STUDY ON PREPARATION OF BLACK SHALLOT DRIED EXTRACTS BY SPRAY DRYING METHOD

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Summary

Objectives: Development of a preparation process for black shallot dry extracts by the spray-drying method. ***Materials and methods:*** Preparation of black shallot extract by hot extraction method with 50% EtOH; preparation of dried extracts from black shallot extract by spray drying method, quantification of cycloalliin by HPLC; investigating the factors affect the yield and quality parameters of dried extracts. ***Results:*** Evaluated and selected suitable conditions to prepare dried black shallot powder by spray drying method, including excipients AE/HPMC E6 (50/50), TD/CR ratio: 25%, temperature spray drying: 130°C, fluid supply speed 30 mL/min, solids/spray fluid ratio: 15%, nozzle compressed air pressure: 2.0 bar. ***Conclusion:*** The process of preparing black shallot dried extracts by spray drying has been developed with 85.90% yield and active ingredients recovery efficiency reaching 92.58%.

* *Keywords: Black shallot; Spray-drying; Cycloalliin; Dried powder.*

INTRODUCTION

Black shallots are a product of fermentation from fresh shallots (*Allium ascalonicum*) over a period of time under the influence of suitable temperature and humidity. Black shallots, after fermentation, have dark a brown color, sweet taste, and light aroma like ripe fruit. Vietnam Military Medical University has been conducting research on fermenting black shallots from locally available raw materials, initially studying the chemical composition and biological effects of black shallot products [1, 2].

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In order to be conveniently and stably used in modern dosage forms, studies on extracts and formulations with a high content of active ingredients are needed [3]. Stemming from the above reasons, we conducted research to develop a process to prepare black shallot dry extracts with high cycloalliin content and recovery efficiency. From there, it is possible *to create dry extracts that can be used in the preparation of tablets, capsules containing* black shallot.

MATERIALS, EQUIPMENT, AND METHODS

1. Materials and equipment

Black shallots are fermented from shallots provided by the Center of Applied Research and Drug Production, Vietnam Military Medical University, to meet In-house standards.

Standard substances: Cycloalliin (CYC) with 99.5% content of Fujifilm Wako Pure Chemical Corporation, Japan; chemicals and solvents: methanol, acetonitrile, phosphoric acid, ETC met analytical standards; Excipients: Maltodextrin (Mal), Aerosil (Aer), Hydroxypropyl methyl cellulose E6 (HPMC E6), Gum Arabic, Lactose, PVP K30 meet pharmaceutical standards.

Equipment: Alliance Waters 2695D High-performance liquid chromatography (HPLC), 4 solvent channels, 2487 UV detector, Empower 2 software, USA, Wakopak Wakosil 5NH2 column (250x4.6 mm; 5 µm); 40-liter reflux hot extractor (Vietnam); LPG-5 spray drying equipment (China) with high-speed centrifugal spray type, spray disc with 24 nozzles (3 mm), same direction spraying, airflow 240 m^3/hour, evaporation rate 5 kg/hour; automatic humidity determination machine ADAM AMB 310 (UK) and some other equipment.

2. Methods

** Preparation of black shallot extract:*

Black shallot extract was prepared by hot extraction method with 50% ethanol, solvent/medicinal ratio is 20/1, temperature: 80°C, 120 minutes, extraction twice. The extracts were combined with removing the solvent at a temperature of 70 ± 5^{o}C, the pressure was reduced (60 - 70 mmHg) until a high ratio of 1:1, then to settle, separated the extraction (DC1) and the residue (C1). The DC1 was added 4 times ethanol 96%, squirrel, settled, and filtered to collect filtrated (DL1). The precipitation was added 2 times ethanol 96%, stirred, settled, filtrated to collect the filtrated (DL2), and removed the precipitation. Combining DL1 and DL2 and recovering solvent under reduced pressure to a high 1:1 ratio (Part 1). Part C1 was added 2 times ethanol 96% and stirred until completely dissolved. Then adding 4 times the volume of hot distilled water, kept stirring and decant to collect the extract, discarding the insoluble precipitation. Concentrated the extract to a high ratio of 1:1 (Part 2). Combine part 1 and part 2, mix well, thicken and adjust to a high 1:1 ratio. Extraction with 2 kg of medicinal herbs/batch.

** Preparing dried black shallot powder:*

Black shallot liquid extracts 1:1 (100g) are mixed with excipients (water can be added) to obtain a spray solution with a solid ratio according to each evaluated condition. Spray drying is carried out on LPG-5 equipment with high-speed centrifugal spray. The evaluated parameters include type of excipients supporting spray drying, a ratio of excipients/solids in spray solution (TD/CR), spray drying temperature (input temperature) and fluid supply speed, and substance ratio solid in spray solution (CR/DP). The evaluation specifications include: Moisture, apparent density, compression index CI, spray drying efficiency, cycloalliin content, and recovery efficiency.

** Evaluating quality criteria of dry extracts:*

- Moisture: Proceed according to the mass loss method due to drying of DVVN V, PL 9.6 (2.0g, 105ºC, 4 hours) [4].

- Hygroscopicity: The spray-dried powder sample (about 2g) was placed in a petri dish and stored in a desiccator at about 25ºC, and relative humidity of 75 ± 2% was created by saturated NaCl solution. After 7 days, re-determine the mass of the powder samples. Hygroscopicity is expressed as the number of grams of water absorbed per 100g of dry solids. The color change in powder was also observed simultaneously [5, 6].

- Powder density (g/mL) and compression index CI (%) [6]:

Weigh approximately 3g of powder (m^2), transfer to a clean dry 25 mL measuring cylinder, read the initial volume of powder (V1), and knock to constant volume (V2). The apparent density after knocking (dT), the apparent crude density (dB) and the compression index (CI) are calculated by the formula:

$$d_B = m/V_1; \qquad d_T = m/V_2;$$

$$CI\ (\%) = (d_T - d_B) \times 100 / d_T$$

Evaluate the flowability of the powder according to USP 38 [0]:

No.	Compression index CI	Flowability
1	< 10	Very good
2	11 - 15	Good
3	16 - 20	Rather
4	21 - 25	Flowable
5	26 - 31	Less flowable
6	32 - 37	Very poor
7	> 38	Very, very poor

- Cycloalliin content:

Quantification of cycloalliin dry powder by HPLC with the following conditions: Wakopak Wakosil 5NH2 column (250x4.6 mm; 5 µm); wavelength: 210 nm; mobile phase: 0.5% phosphoric acid solution in water (A) and 0.5% phosphoric acid solution in acetonitrile (B) were run on a gradient program (0 - 5 min: 84%B, 5 - 15 min: 84 - 80%B, 15 - 20 minutes: 80 - 84%B, 20 - 30 minutes: 84%B); running time: 0 - 30 minutes; flow speed: 1.5 mL/min; sample pump volume 10l [8].

Cycloalliin content was calculated according to the following formula:

$$HL\ (mg/g) = \frac{S_T \times C_C \times V \times n \times 100}{S_C \times M \times (100 - h)}$$

ST: Sample peak area, SC: Standard sample peak area, CC: Standard sample concentration; V: Volume of extract (mL); n: Dilution factor, M: Weight (g), h: Moisture content of the sample (%).

- Active ingredient recovery efficiency (HS) and spray drying efficiency (HSPS): The ratio (%) of active ingredient content or volume of the product obtained compared to the theoretical one.

RESULTS

1. Results of preparation of black shallot extract

Black shallot extraction was carried out at the scale of 2 kg/batch with three different extraction batches. The results are presented in Table 1.

Table 1: Result of black shallot extract 2 kg/batch.

No.	Medicinal herbs (g)	Moisture (%)	Extract (L)		CYC content (mg/g)	Extraction efficiency (%)
1	2000.15	24.66	77.32		70.33	86.99
2	2000.14		76.94		67.59	83.59
3	2000.11		78.46		68.41	84.60
Total	6000.40		232.72	Medium	68.77	85.06
				SD	1.41	1.75

Table 1 results show that: Extract 3 batches black of shallot with a total weight of about 6000.40g (4520.70g of dried herbs) obtained 232.72 liters of extract, extract/material dry ratio is 51.48/1 (mL/g). The content of CYC in the extract is 68.77 ± 1.41 mg/g (calculated according to dried herbs), and extraction efficiency is 85.06 ± 1.75%.

Concentrations and impurities of 230 liters of 1:1 black shallot extract are presented in Table 2.

Table 2: Extraction, concentration, and impurities of black shallot 1:1.

Content	Dried out black shallot	Extract	Dry extract 1:1 without impurities removed	Dry extract 1:1 with impurities removed
Weight, volume	4.47 kg	230.0 liter	4.47 kg	4.47 kg
CYC's Content	80.86 mg/g	1.30 mg/mL	144.12 ± 2.67 mg/g (1)	182.30 ± 3.49 mg/g (1)
CYC's Weight (g)	365.52	297.96	289.94	279.86
Ratio of solid in dry extract			45.03%	34.36%
Extraction and concentration efficiency of dry extract calculated by CYC			79.32%	-
Extraction efficiency, concentration and impurity removal calculated by CYC				76.56%

Note: (1) CYC content in 1 gram of solid in dry extract calculated by drying out.

Results of Table 2 show that: Black shallot liquid extracts 1:1 has 34.36% solids, cycloalliin content is 182.30 mg/g. Black shallot liquid extract 1:1 was used to research and develop a process for preparing black shallot dry extracts by spray drying method.

2. Effect of excipients used in spray drying

Carry out spray drying under the same conditions: TD/CR ratio is 20%; inlet temperature 120ºC, fluid supply speed 20 mL/min; injector pressure 2.0 bar; CR/DP ratio 10%, but with excipients are maltodextrin, Aerosil (Aer), hydroxypropyl methylcellulose E6 (HPMC E6), gum Arabic, lactose, PVP K30 and a mixture of excipients AE/HPMC E6 and AE/gum Arabic is all at 50/50 ratio. Results are presented in Table 3.

Table 3: Effect of excipients on spray drying of black shallots extracts (n = 3).

Sample	Moisture (%)	Hygroscopicity (%)	Apparent specific weight (g/mL)	CI	CYC content (mg/g)	Recovery efficiency of CYC (%)	Spray drying efficiency (%)
CT1 (AE)	4.89 ± 0.06	12.83 ± 0.75	0.545 ± 0.007	24.86 ± 1.36	132.55 ± 1.18	87.25 ± 0.78	66.97
CT2 (HPMC E6)	4.64 ± 0.07	12.52 ± 0.55	0.399 ± 0.010	32.03 ± 1.78	129.69 ± 1.78	85.37 ± 1.17	64.23
CT3 (Gum Arabic)	3.81 ± 0.06	12.40 ± 0.46	0.384 ± 0.008	43.97 ± 1.29	126.78 ± 1.95	83.45 ± 1.29	69.81
CT4 (AE/ HPMC E6)	3.78 ± 0.08	11.70 ± 0.75	0.504 ± 0.009	30.91 ± 1.80	137.94 ± 1.77	90.80 ± 1.17	79.20
CT5 (AE/Gum Arabic)	4.48 ± 0.09	11.79 ± 0.70	0.477 ± 0.014	32.74 ± 1.95	125.06 ± 1.75	82.32 ± 1.15	66.73
CT6 (Mal)	-	-	-	-	-	-	-
CT7 (Lactose)	-	-	-	-	-	-	-
CT8 (PVP K30)	-	-	-	-	-	-	-
CT0 (KTD)	-	-	-	-	-	-	-

Note: (-) product was not obtained

The results of Table 3 show that: Excipients supporting spray drying have a great influence on the quality of black shallot dry extract powder. In the formulas CT0, CT6, CT7, and CT8, dry powder was not obtained, formulas from CT1 to CT5, when adding excipients at the rate of 20%, all obtained extract in the form of dry powder, loose, low-clumping powder with high efficiency. Spray drying efficiency from 64.23 to 79.20%. The highest spray drying efficiency was 79.20% in formula CT4 (excipients AE/HPMC E6) and gradually decreased in formulas CT3, CT1, CT5, and CT2. Moisture and hygroscopicity in formula CT4 are lowest, while formulas CT1, CT2, CT3, and CT5 have higher moisture content and hygroscopicity, but all are low (< 5%). The apparent specific weight of dry powders ranges from 0.384 to 0.545 g/mL and is highest when spray-dried with Aerosil. When evaluating the flowability according to the CI of USP 38, the formula CT1 has a CI in the range of 21 - 25, that is, it is smooth, the formula CT4 has a CI in the range of 26 - 31, that is, it has a poor flow. CT2 and CT5 have a CI in the range of 32 - 37, which means that the smoothness is very poor, and the formula CT3 has a CI greater than 38, which means that the flow is very very poor. The content and recovery efficiency of CYC among the investigated formulations also differed. The formula CT4 gave the highest CYC content and recovery (89.73%) and tended to decrease in the formulas CT1, CT2, CT3, and CT5.

Thus, formula CT4 has high spray drying efficiency, CYC content, and recovery efficiency, low hygroscopicity and moisture. So the AE/HPMC E6 excipient mixture was selected for the next test.

3. Effect of excipients/solids in the spray solution

Spray drying was carried out under the same conditions as CT4 but with the TD/CR ratio of 10, 15, 20, 25, 30, 35, and 40%, respectively. Results are presented in Table 4.

Table 4: Effect of excipients/solids ratio on spray drying black shallots (n = 3).

Sample	Excipients/ Solids ratio (%)	Moisture (%)	Hygroscopicity (%)	Apparent specific weight (g/mL)	CI	CYC content (mg/g)	Recovery efficiency of CYC (%)	Spray drying efficiency (%)
CT9	10	4.60 ± 0.08	13.81 ± 1.05	0.426 ± 0.006	47.64 ± 1.26	147.60 ± 1.28	89.06 ± 0.77	68.87
CT10	15	4.15 ± 0.09	12.30 ± 0.38	0.452 ± 0.008	44.41 ± 1.67	142.06 ± 1.83	89.62 ± 1.15	72.10
CT4	20	3.78 ± 0.08	11.70 ± 0.75	0.504 ± 0.009	30.91 ± 1.80	137.94 ± 1.77	90.80 ± 1.17	79.20
CT11	25	3.43 ± 0.10	11.35 ± 0.46	0.513 ± 0.009	28.91 ± 1.08	137.20 ± 1.19	94.08 ± 0.82	85.70
CT12	30	3.29 ± 0.09	10.36 ± 0.64	0.518 ± 0.008	27.56 ± 1.55	132.97 ± 2.07	94.82 ± 1.48	86.56
CT13	35	3.20 ± 0.07	10.51 ± 0.82	0.524 ± 0.008	24.11 ± 1.69	129.13 ± 1.61	95.63 ± 1.19	89.32
CT14	40	3.12 ± 0.12	10.32 ± 0.62	0.519 ± 0.006	22.56 ± 1.04	125.62 ± 0.74	96.47 ± 0.57	91.24

Results of Table 4 show that: The TD/CR ratio significantly affects dry powder characteristics, spray drying efficiency, and active ingredient recovery efficiency. When the ratio of excipients/solids is increased, powder with better properties is obtained. The higher proportion of excipients, the lower moisture content, and hygroscopicity of powder mass, higher density of dry powder, and lower compressive index CI, which means an increase in flowability. The moisture content of samples was at a low level (< 5%) and met the requirements of moisture content of dry extracts according to Vietnamese Pharmacopoeia V. When increasing the proportion of excipients, there is a tendency to slightly increase the density, but only on average. Samples CT13 and CT14 are flowable, samples CT4, CT11, and CT12 are less flowable, and the remaining samples were at a very poor level of flowability when assessing the ability to flow according to USP 38. When increasing the ratio of excipient/solids, CYC content decreased due to concentration dilution, but the efficiency of active ingredient recovery tended to increase gradually. Because excipients have a certain role in protecting active ingredients during spray drying, spray drying efficiency increased gradually when the excipient ratio was increased, reaching the highest level of 91.24% (CT14). Considering factors of quality criteria, spray drying efficiency, formula CT11 (TD/CR ratio is 25%) is the most suitable, so it should be selected for the next test.

4. Effect of spray drying temperature and fluid supply speed

Carry out spray drying under the same conditions as CT11, but at different spray drying temperatures (inlet temperature) and fluid supply speed. Results are presented in Table 5.

Table 5: Effect of spray drying temperature and fluid supply speed on spray drying black shallots (n = 3).

Sample	Temperature, fluid supply speed	Moisture (%)	Hygroscopicity (%)	Apparent specific weight (g/ mL)	CI	CYC content (mg/g)	Recovery efficiency of CYC (%)	Spray drying efficiency (%)
CT11	120°C, 20 mL/min	3.43 ± 0.10	11.35 ± 0.46	0.513 ± 0.009	28.91 ± 1.08	137.20 ± 1.19	94.08 ± 0.82	85.70
CT15	130°C, 30 mL/min	3.38 ± 0.13	12.06 ± 0.96	0.518 ± 0.005	29.19 ± 0.99	135.92 ± 2.00	93.20 ± 1.37	86.39
CT16	140°C, 30 mL/min	3.28 ± 0.12	12.47 ± 0.38	0.492 ± 0.004	29.50 ± 1.04	134.29 ± 2.25	92.08 ± 1.54	81.21
CT17	150°C, 40 mL/min	3.17 ± 0.08	11.62 ± 0.74	0.491 ± 0.012	30.58 ± 1.11	131.27 ± 2.56	90.01 ± 1.76	76.18

Results of Table 5 show that: When spray drying at a temperature of 120 - 150°C with 3 levels of fluid supply speed of 20, 30, 40 mL/min, they all form a dry, loose powder with low moisture content. (< 5%), but tends to decrease moisture content with increasing spray drying temperature. The density and hygroscopicity of samples were similar, with little variation. Active ingredient content, active ingredient recovery efficiency, and spray drying efficiency tended to increase when decreasing temperature and fluid supply speed, but in formulas, CT11 and CT15 were equivalent.

From the above evaluations, the spray drying temperature is selected as 130°C. Liquid supply speed is 30 mL/min because product had the highest spray drying efficiency, CYC recovery efficiency was equivalent to other samples, low moisture content powder, the obtained powder is dry and loose.

5. Effect of solids ratio in the spray solution

Spray drying is carried out under the same conditions as CT15, but only the solids in the spray solution (CR/DP) are different. Results are presented in Table 6.

Table 6: Effect of solids/spray ratio on spray drying black shallots (n = 3).

Sample	**Solids/ Spray solution (%)**	**Moisture (%)**	**Hygroscopicity (%)**	**Apparent specific weight (g/mL)**	**CI**	**CYC content (mg/g)**	**Recovery efficiency of CYC (%)**	**Spray drying efficiency (%)**
CT15	10	3.38 ± 0.13	12.06 ± 0.96	0.518 ± 0.005	29.19 ± 0.99	135.92 ± 2.00	93.20 ± 1.37	86.39
CT18	15	3.27 ± 0.11	11.41 ± 0.81	0.549 ± 0.008	26.87 ± 0.68	135.02 ± 1.77	92.58 ± 1.21	85.90
CT19	20	3.23 ± 0.08	11.02 ± 0.90	0.567 ± 0.005	25.81 ± 0.72	136.95 ± 1.68	93.91 ± 1.15	81.76

Results of Table 6 show that: When increase percentage of solids in spray solution, there is a tendency to reduce moisture, hygroscopicity, spray drying efficiency and increase the density of powder, while content and recovery efficiency of powder are less affected. Formula CT18 for powder has better mechanical and physical properties than CT15 and higher spray drying efficiency than CT19. Therefore, spray-drying conditions in formula CT18 are the most suitable for preparing black shallot dry powder extracts.

DISCUSSION

Spray drying is one of the most preferred methods for preparing dry extracts from herbal extracts because of its ability to quickly convert from fluid extract to dry powder form, and the resulting product has superior morphology, particle size, the active ingredient content compared with conventional drying methods [9]. However, the most common difficulty in spray drying herbal extracts is the sticky phenomenon that makes the powder particles not dry, adhere to the spray chamber, reduce efficiency and even cannot be dried. Therefore, using excipients as a carrier not only improve the drying process, and increase spray drying efficiency but also improve some physical and mechanical properties, and active ingredient content of the product [10]. Results of preparation of black shallot dry extracts showed that: in formula without excipients (CT0), dry powder was not obtained, but some excipients added increased spray drying efficiency, improved mechanical properties and quality standards of the product. However, each type of excipient has different effects, for black shallot, spray drying with a mixture of excipients AE/HPMC (1:1) is the most suitable.

Increasing the excipients/solids ratio improves physico-mechanical properties, spray drying efficiency, and active ingredient recovery of black shallot dry extracts but reduces active ingredient content due to concentration dilution. Therefore, it is necessary to choose the most appropriate excipients/solids ratio to ensure the product has the appropriate physical and mechanical properties for the preparation of solid drug forms and has a high content of active ingredients. In this study, the excipients/solids ratio of 25% (CT11) was selected as the most appropriate. Spray-drying temperature and fluid supply speed have a decisive influence on the drying ability, efficiency, and quality of the product. When increasing spray drying temperature allow increasing in fluid supply speed. However, with black shallots, spray drying at 140 - 150°C gives lower spray drying efficiency and active ingredient content than at 130°C, while spray drying at 120 - 130°C is equivalent. Therefore, the most suitable spray drying temperature is 130°C because a higher fluid supply speed (30 mL/min) will shorten spray drying time than at 120°C.

The Solids/Spray solution ratio mainly affects the spray drying efficiency and density of the dry powder. The higher percentage of solids in the spray solution, the lower the spray drying efficiency while density increases gradually. When increasing the ratio Solids/Spray solution, spray drying will be more viscous, there is a risk that it is difficult to disperse the spray fluid evenly and it is difficult to form liquid droplets, thus reducing spray drying efficiency. When the Solids/Spray solution ratio is reduced, the spray drying process lasts longer because a larger amount of fluid is required, so it takes more time and energy. In this study, a 15% Solids/Spray solution ratio was selected as the most appropriate.

CONCLUSION

Research has established a process to prepare black shallot dry extracts by spray drying method. From evaluations of influence factors on efficiency and quality dry powder extracts, the appropriate spray drying conditions were selected: Excipients AE/HPMC E6 (50/50), excipient ratio 25%, spray drying temperature 130°C, fluid supply speed 30 mL/min, solids/spray solution ratio 15%, nozzle compressed air pressure 2.0 bar. With selected conditions, black shallot dry powder extracts was obtained with moisture 3.27 ± 0.11%, density 0.549 ± 0.008 g/mL, CI index 26.87 ± 0.68%, cycloalliin content 135.02 ± 1.77 mg/g, active ingredient recovery efficiency 92.58% and spray drying efficiency 85.90%.

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