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STUDIES ON CRUST COLOR. I. THE IMPORTANCE OF THE BROWNING REACTION IN DETERMINING THE CRUST COLOR OF BREAD.[1,2]

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ABSTRACT

Flour milled from certain lots of Dutch wheat, yields loaves with a greyish crust color. The insufficient crust color is not due to shortage of sugars and the caramelization of the sugars does not provide an adequate explanation. The normal crust color of bread is chiefly the outcome of reactions of the Maillard type, i. e., reactions between reducing sugars and proteins or their hydrolytic products. Shortage of the latter may cause an insufficient crust color as well as shortage of sugars.

Adding dried egg white to grey-baking flour improved the crust color. The addition of dried egg white and individual amino acids to wheat starch and reducing sugar produced a stated effect on crust color.

A sufficient supply of steam at the start of the baking process is essential for the development of the crust color.

The literature yields few exact data about the reason(s) for the development of the brown crust color of bread. Blish, Sandstedt and Platenius (3) found a positive correlation between the diastatic power of flour and the crust color. Most manuals state that the brown crust color of bread is due to carmelization of sugars (7, 12) or of sugars and dextrins (2), formed during the fermentation process and the early stages of baking. In accordance with this concept, Kent-Jones and Price (8) enumerate a number of factors causing the absence of a brown crust color, which are all related to shortage of sugars. Certain wheat samples under investigation, however, yielded grey-baking flour, though no shortage of sugars could be established.

Many recent investigations deal with the so-called "browning reaction" in foods, which is characterized by the development of a brown color (10) after keeping the foods for a long time at room temperature or after exposing them to high temperatures such as in baking. The presence of proteins or their hydrolytic products on the one hand and reducing sugars on the other is essential for the "browning reaction".

Maillard in 1912 (9) was the first to describe reactions of this type with amino acids. Barnes and Kaufman in 1947 (1) point out the possible importance of browning reactions as a factor in the formation of color in baking and suspect that reactions of this type may also be

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essential for the development of flavors of many foodstuffs, as for instance the bread crust.

The question arose as to whether the normal brown crust color of bread is the outcome of reactions of the Maillard type and if so, whether the grey-baking properties of certain flour samples under investigation could be explained in accordance with this view.

Materials and Methods

All samples of flour, except one, were milled from Dutch wheat on a Bühler laboratory mill. Some of these wheat samples were obtained from the Dutch milling industry and others were known varieties from experimental-fields. A commercial flour of 78% extraction, milled from a grist of Dutch and import wheat was used as a control to provide bread of normal crust color.

Methods of Analysis. Moisture was determined in a 130°C. air-oven for one hour (after the oven regained 130°C.), ash by incineration at 550°C. to a light grey ash, and crude protein (N × 5.7) by the Kjeldahl method using the Parnas-Wagner apparatus.

Maltose figures were determined according to Van der Lee (See Pelshenke, 13), using however Luff's solution (See Browne and Zerban, 5) instead of Fehling's.

Pre-existing maltose was determined by the same procedure, leaving out the incubation period.

Reducing and non-reducing sugars were determined in the bread crust. The crust was separated from the crumb, dried at 80°C., pulverized and extracted for one hour with distilled water. After treating the extract with Carrez solutions (6) and filtration, reducing sugars were determined in an aliquot of the filtrate using Luff's solution. Another aliquot was subjected to inversion with hydrochloric acid at 70°C. for ten minutes, the total reducing sugars determined, and the non-reducing sugars computed by subtracting the quantity of reducing sugars found before inversion.

The pH value of the baking products was determined with a pH-meter (Radiometer, Copenhagen) after thorough mixing of the crumb with a small amount of distilled water.

Baking Methods. In the experimental baking tests, doughs containing 1.75% yeast and 2% salt (flour basis) were prepared with a consistency of 360 Brabender Units. The doughs were fermented at 30°C. and hand punched after 20, 40, and 55 minutes, then placed in the baking pan and proofed for a sufficient time to yield a standard amount of carbon dioxide, as determined by the S.I.A. recorder[4] on

[4] Manufactured by Aktiebolaget S. J. A., Stockholm, Sweden. This fermentation recorder has been described by Pelshenke (14).

an aliquot of the original dough. The doughs were baked for 25 to 30 minutes in an oven at about 250°C.

Two or three loaves were made from each dough. Their sizes varied from 100 - 400 g. in different experiments, depending upon the amount of material available.

Small-scale baking tests were used in several experiments. In one case 30 g. of flour and 0.6 g. yeast was used and the dough kneaded in the mixograph (15). Round tins (2 cm. high, bottom diameter 4 cm., and top diameter 7 cm.) were used.

A number of "model experiments" were carried out by baking mixtures containing wheat starch, water, sugars, dried egg white or amino acids and, in some cases, of leavening agents.

To distinguish easily small differences in grey-baking properties an oven temperature of 210°C. was used in most experiments. At this temperature normal-baking flour yielded loaves with a light golden brown crust, whereas the abnormal flours gave loaves with a greyish-white crust color in extreme cases.

To avoid a drop in the oven temperature which resulted from the opening of the oven door at regular intervals during the early experiments, the fermentation periods were adjusted in such a way that the loaves were all placed in the oven at the same moment. This was done chiefly at the expense of the three first fermentation periods.

Several experiments with different types of flour proved that lengthening and shortening the three first fermentation periods by 12 minutes at the utmost, did not influence the crust color. In many experiments steam was supplied at the start of the baking process, since this made the surface of the loaves glossy and improved the crust color of those made from grey-baking flours.

Unless indicated otherwise in the description of the baking experiments, no steam was supplied at the start of the baking process and an oven temperature of 210°C. was used.

Results and Discussion

Analysis of Flour Samples. Most experiments were carried out with grey-baking flour No. 1, milled from Dutch wheat of unknown variety. This flour was compared with a commercial flour No. 2, which was used as a normal-baking standard flour. Reducing and non-reducing sugars were determined in the crust of loaves baked from these two types of flour. The data recorded in Table I show that there is a great difference in protein content and a maltose figure. A maltose figure

TABLE I

ANALYSIS OF TWO FLOUR SAMPLES AND OF THE CRUST OF THEIR LOAVES

	Sample No. 1, Giving Grey Crust Color	Sample No. 2, Giving Normal Crust Color
Protein,[1] %	8.1	13.2
Maltose figure,[2] %	0.69	1.05
Pre-existing maltose,[2] %	0.27	0.25
Reducing sugars in bread crust,[1] %	1.40	1.59
Non-reducing sugars in bread crust,[1] %	0.71	0.43

[1] On moisture-free basis.
[2] On flour basis.

of 0.69, however, does not point to sugar shortage in cases of flour milled from Dutch wheat on a Bühler laboratory mill. Of 145 flour samples of the 1947 crop, for instance, which included all the important Dutch wheat varieties, 58 had a maltose figure lower than 0.70 but most of these yielded loaves with a normal crust color. The higher maltose figure of flour No. 2 is undoubtedly connected with the fact that this flour was produced on a commercial mill.

The amount of sugars found in the greyish crust of the loaves from flour No. 1, indicates also that there is no shortage of sugars. The caramelization theory provides no satisfactory explanation for the abnormal crust color in this case.

Addition of Gluten to Grey-Baking Flour. To the dough of a grey-baking flour No. 3, milled from Dutch wheat of the variety Alba, gluten was added. This gluten was in one case made from this grey-baking flour No. 3, in a second case from normal-baking flour No. 4, milled from Hard Winter wheat. The gluten was washed out by hand with tap water from the respective doughs. Table II shows the results of the baking experiments. In both cases a considerable improvement in crust color was obtained after the addition of the gluten.

Addition of Wheat Starch to Normal-Baking Flour. Increasing amounts of commercial wheat starch were added to normal-baking flour No. 2. The resulting crust colors were compared with the crust color of loaves from grey-baking flour No. 1. Table III shows that the addition of wheat starch causes a considerable deterioration of the crust color. In all three cases the crust color, after the addition, was inferior to the color produced by the grey-baking flour, although in two cases the protein content was higher. This proves that the pro-

TABLE II
EFFECT ON CRUST COLOR OF THE ADDITION OF GLUTEN TO GREY-BAKING FLOUR

Flour Used	Weight	Addition to the Dough of gluten from	Protein[1] in Dough	Crust Color
	g.		%	
Normal-baking flour No. 4	180	......	11.2	brown
Grey-baking flour No. 3	180	......	7.1	light cream to grey-white
Grey-baking flour No. 3	170	73 g. flour No. 4	10.9	brown with a grey bloom
Grey-baking flour No. 3	170	125 g. flour No. 3	10.7	slightly lighter brown with a grey bloom

[1] On moisture-free basis.

TABLE III
EFFECT ON CRUST COLOR OF THE ADDITION OF WHEAT STARCH TO NORMAL-BAKING FLOUR

Flour Used	Weight	Wheat Starch	Protein[1] in Dough	Crust Color
	g.		%	
Normal-baking flour No. 2	175	..	13.0	light brown
Grey-baking flour No. 1	175	..	7.9	yellow-cream
Normal-baking flour No. 2	121	54 g.	9.2	grey-white with a bit brown
Normal-baking flour No. 2	105	70 g.	8.1	grey-white with a bit cream
Normal-baking flour No. 2	89	86 g.	7.3	grey-white (like moist chalk)

[1] On moisture-free basis.

tein content of the dough is not the only determining factor in crust color.

Addition of Dried Egg White, Sugars and Individual Amino Acids to Grey-Baking Flour. Although gluten is a complicated mixture, it seems obvious to connect its effect in improving crust color with the proteins, these being the major component of gluten. To verify this, experiments were made with dried egg white,[5] which is less complex than gluten.

Table IV gives the results of these experiments, where dried egg

[5] Dried egg white = "Albumen ovi siccum", according to the fifth edition of the "Nederlandsche Pharmacopee".

TABLE IV

Effect on Crust Color of the Addition of Dried Egg White with or without Sugars to Grey-Baking Flour

Exp.	Flour Used	Weight	Dried Egg White	Sugars Used	Weight	Crust Color
		g.	g.		g.	
A	grey-baking flour No. 1	600	..	....	..	light cream
	grey-baking flour No. 1	600	36	....	..	brown
	grey-baking flour No. 1	600	36	dextrose	12	deep brown
	grey-baking flour No. 1	600	..	dextrose	12	dark cream
	normal-baking flour No. 2	600	..	....	..	light golden brown
B	grey-baking flour No. 1	150	9	....	..	light golden brown
	grey-baking flour No. 1	150	9	dextrose	1.6	golden brown
	grey-baking flour No. 1	150	9	maltose	3	golden brown
	grey-baking flour No. 1	150	9	sucrose	3	golden brown
	grey-baking flour No. 1	150	..	....	..	cream
C[1]	grey-baking flour No. 3	150	9	....	..	dark cream
	grey-baking flour No. 3	150	9	dextrose	1.6	light brown
	grey-baking flour No. 3	150	9	maltose	3	dark cream to light brown
	grey-baking flour No. 3	150	9	sucrose	3	dark cream
	grey-baking flour No. 3	150	..	....	..	grey-white
D	grey-baking flour No. 1	30	..	dextrose	0.32	grey-white to cream
	grey-baking flour No. 1	30	0.3	dextrose	0.32	cream with brown spots
	grey-baking flour No. 1	30	0.6	dextrose	0.32	as many cream spots as brown spots
	grey-baking flour No. 1	30	0.9	dextrose	0.32	light brown with many cream spots
	grey-baking flour No. 1	30	1.2	dextrose	0.32	light brown with few cream spots
	grey-baking flour No. 1	30	1.5	dextrose	0.32	golden brown
	grey-baking flour No. 1	30	1.8	dextrose	0.32	fairly deep golden brown
	grey-baking flour No. 1	30	1.8	....	..	golden yellow
	grey-baking flour No. 1	30	1.8	dextrose	0.08	deep golden yellow
	grey-baking flour No. 1	30	1.8	dextrose	0.16	light golden brown
	grey-baking flour No. 1	30	1.8	dextrose	0.24	golden brown
	grey-baking flour No. 1	30	1.8	dextrose	0.32	fairly deep golden brown
	grey-baking flour No. 1	30	1.8	dextrose	0.64	very deep golden brown
	grey-baking flour No. 1	30	1.8	dextrose	0.96	very deep golden brown
	grey-baking flour No. 1	30	1.8	dextrose	1.28	deep golden brown

[1] No yeast or leavening agent was used in the baking tests.

white and/or sugars were dissolved in water and added to grey-baking flour in making the dough.

Experiment A (Table IV) shows that the addition of dried egg white considerably improves the crust color. This effect is increased distinctly when dextrose is added together with dried egg white, whereas the mere addition of dextrose gives only a slight improvement of crust color.

From the results of experiment B (Table IV) it appears that, added in combination with dried egg white, the color improving effects of equimolecular amounts of dextrose, maltose and sucrose are about the same. The positive effect of sucrose appears to be in contradiction with what is known concerning the browning reaction in other fields. However in doughs, sucrose is rapidly converted to glucose and fructose by the invertase of yeast, as shown by Geddes and Winkler in 1930 (7).

In experiment C (Table IV) no yeast or leavening agent was added to the doughs. Immediately after mixing, the doughs were placed in the baking pans and baked. Under these conditions, where the splitting action of yeast enzymes on the added sugars has been eliminated, dextrose shows a distinct improving effect, maltose a smaller effect and sucrose no positive effect at all. These facts are well in accordance with our knowledge of browning reactions.

In the small-scale baking tests of experiment D (Table IV), increasing amounts of dried egg white in combination with a constant amount of dextrose were added to grey-baking flour No. 1, as well as a constant amount of dried egg white together with increasing amounts of dextrose.

The results demonstrate clearly the importance of both the protein and the reducing sugar for the development of the crust color. Glutamic acid was added to grey-baking flour No. 1 in increasing amounts of 0.5, 1 and 2% on flour basis. The solution of glutamic acid in water was adjusted with sodium hydroxide to a pH of 6.2 and added to the flour on doughmaking. A distinct improvement in crust color, proportional to the amount of the amino acid, was shown.

Addition of 0.5% glycine to grey-baking flour No. 1 also improved the crust color.

"Model Experiments" with Wheat Starch, Different Sugars and Dried Egg White or Amino Acids. Different sugars and dried egg white were dissolved in water and added to wheat starch in doughmaking. Instead of yeast, baking powder was used as a leavening agent. Table V presents the results.

TABLE V

CRUST COLOR DEVELOPED IN BAKING EXPERIMENTS WITH WHEAT STARCH, DRIED EGG WHITE AND DIFFERENT SUGARS

Wheat Starch Used in Doughmaking	Dried Egg White	Sugars Used	Weight	Crust Color
g.	g.		g.	
218	32	..	..	grey-white
215	32	dextrose	2.7	brown
213	32	maltose	5	light brown
213	32	sucrose	5	grey-cream
392	..	maltose	8	greyish

It is apparent from these experiments that a brown crust color is developed when both dried egg white and a reducing sugar (dextrose, maltose) are present, whereas practically no coloring effect appears when only one of these substances has been added to wheat starch or when dried egg white has been added in combination with a non-reducing sugar (sucrose). This proves that under the conditions of these "model experiments" the development of a brown crust color is due to browning reactions. Dextrose has in combination with dried egg white more coloring effect than an equivalent amount of maltose.

In a second series of "model experiments", small-scale baking tests were performed with wheat starch, a constant amount of dextrose and equimolecular amounts of different amino acids. Each dough was made of 25 g. of total solids. With the exception of cystine, all constituent amino acids of egg albumin have been examined (4). When the pH of the amino acid solution was beyond the range 5.1 - 6.3, either diluted hydrochloric acid or sodium hydroxide was added, in order to maintain a narrow pH range of the doughs. No leavening agents were used, these causing ruptures of the crust and local differences in pH. The baking time was fixed at 45 minutes in order to obtain even crust colors. The results are shown in Table VI.

It is apparent from Table VI that the constituent amino acids of egg albumin have different brown-coloring potencies in combination with dextrose.

Influence of Ammonium Compounds on the Crust Color. Browning reactions do not occur only between reducing sugars and amino acids, many other compounds may also be involved. According to Patron (11) for instance, amino acids may be substituted by proteins, polypeptides, amines and ammonium salts.

As indicated before, addition of protein (dried egg white) or amino

TABLE VI

CRUST COLOR DEVELOPED IN BAKING EXPERIMENTS WITH WHEAT STARCH, DEXTROSE AND EQUIMOLECULAR AMOUUNTS OF DIFFERENT AMINO ACIDS

Amino Acid	Weight	Dextrose	Crust Color[1]
	g.		
Lysine-2 HCl	0.135 (0.159)[2]	0.27 g.	light brown to brown to deep brown
Glycine	0.055	0.27 g.	light brown to brown to deep brown
Serine	0.077	0.27 g.	golden yellow to light brown to brown
Alanine	0.065	0.27 g.	golden yellow to light brown to brown
Arginine	0.127	0.27 g.	golden yellow to light brown to brown
Valine	0.086	0.27 g.	yellow to light brown to brown
Leucine	0.096	0.27 g.	yellow to light brown to brown
Glutamic acid	0.108	0.27 g.	golden yellow to light brown
Tyrosine	0.132	0.27 g.	golden yellow to light brown
Tryptophane	0.149	0.27 g.	yellow to light brown
Phenylalanine	0.121	0.27 g.	yellow to light brown
Methionine	0.109	0.27 g.	yellow to light brown
Threonine	0.087	0.27 g.	yellow to light brown
Histidine-HCl	0.14	0.27 g.	yellow to light brown
Aspartic acid	0.097	0.27 g.	yellow with brown bloom
Proline	0.084	0.27 g.	yellow to golden yellow

[1] Steam supplied at start of baking process.
[2] Due to shortage of material, less amino acid has been added than the required amount which has been recorded in brackets.

acids (glutamic acid, glycine) to grey-baking flour No. 1, distinctly improved the crust color. Table VII shows that the addition of different ammonium compounds to grey-baking flour No. 1, also improves the crust color. The pulverized salts were thoroughly mixed with the flour before doughmaking. The added amounts of ammonium chloride–sulfate and–phosphate were equivalent to 0.5% ammonium bicarbonate (based on the nitrogen content).

TABLE VII

EFFECT ON CRUST COLOR OF THE ADDITION OF AMMONIUM SALTS TO GREY-BAKING FLOUR NO. 1

Grey-baking Flour No. 1	Ammonium Salt Added on Basis of Flour	Amount	Crust Color[1]	pH of Crumb
g.		%		
85	....		yellow to golden yellow	5.55
85	ammonium bicarbonate	0.5	golden yellow to light brown	7.49
85	ammonium bicarbonate	1.0	light brown to brown	7.88
85	ammonium chloride	0.34	light brown to brown	5.38
85	ammonium sulfate	0.42	light brown to brown	5.35
85	di-ammonium phosphate	0.42	light brown	6.28

[1] Steam supplied at start of baking process.

In a series of "model experiments", the same ammonium salts in combination with a constant amount of dextrose were added to wheat starch. Three amino acids were also added for comparison. The amounts of ammonium salts and amino acids were based on the nitrogen content equivalent to an addition of 0.5% ammonium bicarbonate. After dissolving the ammonium salts or amino acids in water, dextrose was added and the solution mixed with wheat starch on dough-making. No leavening agents were used. The results are presented in Table VIII.

TABLE VIII
CRUST COLOR IN BAKING EXPERIMENTS WITH WHEAT STARCH, DEXTROSE AND AMMONIUM SALTS OR AMINO ACIDS

Ammonium Salt or Amino Acid[1]	Amount	Dextrose[1]	Crust Color[2]
	%	%	
. .		1.07	light cream
ammonium bicarbonate	0.5	1.07	cream
ammonium chloride	0.34	1.07	cream with light brown spots
ammonium sulfate	0.42	1.07	cream with brown spots
di-ammonium phosphate	0.42	1.07	brown to deep brown
glycine	0.47	1.07	deep brown
serine	0.66	1.07	deep brown
phenylalanine	1.04	1.07	brown with golden yellow bloom

[1] On basis of total solids.
[2] Steam supplied at start of baking process.

Further "model experiments" with ammonium salts, with and without adjusting the pH of the solution of the ammonium salt and dextrose to 5.5, showed that the apparent pH differences did not influence the crust color.

The results of these experiments with ammonium compounds are another indication that browning reactions are involved.

Influence of Steam on the Crust Color. Baking experiments with grey-baking flour No. 1, to which dried egg white and/or dextrose had been added, were performed without steam and with steam supplied immediately at the start of the baking process. The results are shown in Table IX.

Apart from the fact that the crust is dull without steam and glossy with steam, Table IX shows that steam supplied at the start of the baking process considerably improves the crust color of the grey-baking flour with and without dextrose, whereas practically no im-

TABLE IX

EFFECT ON CRUST COLOR OF THE ADDITION OF STEAM IN BAKING EXPERIMENTS WITH GREY-BAKING FLOUR

Grey-baking Flour No. 1	Dried Egg White[1]	Dextrose[1]	Treatment	Crust Color
g.	%	%		
85	..	..	no steam	grey-white with a bit cream, dull
85	..	1.07	no steam	cream with violet bloom, dull
85	6	..	no steam	golden brown, dull
85	6	1.07	no steam	deep brown, dull
85	..	..	steam	cream with golden yellow bloom, glossy
85	..	1.07	steam	light golden brown, glossy
85	6	..	steam	golden brown, glossy
85	6	1.07	steam	deep brown, glossy

[1] On basis of flour.

provement appears if dried egg white, whether or not in combination with dextrose, has been added.

To investigate whether the color improving effect of an immediate steam supply is due to the promotion of a caramelization process or of a browning reaction, baking experiments were carried out using wheat starch to which increasing amounts of dextrose had been added. No yeast or leavening agents were used. Browning reactions were practically eliminated under these conditions. The results appear in Table X.

TABLE X

EFFECT ON CRUST COLOR OF THE ADDITION OF STEAM IN BAKING EXPERIMENTS WITH WHEAT STARCH AND DEXTROSE

Wheat Starch	Dextrose[1]	Treatment	Crust Color
g.	%		
85	..	steam	grey-white
85	1	steam	grey-white
85	2	steam	grey-white
85	4	steam	grey-white

[1] On basis of wheat starch.

These results make it improbable that the color improving effect of steam should be due to the promotion of a caramelization process. If steam supply at the start of the baking process, however, promotes browning reactions, the question arises why this improvement in crust color fails to appear after the addition of dried egg white to grey-baking flour No. 1. A possible explanation is that browning reactions in the loaf surface are inhibited if the moisture content decreases too quickly. An immediate supply of steam will retard the evaporation from the loaf surface, which could explain its positive effect on the crust color in cases of grey-baking flour with or without the addition of dextrose. The addition of dried egg white must, due to its water binding capacities, also retard the desiccation of the loaf surface, thus possibly promoting browning reactions indirectly, apart from its direct role as a reacting compound. On this basis it is understandable that supplying steam at the start of the baking process causes no further improvement in crust color, if dried egg white has been added to grey-baking flour.

The baking experiments referred to in Table X were repeated in one case by fixing the baking time at 50 minutes and in another by using an oven temperature of about 240°C. A slight improvement in crust color, proportional to the amount of dextrose added, could be observed in both cases. The small improvement in crust colors, however, make it clear that caramelization processes play no important part under these conditions.

In further experiments with grey-baking flour, a gradual addition of steam has been compared with an immediate steam supply at the start of the baking process. This gradual addition was obtained by injecting small amounts of steam in the oven, 1, 6, 11, 16 and 21 minutes after the start of the baking process.

The results showed that a gradual addition of steam during the baking process has a much smaller improving effect on the crust color of grey-baking flour than immediate steam supply at the start of the baking process.

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