

Some Effects of Gamma Radiation on the Keeping Quality of Apples

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Storage studies of apples treated with gamma radiation were conducted. The principal immediate effect was a softening by radiation dosages in excess of 10 kilorads, and a transient stimulation of respiratory oxygen consumption. Sauce was prepared from treated apples and evaluated as to quality. The principal effect of irradiation was a loss of "grain," which resulted in no significant change in acceptability or preference of the product. In storage, irradiated fruit softened at a much slower rate than did non-irradiated fruit. Despite the initial softening, the combined effect was fruit firmer than normal. Softening was positively correlated with changes in pectin content. Irradiation produced a marked reduction in the incidence of storage scald, and evidence of reduction in true brown core was suggested. Some slight indication of induced injury to the skin of one picking of one variety was found, and some impairment of flavor was described at high dosages in another variety.

THE STORAGE LIFE of some fresh fruits and vegetables may be extended by treatment with ionizing radiation in the 20,000- to 500,000-rad (20- to 500-krad) dosage range. Examples of specific applications which are in various stages of developmental research include sprout prevention in potatoes (5, 24) and onions (6), retardation of bacterial or mold infestation in strawberries (3, 15), citrus (7, 27), and peaches (2, 8, 14), and delayed ripening in pears (19) and tomatoes (9). The use of such "pasteurizing dosages" is considered to be one of the promising applications of ionizing radiations for food preservation.

However, some undesirable side-effects of this treatment may, in some instances, limit its utilization. These include softening of the tissues (7, 23), development of off-flavors and/or off-colors (9, 78, 20), or initiation of other alterations of familiar characteristics. Of these radiation-induced changes, perhaps the most important is that of tissue softening. In studies concerning this effect, dosages above approximately 25 krads induce tissue degradation which may be correlated with radiation-induced changes in both pectin and cellulose (17). No study of changes occurring in the pectin constitution of tissues subjected to postirradiation storage has been reported previously.

Relatively low dosage of gamma radiation may retard the development of storage scald and brown core in apples (16). To investigate the effectiveness of this treatment and the applicability

from the standpoint of total product acceptability, a study was conducted in which three varieties of apples were irradiated at various degrees of maturity and evaluated for storage longevity from the standpoint of texture, color, flavor, and susceptibility to storage disorders. The quality of the fruit for processing into sauce was also evaluated.

Materials and Methods

These experiments are the results of a 2-year study of the effect of gamma radiation upon the storage longevity of apples. During the first year (1961), McIntosh, Cortland, and Rome Beauty apples were harvested from specific trees during early, medium, and late stages of maturity, the medium date being approximately that of commercial maturity. Immediately following harvest, bushel lots of hand-graded fruit were subjected in duplicate to dosages of 50 and 100 krads of gamma radiation delivered over a 6-hour exposure period from a 4000-curie Co⁶⁰ source in air at ambient temperatures of 72° ± 2° F. (23.5° ± 1.5° C.). For dose-rate comparison, a third treatment of 100 krads was sometimes administered over a 12-hour exposure period. Uniformity of dosage was achieved by placing the fruit in concentric circles so that each individual fruit was equidistant from the source during the exposure period. For further uniformity, the fruit were rotated 180° midway through the irradiation exposure period.

Immediately following irradiation, the fruit were examined for evidence of radiation injury. Firmness was measured with a Magness-Taylor pressure tester on 20 apples from each of duplicate bushel boxes. Soluble solids were measured with a hand refractometer on the juice from 20 apples from each box. An estimate of the ground color was made using the Cornell ground color chart by which fruit are assigned color values from 1 (yellowest) to 5 (greenest) (22). Respiratory activity was also followed for several days following irradiation on duplicate samples of fruit. Oxygen consumption was measured with a Beckman G2 oxygen analyzer using a flow rate of 150 ml. per minute for approximately 1-kg. samples in 1-gallon chambers at a controlled temperature of 70° F. (21° C.).

The fruits were placed in 32° F. (0° C.) and 90 to 95% relative humidity storage for longevity studies. McIntosh and Cortland fruits were removed from storage in February and March, and Rome Beauty, in March and April. After removal from storage, firmness and soluble solids were determined, as previously described. The fruits were then evaluated for scald and other physiological disorders following 1 week at 70° F. (21° C.). Although scald evaluation was somewhat subjective, fruit with over 25% of the surface area heavily scalded was graded as severe. Fruit with less than 25% of the surface area scalded, or if scald was faint, was graded slight.

During the second year (1962), em-

phasis was placed upon the variety Rome Beauty, and the radiation range was increased to include dosages of 5, 10, 50, and 100 krads over a 6-hour exposure, and 10 and 100 krads over a 12-hour period. All methods of irradiation and quality evaluation were performed exactly as before. In addition, samples of the fruit were taken for pectin analysis before and after both irradiation and

storage. For this analysis, extractions were accomplished by a modification of the centrifuge method described elsewhere (10). Briefly, the extractions were made by siphoning off the extract following centrifuging for 15 minutes at 2000 r.p.m. Samples of alcohol-insoluble solids weighing 0.5 gram were extracted with three solvents. The first three extractions were made with 30

ml. of water each at 30° C. Then the residue was extracted three times with a solution containing 0.2% Calgon and 0.8% NaCl at 30° C., and finally three times with 0.1N HCl at 80° C. The water, Calgon, and acid extracts were collected separately and made up to 100 ml. each with enough Calgon and NaCl added to give final proportions of 0.2 and 0.8%, respectively. The extracts

Table I. Effect of Radiation on Storage Life of Rome Beauty Apples

Treatment Krad/s Hours		Before Storage			After Storage						
		Firmness, pounds	Sol. solids, %	Ground color	Firmness, pounds	Sol. solids, %	Scald			Spot scald, %	Brown core, ^a %
HARVESTED 10/13/61											
0	..	21.9	11.2	4	15.5	...	1	99	100	0	0
50	6	19.5	11.1	4	17.7	...	51	34	85	7	0
100	6	17.9	11.1	4	17.9	...	41	13	54	9	0
HARVESTED 10/24/61											
0	..	20.1	12.0	4	15.0	...	6	93	99	0	0
50	6	18.3	12.0	4	16.1	...	25	8	33	2	0
100	6	16.5	12.8	4	16.4	...	14	1	15	16	0
100	12	16.3	12.3	4	16.6	...	5	0	5	30	0
HARVESTED 11/3/61											
0	..	20.1	12.0	3.5	14.9	...	25	21	46	2	0
50	6	18.2	12.0	3.5	16.3	...	13	3	16	15	0
100	6	15.8	11.8	3.5	16.2	...	4	5	9	26	0
100	12	15.5	12.3	3.5	15.1	...	2	0	2	47	0
HARVESTED 10/5/62											
0	..	21.4	10.5	3.0	13.4	11.4	5	95	100	0	32
5	6	21.1	10.8	3.0	13.4	11.7	4	96	100	0	64
10	6	21.0	10.9	3.0	13.6	11.4	4	95	99	0	0
10	12	21.2	10.9	3.0	14.0	11.6	10	90	100	0	4
50	6	19.8	11.0	3.0	16.4	11.5	49	44	93	0	45
100	6	17.3	11.2	3.0	15.5	11.8	52	7	59	0	66
100	12	16.8	11.1	3.0	14.9	11.9	39	5	44	0	88
HARVESTED 10/24/62											
0	..	20.7	11.7	3.0	12.7	11.4	55	24	79	0	0
5	6	20.7	12.2	3.0	12.9	11.8	59	12	71	0	0
10	6	20.3	11.9	3.0	13.2	12.0	47	4	51	0	4
10	12	20.7	11.9	3.0	13.1	11.2	38	1	39	0	0
50	6	19.0	12.4	3.0	15.5	11.9	13	0	13	0	10
100	6	17.7	11.7	3.0	14.6	12.0	17	0	17	0	47
100	12	16.5	11.6	3.0	15.0	11.5	22	0	22	0	80

^a Brown core = flaky, brown tissue at core.

Table II. Effect of Radiation on Storage Life of McIntosh Apples

Treatment Krad/s Hours		Before Storage			After Storage					
		Firmness, pounds	Sol. solids, %	Ground color	Firmness, pounds	Slight, %	Severe, %	Total, %	Spot scald, %	Brown core, ^a %
HARVESTED 9/21/61										
0	..	15.1	10.3	4.0	12.0	39	52	91	71	1
50	6	14.4	10.4	4.0	12.3	6	5	11	11	0
100	6	13.3	10.3	4.0	12.1	0	0	0	22	2
HARVESTED 9/29/61										
0	..	15.0	10.8	3.5	12.0	27	43	70	58	0
50	6	13.2	10.2	3.5	11.8	0	0	0	7	2 ^b
100	6	12.3	10.6	3.5	11.4	0	0	0	10	11
HARVESTED 10/9/61										
0	..	12.9	10.8	2.5	10.1	25	8	33	33	31
50	6	11.3	10.3	2.5	8.5	0	0	0	31	42
100	6	10.4	10.8	2.5	8.1	1	0	1	37	45
100	12	11.3	10.4	2.5	8.8	1	0	1	35	18

^a Brown core = flaky, brown tissue at core.

^b 3% injury on skin.

Table III. Effect of Radiation on Storage Life of Cortland Apples

Treatment		Before Storage			After Storage					
Krads	Hours	Firmness, pounds	Sol. solids, %	Ground color	Firmness pounds	Slight, %	Severe, %	Total, %	Spot scald, %	Brown core, ^a %
HARVESTED 9/26/61										
0	..	15.9	10.6	4	10.8	0	100	100	0	0
50	6	14.5	11.1	4	12.4	13	9	22	0	0 ^b
100	6	13.2	10.8	4	11.9	3	3	6	0	1 ^b
HARVESTED 10/6/61										
0	..	15.0	11.0	2.5	11.0	11	85	96	0	0
50	6	14.0	11.5	2.5	12.6	28	10	38	0	1 ^b
100	6	13.5	11.2	2.5	10.5	9	5	14	0	0 ^b
HARVESTED 10/17/61										
0	..	14.6	11.3	2.5	11.3	27	6	33	0	0
50	6	13.7	12.0	2.5	11.7	1	4	5	0	1
100	6	13.1	11.9	2.5	11.6	1	1	2	0	0 ^c

^a Brown core = flaky, brown tissue at core.

^b Flat taste.

^c Off-flavor.

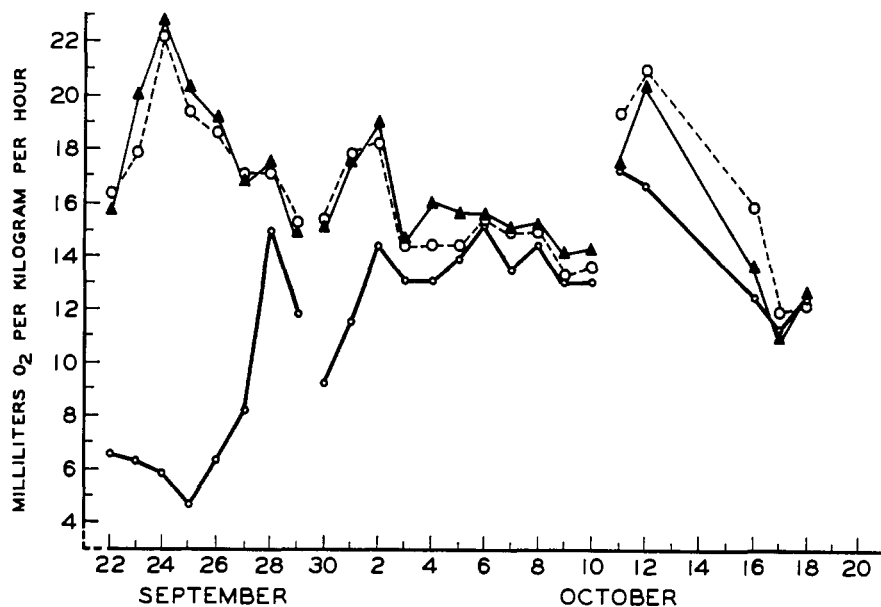


Figure 1. Effect of radiation on respiration of McIntosh apples at 70° F. at different picking dates after harvest in 1961

○ control; ▲ 50 krads; ○ 100 krads

Table IV. Processing Data for Rome Beauty Apples Processed into Sauce Immediately Following Treatment with Various Doses of Gamma Radiation

Treatment		Trimming Waste, %	Sugar Added, ^a %	Water Added, ^b %	Over-all Yield, ^c %
Krads	Hours				
0	...	17.0	10.7	15.5	108
5	6	18.9	10.3	17.8	110
10	6	19.4	11.5	22.5	115
50	6	18.5	9.5	15.2	106
100	6	19.2	9.3	11.5	102

^a Added to bring the soluble solids in the finished product to 19%.

^b Added to bring the Bostwick reading of the finished product to 4.5.

^c Based on the weight of applesauce in the can, including water and sugar added, divided by the weight of fresh apples.

were all adjusted to pH 6.0 ± 0.2. Viscosities of the three separate fractions were determined at 30° C. in Ostwald-Cannon-Fenske pipet. The average specific viscosity values have been obtained by averaging the results obtained on the water, Calgon, and acid extracts, since mixing equal aliquots of each gave a viscosity not different from that of the numerical average of the three. Following viscosity measurements, the extracts were frozen until such time as uronic determinations could be made. Uronic acids were determined by the method of McComb and McCready (13).

In addition, samples of irradiated Rome Beauty apples were processed into sauce immediately following irradiation, according to currently acceptable commercial methods, and evaluated for both processing characteristics and acceptability of the finished product. Briefly, processing involved slicing the peeled and cored apples, cooking by steam injection for 2½ minutes to a temperature of 213° F. (101° C.), finishing, adjusting the consistency with water to a Bostwick reading of 4.6 at 150° F. (66° C.), and packaging in No. 2 cans (12).

Results and Discussion

Prestorage Results. The results of irradiation upon firmness, soluble solids, and ground color of the varieties McIntosh, Cortland, and Rome Beauty as evaluated within 24 hours following treatment are indicated in Tables I, II, and III, respectively. Treatments above 10 krads resulted in progressive softening of the tissues in agreement with data previously reported from this laboratory (4, 11, 23) and elsewhere (17). Dose rate appeared to have little influence upon the softening process, nor did picking time greatly influence the degree of softening attained. The radia-

tion treatments did not seem to result in any consistent effect upon either soluble solids content, or ground color. This is also in accordance with data reported previously (4, 17).

Figures 1 to 3 include the respiration rate data on McIntosh (1961) and Rome Beauty apples (1961 and 1962). Generally, radiation resulted in respiratory activity stimulation. There was a greater stimulation on preclimacteric than on postclimacteric apples—the greater the radiation dosage, the greater was the increase in respiration rate. In 1962, with lower dosage rates used on Rome Beauty, there was a significant reduction in respiration rate with 5-krad treatment from that of the unirradiated control. This was the only inhibitory effect of radiation on respiration. Figure 4 presents data on cumulative respiration or total oxygen consumption over a period of 14 days at 70° F. (21° C.). The inhibitory effect of the 5-krad treatment and the stimulatory effect of dosages of 10 krad or above are readily apparent. The increase in total oxygen consumed by fruit treated with 100 krad was in the order of 50% after 14 days at 70° F. (21° C.).

The results of the processing experiments are indicated in Table IV. Here the major influence of radiation is a variation in the amount of water added to the product necessary to bring the consistency (Bostwick reading) to an acceptable value of 4.5. An increase in water requirement to a maximum at 10 krad followed by a reduction at higher dosages was observed. This property is further reflected in over-all yield. The overriding change in the finished product was a progressive reduction in the grain of the sauce with increasing dose of radiation above 10 krad. Submission of samples to organoleptic preference evaluation produced inconclusive results, however. Although all of the participants could distinguish differences in texture between sauce prepared from the control and from 100-krad treated fruit, there was a lack of agreement as to preference. Of 16 tasters, seven preferred sauce from the nonirradiated fruit, seven preferred sauce from the irradiated, and two could not express a preference.

The influence of gamma radiation upon apple pectin has been discussed elsewhere in more detail from the standpoint of its immediate effect upon tissue composition (17). Briefly, the results indicate (at krad dosages) no significant change in alcohol-insoluble solids content, average specific viscosity, or total anhydrogalacturonic acid residue, but a marked increase in the soluble to insoluble pectin ratio with increasing dosage above 10 krad (Table V). These changes are all in line with the hy-

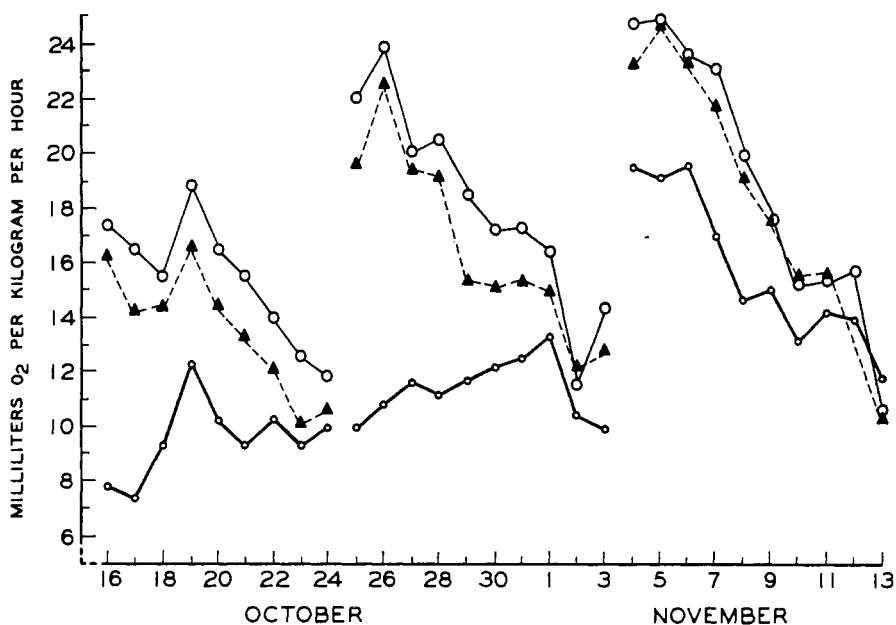


Figure 2. Effect of radiation on respiration of Rome Beauty apples at 70° F. at different picking dates after harvest in 1961

○ control; ▲ 50 krad; ○ 100 krad

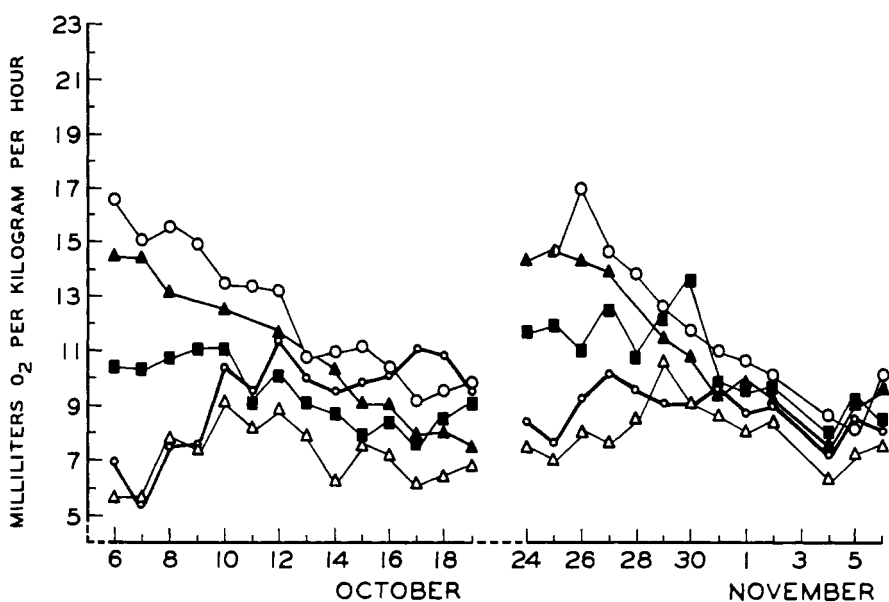


Figure 3. Effect of radiation on respiration of Rome Beauty apples at 70° F. at different picking dates after harvest in 1962

○ control; △ 5 krad; ■ 10 krad; ▲ 50 krad; ○ 100 krad

pothesis that the insoluble protopectin and pectate fractions are broken down to the soluble form by radiation. The soluble to insoluble ratio of these constituents is a good expression of the in vivo changes taking place. The correlation between actual tissue softening (Table I) and changes in soluble to insoluble pectin ratio (Table V) is illustrated.

Poststorage Results. Storage longevity of irradiated fruit, in terms of firmness, is summarized in Tables I, II, and III. In 1961, fruit treated with radiation dosages above 10 krad were more firm following storage than the unirradiated control with Rome Beauty apples, and to a lesser extent, with Cortland apples. The results with McIntosh apples were relatively inconsis-

Table V. Changes in Pectin Fractions of Rome Beauty Apples due to Irradiation and Storage

Treatment		Before Storage						After Storage					
		Specific Viscosity			Anhydrogalacturonic Acid			Specific Viscosity			Anhydrogalacturonic Acid		
Krads	Hours	Fraction	W ^a C + H	Change ^b due to radiation, %	Fraction	W ^a C + H	Change ^b due to radiation, %	Fraction	W ^a C + H	Change ^c due to storage, %	Fraction	W ^a C + H	Change ^c due to storage, %
HARVESTED 10/5/62													
0	..	W 0.144 C 0.127 H 0.568 Av. 0.280	0.207	...	50 42 522	0.091	...	0.263 0.147 0.367 0.259	0.512	+147	209 105 514	0.338	+271
5	6	W 0.152 C 0.158 H 0.616 Av. 0.312	0.194	-6	67 48 540	0.115	+26	0.221 0.133 0.478 0.277	0.362	+87	162 100 508	0.266	+131
10	6	W 0.136 C 0.100 H 0.688 Av. 0.308	0.173	-16	61 37 522	0.110	+21	0.193 0.172 0.457 0.274	0.307	+78	112 106 571	0.182	+66
10	12	W 0.140 C 0.153 H 0.454 Av. 0.249	0.230	+11	49 47 482	0.093	+2	0.201 0.134 0.495 0.277	0.320	+39	138 87 542	0.220	+136
50	6	W 0.308 C 0.190 H 0.458 Av. 0.319	0.475	+129	135 72 503	0.234	+157	0.209 0.195 0.495 0.300	0.303	-36	107 119 581	0.153	-35
100	6	W 0.342 C 0.129 H 0.346 Av. 0.272	0.720	+239	184 67 432	0.368	+304	0.298 0.183 0.274 0.252	0.652	-9	212 122 517	0.332	-10
100	12	W 0.387 C 0.131 H 0.322 Av. 0.280	0.855	+313	188 65 356	0.446	+390	0.275 0.203 0.298 0.259	0.550	-36	210 156 386	0.387	-13
HARVESTED 10/24/62													
0	..	W 0.132 C 0.161 H 0.694 Av. 0.329	0.154	...	46 54 573	0.073	...	0.230 0.160 0.330 0.240	0.469	+205	122 118 514	0.193	+164
5	6	W 0.171 C 0.163 H 0.748 Av. 0.361	0.188	+22	60 44 581	0.096	+31	0.227 0.139 0.425 0.264	0.402	+114	135 91 467	0.243	+153
10	6	W 0.190 C 0.175 H 0.767 Av. 0.377	0.202	+31	68 66 602	0.102	+40	0.253 0.161 0.414 0.276	0.440	+118	135 112 459	0.237	+132
10	12	W 0.203 C 0.176 H 0.650 Av. 0.343	0.246	+55	71 64 557	0.115	+58	0.207 0.143 0.510 0.287	0.317	+29	102 86 581	0.152	+ 32
50	6	W 0.309 C 0.141 H 0.486 Av. 0.312	0.493	+220	140 67 481	0.255	+249	0.234 0.185 0.445 0.288	0.371	-25	147 105 546	0.225	- 12
100	6	W 0.430 C 0.152 H 0.412 Av. 0.331	0.764	+396	166 67 443	0.325	+345	0.320 0.190 0.340 0.283	0.604	-22	197 121 465	0.336	+ 3
100	12	W 0.436 C 0.145 H 0.358 Av. 0.313	0.867	+463	210 75 407	0.435	+495	0.259 0.209 0.424 0.297	0.409	-53	169 126 537	0.256	- 41

^a Fraction W = cold water soluble; C = 0.2% Calgon + 0.8% NaCl soluble; H = 0.05N HCl (80° C.).

^b Expressed in terms of change from control (nonirradiated) ratio.

^c Expressed in terms of change from before storage ratio.

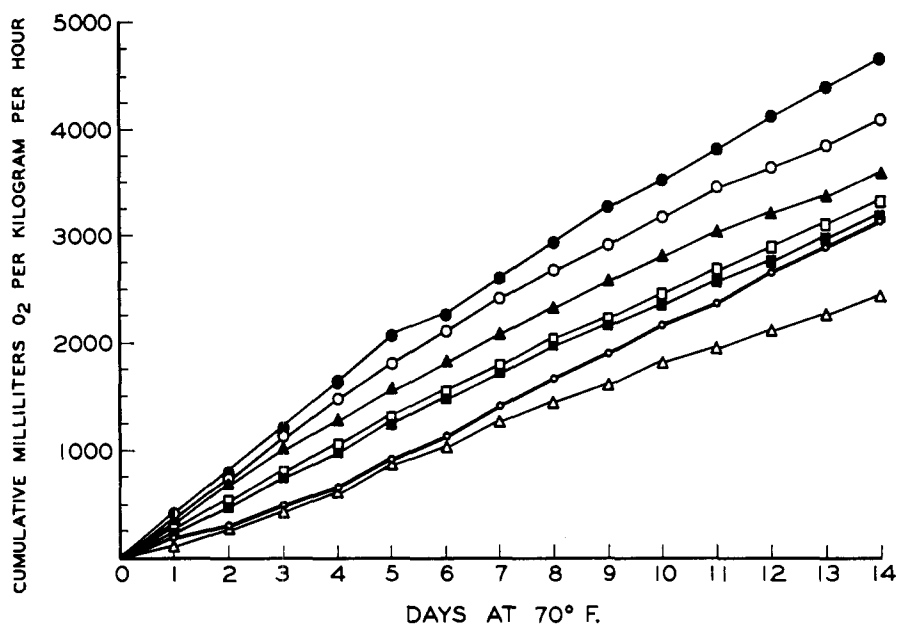


Figure 4. Cumulative respiration of first picking of Rome Beauty apples at 70° F. as influenced by radiation after harvest in 1962

○ control; △ 5 krad, 6 hours; ■ 10 krad, 6 hours; □ 10 krad, 12 hours; ▲ 50 krad, 6 hours; ○ 100 krad, 6 hours; ● 100 krad, 12 hours

tent, but the trend indicated softer than normal fruit. The occurrence of this in-storage decrease in softening rate was totally unexpected, and the phenomenon was examined more closely in 1962.

The results of the second year's tests with Rome Beauty apples again indicated that fruit treated with radiation dosages above 10 krad softened less during storage than did the nonirradiated fruit. As indicated in Table I, despite the softening which occurred during the radiation treatment, irradiation actually resulted in firmer-than-normal fruit at the termination of storage. This modification in softening rate was also reflected in the pectin analyses of this fruit. Although the per cent alcohol-insoluble solids decreased during the storage period, there appeared to be little alteration in change rate due to irradiation. The same also held true of average specific viscosity measurements. Total anhydrogalacturonic acid residues did not change significantly either because of storage or radiation treatment. Again, however, the expected alteration in soluble to insoluble pectin ratio was experienced both as to storage, and as to radiation treatment. However, as indicated in Table V, change in this ratio occurring during storage is the reverse of the immediate change occurring as a result of radiation treatment. The ratio of soluble to insoluble pectins in fruit treated with over 10 krad actually decreased during the storage period as opposed to an obvious increase in the ratio of the unirradiated

controls. This decrease during the storage period was due to a reduction in rate of increase of soluble pectin and in the decrease of insoluble pectin fractions during storage. Thus the normal conversion of insoluble to soluble pectins during storage appears to have been markedly retarded by irradiation.

The radiation treatments resulted in progressively less storage scald with increasing dosage on all three varieties. In 1961, the 100-krad treatment essentially controlled this postharvest disorder except for the first picking of Rome Beauty. Radiation did not control spot scald. If anything, it aggravated the trouble. On the other hand, the scald was so severe on the untreated apples it may have masked this disorder.

Results of the observations on brown core are more difficult to interpret because this term covers a symptom that could be caused by several factors. For example, in McIntosh, true brown core or core flush has a moist, brown discoloration in the core area. In over-mature McIntosh, senile brown core is typified by a brown area at the core with rather dry flesh. These troubles are often hard to differentiate. The data in Table I suggest that radiation reduced true brown core (first picking) in 1961. There was no reduction in senile brown core in the third picking of McIntosh in 1961. In 1962, Rome Beauty had a type of core browning typified by dry, flaky brown tissue at the core region. This trouble was aggravated

by radiation, especially by treatment at high levels.

There were no differences in internal breakdown as a result of radiation in any of the varieties. In 1961, the 50-krad treatment resulted in a small amount of skin injury on the second picking of McIntosh. This injury was typified by a hard, wrinkled, sunken area on the green portions of the skin. In 1961, some of the irradiated Cortlands had a flat taste lacking typical varietal flavor. One treatment (100 krad) resulted in an alcoholic or off flavor on the last picking of this variety.

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Literature Cited

- (1) Beraha, L., Ramsey, G. B., Smith, M. A., Wright, W. R., *Phytopath.* **49**, 91 (1959).
- (2) *Ibid.*, p. 354.
- (3) Beraha, L., Smith, M. A., Wright, W. R., *Ibid.*, **50**, 474 (1960).
- (4) Boyle, F. P., Kertesz, Z. I., Glegg, R. E., Connor, M. A., *Food Res.* **3**, 372 (1957).
- (5) Brownell, L. E., Nehemias, J. V., "Radioisotope Conference 1954," J. E. Johnston, R. A. Faires, R. J. Millett, ed., Vol. 2, Butterworth, London, 1954.
- (6) Dallyn, S. L., Sawyer, R. L., Sparrow, A. H., *Nucleonics* **13**, 48 (1955).
- (7) Glegg, R. E., Boyle, F. P., Tuttle, L. W., Wilson, D. E., Kertesz, Z. I., *Rad. Res.* **5**, 127 (1956).
- (8) Hannan, R. S., "Scientific and Technological Problems Involved in Using Ionizing Radiations for the Preservation of Food," Special Report No. 61, p. 122, Her Majesty's Stationery Office, London, 1955.
- (9) *Ibid.*, p. 146.
- (10) Kertesz, Z. I., "The Pectic Substances," p. 97, Interscience, New York, 1951.
- (11) Kertesz, Z. I., Glegg, R. E., Boyle, F. P., Parsons, G. F., Massey, L. M., Jr., *J. Food Sci.* **29**, 40 (1964).
- (12) LaBelle, R. L., Shallenberger, R. S., Way, R. D., Mattick, L. R., Moyer, J. C., *Food Technol.* **14**, 463 (1960).
- (13) McComb, E. A., McCready, R. M., *Anal. Chem.* **24**, 1630 (1952).
- (14) Nehemias, J. V., Brownell, L. E., Marlin, H. A., *Food Manuf.* **29**, 431 (1954).
- (15) Nelson, K. E., Maxie, E. C., Eukel, W., *Phytopath.* **49**, 475 (1959).
- (16) Phillips, W. R., MacQueen, K. F., "1954-1955 Progress Report on Low Temperature Research (Horticulture Division)," p. 12, Information Division, Canada Department of Agriculture, Ottawa, Ontario, Canada, 1955.

- (17) Phillips, W. R., MacQueen, K. F., Poapst, P. A., *Proc. Intern. Congr. Refrig.* **10th** 3, 176 (1960).
- (18) Pratt, G. B., Ecklund, O. F., *Food Technol.* **10**, 496 (1956).
- (19) Romani, R. J., van Kooy, J., Robinson, B. J., *Food Irradiation* **2**, 11 (1961).
- (20) Salunkhe, D. H., *Econ. Bot.* **15**, 28 (1961).
- (21) Saravacos, G. D., Halzipetrou, L. R., Georgiadou, E., *Food Irradiation* **3**, 6 (1962).
- (22) Smock, R. M., *Cornell Univ. Agr. Expt. Sta. Bull.* **750**, 1948.
- (23) Smock, R. M., Sparrow, A. H., *Proc. Am. Soc. Hort. Sci.* **70**, 67 (1957).
- (24) Sparrow, A. H., Christensen, E., *Nucleonics* **12**, No. 8, p. 16 (1954).

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FOOD FLAVORS

Isolation and Localization of the Precursors of Roasted Peanut Flavor

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This work represents initial attempts to isolate and identify precursors of roasted peanut flavor. The techniques used for these studies revealed useful information concerning the locale and molecular origin of flavor precursors within peanut cotyledons. Flavor does not appear to arise from the large globular proteins nor from carbohydrate material per se. Flavor does arise from one or a combination of ninhydrin-positive compounds and basic compounds. Apparently, flavor originates from rather specific types of micro-molecules rather than from general, macromolecular, cellular components such as the large globulin proteins and the starches.

VERY little is known about the compounds responsible for the typical flavor of roasted peanuts or about the precursors which give rise to flavor during roasting. At the time of a review by Hoffpauir (4), a few of the components of gases given off during roasting had been identified, and a measurable decrease in total sugars during roasting had been noted (6). Gaseous components identified included derivatives of tetrahydrofuran, ammonia, hydrogen sulfide, and diacetyl. The major gaseous component was carbon dioxide. Also, Pickett and Holley (6) had shown that mixtures of amino acids and carbohydrates reacted to produce tetrahydrofuran derivatives along with noticeable browning and discernable aromas. On the basis of the meager information available, Hoffpauir speculated on the precursors responsible for the formation of peanut flavor during roasting. The two main protein components of cotyledons, arachin and conarachin, which have been classified as reserve proteins (7), were implicated as precursors on the basis of their unusually high sulfur content. The implication was made to explain the presence of sulfide in roasting gases. Sucrose was implicated as a precursor on the basis of the loss of total sugars during roasting and on the basis of the appearance of

tetrahydrofuran derivatives. Peanut cotyledons contain about 4.5% total disaccharides of which sucrose is the major component. Protein-bound and free amino acids were implicated for similar reasons. Since 1953, nothing in the literature had added significantly to this sketchy picture.

Work reported here represents initial results of an integrated program to improve peanut quality by identifying the precursors which give rise to typical peanut flavor during roasting.

Reagents

Optical hexane was prepared by distilling high purity *n*-hexane over KOH pellets onto a silica gel column. The hexane collected from the column lacked absorption in the 230- to 260- μ range compared to air.

Sephadex gels, obtained from Pharmacia, Uppsala, Sweden, were swollen, washed, and packed on columns according to the procedures outlined by Fasold, Grundlach, and Turba (3).

Refined cottonseed oil was obtained from the Great Western Foods Company, Fort Worth, Texas.

Carbon tetrachloride, reagent grade, was used for adjustment of the specific gravity of the cottonseed oil.

Spray reagents used for detection of

spots on paper chromatograms were: 0.2% ninhydrin in water-saturated *n*-butanol; equal volumes of 0.1*N* silver nitrate and 5*N* ammonium hydroxide; and 0.4% bromocresol green in 95% ethanol. Chromatograms were developed on Whatman No. 1 filter paper in *n*-butanol-acetic acid-water (8:2:2) solvent.

Procedures

Roasting Studies. Roasting data were obtained by placing 38 grams of raw peanuts in the wire basket of an electrically heated rotisserie (General Electric) preheated to the desired temperature. Heating was continued until roasting was complete as judged by color and taste. If an under- or over-roast was indicated, the process was repeated until a satisfactory roast was obtained. In one case, peanuts of Starr variety were roasted by placing them in the roaster at room temperature and increasing the temperature to the desired level as rapidly as possible. Logarithms of the time necessary for complete roasting of three varieties of Spanish peanuts, Starr, Spantex, and Argentine, were plotted versus the inverse of temperatures used.

Roasting Individual Fractions from Fractionation Studies. Individual frac-