/M_\infty \geq 0.60$ in the Equation 1 which finally yields $M_t/M_\infty = 0.32t^{0.40}$ (Equation 2). Experimental data were calculated against the predicted data derived from the equation. The average absolute deviation of this equation was about 15% indicating that

the influences of the examined variables on the system were minimal. The release of herbal antioxidants during gelation could be satisfactory described with this semi-empirical equation. Base on this equation, encapsulation efficiency of herbal antioxidants by using dripping-gelation method is always determined by the gelation time. Shorter gelation time would be the only most effective process condition to preserve most of the herbal antioxidants within the encapsulated matrix. In this case, the gelation time has to be less than 5 minutes to yield encapsulation efficiency of 50% and above. This is possible if the beads can be harvested immediately, provided the beads have adequate strength for handling.

In general, it was observed that usual trend of higher drying rates as a function of increasing drying temperature, air flow rate, particle size and decreasing air relative humidity (data not shown). Under these conditions, the air is conditioned to higher driving force to promote the transport of water vapor. Besides, the drying rates of encapsulated extract were consistently lower than that of the blank ca-alginate beads under the same conditions. This is because higher bound water content in the encapsulated extract than that of the blank ca-alginate beads and bound water is more resistant to evaporate than free water.

| Variables | | $D_{eff}$ x $10^{-12}$ ($m^2s^{-1}$) | |
|---|---|---|---|
| | | Blank | Encapsulated |
| Drying temperature, ˚C | 45 | 6.54 | 6.54 |
| | 60 | 8.79 | 6.74 |
| | 75 | 10.2 | 6.54 |
| Air flow rate, LPM | 4 | 5.31 | 4.70 |
| | 6 | 8.79 | 6.74 |
| | 8 | 10.0 | 8.79 |
| Air humidity, RH% | 10 | 6.54 | 6.54 |
| | 20 | 5.11 | 4.29 |
| | 30 | 4.09 | 4.29 |
| Particle diameter, mm | 2.2 | 8.79 | 6.74 |
| | 3.3 | 18.9 | 18.85 |

**Table 2: Effect of the process variables on the effective moisture diffusivity, $D_{eff}$ of blank ca-alginate and encapsulated extract**

Calculation of effective moisture diffusivity, ($D_{eff}$) was taken into consideration the first term of drying curves where moisture ratio (MR) < 0.6 (Crank, 1975; Rizvi, 1986). The analysis of the $D_{eff}$ shows that increment of drying temperature resulted in higher $D_{eff}$ values of blank ca-alginate beads (Table 2). Meanwhile, the $D_{eff}$ for the encapsulated extract was not influenced by the increment of drying temperature. It could be due to change of thermal conductivity of the particles after absorption of extract and the drying temperature range of 45-75°C was not adequate to elevate the $D_{eff}$ of encapsulated extract. Based on kinetic theory, hike of drying temperature would shorten the time needed to achieve equilibrium moisture content due to the increased energy of water would promote the transport of the molecules. Higher air flow rate drove the drying curves faster in achieving the equilibrium moisture content. The drying which initially takes place at the surface of the spherical particles is more directly affected by air flow rate particularly for particles arranged in packed bed geometry. In packed bed geometry, the spherical particles form voids in between them which allow the distributive movement of air. The initial surface evaporation is gradually replaced by an evaporation boundary that retreats to the interior of the particles. Drying air with lower relative humidity could enhance the drying efficiency. In fact, decreasing the air RH increased the concentration gradient between water in the drying air and the particles. Besides, air with lower RH could carry higher sensible heat which imply higher driving force of the air and increased the efficiency of the migration of water vapor from the particles to the drying air. Particles with bigger size showed higher drying rate in the packed bed drying system because bigger spherical particles form bigger voidage among themselves when packed together. Void fraction in the packed bed





system for particles with 2.2 and 3.3 mm in diameter were 0.159 and 0.223 respectively. Therefore, in a packed bed consists of particles with bigger diameter had higher void fraction or void volume could eased the movement of air through the bed and consequently boosts the $D_{eff}$ and drying rate.

| Variables | | Preserved antioxidants, % | |
|---|---|---|---|
| | | Mean | s.d. |
| Drying temparature, ˚C | 45 | 82.62 | 0.80 |
| | 60 | 82.77 | 4.06 |
| | 75 | 89.84 | 3.16 |
| Air flow rate, LPM | 4 | 87.51 | 11.04 |
| | 6 | 82.77 | 4.06 |
| | 8 | 94.27 | 4.83 |
| Air humidity, RH% | 10 | 82.62 | 0.80 |
| | 20 | 92.53 | 2.90 |
| | 30 | 95.31 | 1.22 |
| Particle diameter, mm | 2 | 84.02 | 4.36 |
| | 3 | 82.77 | 4.06 |

**Table 3: Effect of the process variables on the preserved antioxidants**

The encapsulated herbal antioxidant contents of the final products were satisfactory preserved (82.6–9 5.3%) and not considerably affected by different drying conditions used in this study (Table 3). On the other hand, more antioxidant contents could be preserved when air with higher RH was used. Similar phenomena were reported in literatures, but there is still lacking of comprehensive explanations for the mechanism behind. Nevertheless, it is speculated that this phenomenon could be resulted from lower oxidative stress of drying air with higher humidity. Consequently, this situation reduces the occurrences of oxidation on the encapsulated extract.

## CONCLUSIONS

By using dripping-gelation, prolonged gelation time resulted in about 10-20% encapsulation efficiency. The release kinetics was primarily driven by the concentration gradient, as verified by the value of diffusional exponent (n). The release of the encapsulated antioxidant can be described with the equation, $M_t/M_\infty = 0.32t^{0.40}$. Drying efficiency of encapsulated extract and blank ca-alginate beads in packed bed system was mainly a function of increasing drying temperature, air flow rate, particle size and decreasing drying air humidity. Drying rates of encapsulated extract were generally lower than that of the blank ca-alginate beads. Overall, the antioxidant contents of the final products were not considerably affected by different drying conditions.

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