INTERRELATION AMONG MEASUREMENTS OF BROWNING OF PROCESSED POTATOES AND SUGAR COMPONENTS¹

SIGMUND SCHWIMMER, C. E. HENDEL, W. O. HARRINGTON, AND R. L. OLSON²

The fact that a highly significant relationship exists between the apparent "reducing sugar" content of potatoes and the extent of nonenzymatic browning of processed potato products such as chips, French fries, and dehydrated dice, has been amply documented in several recent reviews (1, 5, 11). Furthermore, substantial evidence shows that this relation is casual and direct, owing, at least in part, to a typical Maillard reaction between the sugar components and alpha amino groups present in the nitrogenous constituents of the tuber (11) . However, a survey of the quantitative data in the literature reveals that there is considerable scatter in the plots of browning *vs.* reducing sugar. Since it is reasonable to assume a degree of precision well within the limits of this scatter in the measurements of so-called "reducing sugars", the possible sources of this scatter can be sought in the following factors: (a) extraction and clarification procedures for the determination of the sugars; (b) estimates of browning susceptibility; (c) the inclusion (or non-inclusion) of sugars and reducing substances other than the individual reducing sugars; (d) influence of variations in the relative amounts of the components comprising the reducing sugars; (e) alteration of sugar content caused by processing treatment; (f) influence of level and composition of the amino constituents which enter into the browning reactions; (g) variations in levels and relative importafice of other direct or possibly auxiliary factors, the latter of which may profoundly influence but may not directly enter into the browning process.

As part of a comprehensive study of compositional factors influencing browning of processed potato products, several varieties of potatoes, each of which was subjected to a controlled but varied post-harvest history, were analyzed chromatographically for individual sugar components and derivatives by improved methods of extraction, clarification, and separation (13, 14, 19). Since the major sugars found were only sucrose, glucose, and fructose, an effort was made to find statistical interrelationships (including parameter, the "discriminatory index") among levels of these sugars and degrees of soluble colors measured in potato chips and dehydrated potato dice by improved procedures $(3, 4)$. Data are also presented on the course of the accelerated browning of dehydrated dice as affected by potato variety.

MATERIALS AND METHODS

Preparation and Processing of Potatoes. Raw materials included one lot each of the White Rose and Russet Burbank varieties ("western potatoes"). Details of history, storage conditions, *etc.* are given in a previous paper (14). Also used were one lot each of Katahdin and Green Mountain grown in Maine. At specified storage intervals 15- to 20-pound lots of tubers were hand-peeled and a sample of several half tubers was

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²Western Utilization Research Branch, Agricultural Research Service, United States Department of Agriculture, Albany 10, Cal.

prepared. The ends of each tuber were removed and the remainder was sliced transversely. The slices were washed in cold water, blotted with towels, and fried in deep fat.

The remainder of the half tubers were diced mechanically $(3/2) \times 3/2 \times 3/2$ 3/16-inch), intimately mixed, and 400 grams placed in boiling ethanol for subsequent sugar analyses (14).

The remainder of the dice were blanched in steam at 212° F. for 3 minutes on wire-mesh trays at a loading of 1.5 pounds per square foot. The dice were then dried in a cross-flow cabinet drier at an air temperature of 140°F. for 5 hours. The dehydrated dice were stored at 32° F. in nitrogen for subsequent study.

For the preparation of chips, sliced lots (1/16-inch in thickness) containing 75 to 85 grams per batch were fried in hot oil initially at 190 $^{\circ}$ C. until cessation of bubbling was observed (80 to 100 seconds). After draining and cooling, samples of the chips were placed in tightly closed jars and stored at -10°F. for subsequent evaluation. Such a frying technique results in chips with moisture content at about 2.5 per cent, as you will note in tables 3 and 4.

Analyses and Tests. For the measurement of rate of browning, the dehydrated dice were adjusted to 8 per cent moisture content (4, 8) either by addition of the necessary amount of water (dropwise) followed by holding with periodic mixing on each of several successive days, or by removal of water from the dice by placing them in an evacuated desiccator over the appropriate amount of calcium oxide. The moisture-adjusted dice were then canned in 211 \times 300 cans (60 grams per can) and placed in a water bath at 50° C. Dice from the Maine tubers were in evacuated cans, which were tested for loss of vacuum at the end of the browning test. Moisture content of the browned dice was also measured.

The samples, after removal from the water bath at daily intervals, were ground ζ (ζ 2 mm.), extracted with 2 per cent acetic acid, and the filtered extract was mixed with an equal volume of acetone. After filtration, the optical density of this second filtrate was measured in a 5-cm. optical cell at 420 m μ in a Beckman DU spectophotometer. Results are expressed as increase in optical density $(\bar{x}$ 1000) per day under the specified conditions of wave length, length of cell, and concentration referred to the original weight of dice (5 per cent). This measurement is designated as:

> \Box 5% \sim L 5 cm. 420 m μ /day x 10³

The foregoing procedures for heat damage and for measuring degree of browning are similar to those used in other studies at this laboratory $(4, 6, 7)$.

Chip Color and Analytical. The oil content of the chips was determined by the $CCI₄$ extraction procedure (20). The defatted chips were then extracted for 2 hours with 40 times by volume of their weight of 55 per cent (v/v) ethyl alcohol. After filtering (Whatman No. 3 paper) and diluting with an equal volume of water the color was read in a Klett colorimeter with a No. 420 filter. The color values are expressed as Klett

readings. The moisture content of the chips was determined by drying at 70 \degree C. (vacuum) for 40 hours.

Analytical procedures for the determination of the individual sugars of the potatoes are given in a previous publication (14). Total nitrogen of the dice was determined by the Kjeldahl procedure and the amino nitrogen by the Van Slyke procedure.

RESULTS

Browning vs. Composition. Pertinent data on browning, rate and analytical values for the Maine potato varieties are shown in table 1. These data show an interrelationship between specific gravity, reducing sugars, and browning. The browning rates of dehyrated dice from the western potatoes stored at 3 temperatures for varying times are shown in table 2. Sugar data have been published previously (14).

1MFB, determination on the unblanched dice.

 ${}^{2}\Delta E \frac{5\%}{5 \text{ cm}}$ 420 m μ /day \times 10³, as defined in text.

 $1_{\Delta E}$ 5% 420 m μ /day \times 10³ as defined in text (average values).

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TABLE *3.--Color, moisture, and oil content of potato chips, White Rose variety.*

 $11 = \text{Very Good}; 2 = \text{Good}; 3 = \text{Passable}; 4 = \text{Dark}; 5 = \text{Very Dark}.$

 2 Average of stored samples \pm average deviation of a single measurement; initial values not included.

3Overall average including initial sample.

The total and amino nitrogen contents of the dice were relatively constant and thus independent of the temperature or time of storage of the tuber. The total nitrogen for the dice from White Rose and Russet Burbank averaged 1.35 ± 0.03 and 1.53 ± 0.03 per cent on a moisture-free basis (average deviation of a single measurement), respectively. The respective amino nitrogen values were 0.48 ± 0.05 and 0.64 ± 0.03 .

Tables 3 and 4 present data on color, moisture, oil content, and subjective appraisal of the chips prepared from the same lots of western potatoes used for dice dehydration as outlined under "Methods". It will be noted that for both western varieties the color, as expected, was greater at the lower than at the higher raw storage temperatures. Furthermore, the average color developed in both processed products was greater in the White Rose than in the Russet Burbank. The average oil content of the chip was slightly, but also significantly, greater for White Rose (in

TABLE *4.--Color, moisture, and oil content of potato chips, Russet Burbank variety.*

1,2,3See table 3.

agreement with recent findings of Wright (21), but there were no significant differences with respect to raw storage temperatures. The moisture values of the chips exhibited no significant trend and were reasonably uniform over the entire experiment, indicating uniformity of the cooking technique used.

Browning vs. Composition- Conventional Statistical Treatment. A typical plot of the individual sugar content of raw western tubers (Without regard to variety) against the color values of the chip is shown in figure 1. The straight lines drawn through these points are the statistical lines of regression (assuming linear relations between sugar and color) obtained from the statistical constants tabulated in table 5. It will be noted that fair correlation (r greater than 0.90) was obtained in all cases except for sucrose. Good correlation with chip color was obtained for both reducing sugar and glucose. These conclusions are brought out more strikingly by comparison of values of the standard error of estimate $(\sigma y.x)$. Thus the σ y.x for glucose or reducing sugar as a measure of chip color was one-half that of the total sugar or dice color as such a measure. On the other hand,

FIGURE 1.—Plot of soluble chip color against the individual sugar content (and combinations thereof) of the raw tuber (western varieties). The straight lines are the calculated lines of regression (See table 5).

any sugar or combination thereof (with the exception of sucrose) listed in table 5 served almost equally well as an index of dice color, with fructose and reducing sugar yielding a slightly smaller $\sigma y.x$ than glucose or total sugar. However, chip color would seem, from table 5, to be a

Dependent Variable		Statistical Constants ¹					
(x)	$sd\ x$	\boldsymbol{a}	Ь	r	$\sigma y.x$		
		Chips—Soluble Color					
Frucose Red. Sugar Total Sugar Dice Browning	0.63 0.99 1.60 0.63 2.08 \sim	θ 36 6 22 -12 93	244 151 129 96 71 1.19	0.99 0.97 0.53 0.99 0.96 0.94	22 339 136 23 44 44		
		Dice-Rate of Browning X10 ³					
Glucose Red. Sugar Total Sugar Chip Color	0.63 0.99 1.60 0.63 2.08 123	$\frac{5}{9}$ $\frac{8}{7}$ -78	28 18 12 11 8 0.84	0.92 0.94 0.41 0.94 0.93 0.94	$\frac{8}{7}$ 18 8 $\overline{\mathbf{4}}$		

TABLE *5.--Statistical data .on sugar content of raw tubers vs. color of processed western potatoes.*

 $\mathbf{1}y = \mathbf{a} \times \mathbf{b}$; $a = y$ intercept, $b = \text{regression coefficient}$ sd $x = \text{standard deviation}$ of x ; $r = \text{correlation coefficient}$; $\sigma y \times \mathbf{b} = \text{Standard error of } y$ with respect to x.

better index of browning rate of dice than sugar contents. Plots of browning *vs.* sugar did not display any definite grouping of points with respect to the two western varieties.

That these statistical data demonstrate a similar trend for both processed products is shown in figure 2. Here the y-intercepts ("a") and the regression coefficients ("b") of the chips (data of table 5) are plotted against the corresponding constants for dice. The *"a"* values decreased in the order: Fructose reducing sugars, glucose total sugar; and *"b"* values in the order: glucose, fructose, reducing sugar, and total sugar.

To learn whether or not sugar contents could be made to serve as a more reliable index of dice browning rate, sugar analyses were performed on dehydrated dice and compared statistically with rate of drowning of the dice. The data of table 6 (as compared with those in table 5) demonstrate that use of sugar values of western varieties of dehydrated dice did not increase the reliability of sugar values as a measure of browning rate, in comparison with similar statistical values obtained with raw-tuber data. Table 6 also tabulates the statistical constants relating reducing sugar contents of Maine and western varieties to browning rate of dehydrated dice prepared from them. Apparently reflecting the smaller range of sugar values, a relatively low correlation coefficient $(r = 0.90)$ was obtained from the Maine tubers. On the other hand, the values of the standard error of estimate ($\sigma_{V,X}$) were significantly lower than those for western tubers based upon reducing-sugar content of either raw tuber or dehydrated dice.

FIGURE 2.—Interrelation among the statistical constants of the regression line relating chip and dice color to potato sugar content (See table 5).

The "Discriminatory Index" (DI). Since the values for $\sigma y.x$ are expressed in terms of essentially non-comparable units, it is rather difficult to compare the results for chips and dice on the basis of this statistical parameter. One approach to such an intercomparison would be to define a "discriminatory index" (DI) as:

$$
\mathrm{DI} = \frac{\mathrm{R}^1}{\sigma_{\mathrm{y.x}}},
$$

where R_1 is the range of values over the samples studied. In the present cases these ranges are 600 color units for chip color and I00 units for dice browning rate. One can then visualize this range as divided into a series of segments. The size of each segment $(\sigma y.x)$ will depend upon

Varieties	Condition of Potato	Sugar	No. of Samples	a	b	SDx	\mathbf{r}	σ y.x
Western ,, ,, , , ,, , , Maine Maine $+$ Western	Dehydrated ,, ,, , , Raw Raw Raw	Glucose Fructose Sucrose Red. Sugars Total Sugar Reducing Red. Sugar Red. Sugar	24 24 24 24 24 27 14 41	8.3 6.0 -3.9 6.7 0.4 7.1 7.0 6.6	24.7 17.8 19.2 10.6 7.8 11.3 9.7 11.2	0.69 1.08 0.77 1.75 2.37 1.61 0.73 1.34	0.84 0.94 0.72 0.91 0.83 0.94 C.90 0.93	11.4 7.2 14.4 8.6 8.6 6.8 3.6 6.2

TABLE 6.-Statistical data on sugar contents of tuber and of dehydrated *dice vs. browning rate of the dice.*

both the nature of the unitage system and the reliability of the measurement, whereas the number of segments (DI, a dimensionless quantity) will depend only on the "discriminatory" or resolving power of the measurements being used as an index of browning. This parameter is related to the "sensitivity" statistic recently developed by Mandel and Stiehler (9). Table 7 shows values of DI to the nearest integer over the entire color range studied (R_1) for both dice and chips. It will be noted that for $R_1 = R_1$ glucose and reducing sugar constitute more sensitive criteria of chip color than of dice color, whereas fructose and total sugar serve as equally comparable indices (but at a lower level of sensitivity) of both chips and dice. Finally, and possibly most significant from a practical point of view, is the observation that chip color constitutes a more sensitive index of dice browning rate than any one of the sugar components or combinations thereof.

TABLE *7.--Application of the "Discriminatory Index" to measured variables over entire color range* (R_i) and limited *color range (R/').*

Dependent Variable Measured	Discriminatory Index					
	Range $= R_1'$		Range $= R_1''$			
	Chips	Dice:	Chips 9.7 5.5 1.6 9.3 49 4.9	Dice		
				3.8		
				4.3		
Reducing Sugar	26			4.3		
$\begin{minipage}{.4\linewidth} Total Sugar \end{minipage} \begin{minipage}{.4\linewidth} \vspace*{-1em} \begin{minipage}{.4\linewidth} \vspace*{-1em} \begin{minipage}{.4\linewidth} \vspace*{-1em} \vspace*{-1em} \end{minipage} \begin{minipage}{.4\linewidth} \vspace*{-1em} \begin{minipage}{.4\linewidth} \vspace*{-1em} \vspace*{-1em} \end{minipage} \begin{minipage}{.4\linewidth} \vspace*{-1em} \begin{minipage}{.4\linewidth} \vspace*{-1em} \vspace*{-1em} \end{minipage} \begin{minipage}{.4\linewidth}$	14					
Dice Browning Rate	14					
Chip Color \ldots		25		7.5		

It is perhaps more instructive to determine DI values within a narrower range, including only those points one might expect to meet in practice. Thus we may select a value of 2.0 per cent reducing sugar as covering the restricted range of interest, since in commercial practice one very seldom finds higher values. For chips, this reducing-sugar value would correspond to \bar{R}_1 equal to 213 chip color units and to a browning rate equal to 39 units as obtained from the respective calculated regression lines. DI values for these limited ranges are also shown in table $7(R_1'')$.

Here we see the strict unreliability of sucrose as a measure of browning, since a DI value less than 2 would not permit even a qualitative estimate of browning to be made. An intercomparison of these values yields the following conclusions: (a) glucose and reducing-sugar determinations measure chip color twice as reliably as they measure dice browning rate; (b) these two sugar measurements constitute indices of chip color which are about twice as reliable as are fructose, total sugar, or dice browning rate; (c) with the exception of sucrose, each sugar or combination thereof serves equally well as an index of dice browning rate; and (d) chip color is a better index of browning rate than any sugar or combination thereof.

Kinetics of Accelerated Browning of Dehydrated Potato Dice. Figure 3 shows the course of browning for several lots of Katahdin potatoes, assorted according to specific gravity. It will be noted that within the experimental error of the method, a linear relation exists between optical density and time of heat damage at 50° C. It will also be noted, incidentally, that the browning rates are greater for the dice derived from low-specificgravity tubers. Linearity with respect to time was found to hold for the western varieties used in these and in previously reported studies (4,7). On the other hand, the course of browning of dice from the Green Mountain potatoes shows a definite induction period as shown in figure 4.

FIGURE 3.-Increase in optical density of extract from Katahdin dehydrated diced potato w'th time, illustrating the linear nature of the course of the browning reaction.

FIGURE 4.-Increase in optical density of extract from Green Mountain dehydrated diced potato with time, illustrating deviation from linearity of the course of the browning reaction.

To limit ourselves to a useful range of values, an optical density increase of 50 units was used in determining the browning rate. This rate was taken as the slope of the straight line from the origin through the points from browning of 50 units optical density increase. Such a range was selected in a previous publication (7) on the assumption that it may approximate the range of commercial interest. Browning corresponding to this average rate did not differ greatly from the observed browning during the stage of the reaction within this range.

This procedure is considered valid when the deviation from linearity is not very marked as is the case with non-sulfited potato dice. For the cases where the deviation is marked, as when sulfite is present, this procedure would result in considerable error at certain stages of browning; for such cases the procedure used in a previous correlation (6) with sulfited dried vegetables is considered preferable, the reciprocals of the lengths of time for given amounts of browning being taken as the average rates of browning. This procedure was first used by Stadtman *et al.* (18) with sulfited dried apricots.

DISCUSSION

Inspection of the results described herein reveals that although there is a fair degree of statistical interaction (as indicated by the conventional appraisal) among the measured variables (with the exception of sucrose),

the reliability of a single measurement, as evidenced by the "discriminatory index", leaves much to be desired in attempting to place the browningsugar relation on a firm causal basis.

With this viewpoint in mind, let us consider the various factors which might be responsible for variability as enumerated in the Introduction. (a) Extraction and clarification procedures can be eliminated as a source of variability, since these procedures have been shown to recover the sugars quantitatively $(14, 19)$. (b) The estimates of color are based upon carefully worked out procedures for measuring soluble brown color. It is true that the scatter at a high sugar content may be due to inability to measure total color. But elimination of these points by use of a "limited range of interest," R_1 ", table 7, reveals that scatter is still appreciable. (c) Deionization procedures have been shown to eliminate all possible ionic interferers (14). Furthermore, recent sugar analyses on both deionized and undeionized extracts yielded almost identical values as revealed in unpublished results. (d) Although glucose and fructose may apparently contribute to the browning to different extents as revealed here and in model browning systems (14, 15), this discrepancy cannot be the sole factor leading to scatter, since no measurement of fructose or glucose alone was significantly more reliable than the overall reducing sugar content. (c) Although the sugar content of the dice was significantly different from that of the raw tuber, no improvement in the reliability of sugars as a measure of browning rate could be detected. We can therefore eliminate this factor. (f) Variations in total and amino nitrogen were not great enough to be interpreted in terms of variations of browning susceptibility. These values, especially for the Maine potatoes, were characteristic of variety only and were fairly constant with respect to storage temperature.

Although these amino groups are most probably involved in browning (1, 10), under our conditions apparent inability of variations in the level of these groups to reflect corresponding changes in browning may be, at least in part, ascribed to their presence in non-rate-limiting concentrations. Thus it has been calculated that, on the average, the relative molar equivalent concentration of amino nitrogen in potatoes is about 5 times that of the average molar equivalent reducing-sugar concentration. Roughly, it has been estimated that, at this average level of amino nitrogen in the samples herein reported, a 5-fold excess of the latter over reducing sugar in potatoes would correspond to the very low level of reducing sugar of the order of 0.05 per cent (MFB).

We must therefore conclude (g) that unmeasured variations in the levels of other direct or possibly indirect factors (as mentioned in the Introduction) contribute to the formation of soluble brown color. If speculations may be permitted, it may be mentioned that organic acids (18) have been implicated in the non-enzymatic browning of fruits, and that metals (2) and phosphate ion (15) profoundly influence rate of nonenzymatic browning. The implication of phosphate in non-enzymatic browning is in agreement with its detection in appreciable amounts in potatoes (12, 13, 16) and also may serve to explain, in part, recent observations on the decrease in browning of dehydrated dice treated with calcium chloride previous to dehydration (17) . Again using average values, it has been estimated that the total equivalents of phosphorus potentially available as inorganic phosphate are roughly the same as the average of

the equivalents of reducing sugar. Hence there is enough phosphorus available to account for such an effect.

It indeed is not surprising that the chip color constitutes a more reliable index of browning rate of dice than does sugar measurements, since chip color probably represents more closely the resultant effect of the complex factors influencing the Maillard-type reaction in potatoes than does any particular sugar value. The DI value (equal to 7.5) for chips as a measure of dice browning rate within the limited R_1 " of table 7 would indicate that any other measurement which can resolve dice color into at least 8 significance intervals would be at least equally as valid as a measure of dice browning rate. Subjective observation of chip color, especially in comparison with appropriate graded color standards, can probably discriminate among at least 8 graduations. Such subjective tests *e.g.,* see Wright, 21) therefore may constitute as valid a test as any of the more elaborate measurements described herein.

The fact that the plots of accelerated browning *vs.* time were essentially and consistently linear for certain lots of potatoes but consistently curved upwards for other lots, as shown in figures 5 and 6, is evidence that the indicated difference in the shape of the plots is real and not the result of experimental uncertainty. It also indicates that the course of browning (or the relative importance of various contributing reactions) may be different in different lots of potatoes.

SUMMARY

A principal factor in assessment of quality of processed potato products is the Maillard-reaction-induced brown color which develops during processing (chips) or during subsequent storage (dehydrated dice). The present paper constitutes a statistical appraisal of the relationship between this browning and the individual reducing-sugar components of the raw tubers and dehydrated potato dice. Although correlation coefficients were greater than 0.9 (except for sucrose) comparison of standard errors of estimate and application of a parameter referred to herein as the "discriminatory index" reveals that sugar measurements, or combinations thereof, do not yield sufficient information to establish an exclusively complete causal relationship between reducing sugar components and extent and rate of browning. This is true despite the control or elimination of several factors usually overlooked in the pertinent measurements which might lead to the observed scatter. Possible auxiliary factors operating during the browning reaction, other than sugars and amino compounds, are discussed. Application of discriminatory indices to the data reveals reducing-sugar and glucose measurements as a more sensitive index of browning in potato chips than of browning rate of dehydrated potato dice. On the other hand, measurements of fructose and total sugar constitute equally reliable indices of both types of browning but at a lower sensitivity level. From the standpoint of prediction of quality of processed potato products in general, this analysis leads to the conclusion that subjective or semiquantitative appraisal of chip color may constitute at least as adequate an index of quality as any of the more elaborate objective measurements undertaken in the laboratory.

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