Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Color Formation in Wheat Starch Based Glucose Syrups and Use of Commercially Available and Laboratory-Prepared Agricultural Waste-based Activated Carbons for Decolorization

İpek Çelebi[a]; N. Suzan Kincal[a]
[a] Chemical Engineering Department, Middle East Technical University, Ankara, Turkey

To cite this Article Çelebi, İpek and Kincal, N. Suzan(2007) 'Color Formation in Wheat Starch Based Glucose Syrups and Use of Commercially Available and Laboratory-Prepared Agricultural Waste-based Activated Carbons for Decolorization', Separation Science and Technology, 42: 8, 1761 — 1773
To link to this Article: DOI: 10.1080/01496390601174232
URL: http://dx.doi.org/10.1080/01496390601174232

Separation Science and Technology, 42: 1761–1773, 2007

ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390601174232

Color Formation in Wheat Starch Based Glucose Syrups and Use of Commercially Available and Laboratory-Prepared Agricultural Waste-based Activated Carbons for Decolorization

İpek Çelebi and N. Suzan Kincal
Chemical Engineering Department, Middle East Technical University, Ankara, Turkey

Abstract: Glucose syrups were produced from wheat starch using α-amylase at 97°C for 45–90 min liquefaction times, followed by saccharification with amyloglucosidase at 60°C for 18 h to study the color formation. This was followed by decolorization studies using 0.25 to 1.00 g activated carbon per g of syrup of the commercially available NORIT and several activated carbons prepared from apricot stones and hazelnut shells and husks on laboratory scale. Increase in liquefaction time resulted in higher extents of hydrolysis in both maltodextrins and glucose syrups. In maltodextrins, 9–21% and in glucose syrups 72–98% of the linkages were hydrolyzed at 45–90 min liquefaction times. Color levels of glucose syrups increased with increased liquefaction time. The color levels were between 657–1424 ICUMSA units for glucose syrups obtained at 45–90 min liquefaction times. The dosage of activated carbon necessary to decolorize the syrups to the color level lower than 100 ICUMSA units, however, became lower and lower as the liquefaction time (and the level of starting color), was increased; and this behavior was the same for all types of activated carbon studied. Decolorization performances of NORIT, apricot stone (AS), hazelnut husk (HH) and hazelnut shell (HS) based activated carbons were compared by adjusting the activated carbon dosage to be the same as that of NORIT required for 100 ICUMSA residual color. HH was the best giving practically same residual color as NORIT for the decolorization of 90 min liquefied glucose syrups.

Received 1 June 2006, Accepted 11 December 2006
Address correspondence to N. Suzan Kincal, Chemical Engineering Department, Middle East Technical University, Ankara 06531, Turkey. Tel.: +90 (312) 210 2617; E-mail: nskincal@metu.edu.tr

The HS and AS had similar performances, reducing the color to less than 180 ICUMSA units for syrups liquefied for 45 and 90 min.

Keywords: Glucose syrups, activated carbon, decolorization, adsorption

INTRODUCTION

Starch is an important raw material in the food and pharmaceutical industries. Apart from its use in the native or modified form, starch is also used for the preparation of glucose syrups (1), which are purified aqueous solutions of nutritive saccharides, D-glucose, maltose, and oligomers of D-glucose (2). Glucose syrups can be produced by acidic or enzymatic hydrolysis. Acidic hydrolysis produces syrups with several byproducts while enzymatic hydrolysis enables production of syrups with closely controlled composition and properties (3, 4).

Enzymatic hydrolysis is carried out by using amylolytic enzymes which specifically degrade glycosidic bonds in starch. It is typically carried out in two steps—liquefaction and saccharification—involving different enzymes. In the industrial scale, the gelatinization for 1–2 h at high temperatures should be carried out prior to or along with liquefaction in order to make the accessibility of enzymes to starch easier (5, 6). Using batch reactors is the classical way for enzymatic hydrolysis of starch (4). After pH adjustment to 6–7, the starch substrate at 30–35% w/w dry substance concentration (4, 7), is mixed with the enzyme at temperatures higher than 80°C (7) and $CaCl_2$ is added to stabilize the enzyme (8). Heating-up of the suspension, combined with enzyme addition initiates the hydrolysis process. As a result of liquefaction, maltodextrins are formed (7), which find use in several areas in industry. The main factors that affect liqueafaction are starch source and crystallinity, physical-chemical properties of starch; the particle size of starch, enzyme type and concentration, time, pH, and temperature. Recently, hydrolysis of commercial, PIY, and freshly prepared, FWS, wheat starch has been studied (8, 9) for the production of glucose syrups. Liquefaction was carried out using *Bacillus licheniformis* α-amylases from Sigma (6.17 mg protein/ mL) and Orba (1.90 mg protein/ mL) at dosages of 1/1000 (v/w of dry starch). Within the concentrations range of 20–34% (w/w), the highest conversion was obtained with 27% (w/w) at 97°C in batch system. Both enzymes gave similar extents of hydrolysis. The effect of enzyme dosage was determined to be 3.5 times higher than the extent of hydrolysis as a response to 5 times increase in enzyme dosage (9). The extent of hydrolysis under similar conditions was 2.5 lower in the commercial starch PIY, compared to that in FWS (8).

Following liquefaction, saccharification, in which hydrolysis of maltodextrins into lower molecular weight products is achieved by one or several types of enzymes, namely amyloglucosidase, sometimes along with

pullulanase (7). Saccharification is generally performed at 50°C and pH 5–6 (4, 10) which are optimal values. Depending on the final degree of hydrolysis, the products of saccharification will be different (7).

In industry saccharification is followed by a purification step since browning occurs during the processing of glucose syrups, the color of which is represented by ICUMSA units (11). Glucose syrups should have a color level below 200 ICUMSA units (12) defined according to the following equation (11)

$$\text{ICUMSA} = 1000 * \text{A}/(\text{b} * \text{c}), \quad (1)$$

where A = absorbance at 420 nm of the test sample; b = length (cm) of the absorbing path; c = concentration (g sugar/ml) of the test sample.

Color formation in syrups may occur as a result of two types of reactions, namely the Maillard reaction and caramelization, but the latter has a negligible effect on glucose syrups since it requires temperatures higher than 120°C (13). Therefore, color in glucose syrups is thought to be due to the Maillard reaction, occurring at temperatures above 50°C and at pH 4–7 (13). It begins with a condensation reaction between a free amino group and a reducing sugar; the products subsequently undergo Amadori rearrangements followed by cyclizations, dehydrations, isomerizations, and further condensations take place. As a result of all of these reactions, colored intermediates and other reactive precursors form, condense, and polymerize to give brown nitrogenous polymers and co-polymers, known as melanoidins (15). The molecular weights of melanoidins are several thousand Daltons. The lower molecular weight colored compounds, which form during earlier stages, typically contain two-to-four linked rings having extended double-bond conjugations. The relative amounts of these compounds depend on amino acid/protein to sugar ratio, temperature, pH, water activity, heating time, and discrete chromophore groups (14–17). The rate of melanoidin formation increases with temperature (14), and the browning rate was reported to follow an Arrhenius type of dependence on temperature (12, 18, 19). Other authors reported that the rate of melanoidin formation increases in proportion to the length of heating squared at a given temperature (14, 20). Several workers have derived theoretical equations to predict the extent of brown pigment as a function of time but they can not be solved because the rate constants of some intermediary reactions are still unknown (16).

In industrial practice, activated carbons have been used for many years in order to remove color from sugar syrups (21), although ultrafiltration membranes and ion exchange resins were also used. More than 98% of the colorants were removed from a fructose syrups after treatment with 33% (w/v) of activated carbon (22). Physical and chemical properties of activated carbon determine its efficiency for removing sugar colorants. Activated carbons having larger total surface area, a well developed macro- and mesoporosity, along with minimal surface charge (few carboxyl groups)

and low ash content have been reported to be desirable for better sugar decolorization (23).

Not only the properties of activated carbon but also the properties of the adsorbate have importance on adsorption. It has been reported that aromatic compounds are generally more adsorbable than aliphatic compounds of similar molecular size and branched chains are usually more adsorbable than straight chains (24) Little information is available about the adsorption of a wide variety of specific materials by activated carbon (25). But it is known that dissimilar compounds have different adsorption affinities due to having different molecular weights and structures (24). It has been reported that, given the same carbon with different types and varieties of colors, the adsorption isotherms will be different (26).

An increase in size or moleculer weight of organics usually favors adsorption until particle becomes too large to penetrate into the carbon pores (24–29). This can be explained by the lower solubility of adsorbates, and also by the possibility that small molecules, which may become attached at a single point, can be desorbed as soon as this bond is broken while larger molecules can be adsorbed initially by becoming attached through a single bond, the binding becoming strengthened when other points of contact are made (30).

During the production of glucose syrups, the determining step on the color formation is likely to be liquefaction rather than saccharification since it is carried out at temperatures higher than 90°C. Saccharification is carried out at 50–60°C, and its effect on the color formation is considered negligible. The aim of this study was to study the effect of liquefaction time on color formation and to compare performances of commercially available and agricultural waste based activated carbons for color removal in the dosage range 0.25–1 g/100 g syrup.

MATERIALS AND METHODS

Materials

Commercial wheat starch was purchased from the market. The hydrolysis of starch into maltodextrins was achieved using thermo-stable *Bacillus licheniformis* α-amylase from Sigma (liberating 1 mg of maltose from starch in 3 min, optimum pH 6.9; as reported by the company) and the maltodextrins thus obtained were hydrolyzed to glucose syrup using thermo-stable amyloglucosidase (*Aspergillus niger*, liberating 1 mg of glucose from starch in 3 min, optimum pH 4.5; as reported by the company) also from Sigma. NORIT PN 2 3730–1, which is a commercial peat based activated carbon produced by steam activation method, obtained from Norit Company (31), and agricultural waste based activated carbons that had been prepared in studies carried out in the Middle East Technical University Chemical Engineering Department by chemical activation method were used for

decolorization. Hazelnut based activated carbons, HS and HH, were prepared by 2 h carbonizations at 400°C. For HS, hazelnut shells, crushed to 12–18 mesh size were impregnated with 50% H_3PO_4 before carbonization; while for HH, dried hazelnut husks, crushed to 20–50 mesh size were impregnated with 30% H_3PO_4, before carbonization (32). Apricot stone based activated carbon, AS, was prepared by the carbonization for 90 min at 300°C of apricot stones, crushed to 10–18 mesh size and impregnated with 50% H_3PO_4 (33). Some properties of the activated carbons are given in Table 1.

Liquefaction

The hydrolysis of wheat starch into maltodextrins was carried out in a batch system. 15 g samples which contained 27% (w/w) dry wheat starch, in a buffer having a pH value of 6.5 to which $CaCl_2$ was added to give 100 mg Ca^{+2} per litre of solution to increase the thermo–stability of α-amylases were hydrolyzed for 45, 60, 75, and 90 min with α-amylase (0.5% of dry matter), 20 μL, in a shaking water bath having a temperature of 97°C. All experiments were carried out in two or three parallels, in which hydrolysis products were obtained at two or three different flasks at the same time. In experiments on liquefaction only, which were carried out to determine the intermediate extent of hydrolysis, the hydrolysis was stopped by decreasing the temperature by pouring mixture into 10 ml ice-water mixtures at −2°C and increasing pH to 12 by adding 1 ml 1 N NaOH to inactivate the enzyme. Before analysis, the samples were neutralized by 30 ml 37% HCl, diluted and analyzed immediately. In other experiments, the pH was adjusted to 4.5 using 100% acetic acid (0.3 ml).

Saccharification

At the completion of the chosen liquefaction time, 10 ml of hydrolyzed starch was mixed with 3.5 ml of buffer having pH value of 4.5 and were placed in

Table 1. Properties of activated carbons

Activated carbon type	Total pore vol., cm^3/g	Mesopore vol., cm^3/g	BET surface area, m^2/g	Ash content (%)
NORIT	0.50	—	609.2	10.55
HS	0.27[a]	0.03	426.0	1.60
HH	1.05[a]	0.58	1429.0	—
AS	0.22	0.03	450.0	5.40

[a]Mesopore + micropore volume.

shaking water bath at 60°C. Saccharification was started with the addition of amyloglucosidase, 20 μL, at the level of 0.5% of dry matter. At the end of 18 h, 1 ml of the hydrolyzate, glucose syrup, was withdrawn from solution for analysis and put into 8 ml of ice-water mixture at −2°C.

Analysis of Hydrolysates

The extents of hydrolysis in the maltodextrins and glucose syrups were determined by measuring absorbance values at 520 nm according to Nelson-Somogyi Method (34) using a Hitachi U-3200 spectrophotometer.

Decolorization of Glucose Syrups

After saccharification, glucose syrups were centrifuged for 20 min at 1700 g to remove insoluble solids. Before decolourization, the absorbance of centrifuged glucose syrups was determined using Hitachi U-3200 spectrophotometer at 420 nm against water using a 10 mm cell (11) and the color level was expressed in ICUMSA units. A 30 ml sample of glucose syrup was placed in a shaking water bath at 80°C for 5 min to stabilize the temperature. At this point, the type and amount of activated carbon to be studied was added. At the end of 30 min, the decolorized syrup was centrifuged for 20 min at 1700 g and filtered using Schleicher and Schuell Blau Band 589/3 filter paper to completely remove the activated carbon, and the color was measured again.

RESULTS AND DISCUSSIONS

The extent of hydrolysis is defined as the moles of reducing ends produced divided by the total moles of glucose units that would have been obtained from complete hydrolysis of the initial starch.

The reproducibility of extent of hydrolysis in the production of maltodextrins, in 45 min liquefaction, was checked by three experiments, one with two parallels and the other two with three parallels each, resulting in a standard deviation of 0.009 fractional extent of hydrolysis value, or 9.6% variation. The reproducibility of the extent of hydrolysis in 60 min liquefaction time was checked by five experiments, all of which with three parallels, and the standard deviation was found to be 0.007 fractional extent of hydrolysis value, or 5.4% variation.

The results on the extent of hydrolysis values are given (Fig. 1) along with the results of the previous study (8) for comparison. The extent of hydrolysis increased with time, as expected. The results are in agreement with those of (8) when the differences in the hydrolysis behavior of freshly prepared wheat

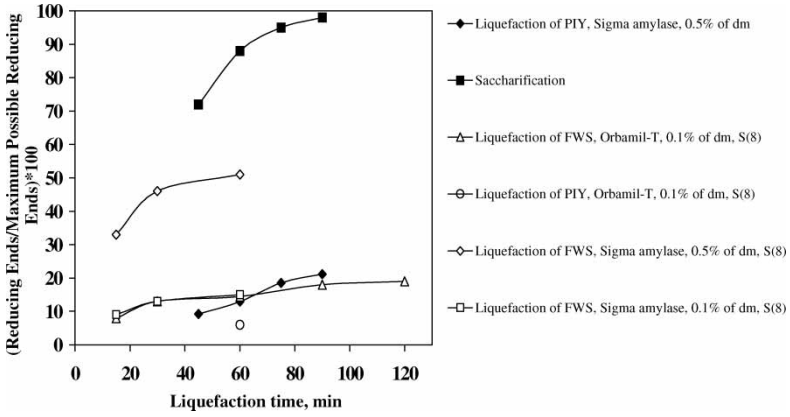

Figure 1. Percentage conversion in the production of maltodextrins and glucose syrups at 27% (w/w) wheat starch.

starch (FWS) and the commercial wheat starch (PIY) are taken into account. As indicated by their results at the enzyme dosage, 0.1% of dry matter, the catalytic effects of Sigma and Orba α-amylases were the same, and the extent of hydrolysis in FWS was much higher than that in PIY. When the enzyme amount increased five times, to 0.5% of dry matter, for the hydrolysis of FWS by Sigma amylase, extent of hydrolysis increased nearly 3.5 times (8). The result of the present work on hydrolysis of PIY for 60 min with Sigma α-amylase, was compared to the one obtained by (8), and it was seen that the conversion increased 2.2 times, due to difference in susceptibility of PIY and FWS to hydrolysis.

The percentage overall extent of hydrolysis as the maltodextrins were saccharified to glucose syrups (Fig. 1) had values between 72–98 for syrup liquefaction times of 45–90 min, as expected.

The final color of glucose syrups (Fig. 2) increased with increasing liquefaction times, as was also expected, since the Maillard reaction starts between reducing ends and amino acids. The highest level of color was obtained in the glucose syrups produced at 90 min liquefaction time as 1424 ICUMSA units and the lowest level was obtained as 657 ICUMSA units in glucose syrups produced at 45 min liquefaction time.

Reproducibility of decolorization percentage of glucose syrups, obtained by 60 min liquefaction time, at 0.5% NORIT was checked by six experiments, and the standard deviation was found to be 0.76% decolorization percentage, or 1.0% variation.

When glucose syrups that were produced at different liquefaction times were decolorized with 0.25–1 g/100 g of NORIT, the results given in Figs. 3 and 4 were obtained. The plots of color removal per dosage of carbon against residual color, or the adsorption isotherms, would have coincided

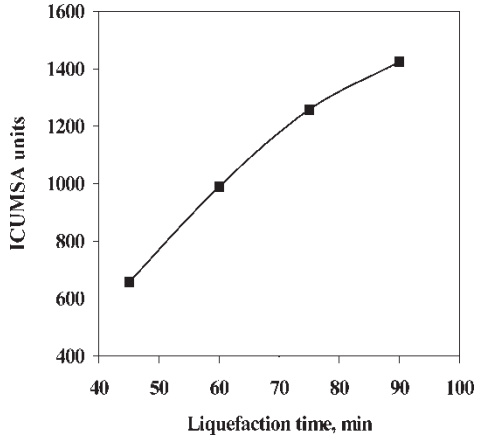

Figure 2. Color of glucose syrups after saccharification.

with each other if the color substances in all syrups were similar. The isotherms in Fig. 3, however, were quite different from each other, even those of the glucose syrups produced at 75 and 90 min liquefaction time which had similar conversion values, were distinct from each other.

The Freundlich isotherm, empirical but very useful in calculations involving activated carbon, is described by the equation (31, 35)

$$X/M = K * C^{1/n} \tag{2}$$

where in the present case X/M, is the amount of adsorbate held per g of activated carbon, is calculated as the difference between the initial and residual levels of color ($C_0 - C$) in ICUMSA units divided by the carbon

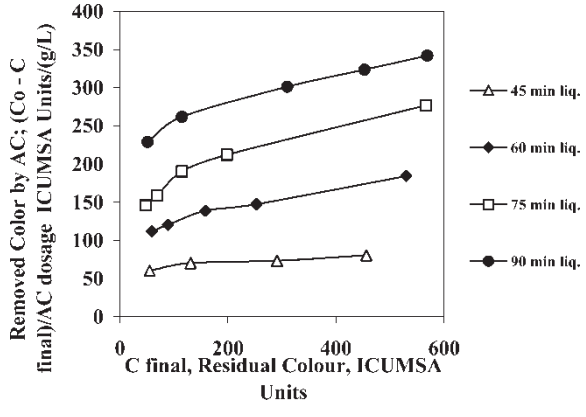

Figure 3. Glucose syrups decolorization isotherms for NORIT.

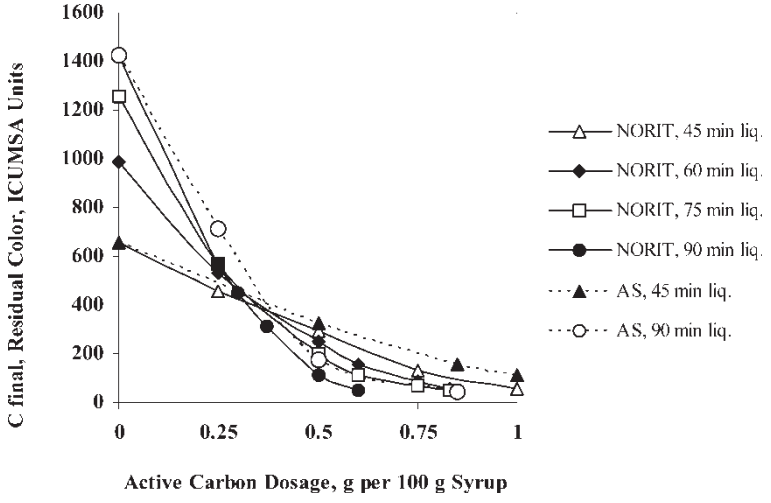

Figure 4. Residual color of glucose syrups after decolorization by NORIT and AS.

dosage in g/L, C is the concentration of adsorbate, i.e. the residual colour at equilibrium in ICUMSA units, and K and n are constants.

All isotherms fitted quite well to the Freundlich equation, and the resulting K, 1/n values and regression coefficients (36) are given in Table 2. In Fig. 4, the residual color units were plotted against the carbon dosage. The residual color remained higher for syrups liquefied for longer times up to about 0.3% dosage, at which all syrups had about 400 ICUMSA units of color. At higher carbon dosages, the level of residual color decreased as liquefaction time increased.

From these results, it was concluded that colored substances produced at different liquefaction times are different from each other in structure, being probably due the complex mechanism of the Maillard reaction, which is beyond of the scope of the present study. But it has been reported by

Table 2. The Freundlich isotherm parameters, K, 1/n and R^2 values

	NORIT			AS		
Liq. time, min	K[a]	1/n	R^2	K	1/n	R^2
Type of activated carbon						
45	36.14	0.13	0.96	21.69	0.20	0.98
60	41.82	0.23	0.98	—	—	—
75	53.58	0.26	0.99	—	—	—
90	120.23	0.16	0.99	81.28	0.20	0.91

[a]Units of K values. $(\text{ICUMSA Units})^{1-(1/n)}\ \text{L g}^{-1}$.

several authors (14, 15, 20) that during Maillard reaction, the molecular weight of colored substances increase due to polymerization reactions as time proceeds. As mentioned before, little information is available about the adsorption of a wide variety of specific materials by activated carbon (25) and it is known that dissimilar compounds have different adsorption affinity due to their different molecular weights and structures (24). Higher molecular weight organics have been reported (24, 25, 27–29) to have higher affinity for activated carbon surfaces, and the present results of lower residual color levels at the same carbon dosage in decolorization of more densely colored syrups is in agreement with these reports.

As has been mentioned before, commercial glucose syrups should have color not exceeding 200 ICUMSA units, and a typical production process target lower values such as 100 to be on the safe side. The NORIT dosages required to reduce the color of syrups liquefied for 45, 60, 75, and 90 min to 100 ICUMSA units are, 0.82%, 0.73%, 0.65%, and 0.53% respectively. The decolorization performances of the agricultural waste based activated carbons were compared with NORIT on this basis. The dosage of activated carbon was adjusted to be the same as that of NORIT required for 100 ICUMSA residual color and the residual color values were determined (Fig. 5). Among the samples studied with syrups liquefied for 90 min., HH was the best, giving a residual color of 101 ICUMSA, practically the same as NORIT. Although HH has large BET surface area and high mesopore volume, its ash content and surface charge might have resulted in lower performance. HS and AS, both of which have considerably smaller mesopore

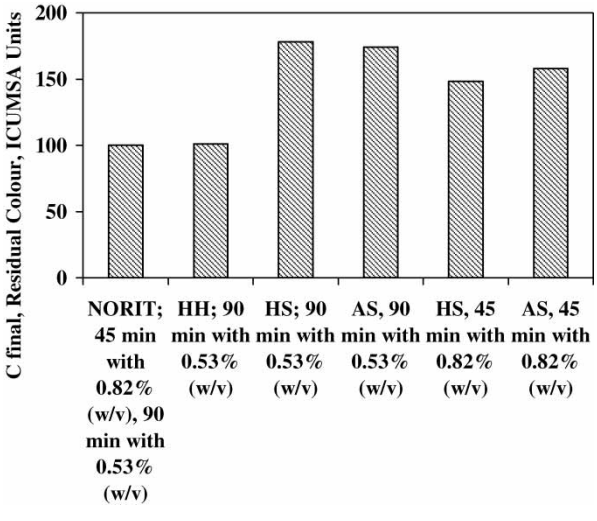

Figure 5. Comparison of performance of activated carbons with that of NORIT at the dosage required by NORIT to reduce the residual color to 100 ICUMSA units.

volume than HH, had very similar performances, reducing the color to less than 180 ICUMSA units for syrups liquefied both for 45 min and 90 min. A target of 100 units would require somewhat higher amounts.

The decolorization by AS being slightly better than HS for decolorization of glucose syrups produced at 90 min liquefaction time, the reverse being the case for the decolorization of glucose syrups produced at 45 min liquefaction time do not seem to have statistical significance. The decolorization of 45 min and 90 min liquefied syrups with different amounts of AS dosage was also carried out, and the shapes of the AS isotherms (Fig. 4) were found to be similar to those observed with NORIT. The numerical values of the Freundlich isotherm parameters for AS (Table 2) were also found to be quite similar to those of NORIT.

CONCLUSIONS

When starch is hydrolyzed at different liquefaction times to give maltodextrin syrups and then for 18 h to give glucose syrups, it was seen that an increase in the liquefaction time resulted in higher conversion values of both products. As expected, color levels of glucose syrups also increased when liquefaction time was increased. Glucose syrups that were produced at 90 min liquefaction time, which had the highest level of color, however, required the smallest amount of activated carbon, for residual color lower than 100 ICUMSA units with all types of activated carbons. This may be due to the increased molecular weight of colored substances during the Maillard reaction, for which the activated carbons were expected have increased affinity. When the adsorption capacities of the commercial product, NORIT, and the hazelnut or apricot stone based activated carbons were compared, it was seen that hazelnut husk based carbons were as good as NORIT, while the adsorption capacities of apricot stone and hazelnut shell based carbons were similar to each other and somewhat less than those of NORIT and hazelnut husk based carbons. It is concluded that agricultural waste based activated carbons are potential alternatives to commercial ones for the decolorization of glucose syrups.

REFERENCES

1. Storz, E. and Steffens, K.J. (2004) Feasibility study for determination of dextrose equivalent (DE) of starch hydrolysis products with near-infrared spectroscopy (NIRS). *Starch/Starke.*, 56 (5): 58–62.
2. Kearsley, M.W. and Dziedzic, S.Z. (1995) *Handbook of Starch Hydrolysis Products and Their Derivatives*; Chapman Hill: London, UK.
3. Kearsley, M.W. and Birch, G.G. (1985) The chemistry and metabolism of starch based sweeteners. *Food Chemistry*, 16: 191–207.
4. Paolucci-Jeanjean, D. et al. (1999) Why on earth can people need continuous recycle membrane reactors for starch hydrolysis? *Starch/Starke.*, 51: 25–32.

5. Baltes, W. (1981) Chemical changes in food by Maillard reaction. *Food Chemistry*, 9: 59–73.
6. Göksungur, Y. and Güvenç, U. (1994) Enzymatic hydrolysis and production of ethanol from potato starch. *Gýda.*, 2: 89–92.
7. Godon, B. (1994) *Bioconversion of Cereal Products*; VCH Publishers: New York.
8. Sakintuna, B. (2001) *Enzymatic Hydrolysis Wheat Starch*; Master's Thesis. METU, Ankara, Turkey.
9. Sakintuna, B. et al. (2003) Hydrolysis of wheat starch fractions and commercial wheat starch using α-amylase. *Chemical Engineering Communications*, 190: 883–897.
10. Akberg, C., Zacchi, G., Torto, N., and Gorton, L. (2000) A kinetic model for enzymatic wheat starch saccharification. *Journal of Chemical Technology & Biotechnology*, 75: 306–314.
11. Anon (1994) ICUMSA Colour Determination Method, Method 2; International Commission for Uniform Methods of Sugar Analysis.
12. Bostan, A. and Boyacioğlu, D. (1997) Kinetics of non-enzymatic colour development in glucose syrups during storage. *Food Chemistry*, 60 (4): 581–585.
13. Kroh, L.W. (1994) Caramelisation in food and beverages. *Food Chemistry*, 51: 373–379.
14. Coca, M., Garcia, M.T., Gonzales, G., Pena, M., and Garcia, J.A. (2004) Study of coloured compounds formed in sugar beet processing. *Food Chemistry*, 86 (3): 421–433.
15. Martins, S.I.F.S., Jongen, W.M.F., and van Boekel, M.A.J.S. (2001) A review of Maillard reaction in food and implications to kinetic modelling. *Trends in Food Science and Technology*, 11: 364–373.
16. Morales, F.J. and van Boekel, M. A. J. S. (1999) A study on advanced Maillard reaction in heated casein/sugar solutions: Colour formation. *International Dairy Journal*, 8: 907–915.
17. Carabasa-Giribet, M. and Ibaraz-Ribas, A. (2000) Kinetics of colour development in aqueous glucose systems at high temperatures. *Journal of Food Engineering*, 44: 181–189.
18. Göğüş, F., Bozkurt, H., and Eren, S. (1997) Kinetics of Maillard reactions between the major sugars and amino acids of boiled grape juice. *Lebensm-Wiss. U. Technol.*, 31: 196–200.
19. Buedo, A.P., Elustondo, M.P., and Urbicain, M.J. (2001) Non-enzymatic browning of peach juice concentrate during storage. *Innovative Food Science and Emerging Technologies*, 1: 255–260.
20. Mauron, J. (1981) The Maillard reaction in food; A critical review from the nutritional standpoint. *Progress in Food and Nutritional Science*, 5: 5–35.
21. Whistler, R.L., Paschall, E.F., and Bemiller, W. (1984) *Starch: Chemistry and Technology*; Academic Press: Orlando.
22. Atiyeh, H.K. and Duvnjak, Z. (2004) Purification of fructose syrups produced from cane molasses media using ultrafiltration membranes and activated carbon. *Separation Science and Technology*, 39 (2): 341–362.
23. Ahmedna, M., Marshall, W.E., and Rao, R.M. (2000) Surface properties of granular activated carbons from agricultural by-products and their effects on raw sugar decolourization. *Bioresource Technology*, 71: 103–112.
24. Hassler, J.W. (1963) *Active Carbon*; Chemical Publishing Company Inc.: New York.

25. Hutchins, R.A. (1994) Activated-carbon systems for separation of liquids. In *Handbook of Separation Techniques for Chemical Engineers*; Schweitzer, P.A. (ed.); Mc Graw-Hill: New York.
26. Mantell, C.L. (1946) *Industrial Carbon, Its Elemental, Adsorptive and Manufactured Forms*; D. VAN Nostrand: New York.
27. Smisek, M. and Cerny, S. (1970) *Activated Carbon Manufacture, Properties and Applications*; Elsevier: New York.
28. Mattson, J.S. and Mark, H.B. (1971) *Activated Carbon: Surface Chemistry and Adsorption from Solution*; Marcel Dekker: New York.
29. Cheremisinoff, P.N. and Ellerbusch, F. (1978) *Carbon Adsorption Handbook*; Ann Arbor Science Pub: Ann Arbor Mich.
30. Hassler, J.W. (1974) *Purification with Activated Carbon*; Chemical Publishing Company: New York.
31. Mudoga, H. (2002) *A Study on the Use of Activated Carbon to Remove Colour in Sugar*; Master's Thesis. METU, Ankara, Turkey.
32. Çuhadar, Ç. (2005) *Production and Characterization of Activated Carbon from Hazelnut Shell and Hazelnut Husk*; Master's Thesis. METU, Ankara, Turkey.
33. Yağşi, N.U. (2004) *Production and Characterization of Activated Carbon from Apricot Stones*; Master's Thesis, METU, Ankara, Turkey.
34. Somogyi, M.J. (1952) *Biol. Chem.*, 195: 19–23.
35. Geankoplis, C.J. (1983) *Transport Processes and Unit Operations*; Allyn and Bacon: Boston.
36. Box, G.E.P., Hunter, W.G., and Hunter, J.S. (1978) *Statistics for Experimenters, An Introduction to design, Data Analysis and Model Building*; John Wiley & Sons: New Jersey.