

The Properties and Performance of Poly AOTM-79; A Nonabsorbable, Polymeric Antioxidant Intended for Use in Foods¹

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ABSTRACT AND SUMMARY

The physical, chemical, and biological characteristics of Poly AOTM-79, a composition selected from a class of polymers prepared by the polycondensation of divinylbenzene and a blend of various sterically hindered phenols and hydroquinone, are discussed. Data are presented indicating that Poly AOTM-79 demonstrates excellent antioxidant activity in stabilizing vegetable oils when compared to monomeric food grade antioxidants [butylated hydroxytoluene (BHT), butylated hydroxyanisole (BHA), and tertiary butylhydroquinone (TBHQ)] as measured by the active oxygen method (AOM) and oxygen bomb test. Stability tests using thermogravimetric analysis indicate that the material is not depolymerized in the presence of air at temperatures up to 300 C while BHT, BHA, and TBHQ are completely volatilized at 150-170 C. Using ¹⁴C-radiolabeled antioxidants, the physical losses of Poly AOTM-79 from frying oil heated to 190 C is nil while 65% and 80% losses are incurred with BHA after 1 and 2 hr holding time, respectively. The physical carry-through of Poly AOTM-79 onto potatoes from frying oil at 190 C is quantitative as compared to ca. 20% losses with BHA and TBHQ. Sensory panel tests reflect that the carry-through of Poly AOTM-79 is in an active form as evidenced by the prolonged shelf-life of chips fried in oil containing Poly AOTM-79 as compared to BHA and TBHQ. Rats and mice orally dosed with ¹⁴C-radiolabeled antioxidants indicate that Poly AOTM-79 is virtually not absorbed from the gastrointestinal tract (ca. 0.2-0.6%) while monomeric antioxidants are readily transported (80-100%). Acute oral and intraperitoneal toxicity tests indicate that Poly AOTM-79 is without effect at the highest dose tested (10 g/kg body weight).

INTRODUCTION

In a previous report, the physical, chemical, and biological properties of a series of oil soluble, sterically hindered phenol-containing polymers were described (1). Data were presented demonstrating that as a class these compositions were (a) effective antioxidants for edible fats and oils, and (b) not absorbed from the gastrointestinal tract as a function of increasing molecular weight. The class of polymeric antioxidants described are such that a great deal of latitude may be employed in the selection of suitable functional species for incorporation into the polymer chain. The synthetic methods also permit a multiplicity of functional species to be employed as comonomers and simultaneously condensed into a chain. In addition, the ratios of the functional species can be varied with respect to one another. This leads to unique compositions in which functional species reacting with singlet oxygen and others acting as effective traps for peroxide and hydroperoxide initiators can be incorporated into a single macromolecule. This flexibility in structural design can result in a product having versatility in numerous applications heretofore

obtainable only by using blends of several chemically discrete monomeric antioxidants.

In this study, the properties and performance of Poly AOTM-79, a polymeric antioxidant selected from numerous examples screened, are described and discussed with relevance to safety and use in foods.

EXPERIMENTAL PROCEDURES

Materials

The monomeric food grade antioxidants utilized for comparative purposes in this study included: butylated hydroxytoluene (BHT), butylated hydroxyanisole (BHA), and tertiary butylhydroquinone (TBHQ) (Eastman Chemical Products, Kingsport, TN). Poly AOTM-79 was synthesized by the aluminum catalyzed polycondensation of divinylbenzene (DVB) with a multicomponent blend of comonomers containing, principally, *p*-tert-butylphenol, *p*-hydroxyanisole and mono-*tert*-butylhydroquinone (2-4).

Glycerol monooleate (GMO) used to prepare solution concentrates of commercial antioxidants and of Poly AOTM-79 was antioxidant-free (Emery Industries, Cincinnati, OH). Vegetable oils, free of added stabilizers, were obtained from various commercial sources; naturally occurring tocopherols were not removed. Freshly distilled, unstabilized lemon essence oil (LEO) was generously supplied by Warner-Jenkinson Mfg. Co. (St. Louis, MO).

Radiolabeling

All monomeric antioxidants were uniformly ring-labeled with C-14 utilizing barium carbonate-¹⁴C as the starting material. The Poly AOTM-79 employed for carry-through studies was radiolabeled by utilizing uniformly ring-labeled 4-hydroxyanisole-¹⁴C as a component in the polycondensation. Alternatively, Poly AOTM-79 utilized for absorption studies was synthesized using a mixture of *meta* and *para* isomers of divinylbenzene (DVB) and ethylvinylbenzene (EVB) labeled in the methylene group with C-14. The specific activity of the antioxidants varied depending on the intended use (see below).

Thermogravimetric Analysis (TGA)

TGA was conducted on powdered samples of each antioxidant using the Rigaku instrument. The temperature programming was 5 C/min; analyses were conducted in air.

Antioxidant Activity

Antioxidant activity in vegetable oils was determined using the Official AOCS Active Oxygen Method (AOM) and results expressed as the number of hours required to reach 100 mEq of peroxide per kg of oil (5). The activity of various antioxidants in stabilizing lemon essence oil (LEO) was determined using the Oxygen Bomb Test (6).

Radiolabeled Carry-Through Tests

The carry-through characteristics of BHA, TBHQ, and Poly AOTM-79 onto potatoes during frying operations were determined using uniformly ring-labeled C-14 radiolabeled antioxidants. The specific activity of the antioxidants were: BHA = 0.5 μ Ci/mg; TBHQ = 9.34 μ Ci/mg; Poly AOTM-79 =

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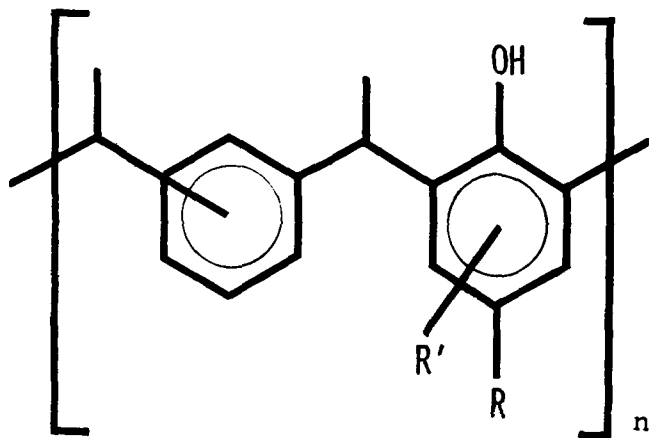


FIG. 1. The general structural formula of Poly AOTM-79. R = -OH, -OMe or alkyl group and R' = H or alkyl group.

4.7 $\mu\text{Ci}/\text{mg}$.

The antioxidants were incorporated into a blended cottonseed/soybean oil at 200 ppm. The oils were heated to 190 C (375 F) and samples of oil radioassayed as a function of time to determine volatility at typical frying temperatures. In a separate experiment, freshly prepared potato sections measuring 1.0 cm in diameter x 0.65 cm thick were fried at 190 C for 3 min; repetitive fryings were conducted to determine the variability of carry-through. After excess oil was drained from the fried sections, the potato was exhaustively extracted with diethyl ether to recover the absorbed oil and antioxidants, the ether evaporated, and the residue weighed prior to radioassay. Subsequently, the residues were dissolved in 10 ml of Insta-Gel[®] (Packard Instruments, Inc., Downers Grove, IL) and counted in the Packard Tri-Carb Model 3385. In this fashion, the antioxidant concentration in the frying bath was compared to the concentration in the oil absorbed by the potato during frying.

Organoleptic Carry-Through Tests

The organoleptic carry-through performance of BHA, TBHQ, and Poly AOTM-79 onto potato chips from frying oil was performed using "cold" (nonradiolabeled) antioxidants. Chips were prepared from freshly sliced potatoes and fried for 3 min at 190 C (375 F) in a 2 liter capacity deep-frier. After excess oil was drained off, the chips were crushed, ground in a Waring blender, and 25 g quantities loaded into 250 ml Erlenmyer flasks fitted with loose-fitting aluminum foil caps. Duplicate samples were subjected to accelerated oven-aging tests at 120 C using the method described by Smouse (7). In the method, sufficient samples (flasks) are prepared and introduced into a forced-draft oven at the beginning of the test to permit the retrieval of at least one sample (preferably two) every 30 min over a period of 4 hr. This represents no less than eight flasks per treatment. In our tests, chips prepared from replicate fryings were each tested in duplicate. After all samples were equilibrated at room temperature, the thermally stressed chips were compared to freshly prepared, unstressed chips for undesirable odor development using a sensory panel consisting of ten members. The samples were presented to each panelist in the same flasks used to conduct accelerated oven-aging tests. The flasks were randomly coded and the contents shielded from view by the panel to avoid the possible influence of color. The unstressed control was identified to the panelists for purposes of establishing a standard of acceptability. The panelists were polled in a forced choice manner and asked to indicate whether the stressed sample was acceptable or unacceptable in odor as compared to the control. Results

from the panel tests were averaged, expressed as hours to reach unacceptable odor development at 120 C, and this related to shelf-life in days at room temperature from a linear correlation found valid up to 3.5 hr at 120 C (7).

Absorption and Acute Toxicity Studies

The absorption of ¹⁴C-radiolabeled Poly AOTM-79 from single oral doses was studied in rats and mice using procedures described previously (1). For these studies Poly AOTM-79 synthesized using a mixture of *meta* and *para* isomers of DVB and EVB radiolabeled in the methylene group with C-14 was employed. The ¹⁴C-Poly AOTM-79 utilized in this study had a specific activity of 0.4 $\mu\text{Ci}/\text{mg}$. This was dissolved in Wesson oil (Hunt-Wesson Foods, Fullerton, CA) at a final specific activity of 4 $\mu\text{Ci}/\text{ml}$ for dosing rats and 1.46 $\mu\text{Ci}/\text{ml}$ for mice. Rats received single oral doses of Poly AOTM-79 in 1.0 ml of oil via stomach tube while mice were dosed with 0.5 ml quantities.

Single dose, oral and intraperitoneal acute toxicity studies were conducted in rats and mice using unlabeled Poly AOTM-79 in oil at a maximum level of 10 g/kg of body weight.

RESULTS AND DISCUSSION

While it is beyond the scope of this study to detail the chemistry and synthesis of the class of polymers represented by Poly AOTM-79, some general comments are nonetheless appropriate for a better understanding of some of the unique characteristics observed. As noted earlier, the polymers are prepared by the aluminum catalyzed polycondensation of divinylbenzene (DVB) with various substituted phenols and hydroquinones (2-6). Critical to the synthesis is the stoichiometry of DVB to the sum of the phenolic or hydroquinone constituents without which polymerization to desirably high molecular weights is not achieved. As illustrated by the general structural formula in Figure 1, typical polymers may contain substituents in which R can be either a phenolic, methoxy, or short chain alkyl group and R' a hydrogen or short chain ethyl group.

It is important to recognize the similarities and unique differences in the nature of the derivatives possible in polymers of this class as compared to monomeric counterparts. In common with some monomeric antioxidants, the hydroxyl group(s) of the phenolic or hydroquinone constituents within the polymer chain are sterically hindered. The use of repeating ethylidene groups as the bridging unit provides steric effects akin to the tertiarybutyl group commonly employed in some monomeric antioxidants. With monomeric antioxidants, the substituted phenolic or hydroquinone moiety is restricted to one component per molecule; two components should the substance be a bisphenol. However, when this is carried forward in a precisely controlled, catalyzed polycondensation with DVB, more than one comonomer can be simultaneously condensed and thereby incorporated into the polymer chain. Further, by strict control of the phenolic or hydroquinone constituents, the ratios can be varied to increase or reduce their concentration relative to one another within the polymer chain. In the case of Poly AOTM-79, the comonomers condensed with DVB are a multicomponent blend containing, principally, *p*-tertiarybutylphenol, *p*-hydroxyanisole, and mono-tertiary butylhydroquinone. Conceptually, this approach leads to interesting compositions in which the antioxidants have the potential for demonstrating multiple modes of action; i.e., species such as hydroquinone which react with singlet oxygen and sterically hindered phenols which are effective traps for initiators leading to the production of undesirable peroxides and hydroperoxides. While this is in part the case, it will be seen that Poly AOTM-79 also demonstrates some

TABLE I

Some Physical/Chemical Characteristics of Poly AOTM-79

Appearance	Off-white powder
Elemental analysis: ^a	
Theoretical	C, 82.77; H, 7.73; O, 9.50%
Typical	C, 83.11; H, 7.88; O, 9.01%
mEq active phenols/g	1.72
Bulk density	>0.5 g/cc
Particle size	180-710 μ
Moisture content; Karl Fischer	<1.0%
UV spectrum:	
λ_{\max}	286 nm
$\epsilon_{\lambda_{\max}}$	13.5 (g/l) ⁻¹ cm ⁻¹
$\epsilon_{264 \text{ nm}}$	6.95 (g/l) ⁻¹ cm ⁻¹
Molecular weight:	
Peak average ^b	ca. 5000
wt % <500 ^c	≤0.5
wt % <10 ⁴	≤30.0

^aCalculated for (C₃₅H₃₉O₃)_n.

^bGel permeation chromatography (GPC) peak MW relative to a polystyrene calibration.

^cSum of monomers, dimers, and trimers.

unique properties exclusively due to its macromolecular character.

The principle physical/chemical characteristics of Poly AOTM-79 are indicated in Tables I and II. Some key features worthy of comment include the following.

- The peak average molecular weight as determined by gel permeation chromatography (GPC) relative to polystyrene calibration is ca. 5000 (1). A typical GPC scan for Poly AOTM-79 is shown in Figure 2. In order to limit gastrointestinal absorption, care is exercised to minimize the weight fraction below molecular weight 500 and to assure control to ≤0.5 wt %. Molecular weight is strictly controlled during polymerization and subsequent processing.
- From the structure (Fig. 1), it is obvious that unlike certain monomeric antioxidants, Poly AOTM-79 contains one equivalent of aromatic moiety which does not contribute antioxidant activity to the polymer since it is void of functionally reactive groups. Accordingly, the total milliequivalents (mEq) of active phenols and hydroquinones per gram of antioxidant, as determined by a combination of proton and oxidative titration with potassium dichromate, is 1.72 mEq/g for Poly AOTM-79 as compared to 4.55 for BHT, 5.56 for BHA, and 6.02 (HQ) for TBHQ. In evaluating Poly AOTM-79 vs. currently approved, food grade antioxidants on a weight basis, the multiples shown in Table III should be taken into consideration. At the 200 ppm level, some 530-700 ppm of Poly AOTM-79 should be considered depending on the monomeric antioxidant to which it is being compared.
- The solubility of Poly AOTM-79 in various solvents is shown in Table II. The solubility in vegetable oils is quite favorable, some care must be exercised in solubilizing powdered Poly AOTM-79 directly in viscous fats and oils since only the surface tends to wet thereby forming gums which reduce the rate of dissolution. For this reason and for expediency in dispensing ppm levels, solution concentrates (typical of industry practice) in which Poly AOTM-79 is solubilized at >10% by weight in 70:30 GMO/Corn oil can be employed. Self-emulsifiable concentrates are readily prepared by solubilizing Poly AOTM-79 in Polysorbate 20.

TABLE II

Solubility of Poly AOTM-79

Solvents	g/100 ml, 25 C
Water	insoluble
Glycerine	insoluble
Propylene glycol (PG)	insoluble
Hexane	0.022
Ethanol	> 20
Diethyl ether	> 100
Chloroform	> 100
Dioxane	> 100
Tetrahydrofuran	> 100
Benzene	> 100
Acetone	> 100
Glycerol monooleate (GMO)	ca. 10
Corn oil	ca. 20
Cottonseed oil	ca. 20
Soybean oil	ca. 20
Cottonseed/Soybean blend	ca. 20
Palm oil, refined	ca. 20
Peanut oil	ca. 1.0
70:30/GMO: Corn oil	> 10
67:29:4/GMO: Corn oil: PG	ca. 5
65:28:4:3/GMO: Corn oil: PG: Citric acid	ca. 5

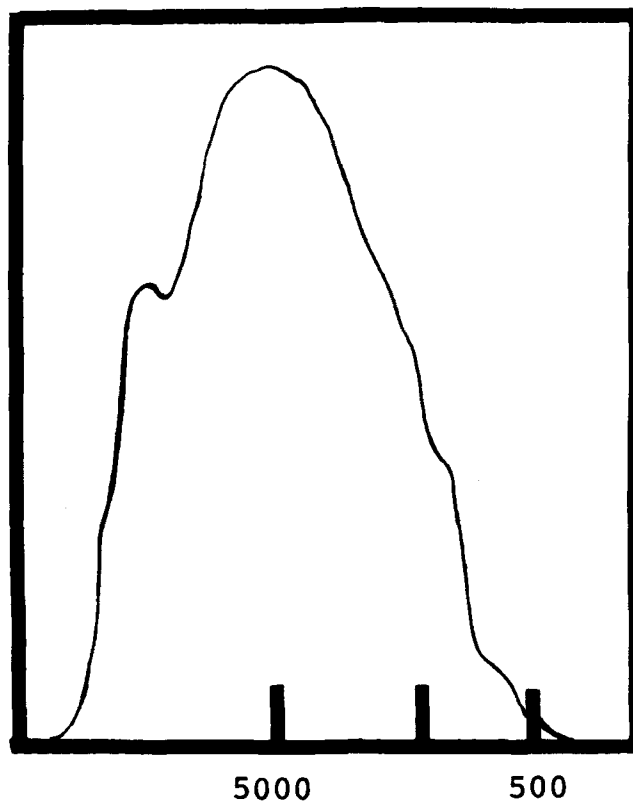


FIG. 2. Typical gel permeation chromatography scan of Poly AOTM-79 relative to polystyrene calibration; refractive index detector.

These form excellent, heat stable emulsions upon addition of water. Finally, the partitioning of Poly AOTM-79 between oil and aqueous phases is essentially nil; it is not extracted from diethyl ether solution by aqueous base (20%).

- As shown in Figure 3, TGA in air indicates that Poly AOTM-79 is exceptionally stable to thermal stress and is not depolymerized at temperatures to 300 C. For comparative purposes, BHT, BHA, and TBHQ show complete volatilization in air at temperatures of 150-190 C.

The activity of Poly AOTM-79 in vegetable oil (Fig. 4)

TABLE III

Equivalency Factors for Comparing Poly AOTM-79
to Other Food Grade Antioxidants on a Weight Basis

Antioxidants ^a	mEq OH/g	Multiples for Poly AOTM-79	ppm Poly AOTM-79 for 200 ppm equivalency
Poly AOTM-79	1.72	-	-
BHT	4.55	2.65	530
BHA	5.56	3.23	646
TBHQ	6.02 (HQ)	3.50	700

^aBHT = butylated hydroxytoluene; BHA = butylated hydroxyanisole; TBHQ = tertiary butylhydroquinone.

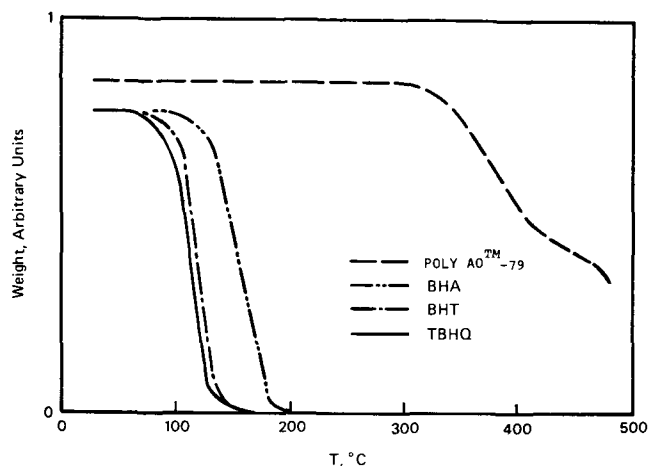


FIG. 3. Comparative thermogravimetric analysis of BHA, BHT, TBHQ, and Poly AOTM-79 in air; heating rate - 5 C/min.

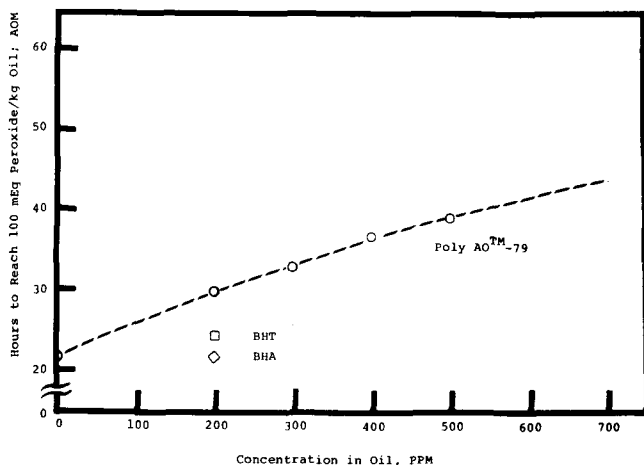


FIG. 4. Comparative antioxidant activity of BHT, BHA, and Poly AOTM-79 in a cottonseed/soybean oil blend; active oxygen method (AOM).

and lemon essence oil (Fig. 5) compares favorably with some of the food grade monomeric antioxidants currently employed by the food industry. At the 200 ppm level in vegetable oil, activity is comparable to BHT and BHA. This is somewhat surprising in view of the lower active phenol content of Poly AOTM-79 and is probably due to the hydroquinone content. As the concentration of Poly AOTM-79 in oil is increased to 500-700 ppm matching 200 ppm BHT and BHA in terms of the mEq of active phenol/gram, activity as measured by the AOM increases significantly. We reason that the particular blend of active constituents leashed to one another in the Poly AOTM-79 polymer chain significantly affects activity and this is not readily explicable in terms of active functional groups.

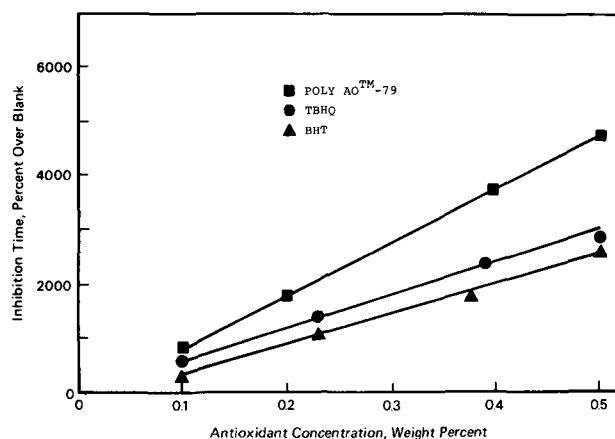


FIG. 5. Comparative activity of BHT, TBHQ, and Poly AOTM-79 in lemon essence oil. Oxygen bomb test at 100 C and 65 psig O₂.

Similar results are obtained using unstabilized LEO as substrate in the oxygen bomb test (Fig. 5). However, in this instance the concentration of polymeric or monomeric antioxidant required to achieve acceptable levels of stabilization are on the order of 0.1-0.5% by weight. The latter concentration of antioxidant (0.5%) is typical of flavor industry practice and within levels prescribed under the U.S. Code of Federal Regulations for BHA (8).

While the AOM and oxygen bomb tests provide objective bench marks for assessing the relative activity of antioxidants, the experimental procedures rarely take into account those stresses encountered in some food processing operations. For example, perhaps the most severe thermal stress imposed on food grade antioxidants is encountered during frying, baking, and high-temperature drying operations. For the most part, such high-temperature thermal stresses result in antioxidant losses due to volatilization. While volatilization losses can be predicted from published vapor pressure data for BHA, BHT, and TBHQ (9,10), more direct evidence is available. During drum drying operations employed in the manufacture of dehydrated potato flakes, 65-85% losses of BHT and BHA are experienced (11). Similar losses are incurred during the baking of pie crust (12). Interestingly, very recent studies show that significant losses of monomeric, food grade antioxidants readily occur during the freeze-drying of emulsions designed to simulate high lipid food (13). The net result is a reduction in process stabilization and subsequently, product shelf-life.

Using the frying of potatoes as a model system in which high-temperature thermal stresses affect both process stabilization and product shelf-life, the performance of monomeric antioxidants and Poly AOTM-79 were compared. As indicated in Figure 6, the depletion of ¹⁴C-BHA as a function of time from vegetable oil held at frying temperatures (190 C) is considerable; ca. 65% loss after 1 hr and 80% after 2 hr. With ¹⁴C-Poly AOTM-79, volatilization losses are nil. Predictably, this situation leads to the loss of

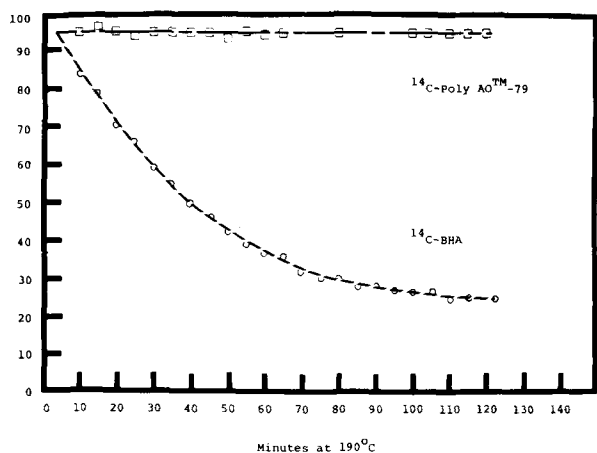


FIG. 6. Depletion of ¹⁴C-BHA and ¹⁴C-Poly AOTM-79 from cottonseed/soybean oil blend heated at 190 C (375 F).

TABLE IV

Carry-Through of ¹⁴C-Radiolabeled Antioxidants onto Potatoes from Frying Oil at 190 C (375 F)

No. of fryings	Percent carry-through of antioxidant ^a		
	¹⁴ C-BHA	¹⁴ C-TBHQ	¹⁴ C-Poly AOTM-79
1	79-84	80-85	96-99
2	83-87	78-81	96-98
3	87-90	83-85	95-96
4	83-85	80-82	95-97
12	-	80-83	-
20	78-82	-	96-98

^aRange; three samples/frying. See Table III for identification of abbreviations.

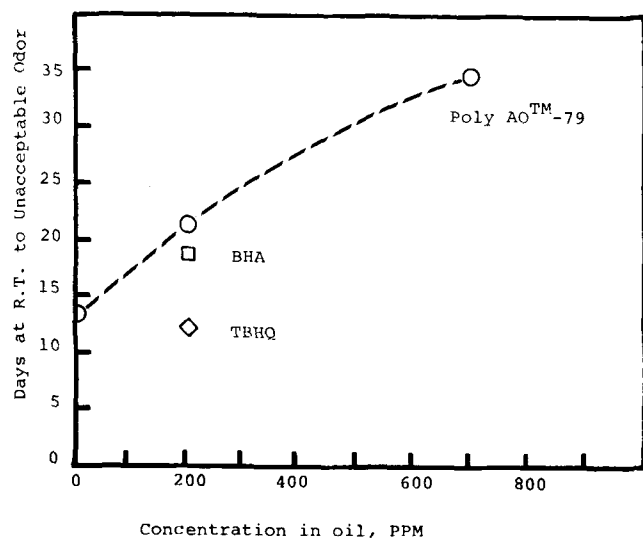


FIG. 7. Functional carry-through performance of antioxidants onto potato chips subjected to accelerated oven-aging at 120 C and related to shelf-life in days at room temperature.

carry-through of BHA and TBHQ onto that portion of vegetable oil absorbed by food products during frying since the water vapor generated at the surface codistills with significant portions of antioxidants. As indicated in Table IV, repetitive frying of potato sections in vegetable oil containing ¹⁴C-radiolabeled antioxidants clearly shows that some 20% losses in physical carry-through of BHA and TBHQ are incurred. With Poly AOTM-79, the carry-through is essentially quantitative.

While radiolabeling studies accurately compare the rela-

TABLE V

Comparative Acute Toxicity of Various Antioxidants

Antioxidants ^a	Route	Species	LD ₅₀ (g/kg)
BHT	Oral	Rat	1.9-2.4
BHA	Oral	Mouse	2.0
	Oral	Rabbit	2.1-3.2
BHA	Oral	Rat	4.1-5.0
	Oral	Mouse	1.2-2.0
PG	Oral	Rat	2.6-3.8
	IP	Rat	0.38
	Oral	Mouse	1.7-2.0
NDGA	Oral	Rat	2.0-5.0
	Oral	Mouse	2.0-4.0
	IP	Mouse	0.55
Poly AOTM-79 ^b	Oral	Rat	>10.0
	IP	Rat	>10.0
	Oral	Mouse	>10.0
	IP	Mouse	>10.0
	Oral	Dog	>10.0

^aPG = propyl gallate; NDGA = nordihydroguaiaretic acid. See Table III for other abbreviations.

^bNo deaths in any of these maximum doses tested.

TABLE VI

Absorption of ¹⁴C-Radiolabeled Antioxidants in Rats

Antioxidant ^a	% Oral dose absorbed
BHT	80-90
BHA	82-100
TBHQ	82-88
Poly AOTM-79	0.2-0.6
	0.4 (mice)

^aSee Table III for identification of abbreviations.

tive "chemical" carry-through efficiency of monomeric antioxidants and Poly AOTM-79 onto food from frying operations, they do not indicate whether the stabilizers have been transferred to the food (potatoes) in an active form suitable for extending product shelf-life. Understandably, the antioxidants have ample opportunity to be spent during the high-temperature processing conditions. By frying potato chips in oil stabilized with various antioxidants (in this instance, unlabeled) and subjecting samples to accelerated oven-aging followed by sensory panel tests, "functional" carry-through can be assessed. As indicated in Figure 7, the functional carry-through of antioxidant at 200 ppm in oil follows the order: Poly AOTM-79 > BHA >> TBHQ. Apparently, TBHQ is readily spent during vigorous frying operations employed in our tests and while TBHQ is no doubt valuable in extending the life of frying oils, very little is left in an active form to subsequently impart product shelf-life. Again, by increasing the concentration of Poly AOTM-79 in oil to 700 ppm in order to match 200 ppm BHA in terms of mEq active phenol/gram, functional carry-through is significantly enhanced.

Turning to preliminary safety tests, the acute toxicity of Poly AOTM-79 appears exceedingly favorable. As indicated in Table V, it was not possible to determine the oral and intraperitoneal LD₅₀ of Poly AOTM-79 since there were no deaths in any of the animal models utilized at the highest dose tested (10 g/kg). By comparison, BHT, BHA, propyl gallate (PG) and nordihydroguaiaretic acid (NDGA) have reported acute toxicities at significantly lower doses. No doubt, the lack of transport of Poly AOTM-79 as compared to monomeric food grade antioxidants (Table VI) has some influence on the lack of observed toxicity. Long term chronic and subchronic feeding studies are planned with the view of establishing a complete safety profile for Poly AOTM-79.

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