Journal of Agricultural and Food Chemistry

APRIL 1991
VOLUME 39, NUMBER 4

Oxidation of Methyl Linoleate Encapsulated in Amorphous Lactose-Based Food Model

Yasunobu Shimada,† Yrjö Roos, and Marcus Karel*

Department of Food Science and Center for Advanced Food Technology, Rutgers University, P.O. Box 231, New Brunswick, New Jersey 08903

A food model containing methyl linoleate encapsulated in an amorphous lactose–gelatin matrix was used to study the effect of physical changes in the amorphous matrix on the rate of lipid oxidation. The glass transition temperature (T_g) and crystallization behavior of the material were determined by using differential scanning calorimetry. The rate of oxidation was measured spectrophotometrically in samples incubated at various temperatures above T_g. The encapsulated oil was released as a consequence of crystallization of the amorphous lactose. The released oil underwent rapid oxidation, while encapsulated oil remained unoxidized. Oxidation of encapsulated fat in dried foods is related to physical changes in the amorphous matrix. The measurement of T_g is important in predicting the onset of deteriorative reactions in foods and should be considered in accelerated shelf life tests.

INTRODUCTION

Dehydration of biological materials often leads to formation of amorphous structures which show thermal and physical properties typical of amorphous materials (White and Cakebread, 1966; Struik, 1978; Roos and Karel, 1991). Retention of volatile compounds, flavors, and oils in drying is based on their encapsulation in the amorphous matrix formed during the dehydration process (To and Flink, 1978). However, amorphous materials undergo a structural change from a "glass" to a "rubber" at the glass transition temperature (T_g) which is specific for each material. Amorphous materials are also metastable, and above T_g various time-dependent changes in the physical properties can be observed (Tant and Wilkes, 1981).

Flink and Karel (1972) showed that mobility of volatiles in freeze-dried carbohydrate structures is related to water content through time-dependent changes in the dried amorphous matrix. Retention of encapsulated volatiles and oxidation in freeze- and spray-dried materials have been related to the collapse temperature (T_c) of the dried matrix and crystallization of amorphous compounds (Flink and Karel, 1972; Chirife and Karel, 1974; Tsourouflis et al., 1976; Gejl-Hansen and Flink, 1977; Omatete and King, 1978; To and Flink, 1978). Roos and Karel (1991) showed that the T_g of amorphous sugars defines their T_c, sticky point (T_s), and the rate of crystallization. In biological and food materials changes in the physical structure at T_g may lead to increased reaction rates and decreased stability. Most dehydrated materials are also extremely hygroscopic, and water plasticization decreases T_g values (Roos, 1987; Slade et al., 1989; Roos and Karel, 1991). Thus, the physical properties are controlled by temperature, moisture content, and time (Roos and Karel, 1991).

Transport mechanisms in amorphous polymers and also in dried biological materials are related to their physical state (Thijssen, 1971; Alexander and King, 1985). Increased permeability and diffusivity of gases above T_g may affect reaction rates and the stability of amorphous foods. The rate of oxidation in dried fat containing food materials is also related to the structural changes in the amorphous matrix and crystallization (Gejl-Hansen and Flink, 1977; To and Flink, 1978). The purpose of this study was to investigate factors leading to oxidation of methyl linoleate encapsulated in a model food powder and to determine the effect of the physical state of the amorphous matrix on the oxidation rate.

† Present address: Morinaga Milk Industry Co., Ltd., Food Research and Development Laboratory, No. 1-83, 5-Chome Higashihara, Zama-City, Kanagawa-Pref., Japan 228.

0021-8561/91/1439-0637$02.50/0

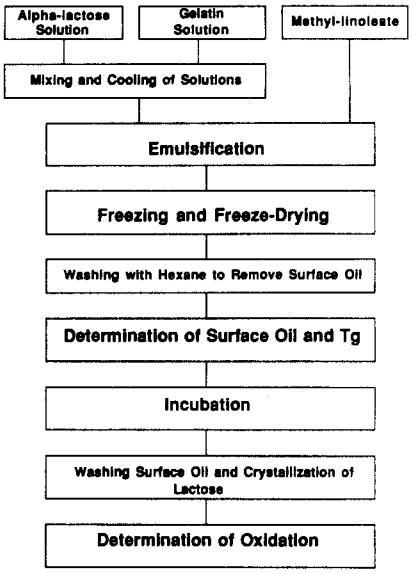

Figure 1. Experimental design for preparation of a food model containing encapsulated methyl linoleate in amorphous lactose–gelatin matrix and determination of the effect of the physical state and temperature on oxidation rate.

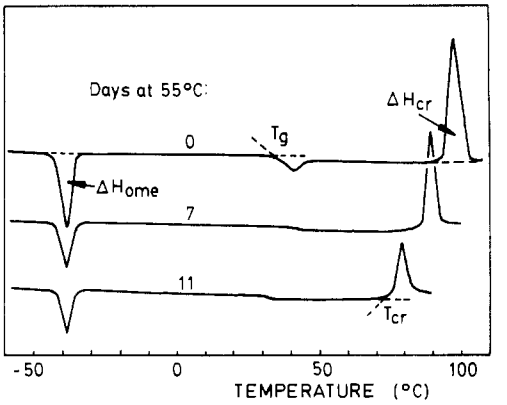

Figure 2. Determination of encapsulated oil content from differential scanning calorimetry data and the effect of partial crystallization of amorphous lactose on glass transition temperature (T_g), crystallization temperature (T_{cr}), and latent heat of crystallization (ΔH_{cr}). Methyl linoleate was found to melt at −43 °C, and encapsulated oil content was obtained by integration of the oil melting endotherm which gave the latent heat of melting (ΔH_{ome}) for encapsulated oil. The encapsulated oil content was obtained by dividing ΔH_{ome} by latent heat of melting of the pure compound (ΔH_{om}). T_{cr} decreased with time because initial crystallization had occurred during incubation, which was also shown by decreased ΔH_{cr} indicating less amorphous lactose per gram of sample.

MATERIALS AND METHODS

Experimental Design. A model system composed of lactose, oil (methyl linoleate), and protein (gelatin) typical of dried fat containing food powders (e.g., milk or baby food powders and coffee whiteners) was developed. The experimental design for the determination of oxidation in this model is shown in Figure 1. Other models with varying lactose, gelatin, and oil contents were tested. Gelatin was found to be necessary for emulsification and encapsulation, and it could not be replaced by maltodextrin or emulsifiers because of poor emulsification or encapsulation, respectively. The model chosen showed thermal behavior similar to that of lactose (Roos and Karel, 1990), and it was considered as most suitable for study of the effect of temperature, moisture content, and time-dependent changes in the physical state on oxidation.

Encapsulation of Methyl Linoleate. α-Lactose (Fisher) was dissolved in HPLC water (Fisher) with slight heating until a clear solution (21.9% lactose) was obtained. Gelatin (Sigma, type A, 60 bloom) was dissolved in boiling water (4.8% solution), and the solutions were mixed and cooled in an ice bath. Methyl linoleate (Sigma) was emulsified in this solution (10 min/50 g, Brinkmann homogenizer Model PT 10/35) in an ice bath. The approximate size of the emulsion oil droplets determined microscopically was 5–15 μm. The emulsion contained 20% "solids" (including methyl linoleate). The solids were composed of 5% gelatin, 25% methyl linoleate, and 70% lactose. The emulsion was frozen (−30 °C overnight, dry ice 3 h) and freeze-dried (Virtis Benchtop 3L, $p < 0.1$ mbar) in open Petri dishes (60-mm dish, 10 mL of emulsion). The dried emulsion was further dehydrated and stored over P_2O_5 at room temperature.

For oxidation experiments, the dried emulsions were broken into powder by use of a mortar and pestle and subsequently washed with hexane (HPLC grade, Fisher) to remove nonencapsulated surface oil. In all washing treatments the powder and hexane were separated in a glass funnel by filtration through filter paper (Whatman No. 1 or 2, washed with hexane before use). Total removal of surface oil was confirmed by additional washing until no absorbance at 234 nm was observed in the extract. This was done to avoid contamination due to oxidation of surface oil. The sample preparation was done at a relative humidity below 40% to keep the samples in their glassy state before the oxidation studies. The amount of washed out oil was determined by weighing the extract after the hexane was evaporated.

Differential Scanning Calorimetry. Differential scanning calorimetry (DSC) was used to analyze the amount of methyl linoleate in the dried samples, their glass transition temperature (T_g, onset of the endothermal change of specific heat), and crystallization behavior (T_{cr}, onset of crystallization exotherm; ΔH_{cr}, latent heat of crystallization) of amorphous lactose. Time to crystallization (θ_{cr}) was determined by using isothermal DSC as reported by Roos and Karel (1990). The instrument used was a Mettler TA 4000 thermal analysis system with TC 11 TA processor, DSC 30S measuring cell and TA 72PS.1 software calibrated for low-temperature operation as reported by Roos (1987). A scanning rate of 5 °C/min was used in all measurements with an empty aluminum pan (Mettler, 40 μL) as reference.

The amount of encapsulated methyl linoleate was analyzed by weighing 5–10 mg of the material in the DSC pan, and the sample was scanned from −100 to 0 °C (Figure 2). The melting peak was integrated to obtain the latent heat of melting for the methyl linoleate in the sample. This heat was divided by the latent heat of melting determined for pure methyl linoleate (134 J/g), which allowed calculation of the amount of encapsulated methyl linoleate in the sample.

T_g was determined for samples dried over P_2O_5 and for samples rehumidified over saturated LiCl, CH_3COOK, $MgCl_2$, and K_2CO_3 solutions (relative humidities 0%, 11%, 23%, 33%, and 43%, respectively) in triplicate by scanning the samples over the glass transition region until crystallization of lactose was completed. T_g and T_{cr} of samples incubated for the determination of oxidation rate were determined to record possible loss of moisture (increased T_g) and crystallization (decreased T_{cr} and ΔH_{cr}) of lactose during incubation. This procedure was necessary to avoid experimental errors caused by variation of the T_g during incubation. The rehumidified moisture content of the samples was obtained by determining weight gain during equilibration over saturated salt solutions as reported by Roos (1987).

Determination of Oxidation. Oxidation of methyl linoleate leads to conjugation of the double bonds, which show increased absorbance at 234 nm. Oxidation was followed spectrophotometrically (Hewlett-Packard, 8452A diode array) in hexane (Fisher, HPLC grade) by determining absorbance at 234 nm. All measurements were made by using a dilution of 0.05% methyl linoleate in hexane. The extinction coefficient of hydroperoxides formed during oxidation of methyl linoleate has been reported to be 29 000 mol^{-1}/L (OD_p) (Privett and Blank, 1962). The extinction coefficient of the unoxidized methyl linoleate (OD_m) used was 83 mol^{-1}/L. This allowed calculation of the amount of oxidized methyl linoleate using eq 1. Since the extinction coefficient of methyl linoleate was small compared to 29 000 mol^{-1}/L, the term (C_mOD_m) could be ignored, and eq 2 was used to calculate percentage of oxidation. (Terms are defined at the end of the text.)

Incubation of Samples. The T_g of the model was close to that of pure lactose. Roos and Karel (1990) showed that raising

$$C_{ox} = \frac{AM}{(C_m OD_p) - (C_m OD_m)} \times 100\% \quad (1)$$

$$C_{ox} = \frac{AM}{C_m OD_p} \times 100\% \quad (2)$$

the temperature of amorphous lactose to about T_g + 50 °C caused instantaneous crystallization, and at temperatures between T_g and T_g + 50 °C crystallization was time-dependent. To study time-dependent changes in oxidation affected by time-dependent changes in the structure of the matrix, it was desirable therefore to incubate samples in the range from T_g to T_g + 50 °C. T_g could be varied by changing the moisture content. Preliminary experiments showed that temperatures over 100 °C caused dehydration, browning, and decomposition. To keep T_g + 50 °C below 100 °C, the rate of oxidation was studied in samples rehumidified over $MgCl_2$ (33% relative humidity) which had the T_g slightly above room temperature at 31 °C and T_g + 50 °C at 81 °C. This allowed handling of samples below their T_g at room temperature and low relative humidities and incubation of samples at different $T - T_g$ values without substantial browning and decomposition. The samples (0.1 g in 22.2-mL vial) were incubated over saturated $MgCl_2$ solution at 37, 45, 50, 55, 70, and 80 °C in vacuum desiccators with respective $T - T_g$ values of 6, 14, 19, 24, 39, and 49 °C. Reference samples were kept over saturated $MgCl_2$ solution at room temperature. During incubation at 70 and 80 °C a small vial (2 mL) containing saturated $MgCl_2$ solution was kept in the sample vial to maintain constant humidity. θ_{cr} was determined for samples rehumidified at 33% relative humidity, and the WLF equation (Williams et al., 1955) was used to predict θ_{cr} of the model as shown in Figure 6 and reported by Roos and Karel (1990).

Determination of Oxidation Rate. One sample incubated at each of the temperatures 37, 45, 50, and 55 °C was washed daily with 20 mL of hexane, and the absorbance at 234 nm was measured. Samples incubated at 70 and 80 °C were analyzed at varying intervals of 30–60 min. After the samples were washed, residual hexane was removed in a vacuum desiccator, and the samples were placed in a vacuum desiccator over saturated KCl solution (84% relative humidity) for 90 min at 25–30 °C. This procedure led to rapid crystallization of amorphous lactose and thus to almost complete release of the encapsulated methyl linoleate. After lactose crystallization, the samples were washed with 20 mL of hexane, and the absorbance of the released oil was measured in this solution. Ten-milliliter aliquots of the solutions containing surface oil and encapsulated oil respectively were evaporated from a 50-mL beaker, and the amount of extracted oil was quantified by weighing. The rate of oxidation for encapsulated oil was compared with the rate of oxidation for nonencapsulated methyl linoleate spread on filter paper with hexane in which case all of the oil had the characteristics of surface oil. The oxidation studies were repeated three times at each temperature.

RESULTS

Encapsulation of Methyl Linoleate. The freeze-dried material was found to contain 4–7% methyl linoleate on the basis of total solids which could be washed out with hexane and was considered as surface oil. The amount of methyl linoleate remaining in the sample after washing was determined by using DSC (Figure 2), and it was found to be approximately 20% of total solids. This was considered as encapsulated oil.

Glass Transition Temperature of the Model. The T_g of the model used was close to that of pure lactose. The effects of the presence of gelatin and of oil on T_g of this model were slight. The T_g of the model as a function of moisture content is shown in Figure 3. Instant crystallization of the amorphous lactose matrix was found to occur at about T_g + 65 °C (Figure 2). However, crystallization could occur at any $T - T_g$ value as a function of time. Transient exposure to high humidity prior to incubation was found to introduce errors because it could cause nucleation and subsequent acceleration of lactose crystallization.

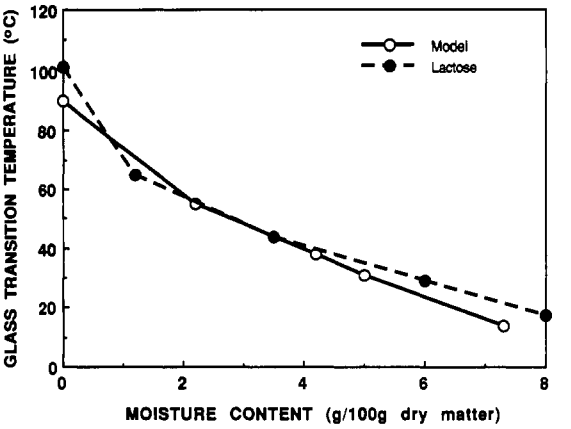

Figure 3. Glass transition temperature of model food containing encapsulated methyl linoleate in amorphous lactose–gelatin matrix and glass transition temperature of amorphous lactose as function of moisture content. The glass transition temperatures and moisture contents for lactose are from Roos and Karel (1991).

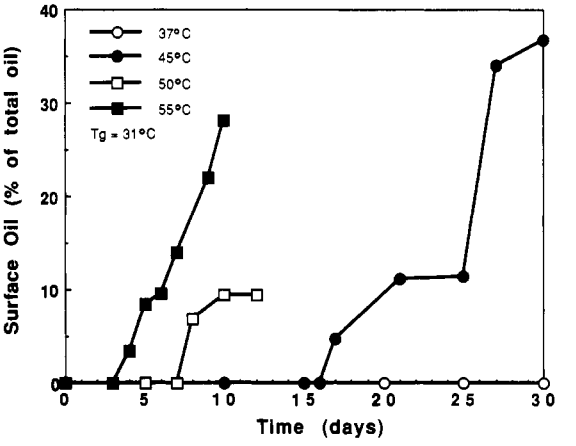

Figure 4. Amount of surface oil of amorphous lactose–gelatin matrix containing encapsulated methyl linoleate as a function of incubation time at varying temperatures above glass transition temperature (T_g). Time to increased surface oil depends on the temperature difference to glass transition temperature ($T - T_g$).

Release of Encapsulated Methyl Linoleate. Time to appearance of surface oil was found to depend on the incubation temperature and to decrease with increasing temperature. As increased amounts of surface oil were detected, the thermograms also showed decreased T_{cr} and ΔH_{cr} values (Figure 2). Thermograms of samples with high surface oil contents showed neither glass transition nor crystallization, and they were considered to be completely crystalline. The amount of surface oil as a function of incubation time at various temperatures is shown in Figure 4.

Oxidation of Methyl Linoleate. The amounts of hydroperoxides in surface and in encapsulated oil as a function of incubation time at various temperatures are shown in Figure 5. Samples incubated at 37 °C did not show oxidation within 40 days. At higher temperatures the increase in surface oil was associated with a rapid increase in absorbance of the extracted oil. Time to increased surface oil was compared with θ_{cr} and θ_{cr} of lactose (Figure 6). Time to increased surface oil correlated with θ_{cr} at high $T - T_g$ values. At low $T - T_g$ values surface oil occurred before the predicted time to crystallization.

DISCUSSION

Changes in structure of metastable amorphous foods may affect rates of physical and chemical deterioration.

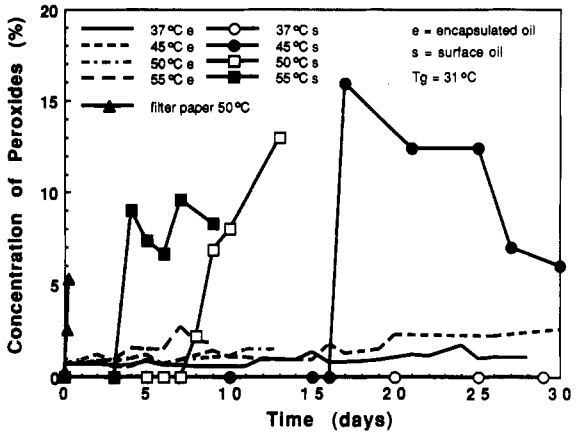

Figure 5. Amount of hydroperoxides as percentage of surface oil and encapsulated oil detected in amorphous lactose–gelatin matrix containing encapsulated methyl linoleate incubated at various temperatures above glass transition temperature (T_g) as a function of incubation time.

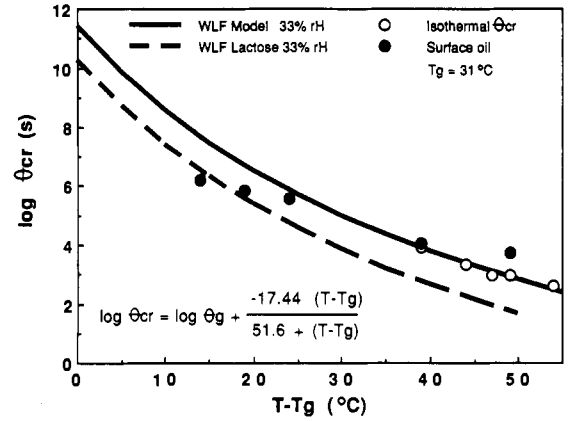

Figure 6. Time to increased amount of hydroperoxides in surface oil of amorphous lactose containing encapsulated methyl linoleate incubated at various temperatures above glass transition temperature (T_g). The appearance of surface oil was related to the crystallization of amorphous lactose which shows WLF type temperature dependence. Time to crystallization in the model food is longer than that of lactose because gelatin delays crystallization and increases time to crystallization.

Among dry foods having a structure containing oxidizable lipids in a lactose–protein amorphous matrix, whole dry milk is an important example (Tamsma and Pallansch, 1964; Sharp and Doob, 1941; Bushill et al., 1965; Raemy et al., 1983). It is known that temperature and moisture content, which control glass transition, affect oxidative stability, but rates of oxidation have not been related to that transition. It is known, however, that diffusion is greatly affected by structural changes which are now known to be related to glass transition (Thijssen, 1971; Omatete and King, 1978; To and Flink, 1978; Rosenberg et al., 1990; Roos and Karel, 1991).

Roos and Karel (1990) used the WLF equation (Williams et al., 1955) to predict time to crystallization of amorphous lactose above its T_g, and a good correlation between time to crystallization and $T - T_g$ was reported. In the present study, we compared time to onset of appearance of surface oil with time to crystallization predicted by the WLF equation (Figure 6). Time to crystallization and appearance of surface oil correlated well at high $T - T_g$ values. At low $T - T_g$ values time to appearance of surface oil was shorter than the time to crystallization predicted by the WLF equation. One reason may be sample preparation, which was found to be critical to the time for appearance of surface oil. This could be due to changes in T_{cr}, which had decreased values if nucleation had occurred. The thermograms of incubated samples showed decrease in T_{cr} and ΔH_{cr} values, indicating incipient crystallization before increased surface oil was noticed (Figure 2). One reason for the longer time to crystallization and to appearance of surface oil than to onset of crystallization predicted from studies on pure lactose (Figure 6) may be the presence of gelatin. We have shown that small amounts of added polymers result in little change in T_g of sucrose but significantly raise T_{cr} (Roos and Karel, 1991). We believe therefore that the main mechanism of oxidation of encapsulated lipid was the release of encapsulated oil, coincident with crystallization of lactose and the resulting exposure to oxygen.

Gejl-Hansen and Flink (1977) and To and Flink (1978) showed that by preparing an amorphous carbohydrate powder containing encapsulated oxidizable lipid and washing away surface oil with hexane a stable nonoxidizing system could be obtained. Exposure to temperatures causing structural collapse (above collapse temperature, T_c) resulted in rapid oxidation of the lipid. The results of the present study show that the release of encapsulated oil and its oxidation are correlated with the onset of crystallization of lactose. Roos and Karel (1991) have shown that crystallization of amorphous sugars occurs above their T_g. At all moisture contents the rate of crystallization was reported to be uniquely determined by the quantity of $T - T_g$. Therefore, oxidation of encapsulated oil is related to the T_g of the amorphous matrix, and the rate of oxidation is determined by both temperature and moisture content.

It is possible that at low $T - T_g$ values oxygen diffusion through the amorphous matrix may lead to some oxidation of encapsulated oils before they are released and become accessible to hexane. In the model studied the time required for oxygen diffusion seems to be substantially longer than the time to crystallization at high $T - T_g$ values. After the initial increase of surface oil and oxidation, the oxidation rate was decreased (Figure 5), probably because of dilution due to the release of additional as yet unoxidized encapsulated oil. The oil released in the early stages of incubation may also temporarily act as a protective barrier, slowing oxidation of oil released later.

Exceeding of T_g and subsequent crystallization of amorphous carbohydrates in food powders may also affect other deteriorative reactions. Saltmarch et al. (1981) showed that browning of spray-dried whey has its maximum rate at the onset of substantial lactose crystallization. When the water liberated by crystallization is absorbed by constant humidity solutions, the moisture content of samples is decreased, and it can be expected that the rate of browning will decrease. However, during storage of food powders in closed containers browning increases the moisture available for plasticization and may increase rates of reactions and crystallization. These factors should be considered in the design of accelerated shelf life tests.

The results of this study show that time-dependent changes in metastable physical state have profound effects on reaction rates, and the changes are sensitive to temperature and moisture content. Rates of some deteriorative changes are governed by WLF kinetics and show exponential increase above T_g. During food processing high humidities may substantially alter rates of crystallization due to nucleation and cause decreased shelf life. The results of this study can be used to evaluate oxidation and release of encapsulated compounds in amorphous food and related materials and to evaluate the importance of physical changes on processing and shelf life of dried foods.

ABBREVIATIONS USED

A, absorbance at 234 nm; C_m, concentration of methyl linoleate (g/L); C_{ox}, concentration of hydroperoxides (%); ΔH_{om}, latent heat of melting methyl linoleate; ΔH_{ome}, latent heat of melting encapsulated methyl linoleate; ΔH_{cr}, latent heat of crystallization; M, molecular weight (g/mol); OD_m, extinction coefficient of methyl linoleate (83 mol^{-1}/L); OD_p, extinction coefficient of hydroperoxides (29 000 mol^{-1}/L); T_c, collapse temperature; T_g, glass transition temperature; T_{cr}, crystallization temperature; T_s, sticky point; θ_{cr}, isothermal crystallization time; θ_g, isothermal crystallization time at glass transition temperature.

ACKNOWLEDGMENT

This is Publication No. D-10535-11-90 of the New Jersey Agricultural Experiment Station supported by Morinaga Milk Industry Co., Academy of Finland, by the State of New Jersey Funds, and by the Center for Advanced Food Technology. The Center for Advanced Food Technology is a New Jersey Commission on Science and Technology Center.

LITERATURE CITED

Alexander, K.; King, C. J. Factors governing surface morphology of spray-dried amorphous substances. *Drying Technol.* **1985**, *3*, 321–348.

Bushill, J. H.; Wright, W. B.; Fuller, C. H. F.; Bell, A. V. The crystallization of lactose with particular reference to its occurrence in milk powder. *J. Sci. Food Agric.* **1965**, *16*, 622–628.

Chirife, J.; Karel, M. Effect of structure disrupting treatments on volatile release from freeze-dried maltose. *J. Food Technol.* **1974**, *9*, 13–29.

Flink, J.; Karel, M. Mechanisms of retention of organic volatiles in freeze-dried systems. *J. Food Technol.* **1972**, *7*, 199–211.

Gejl-Hansen, F.; Flink, J. M. Freeze-dried carbohydrate containing oil-in-water emulsions: Microstructure and fat distribution. *J. Food Sci.* **1977**, *42*, 1049–1055.

Omatete, O.; King, C. J. Volatiles retention during rehumidification of freeze dried food models. *J. Food Technol.* **1978**, *13*, 265–280.

Privett, O. S.; Blank, M. L. The initial stages of autoxidation. *J. Am. Oil Chem. Soc.* **1962**, *39*, 465–469.

Raemy, A.; Hurrell, R. F.; Löliger, J. Thermal behavior of milk powders studied by differential thermal analysis and heat flow calorimetry. *Thermochim. Acta* **1983**, *65*, 81–92.

Roos, Y. H. Effect of moisture on the thermal behavior of strawberries studied using differential scanning calorimetry. *J. Food Sci.* **1987**, *52*, 146–149.

Roos, Y.; Karel, M. Differential scanning calorimetry study of phase transitions affecting quality of dehydrated materials. *Biotechnol. Prog.* **1990**, *6*, 159–163.

Roos, Y.; Karel, M. Plasticizing effect of water on thermal behavior and crystallization of amorphous food models. *J. Food Sci.* **1991**, *56*, 38–43.

Rosenberg, M.; Kopelman, I. J.; Talmon, Y. Factors affecting retention in spray-drying microencapsulation of volatile materials. *J. Agric. Food Chem.* **1990**, *38*, 1288–1294.

Saltmarch, M.; Vagnini-Ferrari, M.; Labuza, T. P. Theoretical basis and application of kinetics to browning in spray-dried whey food systems. *Prog. Food Nutr. Sci.* **1981**, *5*, 331–344.

Sharp, P. F.; Doob, H. Effect of humidity on moisutre content and forms of lactose in dried whey. *J. Dairy Sci.* **1941**, *24*, 679–690.

Slade, L.; Levine, H.; Finlay, J. W. Protein-water interactions: Water as a plasticizer of gluten and other protein polymers. In *Protein Quality and the Effects of Processing*; Phillips, R. D., Finlay, L. W., Eds.; Dekker: New York, 1989; pp 9–124.

Struik, L. C. E. *Physical aging in amorphous polymers and other materials*; Elsevier Scientific Publishing: Amsterdam, 1978; 229 pp.

Tamsma, A.; Pallansch, M. J. Factors related to the storage stability of foam-dried whole milk. IV. Effect of powder moisture content and in-pack oxygen at different storage temperatures. *J. Dairy Sci.* **1964**, *47*, 970–976.

Tant, M. R.; Wilkes, G. L. An overview of the nonequilibrium behavior of polymer glasses. *Polym. Eng. Sci.* **1981**, *21*, 874–895.

Thijssen, H. A. C. Flavour retention in drying preconcentrated food liquids. *J. Appl. Chem. Biotechnol.* **1971**, *21*, 372–377.

To, E. C.; Flink, J. M. "Collapse", a structural transition in freeze dried carbohydrates. III. Prerequisite of recrystallization. *J. Food Technol.* **1978**, *13*, 583–594.

Tsourouflis, S.; Flink, J. M.; Karel, M. Loss of structure in freeze-dried carbohydrates solutions: Effect of temperature, moisture content and composition. *J. Sci. Food Agric.* **1976**, *27*, 509–519.

White, G. W.; Cakebread, S. H. The glassy state in certain sugar-containing food products. *J. Food Technol.* **1966**, *1*, 73–82.

Williams, M. L.; Landel, R. F.; Ferry, J. D. The temperature dependence of relaxation mechanisms in amorphous polymers and other glass-forming liquids. *J. Am. Chem. Soc.* **1955**, *77*, 3701–3707.

Received for review August 17, 1990. Accepted November 6, 1990.

Registry No. Methyl linoleate, 112-63-0; lactose, 63-42-3.