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Extraction, Stabilistiaon and Utilisation of Natural Food Colours from Grape Fruit Rind

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Investigation was carried out to develop natural food colour powder from grape fruit rind (waste) by spray drying. The colour was extracted with water and solvents *viz*., acetone, ethanol and citric acid. The experimental results showed that the 3 % citric acid extraction was best and the filtrate was spray dried at different drying air temperature (180, 200 and 220ºC) and flow rate (25 and 30 ml / min) using 10, 15 and 20 % of maltodextrin (MD). The yield recovery was maximum at 200ºC with a flow rate of 25 ml / min at 20 % maltodextrin. The stability of the colour was analysed by exposing the colour powder in various concentrations of pH, preservative and temperatures followed by packed in aluminium pouch (P_1), brown pet bottle (P_2) and brown glass bottle (P_3). The packed samples were stored in room (R_1) and refrigeration (R_2) temperatures for 3 months and analyzed their colour intensity (L* a* b*) and moisture content. The results revealed that the addition of preservative, increasing temperature, pH and storage periods had affected the stability of colour and moisture content. The samples packed in P_3 had better colour retention than P_2 and P_1 during storage. Colour intensity of developed lollipop and icing with grape skin colour powder had best colour strength and sensory attributes than synthetic food colour (carmoesin).

Key words: Grape skin (waste), Spray drying, Grape skin colour powder, Preservatives

Colouring of food stuff is one of the most important aspects in determining its acceptance and enhancing delicacy. Adding attractive colours enhances the appetizing value of food. In many food systems, colour acts as an indicator of quality, particularly in terms of microbial safety (Kakali Roy *et al.,* 2004; Nuzhet and Ferruh, 2006). Natural colours are generally extracted from fruits, vegetables, seeds, roots and micro-organisms and they are called "bio-colours", because of their biological origin. Anthocyanins are approved as food colourants (Leila *et al*., 2008). Major anthocyanin pigments are found in purple grapes (Bangalore blue), (Sampathu *et al.,* 1981). Bridle and Timberlake (1997) reported that anthocyanin pigments were used in soft drinks, alcoholic drinks, confectionery, preserves, fruit toppings, sauces, pickles, dry mixes, canned, frozen foods and dairy products. They also found that anthocyanin pigments were obtained from black grapes, elder berries, red cabbage, cranberry, black currants, purple corn, blood orange, black chokeberry and sweet potato. In this experiment, the natural colour from the fruit rind of grapes was extracted for use as safe food colours.

Materials and Methods

Standardisation of solvents for extraction of colour

Grape fruit rind was used for the extraction of colour using water and ethanol, acetone and citric acid as given in Table 1. Fresh grapes (Bangalore blue) were washed in tap water thoroughly to remove spray residues and the fruit rind was separated by pulper and used for extraction of colour. One gram of the pulped fruit rind was mixed with 5 ml of solvent and filtered through Whatmann No1 filter paper and kept for 30 minutes, and the colour was read in Spectronic 20 colorimeter at 476nm. Based on the colour value, 3 per cent citric acid was selected as solvent.

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Standardisation of drying parameters for production of grape skin colour using spray dryer

Grape fruit rind was made into pulp by adding 3 per cent citric acid at of 1:1 (grape skin: citric acid w/ v). The pulp was kept at room temperature for 30 minutes to dissolve pigments in solvent. After 30 minutes, the pulp was filtered by using muslin cloth. To this solvent mixture, maltodextrin (10, 15 and 20 %) and 0.01 per cent anticaking agent (calcium stearate) were added. The residue was filtered again through muslin cloth and the filtrate was spray dried at an inlet drying air temperature (180, 200 and 220 ºC) at a feed rate as 25 and 30 ml / min. The colour powder recovery from grape fruit rind is presented in Table 2.

Based on the results, inlet drying air temperature, feed rate and maltodextrin (MD) were optimized as 200ºC, 25 ml / min and 20 per cent maltodextrin

Table 1. Colour extraction media

Solvent	Concentrations (%)
Water	100
Acetone	1,2,3,4, and 5
Ethanol	1,2,3,4, and 5
Citric acid	1,2,3,4, and 5

(MD) respectively for the production of grape fruit rind colour powder in spray drying method.

Sample preparation

One gram of developed spray dried grape fruit rind colour powder was dissolved in 100 ml of distilled water. Then 2ml of above sample was used for the analysis of colour stability against pH, temperature and preservative.

Table 2. Effect of drying air temperature, feed rate and maltodextrin (MD) on spray dried grape fruit rind (waste) colour powder recovery

Drying temperature (ºC)	Feed rate (ml /min)	Powder recovery (g / 100 ml) Maltodextrin (%) (MD)		
		10	15	20
180	25	6.4	12.2	13.3
	30	6.2	11.0	12.1
200	25	8.5	14.8	16.4
	30	8.0	13.1	15.2
220	25	7.5	10.4	15.0
	30	5.4	9.8	14.4

Effect of pH

Stability of colour at different pH (4-9) levels was measured by mixing 2 ml of sample in 10 ml of buffer (pH 4-9). These samples were stored in room (R_1) and refrigeration (R_2) temperatures for a period of seven days and absorbance was measured at 24 hours interval.

Effect of temperature

The degradation of colour was studied by exposing the mixture of 2 ml sample with 10 ml of distilled water at 75,100,125 and 150ºC and the degradation was measured for a period of 10 minutes.

Effect of preservative

Effect of sodium benzoate was studied by mixing an aqueous solution of sodium benzoate (concentration 200 ppm- 1000 ppm) with 2ml samples. These samples were stored in room (R_1) and refrigerator (R_2) temperatures for a period of seven days and absorbance was measured at 24 hours interval by using 2201UV-Visible Double Beam Recording Spectrophotometer at 476 nm (Rekha *et al*., 2007).

Determination of colour intensity (L* a* b*) and moisture

The spray dried grape rind colour powder was packed in aluminium pouch (P_1), brown pet bottle (P_2) and brown glass bottle (P_3) and stored in room (R_1) and refrigeration (R_2) temperatures for 3 months. The colour intensity (L* a* b*) of the packed samples were estimated at monthly intervals by using Hunter Colour Lab System, which consists of a rectangular coordinates system for the definition of colour in term of luminosity (L*), red versus green (a*) and yellow versus blue (b*). The colour intensity (L* a* b*) were carried out by the direct reading of the reflectance of the coordinates L* a* and b* using 45^o / 0^o optical geometry. As a standard, the standard illuminate A incandescent light was used by the method described by Kakali Roy *et al*. 2004. The moisture content of spray dried grape skin colour powder (5g) was estimated at monthly intervals using hot air oven at 110^{o}C for 2 hours as method described in AOAC (1995).

Application of spray dried grape fruit rind colour powder in confectionary products

The developed spray dried powder was added at a concentration of 0.04 g in confectionary food products like jujubes, lollipop and icing and the colour value (L* a* b*) was evaluated and compared with products developed with synthetic food colour (carmoesin).

Statistical analysis

The data obtained were subjected to statistical analysis to find out the impact of packaging materials (P_1, P_2 and P_3) and storage periods on the quality of the prepared natural food colour powder. Factorial Completely Randomised Design (FCRD) was applied for the analysis of the study as described by Rangaswamy (1995). Cost analysis of spray dried grape skin colour powder was computed taking into account the fixed cost, variable cost, interest, depreciation and profit.

Table 3. Extraction of colour from grape fruit rind (waste) by solvents

Solvents	Absorbance at 476 nm	Transparency in percentage
Water extraction (100 %)	0.291	51.15
Ethanol extraction		
Ethanol 1 %	0.742	17.81
Ethanol 2 %	0.121	25.76
Ethanol 3 %	0.191	22.43
Ethanol 4 %	0.384	20.13
Ethanol 5 %	0.322	21.76
Acetone extraction		
Acetone 1 %	1.003	9.93
Acetone 2 %	0.880	13.17
Acetone 3 %	1.422	3.78
Acetone 4 %	1.299	5.02
Acetone 5 %	1.160	6.91
Citric acid extraction		
Citric acid 1 %	1.861	1.17
Citric acid 2 %	1.964	1.24
Citric acid 3 %	2.217	0.64
Citric acid 4 %	1.670	1.05
Citric acid 5 %	1.723	1.13

Results and Discussion

The experimental results revealed that the maximum absorbance (2.217) was found in 3 per cent citric acid and minimum absorbance (0.291) in water at 476 nm in Spectronic 20 colorimeter. Thus grape fruit rind colour was extracted using 3 per cent citric acid (Table 3).

Effect of drying temperature, feed rate and maltodextrin (MD) on spray dried grape fruit rind colour powder

The grape fruit rind colour powder recovery on spray drying is presented in Table 4**.** The maximum powder recovery (16.4 g / 100 ml) was observed at 200ºC drying air temperature at feed rate of 25 ml / min with 20 per cent maltodextrin (MD) compared all other treatments. The powder recovery by spray drying showed highly significant difference between drying temperature, feed rate and addition of maltodextrin. Salvador *et al*. (2009) found that the development of roselle extract (*Hibiscus sabdariffa L.)* powder was the best at inlet air temperature between 190-200 ºC than 150,160 and 180ºC in spray drying and it also minimized the losses of volatile compound.

Table 4. Effect of drying temperature, feed rate and maltodextrin on spray dried grape fruit rind (waste) colour powder

Drying temperature (ºC)	Feed rate (ml /min)(F)	Powder recovery (g / 100 ml)		
		Maltodextrin (%)		
		10	15	20
180	25	6.4	12.2	13.3
	30	6.2	11.0	12.1
200	25	8.5	14.8	16.4
	30	8.0	13.1	15.2
220	25	7.5	10.4	15.0
	30	5.4	9.8	14.4

	SED	CD (0.05)	CD (0.01)
T	0.03954	0.08021	0.10755**
F	0.03229	0.06549	0.08781**
M	0.03954	0.08021	0.10755**
TF	0.05592	0.11343	0.15210**
FM	0.05592	0.11343	0.15210*
TM	0.06849	0.13893	0.18628**
TFM	0.09686	0.19647	0.26344**

Effect of pH on stability of spray dried grape fruit rind colour powder during storage

The experimental results showed that pH had a greater influence on the stability powder during storage. The initial absorbance value of powder was 0.17 at pH 4, which decreased to 0.06 (room temperature) and 0.09 (refrigeration temperature) at the end of storage period of one week. For the samples stored at pH 7 and pH 9, the initial absorbance values were 0.15 at pH 7 and 0.13 at pH 9. Their decrease in absorbent values during storage were significantly higher compared to samples stored at pH 4.

Colour stability at pH 4 was higher compared to pH 7 and pH 9 in all samples during storage in room (R_1) and refrigeration (R_2) temperatures. Samples at pH 7 showed satisfactory stability in all samples during storage at both room (R_1) and refrigeration (R_2) temperatures compared to samples at pH 9 and the stability of the colour was better in refrigeration (R_2) temperature than room (R_1) temperature in all samples. Significant difference was observed between pH and storage period and no significant difference was found with respect to their combined effects. Leila *et al*. (2008) reported that the anthocyanin (grape skin) pigment was more stable (4,103±24 h) at pH 3.0 with temperature of 4±1ºC in dark condition during storage.

Table 5. Effect of preservative on the stability of spray dried grape skin (waste) colour powder during storage

Storage period in days (S)	Optical density values at 476nm									
	P_1		P_2		P_3		P_4		P_5	
	R_1	R_2	R_1	R_2	R_1	R_2	R_1	R_2	R_1	R_2
Initial	0.23	0.23	0.20	0.20	0.19	0.19	0.16	0.16	0.14	0.14
1	0.17	0.18	0.15	0.15	0.14	0.15	0.12	0.14	0.11	0.12
2	0.15	0.17	0.14	0.14	0.13	0.15	0.10	0.13	0.07	0.11
3	0.14	0.15	0.12	0.13	0.10	0.13	0.08	0.11	0.05	0.09
4	0.12	0.13	0.09	0.11	0.07	0.10	0.06	0.07	0.04	0.08
5	0.09	0.10	0.08	0.09	0.05	0.07	0.04	0.05	0.03	0.05
6	0.07	0.08	0.06	0.07	0.03	0.06	0.03	0.04	0.02	0.03
7	0.06	0.07	0.04	0.06	0.02	0.05	0.02	0.03	0.02	0.03

	SED	CD (0.05)	CD (0.01)
S	0.00258	0.00510	0.00673**
P	0.00204	0.00403	0.00532**
R	0.00129	0.00255	0.00337**
SP	0.00577	0.01140	0.01505**
PR	0.00289	0.00570	0.00753**
SR	0.00365	0.00721	0.00952**
SPR	0.00816	0.01613	0.02129NS

Effect of preservative on stability of grape fruit rind colour powder during storage

The minimum colour degradation was observed in 200 ppm of sodium benzoate in all samples during storage in room (R_1) and refrigeration (R_2) temperatures (Table 5) and the colour degradation increased with increase in the concentration of

Table 6. Changes in the colour intensity (L* a* b*) of jujubes, lollipop and icing with synthetic food colour (carmoesin) and spray dried grape fruit rind (waste) colour powder

Products	Synthetic food colour (carmoesin)			Grape fruit rind (waste) colour powder		
	L	a	B	L	a	b
Jujubes	34.09	22.28	8.14	47.91	1.18	5.90
Lollipop	26.38	16.06	2.35	22.35	2.54	7.51
Icing	24.11	15.54	5.76	22.37	2.84	7.63

sodium benzoate (200 ppm to 1000 ppm). Hence the experimental results revealed that addition of sodium benzoate had an adverse effect on the stability of grape fruit rind colour during storage. Statistical analysis revealed that there was significant difference between concentration of preservative and storage periods and no significant difference was found with respect to their interaction between storage period, preservative levels and storage temperature.

Effect of temperature on stability of spray dried grape fruit rind colour powder

A gradual degradation of fruit rind colour was observed at different temperatures based on the optical density (O.D) value. The absorbance value of spray dried grape skin colour powder was 0.19 and on heating at 75ºC the value was reduced to 0.11. At 100ºC, 125ºC and 150ºC the optical density values were 0.09, 0.06 and 0.04 respectively. The experiments revealed that, the loss of colour degradation was higher at elevated temperature (150C). Eliana *et al*. (2007) reported that the half-life of anthocyanin pigment (spinach vine) was decreased from 7.46 h to 2.50 h, when the temperature increased from 40 to 60ºC during storage.

Application of spray dried grape fruit rind colour powder in confectionary products with synthetic food colour (carmoesin)

The products coloured with carmoesin (Table 6) revealed better colour stability compared to products containing spray dried grape fruit rind colour powder. At the same time lollipop and icing with spray dried grape fruit rind colour powder had sufficient L* a* b* values compared to jujubes with spray dried grape skin colour powder. Kiattisak *et al*. (2004) reported that the drink containing carmoesin or SAN RED RC had best colour stability and was acceptable for over 84 days. But the drinks containing roselle anthocyanin powder were least stable in colour and acceptable over 56 days.

Effect of colour value (L* a* b*) of spray dried grape fruit rind colour powder during storage

A gradual reduction of colour value was observed in spray dried grape fruit rind colour powder during storage. The initial a* value of spray dried grape skin colour powder was 38.60 which was reduced to 35.14 respectively in packaging materials P_1, P_2 and P_3 for the samples stored at room (R_1) and refrigeration (R_2) temperatures at the end of 90 days. Minimum a* value changes occurred in the sample stored in P_3 during storage compared to P_2 and P_1 (Table 7). The experiment results indicated that the packaging material (P_3) was better compared to P_1 and P_2.

Table 7. Changes in the colour intensity (L* a* b*) of spray dried grape fruit rind (waste) colour powder during storage

Storage periods	Optical density values at 476nm					
	P_1		P_2		P_3	
	R_1	R_2	R_1	R_2	R_1	R_2
	a*	a*	a*	a*	a*	a*
Initial	38.60	38.60	38.60	38.60	38.60	38.60
30	37.01	37.14	37.21	37.28	37.41	37.50
60	36.24	36.37	36.41	36.62	36.74	36.68
90	35.14	35.40	35.72	35.84	35.87	36.02

Effect of moisture content of spray dried grape fruit rind colour powder during storage

The moisture content was maximum in P_1R_1 (3.61) in P_1 and minimum in P_3R_2 (3.48) in P_3 after 3 months of storage in room (R_1) and refrigeration (R_2) temperatures. The study indicated that the brown bottles (P_3) retained more moisture content in all the samples compared to the other two packaging. This might be due to the permeability of the glass bottle**.** Significant difference was observed between treatments, storage periods and packaging materials. Waquar *et al*. (2000) found that the colour of tomato powder was maintained at the lowest level of water activity during storage.

Cost benefit analysis

The production cost of spray dried grape fruit rind colour powder was comparatively lesser than the market price of synthetic food colour powder (carmoesin). The cost of production of spray dried powder for 10 kg was worked out taking into account the profit, interest and depreciation. The total production cost of spray dried powder was Rs. 530/kg.

Conclusion

Among the solvents, citric acid was found to be suitable for colour extraction from grape skin at 3 per cent. Powder recovery percentage was found to be highest at 200ºC, 25 ml / min feed rate with incorporation of 20 per cent maltodextrin in spray drying. While the colour stability was better at pH 4, addition of preservative (sodium benzoate) and higher processing temperature (150^{o} C) had detrimental effect on stability of colour during storage. The brown glass bottle (P_3) had better colour retention than brown pet bottle (P_2) and aluminium pouch (P_1) during storage at room (R_1) and refrigeration (R_2) temperatures. Lollipop and icing prepared with spray dried powder had best colour strength next to carmoesin during storage and also highly acceptable quality attributes. At the same time powder did not produce colour in jujubes. The cost of the spray dried grape fruit rind colour powder was Rs.530/kg.

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