

# Process Conditions Affect Pigment Quality and Yield in Extracts of Purple Sunflower Hulls

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**ABSTRACT:** Extract of purple sunflower hulls is a potential red food colorant; however, suitable process conditions must still be identified. Selected process variables were studied using bench-scale units to prepare, clarify, concentrate and spray dry extracts. Concentration by evaporation at 32°C and addition of maltodextrin to 15% (wt/dry wt) prior to drying largely eliminated pigment degradation during those steps. Relative to water extracts, extracts prepared with 5 to 15% ethanol in water generally yielded more pigment with similar levels of degradation and loss during subsequent processing. Use of 5% ethanol/2% citric acid reduced yield by about half relative to water, but gave a powder with a lower degradation index and wetting time. Percentage recovery of pigment during concentration and drying was not greatly affected by solvent type or extraction temperature.

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**KEY WORDS:** Anthocyanin, extraction, pigment, processing, purple-hulled sunflower, red food colorant, spray drying, sunflower by-product, sunflower hulls.

Extracts from the purple hulls of certain types of sunflower have been proposed for use as natural red food colorants (1). Their appeal as natural ingredients also carries over to non-food uses, such as textiles (H. Lunde, personal communication, 1994) and cosmetics, even though the extracts may be more expensive to produce and less stable than artificial red colorants. Artificial red colorants are perceived unfavorably by consumers. Red No. 40 is the only artificial red colorant permitted in a wide variety of foods by the Food and Drug Administration (FDA).

An array of other natural red food colorants has become commercially available in recent years (2). The purplish-red pigments in purple sunflower hulls are classified as anthocyanins. Thus, they are chemically similar to pigments in grape skin, red cabbage and some other sources of natural red colorants, and they have similar applications. Purple sun-

flower hull extract is not yet commercially available or approved by the FDA, but it has several desirable features. The pigment is highly stable within the hulls, and it can be extracted under mild conditions. Genotypes have been reported which contain over 2% pigment in the hulls (3). Through conventional plant breeding, the purple-hulled trait could be bred into high-oil sunflower types. Thus, an abundant, inexpensive supply of raw material could become available as a by-product of the sunflower oil industry. A preliminary study of the commercial potential of the sunflower hull extract was favorable (4).

Previously, a bench-scale process was described for the preparation of spray-dried purple-sunflower hull extracts (5). Raw extract was prepared from water and ground hulls in a countercurrent screw-type solvent-extraction unit, clarified by gravity separation and either a filtering centrifuge or batch centrifuge, concentrated with a rising-film evaporator and spray-dried. There are, however, numerous process variables to consider, and the above report was concerned mainly with methods of calculating pigment yield and assessing the effect of citric acid addition at different steps in the process. Subsequently, the goal was to improve pigment yield and quality while reducing total process time. For example, pigment degraded somewhat at each step of the process, but particularly during concentration by evaporation. It was expected evaporation under higher vacuum would expose the extract to milder temperatures and thus should improve pigment yield and quality (6). Gravity separation was slow and cumbersome; a decanter centrifuge with helical conveyor could clarify the extract almost instantly and produce a continuous discharge of both clear extract and sediment (7).

Also of interest in increasing yield and reducing degradation was the use of ethanol solutions during extraction and the use of maltodextrin during spray drying. In the previous report of spray-dried sunflower hull extracts, only water or water adjusted to pH 3.0 with citric acid was used (5); however, researchers who prepared extracts from less than 1-g samples of ground hulls obtained good results with ethanol solutions. Martin (8), using response surface methodology to assess varying levels of ethanol and citric acid in water, reported that the highest pigment yield and quality was

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achieved with 5% ethanol/2% citric acid in water. Holm (9) used a similar approach to assess varying levels of ethanol and tartaric acid in water and achieved optimum results using 18 to 28% ethanol and 2 to 9% acid. Maltodextrin and other water-soluble carbohydrates have been added to liquids prior to spray drying to protect heat-sensitive components. For example, Main *et al.* (10) added a carbohydrate carrier to grape skin and other extracts prior to spray drying, and Wiesenborn *et al.* (5) used maltodextrin previously. Neither work reported a comparison of carbohydrate vs. no carbohydrate addition or of varying levels of addition.

The objectives of this study were to: (i) quantify the effectiveness of certain process modifications, namely the use of evaporation under higher vacuum and a continuous decanting centrifuge in place of gravity sedimentation/filtering or batch centrifugation; (ii) study the effects of solvent type (water, water and ethanol, or water, ethanol and citric acid) and temperature on pigment yield, post-extraction processing and characteristics of spray-dried powder; and (iii) prepare and characterize spray-dried extracts with varying amounts of maltodextrin.

## EXPERIMENTAL PROCEDURES

Seeds from the Neagra de Cluj genotype of purple-hulled sunflower grown in 1991 were dehulled at Red River Commodities Inc., (Fargo, ND). Hulls were stored in paper sacks at ambient outdoor temperatures. The moisture content of the hulls was 7% (wet weight basis) at the time these experiments were completed. No effect of storage on the quantity or quality of pigment was observed. Maltrin M040 maltodextrin from Grain Processing Corp. (Muscatine, IA) was used as a co-drying agent. It contained 6.9% moisture (wet weight basis).

**Extraction.** Pigment was extracted using a continuous, countercurrent screw-type extractor. The extractor design and method of operation were described previously (11). Hulls were first ground in a hammer mill to pass through 1.0-mm perforations. In the study of maltodextrin addition to spray-dryer feed, extractions were done with 70 g/min water at 82°C. The flow rate ratio of water to hulls averaged 4.5 g/g, and the extract collected averaged 11.8 kg per test. In the study of solvent effects, extractions were with 70 g/min of water, 5% ethanol in water, 15% ethanol in water and 5% ethanol/2% citric acid in water at 22, 50 and 70°C. The flow rate ratio of water to hulls averaged 4.3 g/g, and the extract collected averaged 9.0 kg per test.

**First extract clarification.** In the study of maltodextrin addition, suspended solids were removed by gravity sedimentation at 3 to 5°C for 20 h; then extract was decanted from sediment and centrifuged 15 min at 21,500 × g using an RC-5 Superspeed Refrigerated Centrifuge (Sorvall, Inc., Norwalk, CT). In the study of solvent effects, raw extracts were collected and passed through a decanter centrifuge (Westfalia Separator Model CA 150-01-00; Centrico, Inc., Elgin, IL) at a maximum of 3,300 × g and a feed rate of 1.8 L/min then stored at 3 to 5°C for 20 h. Extracts obtained by decantation

and centrifugation or by use of a decanter centrifuge are referred to as once-clarified extracts.

**Evaporation.** Once-clarified extracts were concentrated by a single pass through a steamjacketed rising-film evaporator (fabricated by Standard Industries, Fargo, ND), then clarified a second time by centrifugation for 15 min at 21,500 × g using an RC-5 Superspeed Refrigerated Centrifuge (Sorvall). All evaporator contact surfaces were fabricated from 304 stainless steel. The rising-film heat exchanger section of the evaporator consisted of a vertical tube, 1900-mm long by 48-mm inside diameter. A second concentric tube, 1820-mm long by 97-mm inside diameter, formed the steam jacket. Extract was added continuously at approximately 64 mL/min, and concentrated at 84°C under a vacuum of 380 mm Hg with steam at 107°C. In the study of maltodextrin addition, extract was concentrated 6- to 7-fold; in one case 3-fold. In the study of solvent effects, clarified extracts were concentrated about 6-fold at 32°C under a vacuum of 710 mm Hg with steam at 107°C.

**Spray drying.** Concentrate was dried in a portable spray dryer (Niro Inc., Columbia, MD) at a feed rate of 30 mL/min, using 195–200°C air which exited the dryer at 85 to 90°C. In the study of maltodextrin addition, maltodextrin was added to 0, 15, 25 and 35% wt/dry wt just prior to spray drying. In the study of solvent effects, the amount of maltodextrin was 25%. The total time from extraction through spray drying was about 50 h.

**Analysis of extract.** Dry-solids content of extracts was determined by drying duplicate samples at 130°C for 2 h in a gravity convection oven. Initial sample size was 10 g of liquid extract or 3 g of powdered extract. Anthocyanin pigment concentration in extracts from various stages of processing was determined in duplicate using the pH-differential method described by Wrolstad (12) with modifications as described by Mok and Hettiarachchy (6). Raw extracts were first centrifuged to remove and quantify suspended solids as described above. Absorbance data obtained in the course of the analysis was also used to calculate the degradation index (13). Suspended solids content of liquid extracts was determined by centrifuging duplicate 50 g samples for 15 min at 10,850 × g, decanting off the supernatant, then weighing the wet sediment. Spray-dried solids were first reconstituted in distilled water to 5% (wt/vol) at room temperature. Wetting time was determined using a modification of Niro Atomizer Method No. A 5 (14) for determining wettability of powdered milk, in which 3.0 g powder was added to the quiet surface of 100 mL distilled water at 60°C, and the time required to achieve complete wetting was measured.

**Pigment calculations.** The initial pigment extraction yield (IPY), representing the amount of pigment in once-clarified extract per weight of hulls processed, was calculated from:

$$IPY = (M_{cl} \cdot [P]_{cl}) / (M_h \cdot D_h) \quad [1]$$

where  $M_{cl}$  and  $M_h$  represent the weight in grams of once-clarified extract (before concentration) and ground hulls, respec-

tively,  $[P]_{cl}$ , represents the concentration of pigment in mg/g of once-clarified extract and  $D_h$  represents the dry weight fraction of ground hulls. Percentage recovery,  $R$ , after evaporation and spray drying were calculated from the pigment/dry sunflower hull ratio,  $P/D$ , at each step:  $R = [(P/D)_p / (P/D)_f] \cdot 100\%$ . Subscripts  $p$  and  $f$  denote the product stream and feed stream, respectively, for that unit (5). During clarification of the concentrate (second clarification), the ratio of pigment to water was assumed equal for the concentrate and clarified concentrate. Thus percentage recovery for this step was calculated from the product of weight and weight fraction of moisture,  $M \cdot (1-D)$ , as follows:  $R = \{[M \cdot (1-D)]_{clc} / [M \cdot (1-D)]_c\} \cdot 100\%$ . Subscripts  $clc$  and  $c$  denote the clarified concentrate and concentrate, respectively. The overall percentage recovery was calculated from the percentage recoveries for concentration, second clarification and spray drying. The overall pigment yield, representing the weight of pigment in spray-dried product per weight of hulls, was calculated as the product of  $IPY$  and overall percentage recovery.

## RESULTS AND DISCUSSION

*Effect of maltodextrin during spray drying.* The extract was briefly exposed to high temperatures during spray drying. This may result in degradation of anthocyanin pigments which yields brown-colored polymers or colorless compounds (15). Degradation can be detected by monitoring the degradation index (DI) which increases due to formation of polymers (13), and by monitoring the percentage recovery which by definition is proportional to the change in pigment/dry weight extract. In two dryer tests without maltodextrin, DI increased by 0.11 to 0.16, compared to increases of 0.02 to 0.04 with 15 to 35% maltodextrin (Table 1), confirming the protective effect of maltodextrin. The slightly higher percentage recoveries achieved during drying with maltodextrin, though not statistically significant in these tests, may also indicate a protective effect. Percentage recoveries greater than 100% probably reflect experimental bias or error, rather than an increase in soluble pigment.

Addition of 25 to 35% maltodextrin did not appear advantageous relative to 15%. These higher proportions of maltodextrin have drawbacks, such as increased wetting time (Table 1), dilution of the pigment and raw materials cost. On the other hand, higher levels might result in better storage stability of the dry pigment, but storage stability was not monitored in this study.

Although complete replicates of the maltodextrin/spray-dryer tests were not performed, the preparation of four samples for spray drying was identical to the extent practicable. This allowed an analysis of the variability of process steps prior to spray drying. Initial pigment yield (after the first clarification step) averaged ( $\pm$ SD)  $2.60 \pm 0.34$  mg/g hulls. The factors accounting for this variability were discussed previously (11). Also after the first clarification, the DI averaged  $1.61 \pm 0.03$ , and the pigment per dry weight of extract averaged  $41.8 \pm 4.9$  mg/g. During the subsequent concentration and second clarification steps, percentage recoveries averaged  $68 \pm 4\%$  and  $91.0 \pm 0.6\%$ , respectively. The severe loss of pigment during concentration by evaporation was accompanied by a large increase in DI of  $0.20 \pm 0.06$ .

*Process modifications.* Degradation during concentration for tests in Table 1 was more severe than in our previous report (5), because extracts were concentrated 6- to 7-fold to up to 27% solids vs. only about 3-fold in previous tests. A high level of concentration is desirable, because the energy costs for removing water are much less for evaporation compared to spray drying. As a result of the increased degradation at higher levels of concentration, a liquid-ring vacuum pump was installed after the maltodextrin/spray-drying study. This achieved much higher vacuum during evaporation, resulting in an evaporator temperature of  $32^\circ\text{C}$  vs.  $84^\circ\text{C}$  previously. In 16 subsequent tests during the study of solvent effects, the increase in DI averaged  $0.02 \pm 0.01$ , and the percentage recovery for concentration averaged  $90 \pm 5\%$ . Therefore, the much lower evaporation temperature largely eliminated pigment degradation. The percentage solids ranged from 20 to 30%, but greater concentration may be considered in future tests.

**TABLE 1**  
Effect of Maltodextrin on Spray-Dried Extracts of Purple Sunflower Hulls<sup>a</sup>

| Maltodextrin content (% wt/dry wt) | DI <sup>b</sup> Increase during drying | Percentage recovery during drying | Wetting time (s) | Suspended solids content (%) <sup>c</sup> |
|------------------------------------|--|-----------------------------------|------------------|---|
| 0                                  | $0.135 \pm 0.035$                      | $94 \pm 13$                       | $390 \pm 90$     | $0.86 \pm 0.07$                           |
| 15                                 | 0.02                                   | 100                               | 130              | 0.71                                      |
| 25                                 | 0.04                                   | 106                               | 290              | 0.96                                      |
| 35                                 | 0.04                                   | 101                               | 550              | 1.15                                      |

<sup>a</sup>Results at 15 to 35% maltodextrin are for single tests, whereas results with no maltodextrin are means  $\pm$  SD for duplicate tests, one of which involved only a 3-fold concentration prior to spray drying.

<sup>b</sup>DI is the degradation index of the pigment.

<sup>c</sup>Wet weight basis for powder reconstituted to a 5% solution in water.

Another key process improvement instituted at this time was installation of a decanter centrifuge to rapidly carry out the first clarification step. The suspended solids content of water extracts passed once through the decanter was  $3.8 \pm 1.4\%$ , vs.  $1.0 \pm 0.1\%$  for water extracts which were clarified by the former method (gravity separation followed by batch centrifugation). Although the decanter was not as successful in achieving a clarified extract, it was considered a major advance, in light of its simple operation and rapid process time, and the dryness of the discharged sediment. The decanter was operated with a minimum "pond level" which achieves the dryest sediment but the least clarification (7). Also, the decanter was operated at considerably less than maximum capacity, because of the limited extract volumes provided in this study; the decanter performance might improve under pilot-scale process conditions. Since an additional clarification step was used after evaporation, the final suspended solids content was still comparable to previous samples.

*Effect of solvent type and temperature on yield and quality of clarified extract.* The initial pigment yield in once-clarified extracts was highly dependent upon initial solvent temperature (significant at  $P < 0.01$ ) and type (Table 2). With each solvent, yield increased over 2-fold when extraction temperature was increased from 22 to 70°C. A similar temperature dependence was reported previously in the case of dilute water extracts (11). This suggested that increasing extraction temperature might further increase yield. In the case of solvents which contained ethanol, higher temperatures were not used because the extraction equipment was not designed to adequately contain large quantities of ethanol vapor which would be evolved at or near the boiling point. In the case of water, the results in Table 2 can be extended to 82°C by including extraction results from the maltodextrin tests. There

were, however, negligible differences in pigment yield and content between water extracts at 70 and 82°C.

The use of ethanol-water solutions generally improved initial pigment yield relative to water. For example, in the case of addition of 5% ethanol at 70°C, duplicate tests averaged 20% greater yield compared to water at that temperature. Extraction with 15% ethanol generally did not further improve pigment yield.

Use of solvent containing 5% ethanol and 2% citric acid in water resulted in about half the initial pigment yield achieved with water. The reduction in yield with citric acid is opposite the results of Martin (8) and Holm (9), but similar results were reported earlier in a comparison of citric acid-water solutions vs. water (5). The difference probably was due to the differing solvent-to-hulls ratio, which averaged 4.3 g/g in this study vs. 62.5 mL/g by Martin (8) and Holm (9). The lower pH resulting from citric acid addition may have precipitated protein-bound pigment (16), with this effect becoming more acute in highly concentrated extracts. Increasing the proportion of solvent may increase yield, but concentrating the resulting dilute extract entails higher energy costs associated with evaporating the solvent (11).

Compared to the other solvents, extraction with 5% ethanol/2% citric acid resulted in greatly reduced suspended solids content after the first clarification step, with the sediment being noticeably firmer to the touch. Also, this solvent resulted in a significantly lower DI, either due to a protective effect, or perhaps because partially degraded pigment was less soluble at low pH. The other three solvents were not significantly different from each other with respect to suspended solids content or DI. Suspended solids content generally increased with increasing solvent temperature. The DI might be expected to increase with increasing temperature, but the cor-

**TABLE 2**  
Initial Pigment Yield and Characteristics of Clarified Extracts of Purple Sunflower Hulls Prepared with Alternative Solvent Types and Temperatures<sup>a</sup>

| Solvent                            | Initial solvent temperature (°C) | Initial pigment yield (mg/g hull) | Suspended solids content (%) | Degradation index | Pigment content (mg/g dry wt) |
|------------------------------------|----------------------------------|-----------------------------------|------------------------------|-------------------|-------------------------------|
| Water                              | 22                               | 1.11                              | 2.5                          | 1.58              | 28.2                          |
|                                    | 50                               | 2.13                              | 3.2                          | 1.54              | 39.3                          |
|                                    | 70                               | 2.70 ± 0.10                       | 4.7 ± 1.3                    | 1.55 ± 0.04       | 39.6 ± 3.0                    |
| 5% Ethanol in water                | 22                               | 1.43                              | 2.0                          | 1.50              | 27.0                          |
|                                    | 50                               | 1.98                              | 4.8                          | 1.53              | 41.5                          |
|                                    | 70                               | 3.26 ± 0.19                       | 4.9 ± 1.0                    | 1.59 ± 0.06       | 46.4 ± 8.0                    |
| 15% Ethanol in water               | 22                               | 1.44                              | 2.1                          | 1.52              | 39.5                          |
|                                    | 50                               | 2.84                              | 2.3                          | 1.57              | 50.5                          |
|                                    | 70                               | 3.04 ± 0.37                       | 4.1 ± 1.6                    | 1.61 ± 0.02       | 52.2 ± 0.1                    |
| 2% Citric acid/5% ethanol in water | 22                               | 0.45                              | 1.0                          | 1.45              | 7.2                           |
|                                    | 50                               | 1.05                              | 1.2                          | 1.39              | 13.4                          |
|                                    | 70                               | 1.39 ± 0.21                       | 1.4 ± 0.1                    | 1.43 ± 0.04       | 16.5 ± 0.8                    |

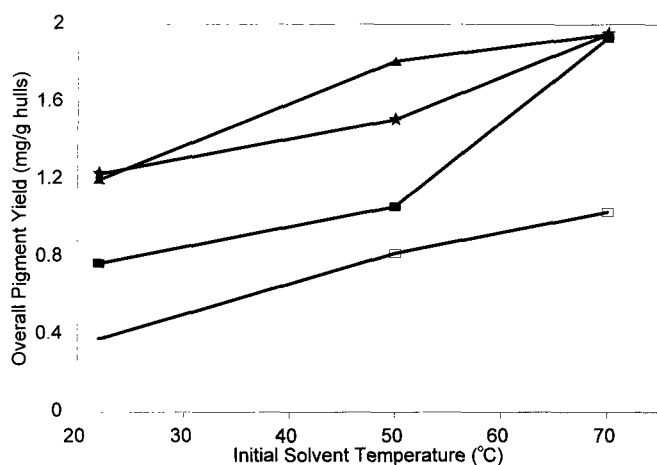
<sup>a</sup>Results at 22 and 50°C are for single tests, whereas results at 70°C are means ± SD for duplicate tests.

**TABLE 3**  
**Processing of Extracts of Purple Sunflower Hulls Prepared with Various Solvents<sup>a</sup>**

|   | Percentage recoveries |                   |              |             | $\Delta$ DI <sup>b</sup> |
|---|-----------------------|-------------------|--------------|-------------|--------------------------|
|   | Concentration         | 2nd Clarification | Spray drying | Overall     |                          |
| Water                                       | 91.2 ± 2.3            | 86.7 ± 6.3        | 87.1 ± 16.0  | 55.4 ± 9.2  | 0.05 ± 0.02              |
| 5% Ethanol<br>in water                      | 87.1 ± 5.5            | 88.9 ± 7.8        | 94.6 ± 4.8   | 61.8 ± 12.1 | 0.06 ± 0.01              |
| 15%<br>Ethanol in<br>water                  | 88.6 ± 5.7            | 90.6 ± 6.8        | 89.9 ± 3.6   | 57.0 ± 9.2  | 0.06 ± 0.02              |
| 2% Citric<br>acid/5%<br>ethanol in<br>water | 92.4 ± 4.4            | 94.4 ± 6.4        | 94.3 ± 2.9   | 65.8 ± 3.5  | 0.04 ± 0.01              |

<sup>a</sup>Mean ± SD for four tests at three solvent temperatures.

<sup>b</sup>Change in DI from first clarification to reconstituted spray-dried powder; see Table 1 for abbreviation.



**FIG. 1.** Overall pigment yield (mg pigment/g hull extracted) in spray-dried extracts of purple sunflower hulls for the solvents: water ■, 5% ethanol in water ★, 15% ethanol in water ▲ and 5% ethanol/2% citric acid in water □. Results at 22 and 50°C are for single tests, whereas results at 70°C are means for duplicate tests. Standard deviations for duplicates were 0.13, 0.09, 0.03 and 0.13, respectively.

relation of DI with temperature was not significant at  $P < 0.05$ . Pigment content (mg pigment/g dry wt) was positively correlated with initial pigment yield ( $P < 0.01$ ). Solvent containing 15% ethanol resulted in the highest pigment content at all three temperatures.

*Effect of solvents on post-extraction processing.* The degradation of the pigment contained in once-clarified extract during subsequent processing might be influenced by the extraction solvent or by various other soluble constituents extracted from the hulls. In the latter case, solvent type and temperature might indirectly influence degradation by their influence on the proportions of these other constituents. However, an effect by solvent temperature on percentage recovery or change in DI was not detected (data not shown). Therefore, the results from the three temperatures were averaged for each solvent (Table 3). Generally, these averages show no significant differences between solvents. The exception is 5%

ethanol/2% citric acid vs. water during the second clarification step. The higher percentage recovery achieved with the ethanol–citric acid solution is partly the result of its lower suspended solids content. DI increased in every test, but this increase was slight due to efforts described above to minimize degradation during evaporation and spray drying. An effect of solvent on change in DI was not observed.

*Effect of solvent type and temperature on yield and quality of spray-dried extract.* Overall and initial pigment yield correlated significantly at the  $P < 0.01$  level of probability ( $r = 0.95$ ;  $n = 16$ ). Thus the overall pigment yield showed a dependence upon solvent type and temperature (Fig. 1) similar to the initial pigment yield. Differences in overall yield apparently resulted during extraction rather than subsequent processing. Consequently, initial pigment yield is a useful predictor of overall yield; however, at 70°C, the overall yield achieved with water was almost identical to that achieved with and 15% ethanol. Given the cumulative variability throughout the entire process, the SD for duplicates at 70°C were less than might be expected.

As with pigment yield, relative values of DI and pigment content were generally unchanged during processing of the once-clarified extracts (Table 4). For example, the superior, low DI of the 5% ethanol/2% citric acid extract and the high pigment content of the 15% ethanol extract were maintained throughout processing. The decrease in pigment contents compared to the once-clarified extracts resulted mainly from addition of 25% maltodextrin.

The ethanol–citric acid extract also had a superior wetting time. In the case of ethanol–water extractions, wetting time increased with increasing extraction temperature, probably a result of differences in the nonpigment solids extracted. All extracts contained similar low levels of suspended solids when reconstituted to a 5% (wt/vol) solution in water.

The advantage of superior quality obtained in ethanol–citric acid extracts was offset by a yield about one-half that obtained with plain water. The added cost of those chemicals and the added care required to confine ethanol vapor and redistill

**TABLE 4**  
**Quality of Spray-Dried Extracts of Purple Sunflower Hulls Prepared with Various Solvent Types and Temperatures<sup>a</sup>**

| Solvent                            | Initial solvent temperature (°C) | Degradation index | Pigment content (mg/g dry wt) | Wetting time (s) | Suspended solids content (%) <sup>b</sup> |
|------------------------------------|----------------------------------|-------------------|-------------------------------|------------------|---|
| Water                              | 22                               | 1.61              | 18.8                          | 330              | 0.81                                      |
|                                    | 50                               | 1.59              | 20.3                          | 340              | 0.79                                      |
|                                    | 70                               | 1.61 ± 0.05       | 29.4 + 2.0                    | 330 ± 150        | 0.89 ± 0.05                               |
| 5% Ethanol in water                | 22                               | 1.55              | 21.1                          | 150              | 0.71                                      |
|                                    | 50                               | 1.59              | 30.4                          | 430              | 0.98                                      |
|                                    | 70                               | 1.65 ± 0.07       | 29.4 ± 2.0                    | 450 ± 90         | 0.94 ± 0.11                               |
| 15%±O Ethanol in water             | 22                               | 1.60              | 28.6                          | 110              | 0.93                                      |
|                                    | 50                               | 1.61              | 32.7                          | 250              | 0.93                                      |
|                                    | 70                               | 1.67 ± 0.01       | 33.4 ± 1.7                    | 350 ± 30         | 0.96 ± 0.04                               |
| 2% Citric acid/5% ethanol in water | 22                               | 1.48              | 5.3                           | 18               | 0.95                                      |
|                                    | 50                               | 1.43              | 8.8                           | 11               | 0.91                                      |
|                                    | 70                               | 1.47 ± 0.03       | 11.6 + 0.0                    | 25 ± 10          | 1.23 ± 0.14                               |

<sup>a</sup>Results at 22 and 50°C are for single tests, whereas results at 70°C are means ± SD for duplicate tests.

<sup>b</sup>Wet weight basis for powder reconstituted to a 5% solution in water.

ethanol in the waste streams should be considered. On the other hand, the yield with ethanol and citric acid might be improved at higher ratios of solvent to hulls than used in this study. The expense of processing with ethanol or citric acid or of using higher ratios of solvent to hulls in a commercial process should be weighed against the yield and quality of the pigment.

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