Radiation-Induced Oxidative Chemical Changes in Dehydrated Egg Products

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Radiation-induced buildup of lipid hydroperoxides (LOOH) and destruction of carotenoids were followed in whole egg powder and egg yolk powder as functions of dose, dose rate, and the presence of oxygen. In the absence of air the formation of LOOH was limited by the available oxygen, while destruction of carotenoids progressed linearly with dose; neither process depended on the dose rate. In the presence of air, the accumulation of LOOH and the destruction of carotenoids were strongly coupled and inversely proportional to the dose rate. The induction dose of 2.5 kGy was observed in air in both whole egg powder and egg yolk powder, independent of the dose rate. The practical consequence is that radiation decontamination can be carried out in the presence of air at the highest available dose rate by a dose not exceeding 2.5 kGy to avoid extensive degradation. This dose is adequate for a 10^3 reduction factor of *Salmonella* and well within the threshold dose of 3 kGy for organoleptic changes.

INTRODUCTION

In view of the increasing awareness of the epidemiological dimensions of poultry and egg salmonellosis (WHO, 1989b), the use of irradiation for the microbial decontamination of eggs continues to receive attention in cases where heat pasteurization is not a feasible option (Schaffner et al., 1989; Slater and Sanderson, 1989).

This work is a continuation of our previous studies of the microbiological (Matić et al., 1990), as well as chemical and organoleptic aspects (Katušin-Ražem et al., 1989) of the irradiation of egg solids for the elimination of *Salmonella*. While demonstrating the efficacy of radiation decontamination, we also reported on peroxidation increase, which was closely related to carotenoid decrease.

Peroxidation products in foods are equally undesirable from the standpoint of toxicity (Kaneda and Miyazawa, 1987), as well as unpalatability (St. Angelo and Bailey, 1987). As a major cause of quality changes (Eriksson, 1987), lipid peroxidation in foods is presently a concern of consumers (WHO, 1989a), food scientists (Simic and Karel, 1980), medical professionals (Simopoulos et al., 1986), and food processors alike (Allen, 1987; Richardson and Findlay, 1985).

Because it is impossible to provide assurance of the absolute safety of any food, whether irradiated or not, the evaluation of wholesomeness remains the only meaningful approach to the safety of irradiated foods. The chemical approach to the evaluation of the wholesomeness of irradiated food, as introduced by the Joint Expert Committee on Food Irradiation (JECFI, 1980), requires quantitative knowledge of radiation-induced chemical changes and the best available information on other potential risk factors.

This paper deals with the quantitative relationships between the radiolytic accumulation of lipid hydroperoxides and the decay of carotenoids in irradiated whole egg powder and egg yolk powder as functions of dose, dose rate, and the presence of oxygen. An understanding of the relationships among these variables is important for comprehending the peroxidative behavior of radiation-sensitive components and forms a basis for actions aimed at mitigating possibly deleterious effects.

While extensive literature deals with autoxidation studies of various lipidic substrates—free fatty acids, triglycerides, phospholipids and their structural organizates, monolayers, micelles, liposomes, and membranes—far less is known about radiolytic oxidation of real food lipids in their natural state. Common features of autoxidation established in these model systems of increasing physicochemical complexity provide a background against which radiation-induced peroxidation in dehydrated egg products will be discussed in the present work. However, efforts to understand radiolytic processes in prospectively more adequate models consisting of egg proteins, lipids, and antioxidants will be the subject of our forthcoming paper.

Because radiation-induced chemical changes in irradiated foods are generally very small and usually difficult to observe, this work bears additional relevance to the development of methods suitable for the identification of irradiated foods (Katušin-Ražem et al., 1990).

MATERIALS AND METHODS

Materials. Commercial samples obtained from two manufacturers were used, several batches from each. The lipid content was determined by extraction with a chloroform–methanol (2:1) mixture in a Soxhlet apparatus or by shaking with a cold deaerated $CHCl_3$–MeOH mixture (Kates, 1986). Lipid content was between 42 and 51% in dry whole egg and about 61% in dry egg yolk. The fatty acid profile in the lipidic component was determined by gas chromatography of free methylated fatty acids on a 2-m glass column, 3 mm i.d., filled with 10% Silar 10C on Chromosorb WHP 80–100 at 185 °C with 30 mL/min N_2 and flame ionization detector (Christie, 1982). The major unsaturated fatty acids, oleic and linoleic, were in the range 21–29% and 2.5–4.2% of the total lipids, respectively.

Sample Preparation, Irradiation, and Dosimetry. Samples weighing 10–50 g were sealed in polyethylene pouches in the presence of air. Samples to be irradiated in the absence of air were evacuated and sealed in laminated aluminum pouches using an industrial vacuum packaging machine. Irradiations were carried out with ^{60}Co γ rays in a panoramic irradiator and a well-type irradiator, both constructed by the Ruđer Bošković Institute, and in a Gammacell 220 (Atomic Energy of Canada, Ltd.). The dose rates varied from 4 Gy/s in the panoramic irradiator, to 0.4 Gy/s in the well-type irradiator, to 0.04 Gy/s in the Gammacell, as determined by ethanol–chlorobenzene dosimetry (Ražem et al., 1985).

Determination of Lipid Hydroperoxides. Hydroperoxides in the lipidic component were determined in deaerated chloroform–methanol mixtures (2:1) by spectrophotometry of the ferric–thiocyanate complex. This complex was formed on the oxidation

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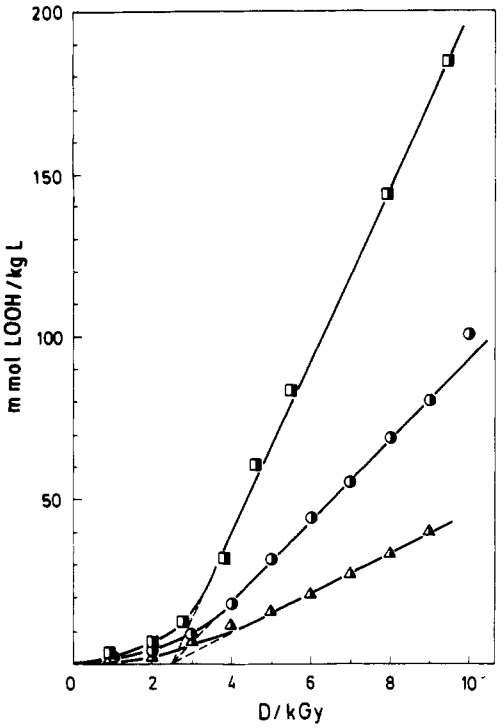

Figure 1. Effect of dose D on LOOH formation in whole egg powder irradiated in air at the following dose rates: (triangles) 4 Gy/s; (circles) 0.4 Gy/s; (squares) 0.04 Gy/s.

of ferrous iron by hydroperoxides and subsequent complexing of ferric ions with thiocyanate, which yields an intense red coloration (λ_{max} = 485 nm, ϵ_{485nm} = 13710 $dm^3 mol^{-1} cm^{-1}$) (Mihaljević, Katušin-Ražem, and Ražem, unpublished data).

Determination of Carotenoids. Carotenoids were determined in a $CHCl_3$–MeOH extract by spectrophotometry against a blank (an extract of whole egg powder irradiated with 20 kGy or of egg yolk powder irradiated with 40 kGy, in which the characteristic absorption of carotenoids was destroyed). Since the optical absorption of the blanks decreased toward longer wavelengths, the absorbance of the longest wavelength peak of carotenoids at 480 nm was used. Taking the molar absorptivity of lutein (the major carotenoid in egg) in $CHCl_3$ (Vogtmann and Prabucki, 1970), the carotenoid content was about 2.5 mg/100 g of whole egg powder and about 3.4 mg/100 g of egg yolk powder.

Normalization of Contents. To compensate for the variability of the lipid and carotenoid contents among the samples, the amount of lipid hydroperoxides (millimoles of LOOH) was expressed per unit mass (kilograms) of lipid L (millimole of LOOH per kilogram of L), and carotenoids were expressed as a percentage of the initial concentration in unirradiated samples.

ESR Measurements. Samples for ESR measurements in vacuum were evacuated in glass ampules with a side-arm fitting the ESR spectrometer cavity. After irradiation, this side-arm was flame annealed and cooled before being filled with the sample. ESR spectra were recorded by a Varian E-109 ESR spectrometer. The amplitude of the first-derivative signal was taken as the measure of spin concentration relative to the amplitude of the standard.

RESULTS

The amount of radiation-induced hydroperoxides as a function of dose in whole egg powder is shown in Figure 1 and that in egg yolk powder in Figure 2. In both materials with free access of air, hydroperoxidation proceeded linearly with dose after an induction dose of 2.5 kGy, as seen in Figures 1 and 2. On the extended irradiation of samples in the sealed pouches, the amount of LOOH decreased with doses above 10 kGy. With free access of air, however, the buildup of LOOH was linear with doses of up to at least 50 kGy. The amount of lipid hydroperoxides also depended on the dose rate and was larger at lower dose rates. This is also illustrated for a fixed dose of 3 kGy by the upper curves in Figure 4a,b.

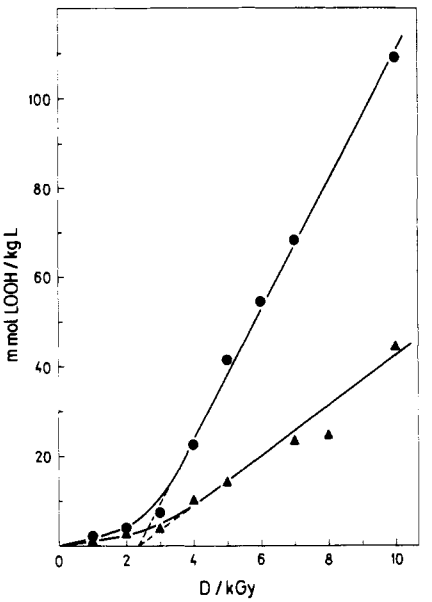

Figure 2. Effect of dose D on LOOH formation in egg yolk powder irradiated in air at the following dose rates: (triangles) 4 Gy/s; (circles) 0.4 Gy/s.

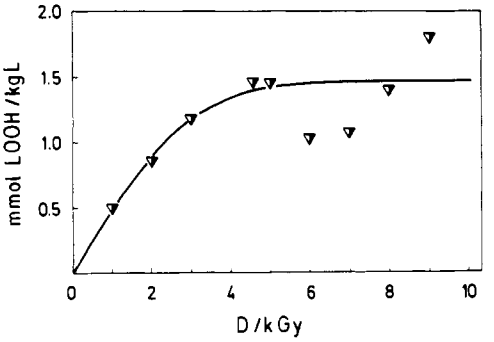

Figure 3. Effect of dose D on LOOH formation in whole egg powder irradiated in vacuum at dose rate of 4 Gy/s.

The radiation-induced change can be expressed in terms of the radiation chemical yield. Radiation chemical yield denotes the number of moles of chemical change produced in irradiated matter by the absorption of one unit of absorbed energy, expressed in joules. For lipid hydroperoxides, G(LOOH) = (μmol of LOOH/kg of L)/J; the value of G(LOOH) at any dose is given by the value of the ordinate of the curves in Figures 1 and 2 at that dose divided by the dose. Because hydroperoxidation does not increase linearly with dose from the beginning of irradiation, the values of radiation chemical yields, G(LOOH), would increase with dose approaching saturation (not shown). Corresponding limiting yields (at infinite doses) are about 20, 10, and 5 (μmol of LOOH/kg of L)/J at dose rates 0.04, 0.4, and 4 Gy/s, respectively, high values indicating that LOOH are formed by chain reactions.

The initial G(LOOH) in evacuated whole egg samples was only about 0.5 (μmol of LOOH/kg of L)/J, while hydroperoxidation did not proceed above 4 kGy (Figure 3). In evacuated samples the amount of hydroperoxides formed did not depend on the dose rate, as shown in samples irradiated with 3 kGy (Figure 4).

Radiation-induced loss of carotenoids in whole egg powder and egg yolk powder was closely related to radiation-induced hydroperoxidation (Figure 5). Caro-

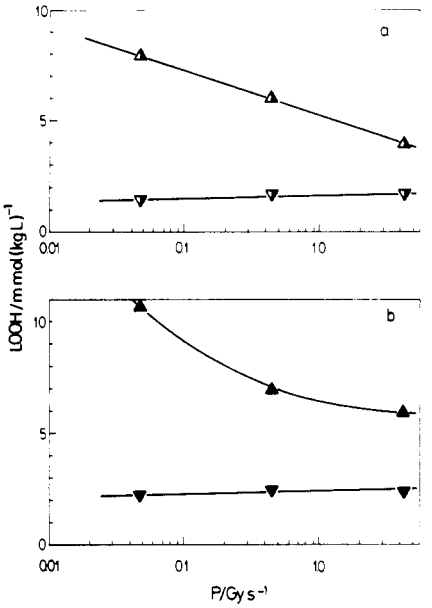

Figure 4. Effect of dose rate P on LOOH formation in whole egg powder (a) and egg yolk powder (b) at 3-kGy dose. (Triangles) Samples in equilibrium with air; (inverted triangles) evacuated samples.

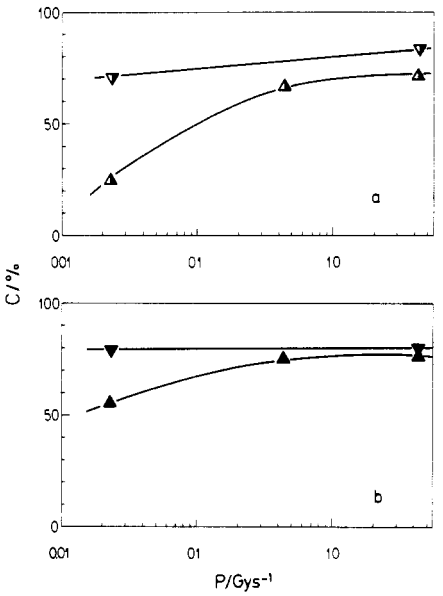

Figure 5. Effect of dose rate P on the carotenoid content of whole egg powder (a) and egg yolk powder (b) at 3-kGy dose. (Triangles) Samples in equilibrium with air; (inverted triangles) evacuated samples.

tenoid loss had many common features with hydroperoxide buildup with dose. In evacuated samples, the destruction proceeded linearly from the lowest doses and larger loss was induced by irradiation at lower dose rates (Figure 6).

In the presence of air after an induction dose between 2 and 3 kGy, linear destruction with dose took place, the dose dependence of which was also inversely proportional to the dose rates for irradiations at 4 and 0.4 Gy/s. The effect of the lowest applied dose rate, 0.04 Gy/s, was indistinguishable from that at 0.4 Gy/s. However, unlike hydroperoxide buildup, the destruction of carotenoids in egg yolk powder was a weaker function of dose than in whole egg powder at the same dose rate. Linear parts of the curves could be extrapolated to the common point on the ordinate, showing that, due to the common induction dose, a common protection level of 35% was achieved in

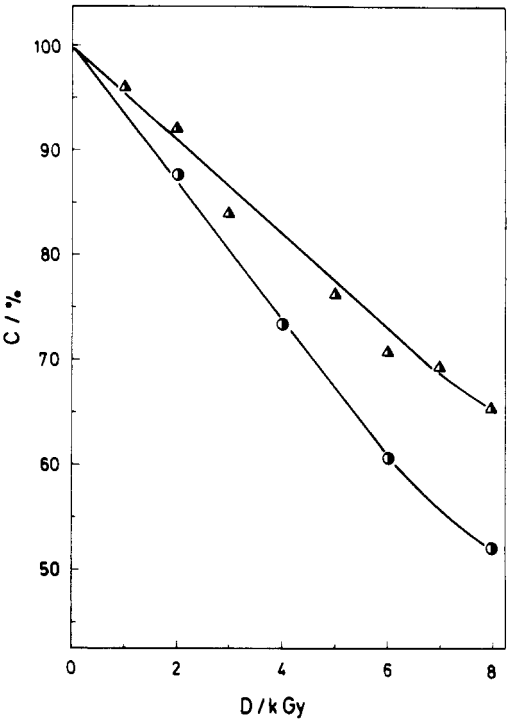

Figure 6. Effect of dose D on the carotenoid content of whole egg powder irradiated in vacuum at the following dose rates: (triangles) 4 Gy/s; (circles) 0.4 Gy/s.

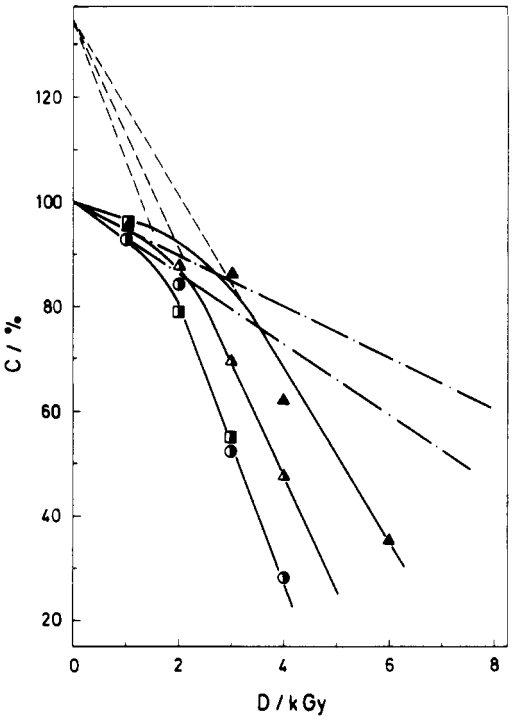

Figure 7. Effect of dose D on the carotenoid content of whole egg (semisolid symbols) and egg yolk powder (solid symbols) irradiated in air at the following dose rates: (squares) 0.04 Gy/s; (circles) 0.4 Gy/s; (triangles) 4 Gy/s. Dash-and-dotted lines are data from Figure 6 and are entered here for comparison.

all samples. Initially slower decays in the induction dose range of samples in air approximately correspond to the decays in evacuated samples at corresponding dose rates (Figure 7).

ESR spectra of irradiated egg solids depend on the presence of oxygen. The major component of the spectrum obtained by irradiation in vacuum is a doublet (Figure 8), while irradiation in the presence of air produced singlet

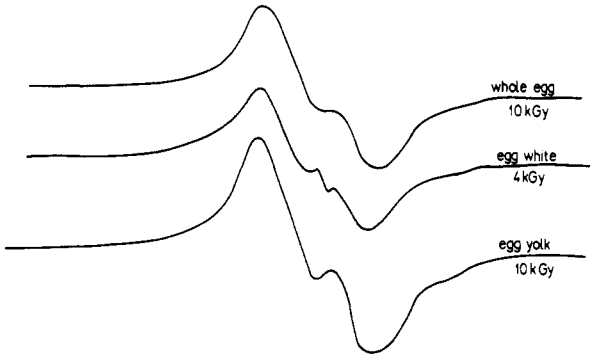

Figure 8. First-derivative ESR spectra of samples irradiated and measured in vacuum at 4 Gy/s.

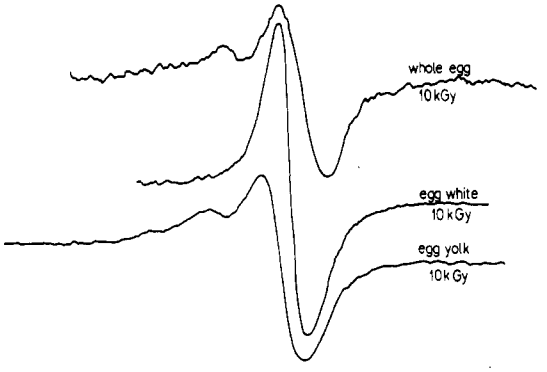

Figure 9. First-derivative ESR spectra of samples irradiated and measured in equilibrium with air at 4 Gy/s.

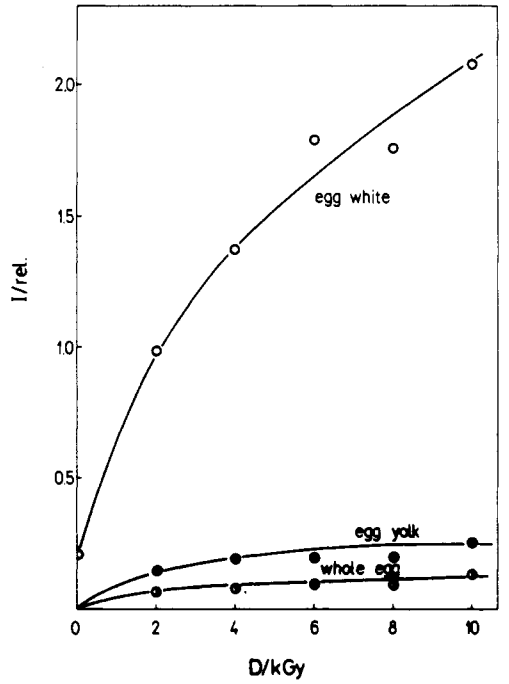

Figure 10. Effect of dose D on the relative intensities of first-derivative ESR spectra of whole egg (semisolid circles), egg yolk (solid circles), and egg white (open circles) irradiated in air at 4 Gy/s.

signals (Figure 9). The relative intensities of signals produced by irradiation in air are shown in Figure 10.

DISCUSSION

The amount of unsaturated fatty acids in yolk lipids is greater than in most animal products (Bergquist, 1979). Nevertheless, lipids in shell eggs, as well as in broken-out form, are surprisingly stable to oxidative deterioration (Pike and Peng, 1985). Dehydrated egg products are more susceptible to peroxidation due to an unfavorably high surface-to-volume ratio (Addis, 1986).

In the following discussion we shall try to present the common features of autoxidation of various lipidic substrates—solutions, monolayers, micelles, liposomes, and membranes—and to examine possible analogies among systems of increasing complexity to better understand radiation-induced oxidative chemical changes in dehydrated egg products.

The rate of autoxidation in homogeneous systems with free diffusion is proportional to the square root of the initiation rate (Barclay and Ingold, 1981; Pryor et al., 1982). This follows if termination is by the bimolecular interaction of the chain-carrying peroxyl radicals. The same mechanism must hold for radiation-induced bulk-phase oxidation of methyl oleate, where the square root dependence of the reaction rate on the dose rate has also been established (Hyde and Verdin, 1968).

The mechanism of radiation-induced peroxidation requires (Spinks and Woods, 1990) that the radiation-chemical yield of lipid hydroperoxides, G(LOOH), be inversely proportional to the square root of the dose rate. This dependence was found in radiation-induced peroxidation of fatty acid micelles (Metwally and Moore, 1987; Raleigh and Kremers, 1978), indicating that free diffusion of radiolysis species is possible in these systems also.

Generally, lower dose rates were found to be more efficient in producing lipid hydroperoxides. This was observed also in dry lipid films (Chatterjee and Agarwal, 1983) and phospholipid liposomes (Petkau and Chelack, 1976), although no inverse square root correlation with dose rate was originally tested in these systems. Our analysis of the published results in egg lecithin liposomes (Raleigh, 1987) and soybean bilayer liposomes (Petkau and Chelack, 1976) showed that the amount of radiation-chemical change was inversely proportional to dose rate

$$G(\text{LOOH}) = a(\text{dose rate})^n$$

where a is a proportionality constant and n is a negative exponent; the exponent n was -0.8 in the former and between -0.8 and -0.9 in the latter case. However, it must be borne in mind that different dose–response curves were obtained by taking different indicators as measures of peroxidation, as well as by taking measurements at different times after irradiation (Corliss and Dugan, 1970), which may distort the results.

Analysis of our own results showed that the radiation-chemical yields of hydroperoxides in the inhibited dose range, up to 3 kGy, were proportional to (dose rate)$^{-1/5}$. At about 4 kGy, where the transition from inhibited to steady-state peroxidation was taking place, the exponent was $-1/4$, whereas at 5 kGy and throughout the steady-state range it was $-1/3$.

We believe this inverse cube root dependence on the dose rate to be the consequence of the temporal and spatial distribution of free radicals created by radiation. At a low dose rate fewer free radicals will be formed in time and space, and more radicals will react with neighboring lipid molecules, thus propagating the chain reaction, rather than recombining with other radicals. The inverse cube root dependence would reflect a reduced recombination probability, depending on the distances between the recombining radicals randomly formed by irradiation in a rigid matrix, as compared with the inverse square root recombination probability in systems with free diffusion.

Radiation-induced peroxidation in whole egg powder and egg yolk powder was inhibited at low doses of radiation,

whereupon at higher doses a linear increase of peroxidation with dose set in. High values of G(LOOH) at all applied dose rates indicated that chain reactions were operative in the range of linear increase.

In the absence of oxygen the formation of lipid hydroperoxides became saturated with dose, as all available oxygen was consumed. Oxygen cannot be completely removed from egg powder samples by evacuation at room temperature. Some molecules remain adsorbed on a very large surface area of the powder and are available for reaction with radiation-induced radicals.

The same induction dose, 2.5 kGy, obtained in both whole egg powder and egg yolk powder, would indicate that the antioxidative protection was due to components that were present in both whole egg and egg yolk. The first candidate substances are carotenoids, the main lipid-soluble antioxidants in egg.

The presence of oxygen is not essential for the radiation destruction of carotenoids, because carotenoids decreased linearly with dose in evacuated samples. It has already been shown that carotenoids in solution can be destroyed by oxidizing free radicals (Peiser and Yang, 1979). However, there have been conflicting results regarding the nature of radicals responsible for carotenoid bleaching. On the one hand, the rate of bleaching in liposome preparations with polyunsaturated fatty acids (PUFA) present in egg phosphatidylcholine (PC) was found to be the same as in saturated dipalmitoyl PC (Krinsky and Deneke, 1982). On the other hand, the addition of unsaturated fats or fatty acids to paraffin solutions of β-carotene accelerated the autoxidation rate (Carnevale et al., 1979; Kanner and Budowski, 1978).

In the presence of air, carotenoids in our samples were initially destroyed by radiation at a rate approximately the same as in evacuated samples. As the amount of lipid hydroperoxides increased with dose, faster carotenoid depletion with dose in the dose range of the steady-state lipid peroxidation occurred, indicating a possible bleaching reaction of carotenoids with LOOH. This second, faster, decay was also linear with dose. The absence of the dose-rate effect on the depletion of carotenoids for the lowest dose rates applied suggests that the availability of carotenoids for chemical reactions leading to bleaching was critical and different from the availability of lipids for radiation-induced peroxidation. This might explain the indifference of carotenoids to bleaching by autoxidized PUFA in liposomes (Krinsky and Deneke, 1982). The same reason, namely the availability for chemical reactions, could also explain the increasing stability of carotenoids with increasing water activity in dehydrated model systems (Kanner et al., 1978) containing antioxidants and apparently conflicting results in the systems not containing water-soluble antioxidants (Zachariev and Kiss, 1985).

By the end of the induction dose for LOOH formation, 2.5 kGy, only one-sixth to one-third of the carotenoids—depending on the dose rate—would be destroyed. This contrasts with the high antioxidative efficiency of α-tocopherol, which was almost completely destroyed before steady-state peroxidation could start in the autoxidation of homogeneous (bulk) methyl oleate (Lips, 1957) and methyl linoleate (Lips, 1957; Niki et al., 1986) systems, as well as in nonhomogeneous systems, linoleate monolayers (Wu et al., 1979) and human erythrocyte ghosts (Yamamoto et al., 1985). On the one hand, this slower depletion of carotenoids indicates their limited availability as antioxidants in our systems. On the other hand, the common breakpoint dose in air indicates that both processes, LOOH accumulation and carotenoid depletion, could be inhibited by the same inhibitor.

A reasonable assumption, which is also supported by the literature (Taguchi et al., 1988), is that egg proteins confer antioxidative protection to both lipids and carotenoids. This is also corroborated by our own finding (Katušin-Ražem et al., 1989) that amino acids in irradiated egg proteins were not destroyed randomly by radiation but that some suffered more damage than did others. The most susceptible amino acids we found in egg proteins (Met, His, Lys) were also powerful antioxidants in the autoxidation of freeze-dried emulsions if added as individual compounds (Riisom et al., 1980). This further demonstrates the ability of radiation damage to be transferred in a rigid system and to become selectively localized. The aspects of radiation damage transfer and localization were studied by the ESR technique.

There is a strong similarity between the radiation effects on ESR spectra and the reactions of proteins with peroxides (Karel et al., 1975). The main features of the ESR spectra of irradiated egg solids are characteristic of the ESR spectra of proteins. The major component of the spectrum obtained by irradiation in vacuum is a doublet, believed to be due to α-carbon radicals of the protein (Schaich, 1980). The presence of lipids is indicated only by the small differences between egg white spectra containing no lipids and spectra of lipid-containing whole egg and egg yolk. However, rather than providing specific spectral information, these spectra appear to be envelopes of many overlapping resonances.

A similar situation exists in the presence of air, whereby singlet signals were obtained after irradiation. Contrary to what one might expect in the presence of easily oxidizable lipids, the strongest signals were obtained in lipidless egg white, while in the presence of lipids additional small downfield resonances appeared in egg yolk and whole egg spectra.

It has already been found (Schaich and Karel, 1976) that peroxidized lipids alone, in the absence of proteins, give no ESR signal, which was explained by the short half-life of the lipid radicals. On the other hand, proteins irradiated alone gave the largest ESR signals. In the presence of lipids, however, the number of radicals in the nonlipid component was reduced, all of which indicated the relative ease of free-radical transfer mediated by the lipid component.

CONCLUSIONS

The dominant radiation-induced chemical changes in whole egg powder and egg yolk powder irradiated in air were degradative changes of lipidic components: the accumulation of lipid hydroperoxides and destruction of carotenoids. An induction dose of 2.5 kGy was observed in whole egg powder as well as in egg yolk powder for both processes. The induction dose itself did not depend on the dose rate, but the extents of degradative changes to both lipids and carotenoids were inversely proportional to the dose rates, higher dose rates producing less degradation at the same doses.

Above the induction dose the accumulation of lipid hydroperoxides proceeded via chain reaction. The transition from the inhibited phase to the chain reaction phase was characterized by the increasingly stronger dependence of hydroperoxidation yield on dose rate P (from $P^{-0.2}$ to $P^{-0.25}$), until an inverse cube root dependence ($P^{-0.33}$) was established. This dependence on the dose rate is weaker, as compared to the inverse square root dependence ($P^{-0.5}$), observed in homogeneous solutions and fatty acid mi-

celles, and even weaker as compared to the almost inverse proportionality dependence ($P^{-0.9}$) observed in liposomes. We believe it to be due to the rigidity of the system, preventing recombination by diffusion and favoring chain propagation.

While removal of oxygen resulted in a significant reduction and ultimate saturation of the amount of lipid hydroperoxides formed in evacuated samples, the destruction of carotenoids in the absence of air proceeded slowly and linearly with dose, indicating a weak direct radiation action. Superimposed on this direct destruction of carotenoids was the additional increased destruction observed in the presence of air above induction dose for hydroperoxidation, attributed to the reaction with oxidized lipids.

From the standpoint of minimizing degradative chemical changes, it would be advantageous to irradiate the products in the absence of air. However, this may not be feasible in practice because the usual packaging of dry egg products—multilayer brown paper sacks—cannot be evacuated. A practical consequence is that irradiation in air should be performed at the highest available dose rate. The dose of 2.4 kGy was adequate for a *Salmonella* inactivation factor of 10^3 (Matić et al., 1990), being at the same time below both the induction dose for producing extensive degradation and the threshold dose of 3 kGy for producing noticeable organoleptic changes (Katušin-Ražem et al., 1989).

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