

Vaporization Behavior of Antioxidant Additive BHT in Polypropylene by Heating

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SYNOPSIS

The vaporization behavior by heating of an antioxidant additive 2,6-di-*tert*-butyl-*p*-cresol (BHT) in polypropylene (PP) was studied. BHT vaporized from PP plates under the stream of air at 70–100°C for 0–3000 min was absorbed to the Tenax GC adsorbent column, and the percentage recoveries of BHT increased linearly with the square root of time: $t^{1/2}$. There were no considerable differences between the vaporization behavior of BHT by heating under the stream of air and that of nitrogen. BHT does not act as an antioxidant at temperature below 120°C. The percentage recoveries of BHT from PP plates at 90–120°C under a nitrogen stream increased linearly with $t^{1/2}$ until about 70% of the recovery. At temperature below 80°C, the linear relations still held. The vaporization also proceeded at the temperature below the melting point of BHT. Both the sample thickness and the temperature greatly influenced the vaporization rate of BHT in PP, and the Arrhenius plots of the logarithmus of the specific rate vs. the reciprocal of the absolute temperature at 70–120°C showed an approximately straight line. Triolein coexisted with interrupted BHT vaporization. However, the PP plates containing 0.2 wt % of olive oil showed the same vaporization behavior of BHT as that of the original PP plates free from oils at 80 and 100°C. The decreasing rate of BHT in PP by heating under air obeyed first-order kinetics. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

To protect polymers from oxidation and decomposition reactions by molecular oxygen, some aromatic hydroxy compounds are usually added to them.¹ 2,6-Di-*tert*-butyl-*p*-cresol (BHT) of relatively high vapor pressure has been generally used as an antioxidant additive of polyolefins. Migration of a very small amount of BHT in polypropylene (PP) to some kinds of foods containing large quantities of fats or oils is well known.^{2,3}

In a previous paper,⁴ the authors reported the concentration distribution of BHT in PP after heating for a long period of time in succession using the rolled film method. The diffusion coefficient and the activation energy were estimated assuming that the diffusion process obeys Fick's law.

It would be, thus, valuable to study whether BHT is removed by vaporization from the polymer surface without decomposition or whether BHT is partly transformed to other ones due to the thermal oxidation by air penetrated from the polymer surface.

In this paper, we report on the time course of vaporization of BHT initially uniformly distributed in PP under air or an inert gas, nitrogen, using an apparatus equipped with a Tenax GC adsorbent column. We also report on the factors affecting the rate in BHT vaporization. The decreasing rate of the mean BHT concentration in PP by heating was determined in air at a few temperatures in order to make clear the mechanism of the lowering of the BHT content in PP.

EXPERIMENTAL

Reagent

An antioxidant additive BHT as a standard material for the gas-liquid chromatographic determination

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was purchased from Tokyo Kasei Kogyo Co. and was used without further purification.

A triolein was purchased from Tokyo Kasei Kogyo Co. The olive oil free from BHT was commercial grade. The commercial PP pellet, Noblen W101 HEZ 103, which was used to make the PP plate, was purchased from Sumitomo Chemical Industry Co. The PP was a homopolymer and contained BHT.

The Tenax GC adsorbent, 80–100 mesh, Lot. No. 0606-C17, was purchased from Gasukuro Kogyo Co. The commercial PP film of 0.003 cm in thickness was supplied by Tonnen Film Co. This film initially contained BHT of $7.1 \times 10^2 \text{ mg kg}^{-1}$ and had a crystallinity of 0.54. The X-ray profile of this film did not show any orientation. Other reagents were all analytical grade.

Apparatus

The gas-liquid chromatograph used in this study was a Shimadzu Model GC 7A gas chromatograph equipped with a flame-ionization detector. The vaporization-collection system shown in Figure 1 was put into a heating oven equipped with a pass of dried air or nitrogen. This oven was controlled within $\pm 0.1^\circ\text{C}$. One of the carrier gases, dried air, was led from a Hitachi Model YEFOU air compressor equipped with the silicagel drying chamber, and nitrogen gas was led from the cylinder. The other heating oven used to heat the PP plates was a Sanyo model MOV heating oven (temperature precision within $\pm 1^\circ\text{C}$).

Processing of PP Pellet to Plate

The PP plates of $150 \times 150 \times 2 \text{ mm}$ were prepared by operating a Nissei Jushi Industry Co. Model PS 60E SE injection-molding machine. The processing conditions were as follows: resin temperature, 235°C ; injection pressure, ca. 1200 kg cm^{-2} ; mold temperature, 40°C ; and injection speed, 1 cycle/40 s.

Plates of $110 \times 6 \times 2 \text{ mm}$ (A) and $55 \times 110 \times 2 \text{ mm}$ (B) were prepared from the plates. The content of BHT was determined as $9.7 \times 10^2 \text{ mg kg}^{-1}$ in accordance with our method.⁵

Vaporization of BHT

1. The two plates of sample (A) that were inserted into a vaporization tubing as is shown in Figure 1. The plates untreated and those preliminary immersed into the olive oil at 20°C for 44 days in the dark were used. A 2 g of the PP films of 0.003 cm in thickness and of $20 \times 10 \text{ mm}$ in size was inserted into the apparatus.

After equipping with the Tenax GC column activated at 250°C for 24 h under a nitrogen stream, the glass tubing was put into the heating oven at $55\text{--}120^\circ\text{C}$, controlled within $\pm 0.1^\circ\text{C}$, and immediately allowed to pass nitrogen gas or dried air at a flow rate of 30 mL min^{-1} . BHT vaporized under these conditions was collected quantitatively in the Tenax GC adsorbent column. A series of va-

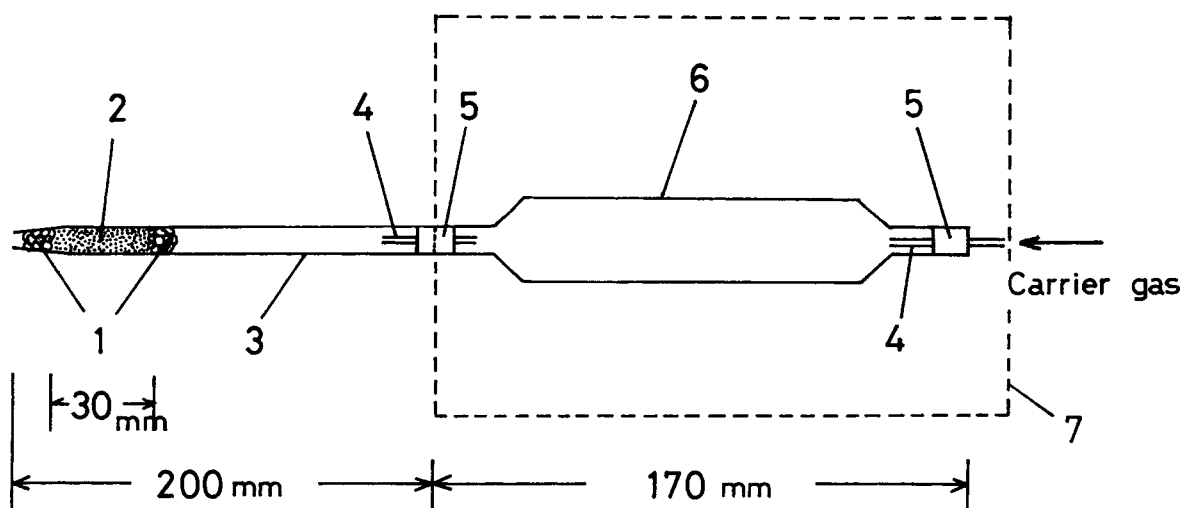


Figure 1 Vaporization-collection system of BHT in PP or oil: (1) glass wool; (2) Tenax GC adsorbent; (3) glass tubing; (4) stainless-steel tube; (5) silicone gum; (6) glass sample tubing; (7) heating oven.

porization processes were successively carried out by exchanging with a newly activated one.

2. One milliliter of the *n*-hexane solution containing 3 mg of BHT and 0, 0.03, 0.3, 3.0, 30, and 300 mg, respectively, of triolein was taken into the vaporization tubing, and *n*-hexane was removed by heating indirectly with the hot air. After equipping with the Tenax GC column, the vaporization procedure of BHT in triolein was done similarly to that for the PP plate or the film at $55\text{--}100 \pm 0.1^\circ\text{C}$.
3. About 10 plates of sample (B) were put into the heating oven at 80°C for 52, 104, 195, 408, and 600 h, and at 100°C for 25, 50, 117, and 200 h, respectively, in air. After heating, the content of BHT in a sample was determined by gas-liquid chromatography.⁵

Determination of Antioxidant Additive BHT Content

The BHT solution for a solid PP material was prepared according to the previous paper.⁵ A PP plate cut into about 5 mm square was refluxed with 100 mL of *n*-hexane for over 2 h at 80°C , and the filtrate was evaporated under vacuum until just before *n*-hexane completely disappeared and then was dis-

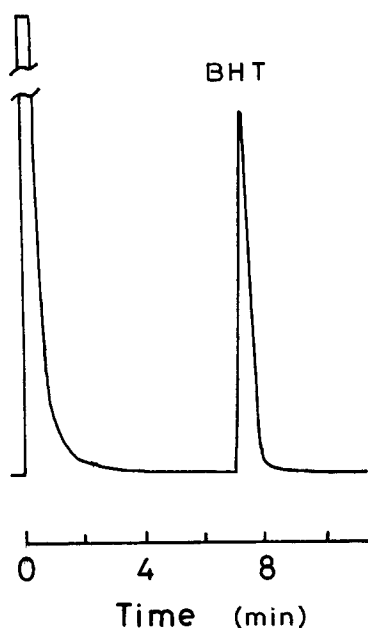


Figure 2 Typical gas-liquid chromatogram of BHT solution. Operating conditions: column, $2\text{ m} \times 3\text{ mm}$ i.d., glass; packing reagent, 10% silicone SE-30 on Gaschrom Q, 60-80 mesh; carrier gas, nitrogen, 50 mL min^{-1} ; chart speed, 5 mm min^{-1} .

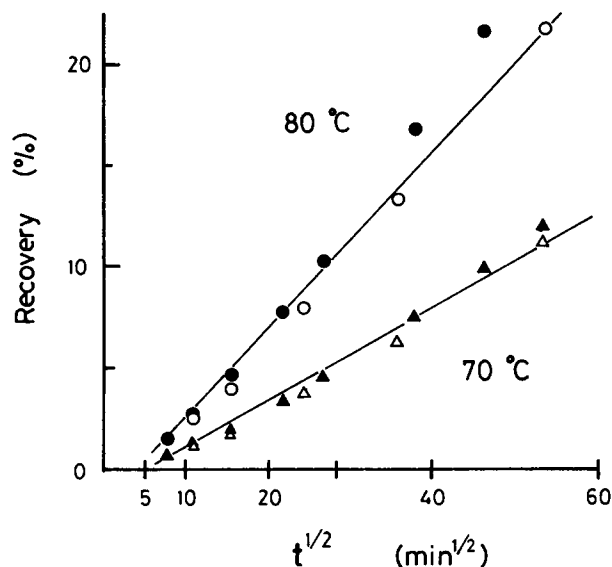


Figure 3 Relations between percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column and heating time ($t^{1/2}$) when PP plates of $110 \times 6 \times 2$ mm in size were heated at 70 and 80°C under air or nitrogen: (—●—), (—▲—) under air; (—○—), (—△—) under nitrogen.

solved in 2 mL of dichloromethane. The BHT solution from the Tenax GC column was prepared as follows: The Tenax GC column in Figure 1, which absorbed BHT, was allowed to flow in 20 mL of a 5% ether/*n*-hexane mixture. The eluate was evaporated using the same procedure as that of a solid material and then was dissolved in 2 mL of *n*-hexane.

The two columns, 3.0 mm i.d. \times 2000 mm in length, glass tubing packed with 10% SE-30 on Gaschrom Q (60-80 mesh), and 5% Silicone OV-17 on Shimalite W (80-100 mesh), respectively, were isothermally operated at 160°C . The injector was maintained at 220°C . Nitrogen as a carrier gas was allowed to flow at 50 mL min^{-1} . Figure 2 shows the typical gas chromatogram for these sample solutions.

RESULTS AND DISCUSSION

BHT Vaporization from PP under Air and Nitrogen

Generally, the PP products are processed and utilized in air. In a previous paper,⁴ the concentration distribution of BHT in PP after heating was also obtained in air.

Figures 3 and 4 show the relations between the square root of the time, $t^{1/2}$, and the percentage re-

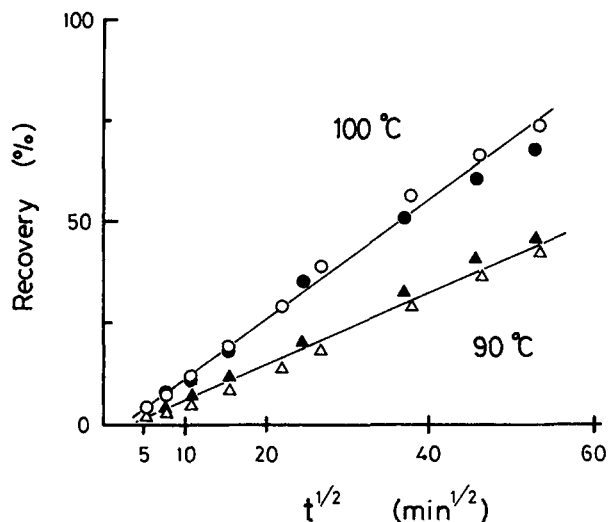


Figure 4 Relations between percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column and heating time ($t^{1/2}$) when PP plates of $110 \times 6 \times 2$ mm in size were heated at 90 and 100°C under air and nitrogen: (—●—), (—▲—) under air; (—○—), (—△—) under nitrogen.

covery of BHT vaporized from the PP plate (A) that was heated under air at 70, 80, and 90, and 100°C, respectively. With this temperature, the percentage recoveries of BHT increased linearly with $t^{1/2}$ and the slope also increased steeply at higher temperature.

On the other hand, BHT reacts with molecular oxygens to yield 1,2-bis-(3,5-di-*tert*-butyl-4-hydroxyphenyl)-ethane via the corresponding benzyl radicals.

This compound is readily oxidized to 3,5,3',5'-tetra-*tert*-butylstilbene-4,4'-quinone,⁶⁻⁸ which is not further oxidized. Therefore, the oxidative reaction of this sterically hindered phenol, which has a very reactive functional group, OH, depends largely on the ambient gas. It is, therefore, interesting to determine the influence of oxygen in air to BHT vaporization whenever the PP samples were heated under air. The quinone was not found through GC experiments for the sample absorbed to the Tenax GC adsorbent at 70–100°C under air.

The time course of BHT vaporization under the inert gas, nitrogen, are shown in the same figures to determine the influence of ambient air on the BHT vaporization behavior. The percentage recoveries of the BHT vaporization under nitrogen increased linearly, like that of the vaporization under air, and also increased steeply faster at higher temperature. Both plots obtained under the different ambient gases are exactly on the same straight line for each

temperature. The percentage recoveries of BHT are independent of heating under air or nitrogen.

It was, thus, apparent that there were no considerable differences in the vaporization behavior of BHT in PP by heating under air or nitrogen.

Influence of Sample Thickness on the Rate of BHT Vaporization

Figure 5 shows the relations between the square root of heating time, $t^{1/2}$, and a percentage recovery of BHT collected in the Tenax GC adsorbent column when two plates (A) were heated at 70–120°C, respectively, under nitrogen.

In the temperature range of 90–120°C, the recoveries of BHT increased linearly with $t^{1/2}$ until the percentage of 70%, and thereafter the plots deviated from the straight line downward. The vaporization rate increased steeply at higher temperatures. The deviations from the straight line are attributed to the considerable decrease of BHT concentration in PP. On heating at 70 and 80°C, on the other hand, still, linearity was established until maximum heating time.

Figure 6 shows the relations between $t^{1/2}$ and a percentage recovery of vaporized BHT on PP films of 0.003 cm in thickness at the temperature range of 55–100°C under nitrogen. These films contained initially 7.1×10^2 mg kg^{-1} of BHT.

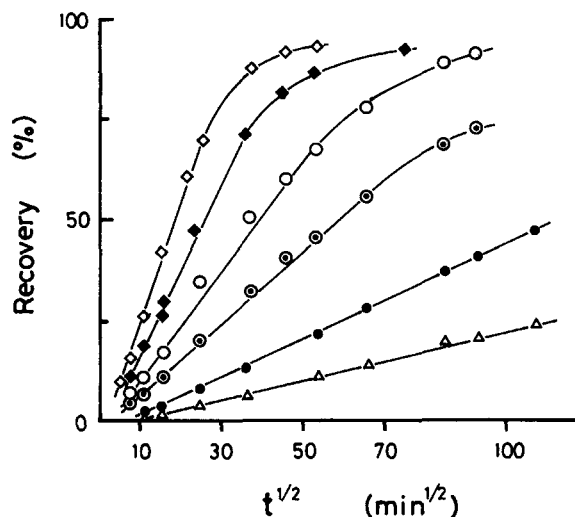


Figure 5 Relations between percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column and heating time ($t^{1/2}$) when PP plates of $110 \times 6 \times 2$ mm in size were heated at 70–120°C under nitrogen: (—◇—) at 120°C; (—◆—) at 110°C; (—○—) at 100°C; (—○—) at 90°C; (—●—) at 80°C; (—△—) at 70°C.

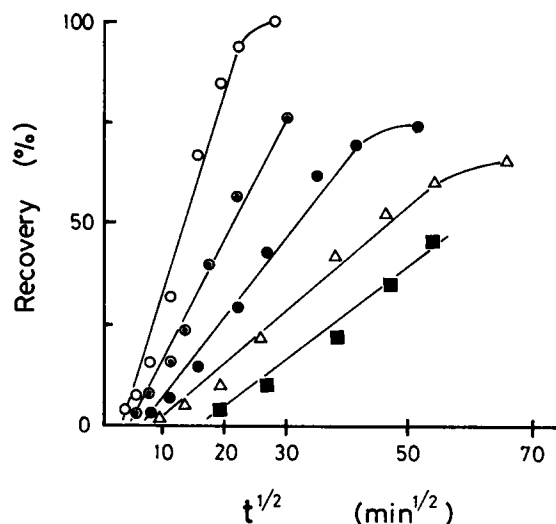


Figure 6 Relations between percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column and heating time ($t^{1/2}$) when PP films of 0.003 cm in thickness were heated at 55–100°C under nitrogen: (—○—) at 100°C; (—○—) at 90°C; (—●—) at 80°C; (—△—) at 70°C; (—■—) at 55°C.

Similar relations as with the PP plates have been found. The percentage recoveries of BHT at 100, 90, 80, and 70°C increased linearly with $t^{1/2}$ until the recovery of about 90, 80, 70, and 60%, respectively, and thereafter the plots deviated from the straight line downward. The vaporization rate also increased at higher temperature. However, two curves for PP samples having different thicknesses, as shown in Figures 5 and 6, had considerable differences in the slope at the same temperature, so that the vaporization rate from thin films is apparently higher than that from the thick plates.

For example, as seen in Figure 5, when the plates were heated at 70, 80, 90, and 100°C for 13 h, the percentage recoveries of BHT vaporized were 4, 9, 23, and 39%, respectively. These values are lower than those obtained for the thin films that showed the recoveries of 25, 45, 71, and 99.9%, respectively, at the same conditions. The difference between these two data sets becomes larger with falling temperature.

If the crystallinity, the orientation of PP molecules, the concentration distribution of BHT, and temperature are the same for both, and if there are no other coexistent substances that may influence the BHT vaporization, the mobility of BHT in PP will be independent of the sample thickness. On the other hand, the transferring time of a BHT molecule in PP from center to the surface will depend on the

distance, i.e., the thickness. Thus, the thicker the film is, the later the vaporization is.

Even if the heating is performed at 55°C, a much lower temperature than the melting point of BHT, 70°C, the BHT vaporization proceeded like that at the temperature above the melting point. This fact suggests that the BHT molecules are able to migrate quantitatively from the PP material to foods and other products even if the temperature is relatively low.

In a previous paper,⁴ the concentration distribution of BHT in PP was found to obey Fick's law on the diffusion process. Consequently, an amount of BHT vaporized from the polymer surface in this study naturally increased linearly with the square root of time, $t^{1/2}$, depending on the diffusion processes in PP. The method for the influence of temperature on vaporization velocity can be derived by plotting the logarithm of the specific rate of vaporization, $\text{Log } \% / t^{1/2}$, against the reciprocal of the absolute temperature $1/T$ by means of eq. (1) of the following form:

$$\text{Log } k = \text{Log } A - E/2.303RT \quad (1)$$

where k is the specific rate of vaporization, $\% / t^{1/2}$; A , a constant; E , the activation energy; R , the gas constant; and T , the absolute temperature.

These Arrhenius plots, as shown in Figure 7, described approximately straight lines at 70–120°C for both data sets, except for the point at 55°C, which did not fall on the same line.

The constant E , which is characteristic of the BHT vaporization from the polymer and determines the influence of temperature on the vaporization rate, is considered to correspond to the activation energy of Fickian diffusion of BHT in PP.⁴ The values of E for the thicker plate and the thinner film could be calculated as 52.9 and 48.6 kJ mol⁻¹, respectively, from the slope. Both values can be interpreted as approximately the same values as those of the other depending on the sample thickness and about half of the value, 105 kJ mol⁻¹, obtained from the rolled film method.⁴ It is thought that a flow of nitrogen gas at 30 mL min⁻¹ lowers the activation energy of the diffusion.

Influence of Coexistent Oils on the Rate of BHT Vaporization

As the PP lunch wares, which have been widely used at the mass lunch facilities, are frequently in contact with moisture and many kinds of food components, such as oils, proteins, and saccharides, the properties

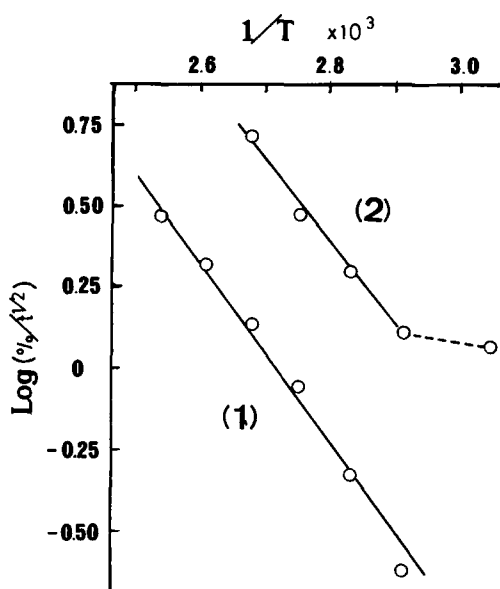


Figure 7 Relations between logarithmus of specific rate of BHT vaporization ($\text{Log } \% / t^{1/2}$) and reciprocal of absolute temperature ($1/T$) for (1) PP plates and (2) films.

of polymer products and the behavior of additives would be affected by these components. Especially, the oils are well known to be migrated into the inside of the PP materials, and the surfaces are frequently still wet and smeary after using for a long period of time. In addition, BHT is now widely employed as one of the most effective antioxidant additives for refined vegetable oils because BHT readily dissolves in oils.

It is, therefore, interesting to study the influence of the coexistent oils on the vaporization rate of BHT in PP. At first, the vaporization rates of BHT in mixtures of various ratios of triolein and BHT were studied. Figure 8 shows the relations between the heating time at 55 and 80°C and a percentage recovery of BHT vaporized from the mixture in which the composition was varied.

On heating at 55°C, the plots for each mixture were apparently different from that of BHT alone, which is free from oils, and the vaporization rate decreases with increasing content of triolein. At 80°C, the mixture containing triolein of the ratio of one-hundredth to BHT showed almost the same curve as that of BHT alone. However, the rate decreased remarkably in proportion to the ratio of triolein to BHT in the range from one-tenth to 100 times.

Ishitani and co-workers⁹ reported that after heating the liquid paraffin containing 50 mg/250 mL of BHT at 140 and 160°C for 1 h, 10 and 17% of BHT in the samples were removed, respectively.

In our experiments, when the mixtures of 1 : 100, 1 : 10, 1 : 1, 10 : 1, and 100 : 1 in ratios (oil:BHT) were heated at 80°C for 1 h under nitrogen gas, 98.5, 99.5, 97.7, 57.0, and 11.4%, respectively, of BHT were transferred to the Tenax GC column. In other words, these values correspond to the total percentage of vaporized BHT. If the ratio of triolein to BHT rose to 5000 times as in Ishitani et al.'s report, the vaporization rate might further decrease. Thus, the amount of coexistent oils were proved to influence considerably the vaporization rate of BHT.

The influence of coexistent oils was studied for the PP plate immersed into olive oil at 20°C for 44 days and heated at 80 and 100°C under nitrogen gas. The samples contained 0.2 wt % of olive oil by the gravimetric method, which is about two times higher than the BHT content. Figure 9 shows the time course of the BHT vaporization for the sample

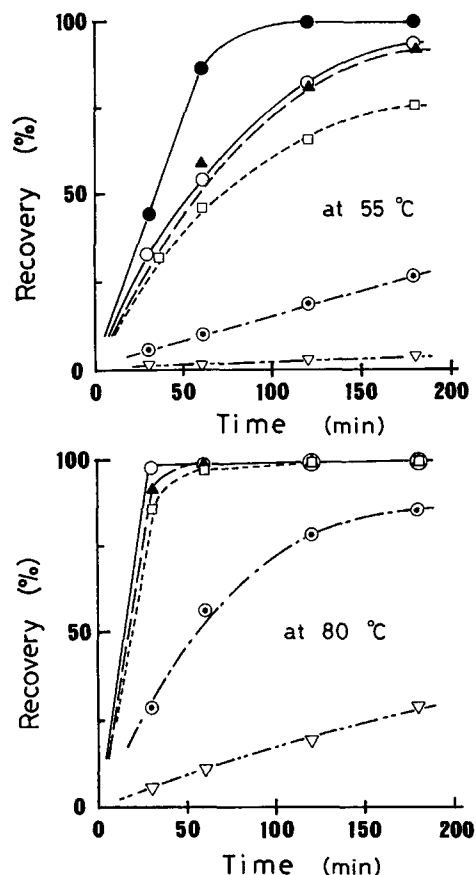


Figure 8 Relations between heating time and percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column when mixtures of various contents of triolein were heated at 55 and 80°C under nitrogen: (—●—) without triolein; (—○—) triolein: BHT = 1 : 100; (—▲—) = 1 : 10; (—□—) = 1 : 1; (—○—) = 10 : 1; (—▽—) = 100 : 1.

containing olive oil and the original one without immersion.

The two curves are approximately similar, showing no influence of the coexistent oil on the BHT vaporization. The concentration of 0.2 wt % is thus too low to influence the BHT vaporization.

In our experiments, however, it is necessary to immerse the PP plates into olive oil at as low a temperature as possible to avoid the migration of BHT into the oil. The immersion was thus preliminary done at 20°C for 44 days. In this condition, there is no evidence for the decrease of the BHT concentration in PP. The higher level of the oil content in the polymer is of great interest in the thermal behavior of the additive.

If the oil migrates into PP according to the diffusion process that follows Fick's law, as for BHT,¹⁰ the oil molecules are considered to transfer across voids in the amorphous regions in PP and there produce a concentration distribution of oil in PP. The distribution would also occur in PP lunch wares in the same way.

At present, there are few studies on the quantitative analysis of the oils migrated into PP, though a qualitative observation is well examined by thin-layer chromatography. It is necessary to study the diffusion behavior of the oil in PP in detail hereafter, involving in the influence of BHT vaporization.

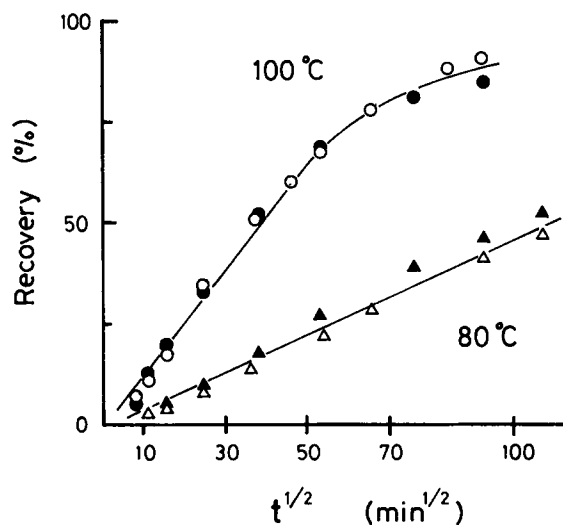


Figure 9 Relations between heating time ($t^{1/2}$) and percentage recoveries of BHT (%) collected on the Tenax GC adsorbent column when PP plates of $110 \times 6 \times 2$ mm in size, preliminary immersed into olive oil at 20°C for 44 days or not, were heated at 80 and 100°C under nitrogen: (—○—), (—△—) without immersion; (—●—) (—▲—) preliminary immersed into olive oil at 20°C for 44 days.

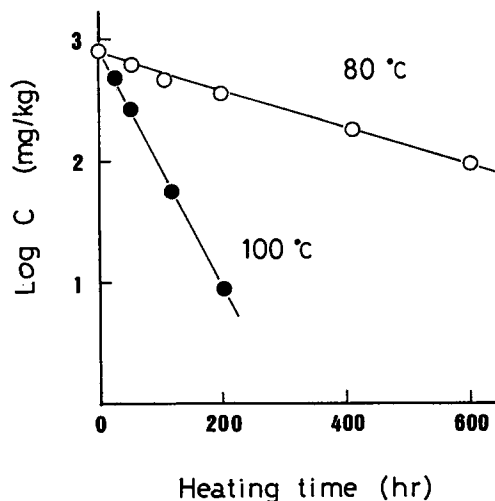


Figure 10 Relations between heating time and mean BHT concentration in PP plate, $\text{Log } C$, which were heated at 80 and 100°C, respectively, under air.

The authors¹¹ and other researchers^{12,13} noted that BHT was detected frequently in PP widely used as a material in the lunch wares at mass lunch facilities in Japan, and the concentration tended to decrease with time when the lunch wares were used for a long period of time. In the mass lunch facilities, the lunch wares are heated at around 80°C for a few hours to dry and sterilize. It was, therefore, considered that the residual BHT concentration held in PP largely depended on the heating conditions.

Decreasing Rate of BHT Concentration in PP by Heating

Figure 10 shows the relations between the logarithm of BHT concentration, $\text{Log } C$, in PP and the heating time when the PP plate (B) containing initially $9.7 \times 10^2 \text{ mg kg}^{-1}$ of BHT were heated in an oven at 80 and 100°C, respectively, in air.

The mean BHT concentration in PP decreased following eq. (2) and the decreasing rate depended on the temperature. The amounts of BHT in PP after being treated at 100°C was lower than that treated at 80°C in all cases. Equation (2) is differentiated to give eq. (3), which obeys first-order kinetics:

$$\text{Log } C = -k_1 t + \text{const} \quad (2)$$

$$-dc/dt = k_1 c \quad (3)$$

Here, c is the mean BHT concentration in PP; t , the heating time; and k_1 , the rate constant.

Similar relations have been reported by Matsumoto.¹⁴ He showed that the 4,4-thio-bis(3-methyl-6-*tert*-butyl phenol) concentration in PE films of 155 μm in thickness, aged in air or nitrogen at constant temperature between 70 and 150°C, decreased in accordance with eq. (2). A 40% of total BHT was yet held in the PP plate after heating at 80°C for 200 h, whereas it nearly disappeared after heating at 100°C for the same period of time.

In an early stage of a series of our works, it was supposed that the heat treatment of PP under air at the temperature below 120°C would result in the decrease of the BHT concentration due to partly transforming to other compounds in the thermal oxidation process of PP for a long period of time. No apparent proof for this was shown in Figure 10. However, as seen in Figures 3 and 4, the vaporization of BHT in PP by heating at 70–120°C was obviously found to result in the decrease of BHT concentration.

Now, when the PP plates were heated at 100°C for 30, 40, 80, and 120 h, the percentage recoveries of the BHT vaporization were determined as 56.0, 62.9, 79.9, and 88.0%, respectively, from the curve shown in Figure 5.

On the other hand, the curve at 100°C in Figure 10 gives residual BHT content of 43.3, 34.0, 14.0, and 5.1% for the corresponding times. Each summation of both at the same heating hours is 99.3, 96.9, 94.3, and 93.1%, respectively. The majority of the BHT molecules in PP, thus, decreased without decomposition by vaporizing from the polymer surface to the outside at the temperatures below 120°C. Furthermore, the ambient air does not influence the BHT vaporization since the time course was almost similar to that in inert gas until 50 h. However, when the heating extended to above 50 h, the sums tended to decrease gradually with time. The decrease is considered to be due to the lowering of the ability of the BHT adsorption in the Tenax GC adsorbent.

Since the polymer segments vibrate thermally with a large amplitude depending on heating temperature, voids are produced in amorphous regions in which BHT molecules are able to pass in a moment. The numbers of the voids will increase with rise in temperature. Moreover, the mobility of BHT in PP may also be affected by the ratio of amorphous regions to crystal ones because priority is given to the amorphous regions for the moving of BHT molecules. Therefore, BHT adsorbed to the Tenax GC adsorbent column is due to BHT molecule that passed through the voids in the amorphous regions produced during the thermal vibration of polymer segment in the way just mentioned.

CONCLUSION

The vaporization–collection system equipped with the Tenax GC adsorbent column was applied to verify further that the concentration distribution of BHT in PP by heating under air was due to the vaporization of BHT from the polymer surface. The effect of air on the BHT decomposition was ascertained through comparison with the vaporization behavior under nitrogen. The results are as follows:

1. BHT vaporized from the PP plates under the air stream at 70–120°C was absorbed to the Tenax GC adsorbent column, and the percentage recoveries of BHT increased linearly with the square root of time, $t^{1/2}$, and the slope also increased steeply at higher temperatures. There was no considerable difference between the vaporization behavior of BHT by heating under the stream of air and that of nitrogen.
2. The percentage recoveries of BHT from the PP plates at 90–120°C under the nitrogen stream increased linearly with $t^{1/2}$ until about 70% of the recovery. On heating at the temperatures below 80°C, the linear relations still held.
3. Both the sample thickness and the temperature greatly influenced the vaporization rate of BHT in PP. The Arrhenius plots that were derived by plotting the logarithm of the specific rate of vaporization, $\text{Log } \% / t^{1/2}$, against the reciprocal of the absolute temperature showed an approximately straight line at 70–120°C for the thicker plate and thinner film except for the plot at 55°C. The activation energies were estimated as 52.9 and 48.6 kJ mol^{-1} and were about half of the value obtained from the rolled film method.
4. Triolein coexisted with interrupted BHT vaporization, the rate of which decreased remarkably with increasing the triolein content from one-tenth to 100 times. However, the PP plates that contained 0.2 wt % of olive oil by immersing in olive oil at 20°C for 44 days showed the same vaporization behavior of BHT as did the original sample free from oils in PP. The oil concentration of 0.2 wt % is, thus, too low to influence the vaporization.
5. The mean BHT concentration in PP decreased with the heating time at 80 and 100°C. The decreasing rate of BHT that depended greatly on the temperature obeyed first-order kinetics.

It is necessary to study the diffusion behavior of the oils in PP in detail hereafter, involving in the influence of the BHT vaporization.

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