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# Accelerated Shelf Life Testing of Whey-Protein-Coated Peanuts Analyzed by Static Headspace Gas Chromatography

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Four different formulations of whey-protein-based coatings were used to coat peanuts. Four controls were used to investigate the effects of different ingredients in the coating formulation on the peanut shelf life. Untreated peanuts were designated as the reference. The peanut samples were stored in duplicate at 40, 50, and 60 °C for storage durations of up to 31 weeks. The analysis of hexanal indicated that the coated samples were oxidized significantly slower than the reference; hence, the predicted shelf life was longer for the coated samples. However, the investigation of the control ingredients revealed that even when only water was applied onto the peanuts the oxidation was delayed.

KEYWORDS: Lipid oxidation; peanuts; whey protein; edible coatings; shelf life

#### INTRODUCTION

Peanuts are high in oil and fat consisting mainly of unsaturated fatty acid, providing high vulnerability to oxidative rancidity when combined with the high heat treatment of the roasting process (1-3). For roasted peanuts, autoxidation is the major cause of oxidative rancidity (4-6). Autoxidation in peanuts results in "flavor-fade" and off-flavor development, which may be due to the masking of pyrazines by large amounts of lowmolecular-weight aldehydes such as hexanal (7, 8). Hexanal, which is a major breakdown product of linoleic acid oxidation (9), has been shown to be a good indicator of oxidative rancidity in peanuts (10, 11).

Films based on heat-denatured whey-protein-isolate (WPI) have been found to be excellent oxygen barriers (12). Moreover, whey protein coatings applied by a bench-scale coating method have been shown to provide significant protection against oxidative rancidity in peanuts when analyzed by chemical and instrumental methods (10, 13). In our previous study (11), peanuts coated with WPI-based formulations using a commercial coating method showed significant reduction in oxidative rancidity, measured by both sensory rancidity rating and instrumental hexanal level, when compared with that of untreated peanuts. However, the control peanut sample, which was absent the film-forming coating material WPI, also exhibited protection against oxidative rancidity. Thus, the first objective of the present study was to investigate further the antioxidant effect of the ingredients in the control peanut sample of the previous study.

Shelf life of peanuts is influenced by innate factors such as maturity (14), fatty acid composition (15), and variety (16). Because the shelf life of peanuts is limited mainly by lipid oxidation, it can be significantly increased by special packaging techniques such as N<sub>2</sub> flushing, vacuum packaging, highoxygen-barrier packaging material, and oxygen-absorbing sachets or film ingredients. Shelf life of peanuts can also be enhanced by direct external treatments such as oxygen-barrier edible coatings. When conducting a shelf life test, the storage conditions may be modified to accelerate the reaction in order to expedite obtaining the results. When the temperature is raised to conduct accelerated shelf life testing in food systems, the results of such tests can be modeled using a temperature vs reaction rate relationship (17) to predict the shelf life at normal storage temperature. Thus, the second objective of this study was to determine the degree of WPI-coated and uncoated peanut oxidation at three accelerated shelf life test temperatures and then use this information to model the shelf life of these peanuts at ambient conditions. In this study, both the Arrhenius model (18) and the linear model (19) were used to predict the shelf life of peanut samples at ambient conditions.

## MATERIALS AND METHODS

**Raw Materials.** The variety of peanuts used for this experiment was "Runner". The peanuts contained 48-52% lipid, 22-30% protein, 3-5% sugar, and less than 2% moisture ( $a_w \approx 0.25$ ).

The whey protein coatings included WPI (Bipro, Davisco Foods International, Lesuer, MA), glycerol (USP/FCC, Fisher Scientific Inc., Fair Lawn, NJ) as a plasticizer, lecithin (Centrolene A, Central Soya Company, Fort Wayne, IN) as a surfactant, and methyl paraben (NF/ FCC, Fisher Scientific Inc., Fair Lawn, NJ) as an antimicrobial agent. Vitamin E (Nature's Life, Gardengrove, CA) was added to some of the coating formulations to test its antioxidant properties.

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Table 1.	Compositions	of the	Nine	Sample	Treatments
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sample treatment	composition
heat-denatured WPI with vitamin E	heat-denatured WPI + vitamin E + glycerol + lecithin + methyl paraben + water
heat-denatured WPI without vitamin E	heat-denatured WPI + glycerol + lecithin + methyl paraben + water
native WPI with vitamin E	native WPI + vitamin E + glycerol + lecithin + methyl paraben + water
native WPI without vitamin E	native WPI + glycerol + lecithin + methyl paraben + water
control 4	glycerol + lecithin + methyl paraben + water
control 3	lecithin + methyl paraben + water
control 2	methyl paraben + water
control 1	water
reference	untreated

Sample Treatments and Storage Conditions. Both native and heatdenatured WPI were included in the study because they produce films with different solubility, tensile strength, and oxygen-barrier properties (20, 21). Table 1 shows the nine sample treatments investigated in this study. All coating solutions contained 10% WPI (w/w). The heatdenatured solutions were prepared by heating 10% WPI solution (w/ w) for 30 min in a water bath at 90 °C (22). The denatured solutions were then cooled to room temperature (~25 °C) in an ice bath. Glycerol was added to all coating solutions at a 1 to 1 ratio of WPI to glycerol. Lecithin and methyl paraben were added to all coating solutions at 0.05% and 0.1% of the coating solution (w/w), respectively. When vitamin E was added, it was at 0.5% of the coating solution (w/w). The same amount of ingredient(s) was added to water (Arrowhead Mountain Spring Water Company, Brea, CA) to make each of the 4 controls. WPI-vitamin E emulsions were made using a Microfluidizer homogenizer (HC 5000, Microfluidics International Corp., Newbury, MA). The hot liquid was passed through the homogenizer 6 times using a homogenizing pressure of 6000 psig. The resulting emulsion had a normal particle-size distribution, and the mean particle size was approximately  $0.6 \,\mu\text{m}$ . After all the ingredients were mixed, the solution was strained with 2 layers of cheesecloth and stored for 1 to 3 d at a refrigeration temperature (5-10 °C) until the coating process took place.

A commercial coater (Labcoater II system, O'Hara Manufacturing, Ltd., Toronto, ON) was used to coat the peanuts with WPI solutions and control 4. This coater is a simultaneous spray-jog-dry type of coater which is mainly used to coat pharmaceuticals and nutritional supplements. For our peanut coating, we separated the spray phase and the dry phase of the process.

For the spray phase, the conditions were the following: air inlet temperature, 30 °C; air exhaust temperature, 15 to 22 °C; pan rotation rate, 12 to 18 revolution/min; and air flow, 200 cfm. For the dry phase, the conditions were the following: air inlet temperature, 65 °C; air exhaust temperature, 19 to 49 °C; pan rotation rate, 1 revolution/min; and air flow, 750 cfm. The total amount of peanuts coated was 10 kg. The amount of coating solution applied was aimed at a 5% weight gain of the peanuts after the coating was completely dried. The spray rate was set at 300 g/min per gun, and the actual spray rate was approximately 580 g/min for two guns. The spray phase took approximately 5 min. The duration of the spray phase was calculated according to the spray rate measured just before each spray phase. The drying phase lasted approximately 30 min, and then the peanuts were cooled to room temperature (~25 °C) before they were taken out of the pan. Close visual observation of the coated peanuts revealed smooth, glossy coatings without cracks or holes. For controls 1, 2, and 3, the coating procedures were done in a lab bench pan coater with pan diameter of 16 in. (LP16, LMC International, Elmhurst, IL). This coater is a conventional coater used in the confectionery industry. With the confectionery coater, the solution was ladled onto the samples, rather than sprayed on as in the pharmaceutical coater. After the control solutions were applied onto the samples they were dried for approximately 30 min, and then cooled to room temperature before they were taken out of the pan.

After they were treated in the pan, all the peanut samples were laid out at room temperature for  $\sim 24$  h prior to packaging them into oxygenbarrier bags. They were then held at -24 °C until they were taken out of the freezer to be stored at various storage conditions for headspace GC analysis.

For storing at various conditions, peanut samples weighing 180 g were placed into wide-mouth 473 mL mason jars (Ball, Alltrista Corp., Muncie, IN). The storage temperatures were 40, 50, and 60 °C; and the  $a_w$  values of the coated and the control peanuts were adjusted to the range 0.29-0.38 using moisture-absorbent sachets (silica gel pillow pack, Desiccare Inc., Santa Fe Springs, CA). The number of moistureabsorbent sachets added into the mason jars was calculated based on the initial  $a_w$  of the coated and the control peanuts and the capacity of the moisture-absorbent sachets to absorb a certain amount of water. The  $a_w$  values of the peanuts were used to calculate the amount of moisture necessary to absorb in order to achieve the  $a_w$  of the reference sample, using a peanut moisture isotherm (23). The  $a_w$  of the reference was in the range 0.25-0.3. The temperatures and the relative humidities of the chambers were monitored using a data logger (model TL 120, Dickson Company, Addison, IL). The samples were stored for up to 31 w at the three temperatures.

Headspace Gas Chromatography Analysis. Lipid oxidation was evaluated by measuring the hexanal content of the peanut samples by static headspace gas chromatography (GC) (Perkin-Elmer autosystem with HS-40 autosampler, Norwalk, CT). The GC analysis used a capillary DB-1701 column (30 m (l)  $\times$  0.32 mm (i.d.), 1  $\mu m$  thickness, J & W, Folsom, CA); HS sampler temperature, 60 °C; oven temperature, 65 °C; injector temperature, 180 °C; and detector temperature, 200 °C. Peanut samples weighing 5 g were ground for 8 s using a grinder (Braun coffee bean grinder KSM2(4), Braun Inc., Woburn, MA). Duplicate 0.5-g ground peanut samples were placed into 22-mL headspace sample vials, which were immediately sealed with silicone rubber Teflon caps. The vials were then inserted into the headspace sampler at 60 °C for 15 min and pressurized with carrier gas (He) for 30 s. An aliquot of gas phase was injected directly into the GC through the stationary injection needle. The hexanal content of samples was measured for peanuts stored for 0, 5, 15, 28, 45, 56, 84, 112, 140, 161, 175, 189, and 217 d at 40 °C, and for peanuts stored for 0, 5, 15, 28, 45, 56, 84, 112, 140, and 154 d at 50 and 60 °C.

#### **RESULTS AND DISCUSSION**

Most food quality deterioration has been found to fit either a zero- or first-order mathematical expression (24):

$$-\left(\frac{\mathrm{d}A}{\mathrm{d}t}\right) = k(A)^n$$

where A = a quality attribute measured in some units, n = the reaction order, and k = the rate constant. For either zero- or first-order deterioration, it can be shown that (19)

$$k_1 t_{S1} = k_2 t_{S2}$$

where  $k_1$  = rate constant at  $T_1$ ,  $k_2$  = rate constant at  $T_2$ ,  $t_{S1}$  = shelf life at  $T_1$ , and  $t_{S2}$  = shelf life at  $T_2$ . The Arrhenius relationship (24) or the linear model (19) can be utilized for describing how much faster or slower a reaction will go if the sample is held at some other temperature (i.e., effect of temperature on k). One can use these models to extrapolate shelf



Figure 1. The extent of oxidation for peanuts coated with denatured WPI with Vit. E stored at 40 °C.

Table 2. Regression Equations and R <sup>2</sup> Values for Initiation and
Propagation Periods of Lipid Oxidation and the Estimated Initiation
Periods for Peanut Samples Stored at 40 °C

peanut sample treatments	regression equation and <i>R</i> <sup>2</sup> for initiation period	regression equation and R <sup>2</sup> for propagation period	estimated initiation period (days)
denatured WPI with vitamin E	y = 3.06x - 48.32 $R^2 = 0.74$	$y = 45.20x - 5765.4$ $R^2 = 0.96$	136
denatured WPI without vitamin E	y = 2.92x - 43.20 $R^2 = 0.52$	$y = 65.78x - 8666.5$ $R^2 = 0.99$	137
native WPI with vitamin E	y = 2.22x - 10.96 $R^2 = 0.70$	y = 132.65x - 16917 $R^2 = 0.9986$	130
native WPI without vitamin E	y = 1.59x - 10.73 $R^2 = 0.81$	y = 124.67x - 17271 $R^2 = 0.96$	140
control 4 <sup>a</sup>	y = 1.48x - 4.55 $R^2 = 0.70$	y = 39.48x - 5698.6 $R^2 = 0.94$	150
control 3	y = 1.12x + 1.51 $R^2 = 0.5754$	y = 57.40x - 9280.8 $R^2 = 0.94$	165
control 2	y = 1.37x - 14.71 $p^2 = 0.56$	y = 31.79x - 4915 $p^2 = 0.04$	161
control 1	y = 0.58x + 12.77 $p^2 = 0.70$	y = 39.48x - 5698.6	153
reference	$x^2 = 0.70$ y = 4.17x + 29.94 $R^2 = 0.93$	$y = 51.74x - 3286.5$ $R^2 = 0.88$	70

<sup>a</sup> Control and reference samples are described in Table 1.

life results from accelerated tests at higher storage temperatures to estimate shelf life results under ambient storage conditions.

The rate of oxidation for peanut samples was determined by plotting the hexanal level vs storage time. When the same peanut samples were evaluated by both sensory and instrumental methods, the hexanal level correlated well with the rancid attribute (11). Hexanal level was also found to be a good indicator of oxidative rancidity in many other food systems (25-28). From the hexanal vs time plots, linear regressions were done with the data points of the initiation period and the propagation period. The x-value of the intercept of these linear regressions was determined to be the estimated end of initiation period. This method of analysis was performed for all the samples at each temperature. Figure 1 shows the hexanal vs time plots for peanuts coated with denatured WPI containing vitamin E solution and then stored at 40 °C. The estimated initiation period at this temperature was 136 d. The same method was used to estimate the initiation period at 50 and 60 °C.

Table 3. Regression Equations and R <sup>2</sup> Values for Initiation and	nd
Propagation Periods of Lipid Oxidation and the Estimated Init	ation
Periods for Peanut Samples Stored at 50 °C	

peanut sample treatments	regression equation and <i>R</i> <sup>2</sup> for initiation period	regression equation and <i>R</i> <sup>2</sup> for propagation period	estimated initiation period (days)
denatured WPI with vitamin E	y = 2.32x + 3.10 $R^2 = 0.77$	$y = 60.31x - 4803.5$ $R^2 = 0.98$	83
denatured WPI without vitamin E	y = 2.04x - 5.06 $R^2 = 0.59$	$y = 66.84x - 5703.9$ $R^2 = 0.95$	88
native WPI with vitamin E	y = 4.82x - 31.75 $R^2 = 0.80$	$y = 68.67x - 5191.4$ $R^2 = 0.95$	81
native WPI without vitamin E	y = 2.67x - 3.78 $R^2 = 0.6$	$y = 68.82x - 5595.3$ $R^2 = 0.94$	85
control 4 <sup>a</sup>	y = 0.65x + 43.18 $R^2 = 0.76$	$y = 60.88x - 5223.1$ $R^2 = 0.88$	87
control 3	y = 0.92x - 13.72 $R^2 = 0.97$	$y = 71.42x - 7487.4$ $R^2 = 0.97$	106
control 2	y = 0.57x + 1.8 $R^2 = 1$	y = 68.53x - 6945.1 $R^2 = 0.86$	102
control 1	y = 0.80x - 9.19 $R^2 = 0.97$	$y = 54.53x - 5303.5$ $R^2 = 0.82$	99
reference	$y = 11.92x - 5.65$ $R^2 = 0.91$	y = 58.16x - 2196.7 $R^2 = 0.88$	47

<sup>a</sup> Control and reference samples are described in Table 1.

Because the rate of oxidative rancidity accelerates at the onset of the propagation period, the estimated initiation period could be recognized as a conservative shelf life ( $t_s$ ) at the three accelerated-storage temperatures.

**Tables 2, 3,** and **4** show regression equations and  $R^2$  values for the initiation and propagation periods and corresponding estimated initiation periods ( $t_s$ ) in days for all the sample treatments, at storage temperatures of 40, 50, and 60 °C, respectively.

The general mathematical expression for the Arrhenius relationship is as follows (24):

$$k = k_0 \,\mathrm{e}^{-E_{\mathrm{A}}/RT} \tag{1}$$

where k = rate constant for deteriorative reaction at temperature T,  $k_0 =$  constant, independent of temperature (also known as the Arrhenius, preexponential, collision or frequency factor),  $E_A$  = activation energy (J/ mole), R = ideal gas constant (8.314)

**Table 4.** Regression Equations and  $R^2$  Values for Initiation and Propagation Periods of Lipid Oxidation and the Estimated Initiation Periods for Peanut Samples Stored at 60 °C

peanut sample treatments	regression equation and <i>R</i> <sup>2</sup> for initiation period	regression equation and <i>R</i> <sup>2</sup> for propagation period	estimated initiation period (days)
denatured WPI with vitamin E	y = 0.66x + 31.49 $R^2 = 0.40$	y = 32.02x - 1467.6 $R^2 = 0.96$	48
denatured WPI without vitamin E	y = 1.52x + 26.29 $R^2 = 0.79$	$y = 42.59x - 2437.2$ $R^2 = 0.99$	60
native WPI with vitamin E	y = 2.56x + 14.49 $R^2 = 1$	y = 42.45x - 2006 $R^2 = 0.97$	51
native WPI without vitamin E	y = 1.92x + 27.54 $R^2 = 0.72$	y = 42.09x - 2170.2 $R^2 = 0.98$	55
control 4 <sup>a</sup>	y = 2.93x + 10.69 $R^2 = 0.91$	y = 52.77x - 3808.6 $R^2 = 0.99$	65
control 3	y = 7.31x - 229.96 $R^2 = 0.81$	y = 40.42x - 2620.8 $R^2 = 0.91$	72
control 2	y = 4.92x - 140.28 $R^2 = 0.82$	y = 40.94x - 2708.2 $R^2 = 0.86$	71
control 1	y = 5.82x - 152.91 $p^2 = 0.02$	y = 42.68x - 2762.5 $p^2 = 0.86$	71
reference	$y = 20.02x + 26.04$ $R^2 = 1$	$y = 77.41x - 1789$ $R^2 = 0.88$	32

<sup>a</sup> Control and reference samples are described in Table 1.

 $JK^{-1}$  mol<sup>-1</sup>), and T = absolute temperature (K). From eq 1, it can be seen that

$$\log k = \log k_0 - \frac{E_{\rm A}}{2.3RT} \tag{2}$$

and, since  $k_1t_{S1} = k_2t_{S2}$ , it can be shown that

$$\log\left(\frac{t_{S1}}{t_{S2}}\right) = \frac{E_{A}}{2.3R}\left(\frac{1}{T_{1}} - \frac{1}{T_{2}}\right)$$
(3)

where  $k_1$  = rate constant at  $T_1$ ,  $k_2$  = rate constant at  $T_2$ ,  $t_{S1}$  = shelf life at  $T_1$ , and  $t_{S2}$  = shelf life at  $T_2$ .

The plot of eq 3 was made by converting the estimated initiation period  $(t_s)$  at each of the three storage temperatures to  $\log(t_s)$  and the storage temperature to 1/(absolute temperature of the storage temperature, *T*), and plotting  $\log(t_s)$  vs 1/*T*. Figure 2 shows this plot for peanuts coated with the denatured WPI

Table 5. Arrhenius Shelf Life Equation,  $R^2$ , and the Estimated Shelf Life ( $t_s$ ) at 25 °C

peanut sample treatments	Arrhenius equation	R <sup>2</sup>	estimated shelf life at 25 °C (days)
denatured WPI with vitamin E	y = 2358.5x - 5.40	0.998	330
denatured WPI without vitamin E	y = 1873.2x - 3.85	0.999	273
native WPI with vitamin E	y = 2129.9x - 4.69	1	287
native WPI without vitamin E	y = 2131.2x - 4.67	0.999	307
control 4 <sup>a</sup>	y = 2035.3x - 4.31	0.996	332
control 3	y = 1870.1x - 3.76	1	328
control 2	y = 1846.3x - 3.70	0.998	316
control 1	y = 1749.2x - 3.41	0.996	289
reference	y = 1788.2x - 3.87	0.999	136

<sup>a</sup> Control and reference samples are described in Table 1.

with vitamin E treatment solution. From this plot, an Arrhenius shelf life equation was determined using regression analysis, and the shelf life for this treatment at 25 °C was predicted. The same method of analysis was used to predict the shelf life for each coating treatment at 25 °C. This information is shown in **Table 5**. This table shows that the Arrhenius shelf life equation was a good fit for all the samples, with  $R^2$  of above 0.99.

The linear model (19) is another model which can be used to extrapolate the shelf life to different temperatures. The general mathematical expression for the linear plot is as follows:

$$k = k_0 e^{b(T - T_0)}$$

where  $k_0$  = rate at temperature  $T_0$  (°C), k = rate at temperature T (°C) and b = a constant characteristic of the reaction (19). When only a small temperature range is used, there is little error in using the linear plot rather than the Arrhenius plot (24). Again, since  $k_1t_{S1} = k_2t_{S2}$ , it can be shown using the linear model that

$$\log\left(\frac{t_{\rm S1}}{t_{\rm S2}}\right) = \frac{b}{2.3}(T_1 - T_2) \tag{4}$$

The linear model shelf life plot, also known as the shelf life plot (29) was made by plotting  $\log(t_s)$  vs *T* (°C). **Figure 3** shows the linear model shelf life plot for peanuts coated with the



1/T (1/ºK)

Figure 2. Arrhenius shelf life plot and equation for peanuts coated with denatured WPI with vitamin E.



Figure 3. Linear shelf life plot and equation for peanuts coated with denatured WPI with vitamin E.

Table 6.	Linear	Model	Shelf	Life	Equation,	R <sup>2</sup> ,	and	the	Estimate	d
Shelf Life	(t <sub>s</sub> ) at	25 °C								

peanut sample treatments	linear model equation	R <sup>2</sup>	estimated shelf life at 25 °C (days)
denatured WPI with vitamin E denatured WPI without vitamin E native WPI with vitamin E native WPI without vitamin E control 4 <sup>a</sup>	y = -0.0226x + 3.0426 y = -0.018x + 2.8514 y = -0.0204x + 2.9304 y = -0.0204x + 2.9592 y = -0.0195x + 2.9721	0.999 0.998 1 0.998 0.993	300 252 263 281 305
control 2 control 1 reference	y = -0.0179x + 2.9305 y = -0.0177x + 2.9081 y = -0.0168x + 2.8478 y = -0.0172x + 2.5313	0.998 0.996 0.993 1	304 292 268 126

<sup>a</sup> Control and reference samples are described in Table 1.

denatured WPI with vitamin E solution. The mathematical expressions and  $R^2$  values for the linear model shelf life equations for all the samples are shown in **Table 6**, along with the predicted shelf life at ambient temperature. A significant difference was not shown between the fit of the data to the linear model and that of the Arrhenius model. The data of this study fit the two models very well, with  $R^2$  values of above 0.99 in each case.

With shelf life data at two temperatures 10 °C apart, the  $Q_{10}$  factor can be calculated. The  $Q_{10}$  factor is defined as the rate of reaction at temperature (T + 10) divided by the rate of reaction at temperature (T), which is simply the inverse of the ratio of shelf life at two temperatures 10 °C apart (18).  $Q_{10}$  can be calculated using the linear model shelf life plot as follows (19):

$$Q_{10} = e^{10b}$$

where b = a constant characteristic of the reaction  $= 2.3 \times (-$  slope of the linear model plot). **Table 7** shows the  $Q_{10}$  factor for each sample calculated with the linear model. The  $Q_{10}$  values were in the range of 1.5–2.0, which was reported to be the range for lipid oxidation in various food products (30, 31).

The nature of the lipid substrates and the nature of the system where vitamin E is added have a significant impact on vitamin E's potential to be an antioxidant or a prooxidant, as well as the level of vitamin E added. The literature reports vitamin E

 Table 7. Q<sub>10</sub> Values for Each Peanut Sample Using the Linear Model

peanut sample treatments	$Q_{10}$ from the linear model
denatured WPI with vitamin E	1.68
denatured WPI without vitamin E	1.51
native WPI with vitamin E	1.60
native WPI without vitamin E	1.60
control 4 <sup>a</sup>	1.57
control 3	1.51
control 2	1.50
control 1	1.47
reference	1.49

<sup>a</sup> Control and reference samples are described in Table 1.

levels where it can be an antioxidant or a prooxidant for oils and model emulsion solution systems (32-34). However, there are no reported levels of vitamin E in coating systems where it may act as an antioxidant or a prooxidant. And the results in this study indicated that the vitamin E level used in this study did not affect oxidation significantly either way.

Oxidation in the coated samples and the controls was delayed compared to that of the reference by 2-fold, shown by increase in the shelf life by 2-fold. This indicated that the WPI-based coatings provided protection against oxidation. However, the same degree of protection was also exhibited by the control treatments, even with control 1, which only had water treatment. Three possible explanations can be posed.

First, results may have been influenced by the  $a_w$  differences among the samples. Moisture absorbent sachets were used to adjust the  $a_w$  levels of the coated and the control samples to the  $a_{\rm w}$  level of the reference sample. However, the final  $a_{\rm w}$  of different treatments varied in the range of 0.25-0.38. The reference sample had the lowest  $a_w$  range of 0.25–0.3, the controls had a  $a_w$  range of 0.29–0.33, and the  $a_w$  values for the coated samples were in the range of 0.33-0.38. Water is reported to act as a prooxidant at very low and very high water activities, and to act as an antioxidant between these two extremes (35). This finding was confirmed in a peanut butter system (36). It has also been reported that oxidation was more rapid for unblanched salted roasted peanuts stored at low (1.4%) and high (3.9%) moisture contents than for peanuts stored at intermediate moisture contents (2.2% and 2.9%), with 2.9% moisture content peanuts being the least oxidized (37). The

samples in our study may have exhibited differences in the rate of oxidation due to the effect of  $a_w$  differences, consistent with results shown in the study by Evranuz (37). The coated samples with higher  $a_w$  than the control samples with intermediate moisture contents likely had a tendency to oxidize more rapidly. Thus, the protective effect of the coating was compromised by the high  $a_w$  of the coated peanuts, resulting in oxidation rates similar to those of the control peanuts.

The second possible explanation is that the control results may be due to physical modification of the surface when treated with the aqueous solution. The surface pore structure may have collapsed when the peanuts were treated with the solution, which upon drying formed into a more densely packed surface less permeable to oxygen than the original peanut surface. If this effect is more dominant than the effect of the edible coating, then the coated samples and the control samples would likely exhibit similar degrees of oxidation which would be significantly less than that of the reference sample. Previous researchers have found water blanching or immersion treatment of peanuts can either physically modify the surface to form a glaze to protect from oxidation or chemically modify the surface protein and lipid composition to delay the oxidation in peanuts (38-41), which supports our surface modification postulation.

The third possible explanation is that either the WPI coatings were not effectively formed on the peanut surfaces or the accelerated temperatures in which the samples were stored were damaging to the WPI coatings, resulting in cracking of the coating. Thus, the WPI coatings were not significantly different from the control samples in protecting peanut samples from oxidation.

To test the first possible explanation, peanuts with different  $a_w$  values in the range of 0.25–0.4 should be stored and evaluated for oxidation. To test the second possible explanation, peanuts with the same  $a_w$ , but one with water treatment and one without water treatment, should be stored and evaluated for oxidation. To test the third possible explanation, peanut samples stored at ambient conditions should be tested for oxidation.

## CONCLUSIONS

The results from accelerated shelf life testing conducted at higher temperatures can be extrapolated to predict the shelf life at normal storage conditions. This study used the Arrhenius and the linear models with the accelerated test results to predict the shelf life at room temperature (25 °C), with similar results. The predicted shelf life of this study will be compared to the actual shelf life of the peanut samples stored at ambient conditions in a future study to test the validity of the models used. The coated peanuts exhibited a significant protection effect from oxidation. However, the controls also showed protection from oxidation compared to the untreated peanuts. Several possible explanations need to be investigated further to clarify the mechanism of inhibiting effect on oxidation in the control peanuts.

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