CAFEi2016-71

Response surface optimization of grinding temperature for cryogenic grinding of black pepper

B.M. Ghodki and T.K. Goswami[a]

Agricultural and Food Engineering Department, Indian Institute of Technology Kharagpur, 721302, India

Abstract

The cryogenic grinding temperature for grinding of black pepper was optimized within the domain of -120 to 40°C using response surface methodology (RSM). The aim of the study was to maximize the flowability and volatile oil content along with minimal water activity, total color difference (ΔE), specific energy consumption and particle size of the powder. A one factor-five-level RSM design was applied for the optimization. The linear and quadratic polynomial models for the computed responses were used for their prediction during the grinding process. The numerical optimization indicated that cryogenic grinding of black pepper sample of 3.90% dry basis with a feed rate of 1.5 kg h^{-1} and at a peripheral speed of 26 m s^{-1} would be optimally ground at -42°C with the highest desirability value of 0.583. Further, the optimized grinding condition was validated at the closest possible condition within the domain. At the optimal grinding temperature, water activity of 0.55 and 99.34 μm particle size with 38.85° angle of repose and Hausner ratio of 1.17 containing 1.23 mL (100 g)$^{-1}$ of volatile oil was recorded. Also, the specific energy consumption of 478 kJ kg^{-1} with the least total color difference (ΔE<0.25) was observed in the optimized sample.

Keywords: cryogenic grinding, black pepper, response surface methodology, optimization, water activity

INTRODUCTION

Black pepper is one of the most demanding spices worldwide due to its distinct aroma and taste; hence, also called "The King of the Spices." The dried fruits (partially ripened berriesA, moisture, fats, and fiber content (Pesek and Wilson, 1986; Murthy and Bhattacharya, 2008; Ghodki and Goswami, 2015). The difficulties of the conventional grinding process can be overcome by cryogenic grinding (cryo-grinding) technology. Grinding of spices at a lower temperature usually in the presence of a cryogen like liquid nitrogen (LN$_2$) is termed as the cryo-grinding of spices. The optimal processing conditions should be determined to produce the higher quality of end product with consumer satisfaction (Mao and Danzart, 2008). Response surface methodology (RSM) is most widely used and acceptable method to optimize process conditions (Banga et al., 2003). Hence, the objective of this study is to optimize the grinding temperature for cryo-grinding of black pepper based on the quality of black pepper powder using RSM as a numerical optimization technique.

MATERIALS AND METHODS

Raw material

Black pepper (var. *panniyur-1*) seeds were collected from Indian Institute of Spice Research, Marikunnu, Calicut, India. The best quality of black pepper seed of 4.90 ± 0.30 mm geometric mean diameter, 3.90 ± 0.10% dry basis (d.b.) moisture content, and sphericity of 0.98 ± 0.03 were taken for cryo-grinding experimentation.

---

[a] E-mail: tkg@agfe.iitkgp.ernet.in

Experimental design

Cryo-grinding experiments were performed using a cryo-grinding system whose details were discussed in author's previous work (Ghodki and Goswami, 2015) to obtain ground black pepper samples using LN_2 as a cryogen. The mill had a rotor of 186×10^{-3} m diameter having 4 number of fixed rotor ribs, with a maximum peripheral speed of 29.20 m s^{-1}. Statistically designed experiments were used to optimize grinding temperature (independent parameter) based on the observed values of critical responses (dependent parameters) viz., specific energy consumption, total color difference, flowability parameters (Hausner ratio: HR and angle of repose: α), water activity, volatile oil content and particle size of ground pepper. While all the other grinding parameters: feed rate of 1.5 ± 0.02 kg h^{-1}, 200 g of black pepper seed (fed to mill), peripheral speed of 26 ± 0.14 m s^{-1}, moisture content of $3.90 \pm 0.10\%$ d.b., four number of rotor ribs, grinding time of 10 minutes (90% of ground pepper obtained) and sieve size of 500 μm were kept constant throughout the experiment. A similar cryo-grinding procedure was followed as defined for cryo-grinding of cassia (Goswami and Ghodki, 2015; Ghodki and Goswami, 2015).

Design-Expert software (version 7.0.0 (Dx7) Stat-Ease, Inc., Minneapolis, MN, USA) was used to optimize a five-level-one-factor, with one-factor design and RSM. The experimental design includes five levels of grinding temperature with a total of seven experimental runs to optimize the grinding temperature for cryo-grinding of black pepper seeds. The experimental runs consisting of one replica each at the centre point (0) and -0.5 and +0.5 levels as well as two replicas each at the extreme levels (-1 and +1) of the design to evaluate pure error. The extreme levels of the experimental domain have been selected according to the literary knowledge and preliminary experiments conducted (Table 1). A particular standard order was allotted to each experimental run, and corresponding samples were treated in triplicate. Once the experiments were performed, the response variables were fitted with appropriate linear and quadratic models to correlate the response variables with the independent variable (Halim et al., 2009; Ghodki and Goswami, 2015). The quadratic model for predicting the optimal design conditions can be expressed according to following equation (Ghafari et al., 2009):

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i \cdot X_i + \sum_{i=1}^{k} \beta_{ii} \cdot X_i^2 + \sum_{i \leq j}^{k} \sum_{j}^{k} \beta_{ij} \cdot X_i \cdot X_j + \cdots + e \tag{1}$$

where *i* is the linear coefficient, *j* is the quadratic coefficient, *β* is the regression coefficient, *k* is the number of factors studied and optimized in the experiment and *e* is the random error.

Table 1. Experimental range and levels of grinding temperature indicating the actual and coded value.

Independent parameter (Factor)		Levels				
Grinding temperature[1] (°C)	Coded	-1	-0.5	0	0.5	1
	Actual	-120	-80	-40	0	40

[1]Standard deviation of ±3°C.

The models were further analyzed using analysis of variance (ANOVA). The statistical significance of the factors toward the selected responses of the process was examined using F-test (Ahmad et al., 2005; Ghodki and Goswami, 2015) at desired significant level of 0.05. Moreover, the significant model was chosen. Further, the accuracy of the developed regression model was examined by comparing the experimental values of the foretold quality parameters of the powder with the data predicted by polynomial models.

Measurement of parameters affecting quality of black pepper powder

Different critical quality parameters influenced by the grinding temperature were screened out and were analyzed according to the available literary knowledge. Energy consumed during grinding of the sample was measured as specific energy consumption (kJ kg^{-1}) following the method discussed for cryo-grinding of cassia (Ghodki and Goswami, 2015).

Hand-pounded black pepper sample with a particle size less than 500 μm was taken as a standard to evaluate the total color difference. The Hunter Lab color scale: L, a, b values were measured using a chromameter (Model CR-400, Konica Minolta Sensing Inc., Japan). Flowability of the black pepper powders was analyzed by applying HR and α test (Liu et al., 2008). Aqualab Water Activity Meter (Model CX2, Decagon Devices Inc., USA) was used to measure the water activity following the method defined for gac fruit aril powder (Kha et al., 2010). The volatile oil content of the samples was determined by the hydro-distillation technique as described for black pepper powder (Goswami and Ghodki, 2015). The particle size (volumetric mean diameter) of the representative samples were examined by the laser particle size analyzer (Model Analysette 22 MicroTech Plus, Fritsch, Germany). The average value of triplicates for each quality parameters is reported.

Optimization and validation of process parameter for cryo-grinding of black pepper

Numerical optimization feature of Design-Expert 7.0.0 software was applied to optimize the process parameter (grinding temperature) for obtaining the powder with desirable quality attributes. The software searches for the best combination of the independent variable that simultaneously satisfy the requirements placed on each of the response and independent variable. The optimum values were selected by choosing the highest desirability level in the numerical optimization method. Further, the experiments were performed at the nearest possible process condition to validation the predicted optimized values.

RESULTS AND DISCUSSION

The average values of responses: specific energy consumption, total color difference, flowability, water activity, volatile oil content and particle size of the powder were recorded from seven experimental runs were recorded from seven experimental runs (Table 2). The observed values of responses were further statistically analyzed (ANOVA and F-test) at every point based on the experimental design, and testing of the fit summary output revealed that the linear model was statistically significant for particle size and HR, while the quadratic model was found to be statistically significant for total color difference, specific energy consumption, water activity, angle of repose and volatile oil content (Table 3); hence, the models were used for further analysis. The accuracy of data predicted (predicted value) by the models was compared with the new set of experimental data (actual value) observed at all the five levels of grinding temperature. The actual versus predicted values for all the models are within the acceptable limit of 5%. The results observed are consistent with the report of cryo-grinding of cassia (Ghodki and Goswami, 2015).

Table 2. Design matrix of seven experimental runs showing parameter and responses used for optimization analysis.

| X[1] | Y[2] | Factor | Responses | | | | | | |
		T[3]	E[4]	ΔE[5]	α[6]	HR[7]	a_w[8]	V[9]	PS[10]
1	1	-120	98.57	1.33	42.34	1.20	0.64	1.79	54.38
2	5	-120	99.04	1.29	42.28	1.20	0.63	1.80	54.21
3	2	-80	377.14	0.37	39.93	1.18	0.59	1.55	55.45
4	4	0	582.86	0.41	38.10	1.15	0.53	0.98	133.45
5	3	40	771.43	0.91	36.28	1.14	0.52	0.90	141.56
6	6	40	770.18	0.93	36.60	1.14	0.52	0.92	142.15
7	7	-40	474.1	0.33	38.67	1.17	0.54	1.21	121.43

X[1]: Standard order; Y[2]: Run; T[3]: Grinding temperature (°C); E[4]: Specific energy consumption (kJ kg[-1]); ΔE[5]: Total color difference; α[6]: Angle of repose (°); HR[7]: Hausner ratio; aw[8]: Water activity; V[9]: Volatile oil content (mL (100 g)[-1]); PS[10]: Particle size (μm).

Table 3. Model equations for the responses with statistical parameters.

Y^1	Model (F-value)[9]	Equation (in terms of actual factors)	Significant model terms (F-value)[9]	R^2	Adj. R^2	Pred. R^2	Adeq. precision	CV (%)
E^2	Quadratic (120.46)	$E = 3.40T - 7.71 \times 10^{-3}T^2 + 633.47$	T (239.37), T×T (1.54)	0.984	0.976	0.962	22.28	9.72
ΔE^3	Quadratic (49.81)	$\Delta E = 8.79 \times 10^{-3}T + 1.36 \times 10^{-4}T^2 + 0.37$	T (12.01), T×T (87.61)	0.961	0.942	0.899	15.38	12.99
α^4	Quadratic (115.83)	$\alpha = -0.03T + 9.33 \times 10^{-5}T^2 + 37.53$	T (228.83), T×T (2.83)	0.983	0.975	0.958	21.79	1.01
HR^5	Linear (567)	$HR = -3.75 \times 10^{-4}T + 1.15$	T (567)	0.991	0.990	0.985	42.00	0.23
a_w^6	Quadratic (205.54)	$a_w = -3.38 \times 10^{-4}T + 4.80 \times 10^{-6}T^2 + 0.53$	T (381.55), T×T (29.54)	0.991	0.986	0.970	28.13	1.11
V^7	Quadratic (245.25)	$V = -4.08 \times 10^{-3}T + 2.03 \times 10^{-5}T^2 + 1.03$	T (479.83), T×T (10.67)	0.991	0.988	0.981	31.55	3.38
PS^8	Linear (48.27)	$PS = 0.60T + 124.17$	T (48.27)	0.906	0.887	0.855	12.25	14.47

Y^1: Responses; E^2: Specific energy consumption (kJ kg^{-1}); ΔE^3: Total color difference; α^4: Angle of repose (°); HR^5: Hausner ratio; a_w^6: Water activity; V^7: Volatile oil content (mL $(100 g)^{-1}$); PS^8: Particle size (µm); (F-value)[9]: Significant model and model terms are selected based on Fisher F-test with 95% confidence level (F, mean square regression/mean square residual) and a low probability value [p value i.e. (Prob > F) < 0.05]; additionally, the model with non-significant lack of fit was selected; T: Grinding temperature (°C).

The optimization performed by numerical optimization technique (Design-Expert 7.0.0 software) was targeted for the minimal values of specific energy consumption, total color difference, angle of repose, HR, water activity and particle size together with maximum retention of volatile oil for a grinding temperature domain of -120 to 40°C. The outcome of numerical optimization indicated that the black pepper sample would be optimally cryo-ground at -42°C with the highest desirability level of 0.583. At the optimal grinding temperature, the predicted value of responses: 0.55 water activity, 99.34 µm particle size, 38.85° angle of repose, 1.17 HR, 1.23 mL $(100 \text{ g})^{-1}$ volatile oil, 478 kJ kg^{-1} specific energy consumption with the least total color difference of 0.24 was observed. Further, the optimized grinding condition was validated by conducting the experiment at the closest possible condition within the domain, and it was noted that experimental outcomes were in close agreement with the predicted values of responses.

CONCLUSIONS

The following conclusions can be drawn from the study:
– The predicted models for specific energy consumption, total color difference, flowability (Hausner ratio: HR and angle of repose: α), water activity, volatile oil content and particle size of the powder can be used to navigate space defined in this study.
– The cryo-grinding of black pepper should be done at -42°C to obtain the excellent quality of ground pepper.
– The optimized condition will be beneficial for confining the range of grinding temperature prior to scale-up studies for cryo-grinding of black pepper.

ACKNOWLEDGEMENTS

Literature cited

Ahmad, A. L., Ismail, S., and Bhatia, S. (2005). Optimization of coagulation-flocculation process for palm oil mill effluent using response surface methodology. Environ. Sci. Technol. *39*(8), 2828-2834. http://dx.doi.org/10.1021/es0498080.

Banga, J.R., Balsa-canto, E., Moles, C.G., and Alonso, A.A. (2003). Improving food processing using modern optimization methods. Trends Food Sci. Tech. 14(4), 131–144 http://dx.doi.org/10.1016/S0924-2244(03)00048-7.

Ghafari, S., Aziz, H. A., Isa, M. H., and Zinatizadeh, A. A. (2009). Application of response surface methodology (RSM) to optimize coagulation–flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum. J. Hazard. Mater. 163(2-3), 650-656 http://dx.doi.org/10.1016/j.jhazmat.2008.07.090.

Ghodki, B.M., and Goswami, T.K. (2015). Optimization of cryogenic grinding process for cassia (cinnamomum loureirii nees L.). J. Food Process. Eng. http://dx.doi.org/10.1111/jfpe.12258.

Ghodki, B.M., and Goswami, T.K. (2016). Thermal and mechanical properties of black pepper at different temperatures. J. Food Process. Eng. http://dx.doi.org/10.1111/jfpe.12342.

Goswami, T.K., and Ghodki, B.M. (2015). Cryogenic grinding of cassia. Paper presented at: International Conference on Advances in Chemical, Biological & Environmental Engineering (ACBEE), March 29–30, pp. 242–249, Singapore.

Halim, S.F.A., kamaruddin, A.H., and Fernando,W.J.N. (2009). Continuous biosynthesis of biodiesel from waste cooking palm oil in a packed bed reactor: Optimization using response surface methodology (RSM) and mass transfer studies. Bioresour. Technol. *100*(2), 710–716 http://dx.doi.org/10.1016/j.biortech.2008.07.031.

Kha, T.C., Nguyen, M.H., and Roach, P.D. (2010) Effects of spray drying conditions on the physicochemical and antioxidant properties of the Gac (Momordica cochinchinensis) fruit aril powder, J. Food Eng. *98*(3), 385–392 http://dx.doi.org/10.1016/j.jfoodeng.2010.01.016.

Liu, L.X., Marziano, I., Bentham, A.C., Litster, J.D., White, E.T., and Howes, T. (2008). Effect of particle properties on the flowability of ibuprofen powders. Int. J. Pharm. *362* (1-2), 109–17 http://dx.doi.org/10.1016/j.ijpharm.2008.06.023.

Mao, M., and Danzart, M. (2008). Multi-response optimization strategies for targeting a profile of product attributes with an application on food data. Food Qual. Pref. *19* (2), 162–173 http://dx.doi.org/10.1016/j.foodqual.2007.08.003.

Murthy, C.T., and Bhattacharya, S. (2008). Cryogenic grinding of black pepper, J. Food Eng. *85* (1), 18–28 http://dx.doi.org/10.1016/j.jfoodeng.2007.06.020.

Pesek, C.A., and Wilson, L.A. (1986). Spice quality: effect of cryogenic and ambient grinding on color. J. Food Sci. *51* (5), 1386–1386 http://dx.doi.org/10.1111/j.1365-2621.1986.tb13135.x.